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Eduardo Carrera-Guilarte

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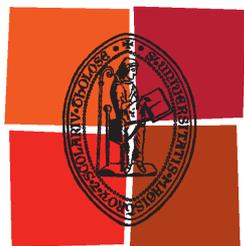
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de Toulouse

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This thesis is dedicated to the memory of my grandmother. From where she is, she must be proud of her grandson.

ABSTRACT

This work presents a methodological design framework for Energy Supply Chains (HMSC), focusing on Power-to-Gas (PtG) concepts, and their interactions with other technologies, and energy vectors (i.e., Stream Methane Reforming – SMR, and natural gas). The overall objective of this work is to perform mono-objective and multi-objective optimizations of the Hydrogen and Methane Supply Chains (HMSC) to provide effective support for the study of deployment scenarios. The methodology is applied to the case study of Occitania (France). The main technological components of an HMSC are presented to explore the diversity of options that can be used in the design. Specific focus is paid to the design and deployment system to capture the required level of technological and spatial detail needed, as well as the methods and tools used in order to obtain the solution of the mono and multi-objective optimization problems (i.e., problem formulation, optimization framework and tools for multi-criteria decision making (MCDM) steps). The methodological framework developed is based on a Mixed Integer Linear Programming (MILP) approach with augmented ε -constraint implemented in the GAMS environment according to a multi-period approach (2035-2050). Several available energy sources (wind, PV, hydro, national network, and natural gas) for hydrogen production through electrolysis and SMR are included. Carbon dioxide sources stem mainly from methanization and gasification processes, which are used to produce methane through methanation. Methane demand can be also met by importing natural gas. Hydrogen demand is determined based on the expected use of fuel cell electric vehicles (FCEVs) whereas the demand for renewable methane includes the residential, industry, transport, and service sectors. The optimization variables involve the number and size of production and storage units, the number of tanker-trucks for hydrogen transport as well as the flows of imported/exported hydrogen from one grid to another one. Four objective functions are involved: Total Annual Cost (TAC), Greenhouse Gas (GHG) emissions, Total Renewable Methane Production (TMP), and Total Exergy Lost and Destroyed (TELD). Some of the key issues to be investigated are the simultaneous satisfaction of hydrogen and renewable methane from PtG systems, the effect of environmental regulation (i.e., carbon price), and the interaction with fossil energy sources (Steam Methane Reforming – SMR, and natural gas). Some indicators as the Levelized Cost of Energy (LCOE) and the greenhouse gas emissions for each energy carrier are also computed. Sensitivity assessments are carried out to study the influence of key parameters on the LCOE (i.e., discount rate, cost of electricity, and carbon price). The results show that renewable hydrogen from PtG can be competitive with SMR through the implementation of carbon prices below 0.27 €/kgCO₂. In the case of synthetic methane, the available resources can meet the demand through PtG, and even if synthetic methane for grid injection is thus far from competitiveness with natural gas, PtG technologies have the most potential to decarbonize the fossil economy and

achieve circular economy via CO₂ valorization. Furthermore, coupling with other energy systems and processes is essential to increase the exergetic performance of the HMSC.

KEYWORDS: Power-to-Gas, hydrogen, methanation, MILP, supply chain optimization, exergy analysis

Ce travail présente un cadre de conception méthodologique pour des chaînes logistiques « énergie », en se concentrant sur les concepts Power-to-Gas (PtG), et leurs interactions avec d'autres technologies, et vecteurs énergétiques (c'est-à-dire reformage à la vapeur et gaz naturel). L'objectif général de ce travail est de réaliser des optimisations mono-objectifs et multi-objectifs de la chaîne logistique de l'hydrogène et du méthane (HMSC) afin d'apporter une aide efficace à l'étude des scénarios de déploiement. La méthodologie est appliquée à l'étude de cas de l'Occitanie (France). Les principales composantes technologiques d'une HMSC sont présentées afin d'explorer la diversité des options qui peuvent être utilisées en conception. Une attention particulière est accordée au système de conception et de déploiement afin de prendre en compte le niveau de détail technologique et spatial approprié, ainsi que les méthodes et outils utilisés pour obtenir la (les) solution(s) des problèmes d'optimisation mono et multi-objectifs (c'est-à-dire la formulation du problème, le cadre d'optimisation et les outils pour les étapes de prise de décision multicritères (MCDM)). Le cadre méthodologique développé est basé sur une approche de programmation linéaire en nombres entiers mixtes (MILP) avec la méthode *augmented ϵ -constraint*, mise en œuvre dans l'environnement GAMS selon une approche multi-période (2035-2050). Plusieurs sources d'énergie disponibles (éolienne, photovoltaïque, hydraulique, réseau national et gaz naturel) pour la production d'hydrogène par électrolyse et par reformage à la vapeur sont incluses. Les sources de dioxyde de carbone proviennent principalement des procédés de méthanisation et de gazéification, qui sont utilisés pour produire du méthane par méthanation. La demande en méthane peut également être satisfaite par l'importation de gaz naturel. La demande d'hydrogène est déterminée en fonction de la prévision de la demande en véhicules électriques à pile à combustible (FCEV), tandis que la demande de méthane renouvelable comprend les secteurs résidentiel, industriel, des transports et des services. Les variables d'optimisation concernent le nombre et la taille des unités de production et de stockage, le nombre de camions-citernes pour le transport de l'hydrogène ainsi que les flux d'hydrogène importés/exportés d'un réseau à l'autre. Quatre fonctions objectives sont concernées : coût annuel total (TAC), émissions de gaz à effet de serre (GES), production totale de méthane renouvelable (PTM) et exergie totale perdue et détruite (TELD). Parmi les aspects clés à étudier, figurent la satisfaction simultanée de l'hydrogène et du méthane renouvelable provenant des systèmes de PtG, l'effet de la réglementation environnementale (c'est-à-dire le prix du carbone) et l'interaction avec les procédés sources d'énergie fossiles (reformage du méthane à la vapeur et gaz naturel). Certains indicateurs tels que le *Levelized Cost of Energy* (LCOE) et les émissions de gaz à effet de serre pour chaque vecteur d'énergie sont également calculés. Des évaluations de sensibilité sont effectuées pour étudier l'influence des paramètres clés sur le LCOE (taux d'actualisation, le coût de l'électricité et le prix du carbone). Les résultats montrent que l'hydrogène renouvelable provenant du PtG peut être compétitif par rapport au

reformage à la vapeur grâce à la mise en place d'un prix du carbone inférieur à 0.27 €/kgCO₂. Dans le cas du méthane de synthèse, les sources disponibles peuvent répondre à la demande à travers le PtG et même si le méthane de synthèse destiné à l'injection dans le réseau est loin d'être compétitif avec le gaz naturel, les technologies PtG ont un plus grand potentiel pour décarboner le système énergétique et atteindre une économie circulaire via la valorisation du CO₂. En outre, le couplage avec d'autres systèmes et procédés est essentiel pour augmenter les performances exergétiques du HMSC.

MOT CLES: Power-to-Gas, hydrogène, méthanation, MILP, optimisation de chaînes logistiques, analyse d'exergie.

CONTENT

CONTENT	10
1 INTRODUCTION AND PROBLEM DEFINITION.....	15
1.1 Introduction	15
1.2 Research problem and research questions.....	19
INTRODUCTION ET DEFINITION DU PROBLEME.....	25
Introduction	25
Problématique de recherche et questions de recherche.....	29
2 DEPLOYMENT AND DESIGN OF HYDROGEN AND METHANE SUPPLY CHAINS BASED ON PTG: HOW TO CAPTURE THE REQUIRED LEVEL OF TECHNOLOGICAL AND SPATIAL DETAIL IN MODELING	37
ABSTRACT	37
RÉSUMÉ.....	37
2.1 Introduction	38
2.2 Main technological bricks involved in hydrogen and methane supply chains.....	41
2.2.1 Hydrogen supply	41
2.2.2 Methanation for Power-to-Gas.....	48
2.2.3 Storage and transport infrastructure	51
2.2.4 End-uses	54
2.2.5 Feedstock	55
2.2.6 Conclusions	57
2.3 A first taxonomy of optimization-based HMSC models	60
2.3.1 Energy system optimization models (ESOM).....	61
2.3.2 Geographically explicit optimization models (GEOM)	61
2.3.3 Location or partial optimization models (LoPOM).....	62
2.4 Conclusion.....	63
3 METHODS AND TOOLS FOR MULTI-PERIOD, MULTI-CRITERIA AND MULTI- ECHELON HMSC DESIGN	65
ABSTRACT	65
RÉSUMÉ.....	65

3.1	Introduction	66
3.2	Optimization framework for GEOM.....	67
	The development of the optimization framework for HMSC involves considering the following key items... 67	
3.2.1	Decisions and planning time horizon	67
3.2.2	Strategic decisions.....	67
3.2.3	Multi-period approach.....	67
3.2.4	Performance measures.....	68
3.2.5	Multi-objective approach	69
3.2.6	Integration in the GEOM model.....	69
3.3	Solution methods	70
3.3.1	Mono-objective problems	70
3.3.2	Multi-objective problems	72
3.4	Multi-criteria decision making (MCDM): decision aid methods	75
3.4.1	ELECTRE	75
3.4.2	TOPSIS	76
3.4.3	M-TOPSIS	77
3.5	Conclusion.....	78
4	CASE STUDY FOR THE HMSC OPTIMIZATION	79
	ABSTRACT	79
	RÉSUMÉ.....	79
4.1	Introduction	80
4.2	Energy transition in the Occitania region	81
4.3	Collection of techno-economic data	81
4.4	Geographical division.....	85
4.5	Demand estimate	85
4.6	Feedstock estimate.....	86
4.7	Conclusion.....	88
5	AN OPTIMIZATION FRAMEWORK FOR POWER-TO-METHANE SYSTEM DESIGN AND DEPLOYMENT	91
	ABSTRACT	91
	RÉSUMÉ.....	91
5.1	Introduction	92
5.2	Problem definition.....	95
5.3	Mathematical model.....	98
5.3.1	Demand constraints	101
5.3.2	Production facilities constraints	102
5.3.3	Transportation constraints	102
5.3.4	Storage facilities constraints.....	103
5.3.5	Determination of the number of units	104
5.3.6	Energy constraint	105
5.3.7	CO ₂ source constraint.....	105
5.3.8	Cost objective.....	105

5.4	Results and discussion	109
5.4.1	Analysis of scenarios	109
5.4.2	Sensitivity analysis	115
5.5	Conclusions	116
6	BI-OBJECTIVE OPTIMAL DESIGN OF PTH AND PTM SUPPLY CHAINS	117
	ABSTRACT	117
	RÉSUMÉ	118
6.1	Introduction	119
6.2	Problem definition	121
6.3	Mathematical model	123
6.3.1	Hydrogen demand	125
6.3.2	Refueling station constraints	125
6.3.3	Number of refueling stations	125
6.3.4	Refueling station capital cost	125
6.3.5	Environmental objective function and indicators	125
6.3.6	Physical allocation for cost, and environmental indicators	126
6.4	Solution strategy	128
6.5	Results and discussions	130
6.5.1	Mono-objective optimizations	130
6.5.2	Multi-objective optimization and comparison	131
6.5.3	Sensitivity analysis	139
6.6	Conclusions	141
7	BEYOND RENEWABLE POWER-TO-GAS: COMPETITION WITH FOSSIL SOURCES	142
	ABSTRACT	142
	RÉSUMÉ	143
7.1	Introduction and context	144
7.2	Methodology and formulation of the HSC problem	145
7.2.1	Problem definition	145
7.2.2	Methane demand	147
7.2.3	Distinction between renewable and fossil energy sources	148
7.2.4	Cost objective function	148
7.2.5	Environmental objective function	148
7.2.6	Methane production objective function	148
7.3	Solution strategy	149
7.4	Results and discussion	151
7.4.1	Mono-objective optimizations	151
7.4.2	Multi-objective optimization and comparison	153
7.4.3	Sensitivity analysis	157
7.5	Conclusion	159
8	COST, ENVIRONMENTAL, AND EXERGY OPTIMIZATION OF HYDROGEN AND METHANE SUPPLY CHAINS	162

ABSTRACT	162
RÉSUMÉ.....	163
8.1 Introduction	163
8.2 Exergy definition	164
8.3 Methodology and formulation of the HSC problem.....	166
8.3.1 Exergy objective function	166
8.4 Results and discussion.....	167
8.5 Conclusions	169
9 GENERAL CONCLUSIONS AND PERSPECTIVES.....	170
9.1 Conclusions	170
9.2 Lessons learned.....	172
9.3 Perspectives.....	173
CONCLUSIONS ET PERSPECTIVES GÉNÉRALES.....	176
Conclusions	176
Leçons tirées.....	178
Perspectives.....	179
DATABASE.....	183
REFERENCES.....	188
LISTE OF FIGURES.....	207
LISTE OF TABLES	211

1 INTRODUCTION AND PROBLEM DEFINITION

1.1 Introduction

The Paris Agreement adopted at the Paris climate conference (COP21) in December 2015 is a milestone in international climate politics. It stipulates that global temperature rise should be held below 2°C, if possible at 1.5°C above pre-industrial levels, to mitigate the unmanageable and irreversible consequences of climate change (United Nations, 2015). In this regard, the EU has set as objective to reduce greenhouse gas emissions by at least 40% by 2030 compared to 1990. To reach such objectives, the energy network requires a radical transformation, since 87% of the equivalent CO₂ emissions come from sectors associated with energy transformation: 53% from fuel combustion and fugitive emissions from fuels (without transport), 25% from transport (including international aviation) and 9% from industrial processes and product use (European Commission, 2015). In this regard, the use of renewable energy sources (RES) is one pillar of the transition towards a better and more sustainable energy future. Hence, RES. According to the International Renewable Energy Agency (IRENA), renewables would then make up two-thirds of energy consumption and 85% of power generation in Europe by 2050

(IRENA, 2018a). One of the most important challenges in the integration of renewable energy sources into the power grid lies in the “intermittent” nature of sources like wind and solar. To drive their penetration into the energy mix, the Power-to-Gas (PtG) concept is a promising solution, in which electrical power and CO₂ are used to produce gas (i.e., hydrogen or methane), offering several services:

1. dealing with the intermittency of renewable energy sources by recovering large amounts of overproduced electricity (Buttler and Spliethoff, 2018; Götz et al., 2016);
2. flexibility and greater power grid integration by providing a physical, bidirectional interaction between the gas network and the electricity network (Buttler and Spliethoff, 2018; Khalilpour, 2018);
3. low-carbon fuel supply (Buttler and Spliethoff, 2018; E&E consultant et al., 2014; Koj et al., 2019) with CO₂ capture and recovery (Götz et al., 2016).

In this regard, PtG is the only technology currently available that offers the possibility of storing energy seasonally, in large quantities, and also the potential of decarbonizing other sectors such as heat production, transport, and some industrial applications (ADEME, 2018; E&E consultant et al., 2014).

Before going further in this work, the “Power-to-Gas” concept needs to be precisely defined in order to determine the scope of investigation of this Ph.D. thesis. In fact, in the literature, Power-to-Gas can refer to Power-to-Hydrogen for a range of different applications, sometimes even including fuel for mobility that has been the core of previous works in our research team in the “Laboratoire de Génie Chimique” (De-León Almaraz, 2014). In other definitions, PtG corresponds to the process that links the power grid with the gas grid by converting surplus power into a grid compatible gas via a two-step process: H₂ production by water electrolysis and H₂ conversion with an external CO or CO₂ source to CH₄ via methanation (Götz et al., 2016). Thus, hydrogen obtained from PtG can mainly be:

1. stored and then transformed back into electricity (Dagdougui, 2012)
2. injected into the natural gas grid (according to the limitations of the local natural gas network, i.e. 6% in France) (E&E consultant et al., 2014).
3. used to meet some of the demand from the transport sector or industry (Buttler and Spliethoff, 2018).
4. used in a second step to produce methane through methanation (Sabatier reaction) (Buttler and Spliethoff, 2018; Götz et al., 2016).

Methane (i.e, substitute natural gas or synthetic methane) offers certain advantages since it can be stored and injected without technical restrictions into the well-developed natural gas infrastructure (Khalilpour, 2018) or gas storages, used as a motor fuel, it can also easily be used in industrial facilities. Despite

these benefits, PtG presents certain challenges that need to be addressed before it can play an important role in the energy mix. Among them we can mention that:

- energy losses could be significant along the supply chain (Thema et al., 2019).
- in the case of hydrogen, the blending limit is currently up to 10% (volume basis) with most of the countries below 2% (Quarton and Samsatli, 2018).
- methane could be injected directly into the natural gas grid, but the methanation stage implies additional costs (Blanco and Faaij, 2018; E&E consultant et al., 2014).

To overcome these ambiguities and lack of semantic precision, this dissertation is instead promoting the terms “Power-to-Hydrogen” and “Power-to-Methane”. The term PtG will include both concepts. We also mention their corresponding supply chains (SC) with the explicit terms HSC and MSC terms, respectively for Hydrogen and Methane. The HMSC term (Hydrogen and Methane Supply Chain) will refer to the dual production of both vectors. This keeps “hydrogen” at the center and thus emphasizes its flexibility as an energy carrier. Besides, the conversion of electricity into hydrogen and its reconversion into electricity is called "Power-to-Power".

It must be emphasized at this level that the energy supply chain is more generic in its definition than the PtG concepts in the sense that sources other than power can be used for the production of hydrogen. Today, hydrogen is mainly produced via steam methane reforming (SMR) which is highly carbonized, since natural gas is used as feedstock (i.e., gas-to-gas). Its production with minimal greenhouse gas (GHG) emissions, for example through electrolysis powered by renewable electricity or from bioenergy or fossil fuels with carbon capture and storage (CCS) (Hydrogen Council, 2020) represents a very interesting alternative. The set of options is summarized in Figure 1.1.

The potential services offer by PtG systems, in general, are in line with the EU Strategy for Energy System Integration (European Commission, 2020a). The cumulative investments in renewable hydrogen in Europe could be up to €180-470 billion by 2050. It would help position the EU as a global leader in clean hydrogen technologies manufacturing, since it is expected that clean hydrogen could meet 24% of world energy demand by 2050, with annual sales in the range of 630 billion € (European Commission, 2020b). In this regard, several energy transition strategies of European countries have already integrated PtG systems as part of the set of technologies to achieve their goals (ADEME, 2018; E&E consultant et al., 2014; Gigler and Weeda, 2018; Thomas et al., 2016), in particular in Germany where PtG technology is seen as a cornerstone of its future energy system (Schiebahn et al., 2015; Varone and Ferrari, 2015). These studies are consistent with the major role that PtG systems are expected to play from 2030 with a projection of 80 to 100% renewable electricity production in 2050 (Belderbos et al., 2015). They highlight that the profitability of PtG depends on local circumstances and market conditions (Holstein and van den Noort, 2018).

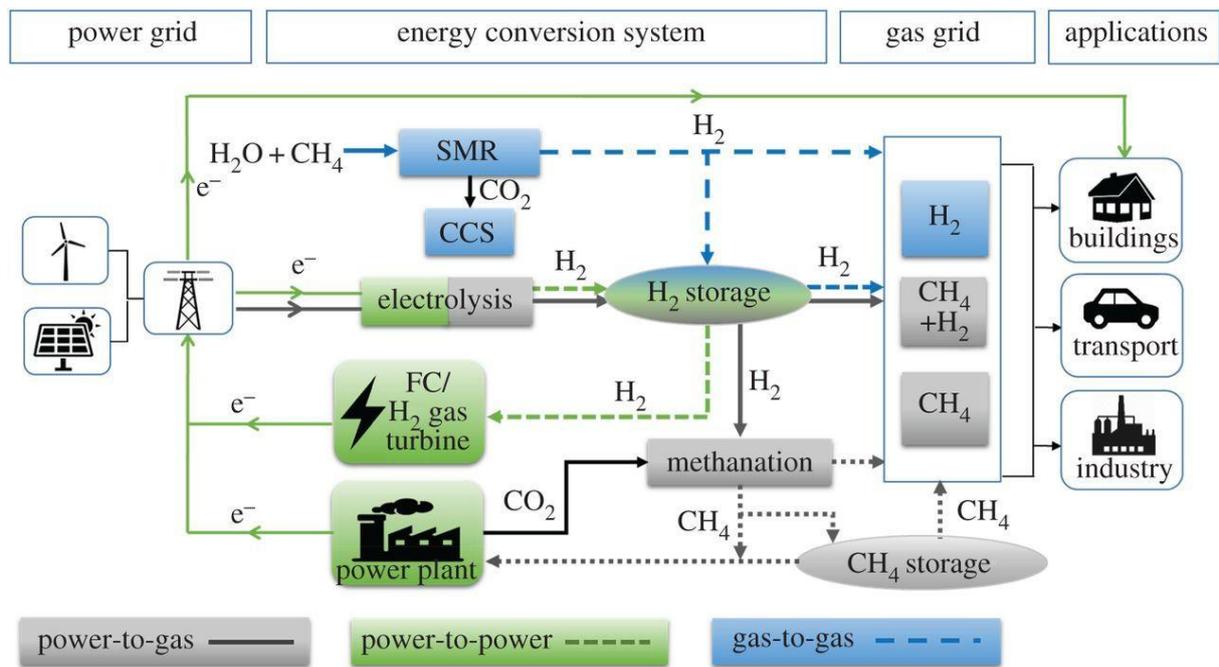


Figure 1.1. Main energy conversion pathways (power-to-gas, power-to-power, and gas-to-gas) in a renewable energy integrated energy system (Brandon and Kurban, 2017)

In the case of France, renewable hydrogen as defined in the French law of 18 August 2015 and in the French Energy Research Strategy (SNRE) (Ministère de l'Économie, des Finances et de la Relance, 2020) and syn-methane will take an important place in its future energy mix (ADEME, 2018; E&E consultant et al., 2014; Fuel Cells and Hydrogen Joint Undertaking (FCH JU), 2020). The French natural gas network is the longest in Europe and has significant gas interconnections because of its borders with five European countries. Besides, its storage capacity is the third in Europe with 132 TWh (H2V INDUSTRY, n.d.; Ministère de la Transition Écologique et Solidaire, 2018). Eight pilot projects dedicated to PtG systems have been implemented in France from 2000 to 2015 (Thema et al., 2019), and a new PtG pilot will be installed in the north of France, with a capacity of 610 MW_{el} (H2V INDUSTRY, n.d.). The French Agency of Ecological Transition (ADEME) ("Agence de la transition écologique," n.d.) has developed a series of studies on PtG systems in the French context, taking into account the availability of resources, possible applications, costs, and limitations (ADEME, 2018; E&E consultant et al., 2014). In addition, hydrogen is seen as an essential part of energy transition plans in island territories and for the deployment of competitive clusters as an effective tool for leveraging the growth of renewables. A flagship example is the MYTRA project in Corsica, where emerging hydrogen technologies are tested, involving electrolyzers, fuel cells, hydrogen storage, and strategies for coupling with RE sources (Cristofari et al., 2014).

At the regional level, the Occitanie Region has announced the creation of a techno-campus dedicated to hydrogen technologies in close connection with the Hyport project by 2024 at Francazal in Toulouse. It will host the largest European center for research, testing, and technological innovation dedicated to

green hydrogen in partnership with the CNRS and its Research Laboratories (Plasma and Energy Conversion Laboratory (Laplace), Cirimat, the Chemical Engineering Laboratory (LGC), and the Toulouse Institute of Fluid Mechanics (IMFT), the University of Toulouse, ONERA, research laboratories but also major groups such as Safran, Airbus, Liebherr, and Vitesco Technologies.

1.2 Research problem and research questions

Over the last years, scientific interest in hydrogen and methane supply chains (HMSC) has progressively increased, as can be seen in Figure 1.2. Thirty papers were identified from 2005 to 2020, based on an initial search in Web of Science (WoS) using the keywords “renewable hydrogen and methane supply chains”. Given the set of technologies, energy sources, and services involved in the HMSC, the elements linked to PtG systems have been extensively addressed in the literature (Buttler and Spliethoff, 2018; Götz et al., 2016), especially the hydrogen supply chain (HSC) (Dagdougui, 2012; Li et al., 2019). In this regard, the number of publications with keywords as “hydrogen supply chain” and “power-to-gas” is 720 and 1198, respectively.

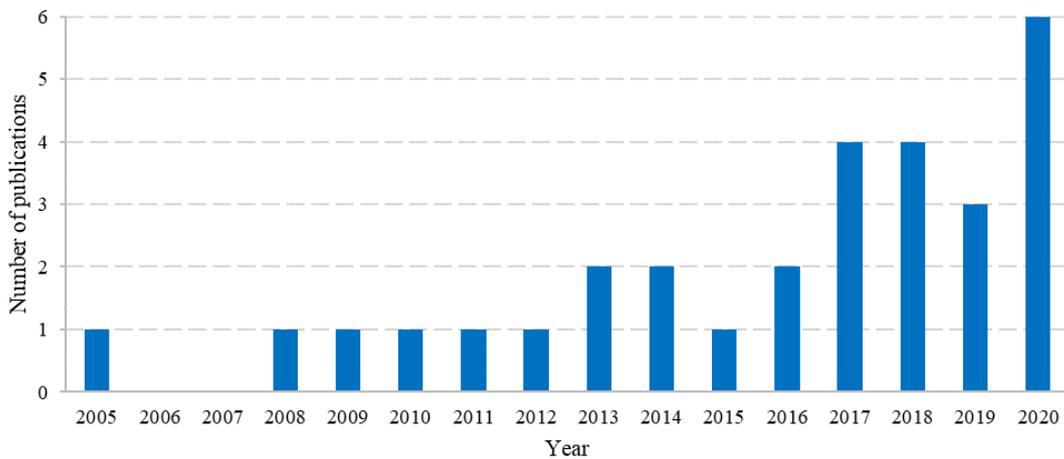


Figure 1.2. Number of papers based on an initial search in Web of Science™ (WoS) using the keywords “renewable hydrogen and methane supply chains” (December, the 5th 2020)

Energy system models form the basis of most quantitative energy scenarios. A vast number of energy system modeling tools exist and can be categorized in different ways, including simulation vs. optimization, top-down vs. bottom-up, etc. A review of computing tools for energy systems (Connolly et al., 2010) identified 68 different energy system modeling tools. Following the same taxonomy, a detailed and recent review of 24 energy systems (Lopion et al., 2018) found a clear trend towards techno-economic bottom-up optimization models to answer current research questions. The most difficult task for modern-day energy systems models is to capture the full degree of variability and complexity that exists in energy systems in an integrated manner. To ensure that energy system models not only provide

an accurate representation of energy systems but also do not miss the potential of new technologies such as hydrogen-based technologies, they must dynamically capture the required level of temporal, spatial, technological, (Poncelet et al., 2016) and inter-sectoral detail (Lucchese et al., 2018). *This Ph.D. work is framed within this perspective for PtG systems.*

A key point in the development of the PtG supply chain is the demonstration of the feasibility of its infrastructure while many technical, economic, and social obstacles must be overcome. Some strategic roadmaps were currently published about the energy potentials of hydrogen that is the core of PtG systems at European, national and regional levels. Their main objective is to evaluate some industrial, technological, environmental, and social issues and to identify the main obstacles associated with the hydrogen economy.

The literature survey has shown that numerous researches have been carried out considering the various usages, main technological bricks, the energy and CO₂ sources involved in PtG (Götz et al., 2016, 2014; Lewandowska-Bernat and Desideri, 2018; Li et al., 2019; Rönsch et al., 2016; Wulf et al., 2018). Regarding the logistic aspects, the hydrogen supply chain has been addressed in several studies as highlighted for instance in two review papers at 8-year intervals (Dagdougui, 2012; Li et al., 2019). However, only a few studies have addressed the logistical aspects involved PtG supply chains, in particular the dual production of hydrogen and methane with their associated issues, i.e. determining the location, size, and the number of production, storage, and transport units, as well as of hydrogen refueling stations.

Recent dedicated scientific publications agree on the need to develop systemic studies to demonstrate the feasibility of the sector, to validate the technical and economic interest in the production and recovery of hydrogen produced from renewable sources. Such works involve the development of models based on economic scenarios for PtG deployment.

To bridge this gap, the scientific objective of this work is to propose a comprehensive methodological framework for the multi-objective optimal design and deployment of Hydrogen and Methane Supply Chains (HMSC), focusing on Power-to-Gas Systems and their spatio-temporal integration considering the regional potentials of Renewable Energy Source (RES). This framework will lead to a decision support system for scenario analysis.

According to (Fuel Cells and Hydrogen Joint Undertaking, 2019), hydrogen supplies could come from a mix of ultra- low-carbon sources. While the exact split of production methods could differ among applications, and depend on technology and cost developments, both electrolysis and steam methane reforming/ auto-thermal reforming with carbon capture and storage (SMR/ATR with CCS) will most likely play key roles. Electrolysis could provide the sector coupling mechanism required for the

integration of renewables and a major technological brick of PtG. Currently, electrolyzers are available at a small scale (< 1 MW) production, with demonstration projects for larger scales (up to 10 MW) are underway. Steam Methane Reforming (SMR) is the most common large-scale hydrogen production technique in use today and could be decarbonized with Carbon Capture and Sequestration (CCS). Scenarios relying on only one of these two production pathways seem unrealistic and would fall short of the required deployment. The coupling of both technologies needs to be explored to see if SMR can be viewed as a necessary transition technology.

The main research questions tackled in this work can be stated as follows.

Research Question 1:

How could a hydrogen and methane supply chain mainly based on Power-to-Gas systems be designed and spatially integrated into a territory? How could it be deployed to capture the required level of temporal, spatial, technological detail?

The HMSC based on Power-to-Gas systems has been scarcely addressed. Several aspects such as the change of configuration over time and variation according to different criteria have not been tackled. The main scientific objective of this Ph.D. work is to develop a methodological framework for the optimization of HMSC produced from Power-to-Gas systems, i.e. determining the location, size, and the number of production, storage, and transport units, as well as hydrogen refueling stations.

Research Question 2:

What are the key performance indicators to be used for HMSC design? How would the configuration of a hydrogen and methane supply chain change according to different decision criteria? How considering the multicriteria aspects?

The process of energy and fuel production is closely related to sustainability. Sustainable supply chain design has emerged as a growing subject that now receives increasing interest from practitioners and academicians. The use of appropriate metrics for determining tradeoffs is a critical part of decision making. Many potentially competing environmental indicators could be used to assess the environmental impact of an energy system, following the guidelines of Life Cycle Assessment framework (Hauschild et al., 2018). All of these are important, however, GHG metrics are considered here as relevant to PtG goal of mitigating climate change. In addition, exergy analysis is a powerful thermodynamic tool that will also be employed to analyze and identify the inefficiencies in a process or a system and the potential for improvement.

In this Ph.D., multi-objective optimization methods and multi-criteria decision techniques will be applied for the selection of optimal and sustainable energy supply chains.

Research Question 3:

How could the methodological framework be used? How defining scenarios?

As seen above, Power-to-Gas concepts have been studied over the past decades and offer several key services to achieve decarbonization of the energy network. In this sense, the study of this potential in the French context is crucial for the development of this work. In particular, the proposed methodology could be helpful to study the effect of environmental regulations (i.e., carbon price) on the configuration of an HMSC.

Research Question 4:

How would the configuration of a hydrogen and methane supply chain change by considering the interaction with other technologies?

Although Power-to-Gas concepts offer several advantages, they also present some challenges to be overcome in comparison to other types of technologies and energy vectors such as SMR and natural gas (especially in terms of costs). In this sense, the interaction with other technologies needs to be investigated.

The doctoral research presented in this manuscript was conducted at the Laboratory of Chemical Engineering, LGC UMR CNRS 5503 INPT/UPS in the “Process Systems Engineering” (PSE) department. The project is part of the complementary work carried out within the Process and Systems Engineering (PSE) department, in particular, the Ph.D. works of:

- Sofia De León Almaraz, *Multi-objective optimization of a hydrogen supply chain*, 2014.
- Jesus Ochoa Robles, *Multi-objective optimization strategies for design and deployment of hydrogen supply chains*, 2018.

both dedicated to the hydrogen supply chain design. In this sense, the work carried out during this doctoral project aims to make a scientific contribution to the field of energy supply chain management.

The Ph.D. manuscript is organized into eight chapters. A brief description of the content of each chapter is presented below:

Chapter 1 has presented the aims and objectives of this research.

Chapter 2 explores the main technological components of an HMSC and the diversity of options that can be used in HMSC design. Specific focus will be paid to the design and deployment system to capture the required level of, technological and spatial detail needed.

Chapter 3 describes the multi-period and multi-objective approach, as well as the main methods and tools used in this study for HMSC modeling, optimization, and decision-making according to different criteria.

Chapter 4 presents the case study of the Occitania region in France that will serve as a guideline for the application of the developed methodology.

Chapter 5 is dedicated to the development of a methodological framework for the design and deployment of synthetic methane supply chains based on Power-to-Gas systems, through a mono-objective optimization formulation. The spatio-temporal integration of the technological bricks constituting the supply chains into the territory forms the basis of the modeling approach.

All the hydrogen produced is used as a feedstock to meet the demand for methane. The objective to minimize is the *total annualized cost* of the entire MSC over the total horizon studied. The Levelized Cost of Hydrogen and Methane can thus be calculated. Some key optimization variables are the number and size of plant units and hydrogen transport. This chapter paves the way for the three following chapters.

Chapter 6 will consider HMSC optimal design with a dual demand for renewable methane and hydrogen to be satisfied. In this case, both cost and environmental impacts are considered as objective functions.

Chapter 7 extends the HMSC in which renewable and non-renewable energy sources interact. In this sense, PtG, Steam Methane Reforming (SMR) concepts, and the possible import of natural gas are taken into account to satisfy the demands for hydrogen and methane. Besides, the use of carbon price is also considered as an environmental regulatory measure, which can be a driver for increased use of renewable sources. Three objective functions will be considered, i.e., cost, environmental impact, and production of renewable methane.

Chapter 8 incorporates exergy analysis as part of the set of decision criteria. The destruction and loss of exergy throughout the HMSC are then assessed, which allows better use of the useful work available. This analysis provides the basis for detecting possible links with other systems and thus reducing these losses.

Finally, **Chapter 9** presents the general conclusions related to the proposed strategy as well as perspectives for future work that derives from these findings.

This Ph.D. work led to the following publications:

A methodological design framework for Methane Supply Chains based on Power-to-Gas systems: application to the Occitania region in France. Submitted for publication in Applied Energy from ELSEVIER

A methodological design framework for hybrid “Power-to-Methane” and “Power-to-Hydrogen” supply chains: application to Occitanie region, France. Published in the 30th European Symposium on Computer Aided Process Engineering - Computers & Chemical Engineering from ELSEVIER.

Bi-objective optimal design of hydrogen and methane supply chains based on Power-to-Gas systems. Submitted for publication in Chemical Engineering Science from ELSEVIER.

A methodological design framework for hydrogen and methane supply chain with special focus on Power-to-Gas systems: application to Occitania region, France. Submitted for publication in Computers & Chemical Engineering from ELSEVIER.

Cost, environmental and exergy optimization of hydrogen and methane supply chains: application to Occitania region, France. Accepted for publication in the 31st European Symposium on Computer-Aided Process Engineering - Computers & Chemical Engineering from ELSEVIER.

INTRODUCTION ET DEFINITION DU PROBLEME

Introduction

L'Accord de Paris adopté lors de la Conférence de Paris sur le Climat (COP21) en décembre 2015 est une étape importante de la politique climatique internationale. Il stipule que l'augmentation de la température mondiale doit être maintenue en dessous de 2 °C, si possible à 1,5 °C au-dessus des niveaux préindustriels, afin d'atténuer les conséquences imprévisibles et irréversibles du changement climatique (United Nations, 2015). À cet égard, l'UE s'est fixée comme objectif de réduire les émissions de gaz à effet de serre d'au moins 40 % d'ici 2030 par rapport à 1990. Pour atteindre ces objectifs, le réseau énergétique doit subir une transformation radicale, puisque 87 % des émissions de CO₂ équivalentes proviennent des secteurs associés à la transformation de l'énergie : 53 % proviennent de la combustion de combustibles et des émissions volatiles de combustibles (sans les transports), 25 % des transports (y compris l'aviation internationale) et 9 % des processus industriels et de l'utilisation des produits (European Commission, 2015). À cet égard, l'utilisation des sources d'énergie renouvelables (SER) est l'un des piliers de la transition vers un avenir énergétique durable. Selon l'Agence Internationale pour les Énergies Renouvelables (IRENA, en anglais), les SER représenteraient deux tiers de la consommation d'énergie et 85 % de la production d'électricité en Europe d'ici 2050 (IRENA, 2018a). L'un des défis les plus importants dans l'intégration des SER dans le réseau électrique réside dans la nature "intermittente" de sources comme le vent et le soleil. Pour favoriser leur pénétration dans le mix énergétique, le concept de Power-to-Gas (PtG) est une solution prometteuse, dans laquelle l'énergie électrique et le CO₂ sont utilisés pour produire du gaz (c'est-à-dire de l'hydrogène ou du méthane), offrant plusieurs services:

1. Faire face à l'intermittence des sources d'énergie renouvelables en récupérant de la surproduction de grandes quantités d'électricité (Buttler and Spliethoff, 2018; Götz et al., 2016);

2. Apporter flexibilité en favorisant une plus grande intégration du réseau électrique à travers une interaction physique bidirectionnelle entre le réseau gazier et le réseau électrique (Buttler and Spliethoff, 2018; Khalilpour, 2018);
3. Assurer un approvisionnement en carburant à faible teneur en carbone (Buttler and Spliethoff, 2018; E&E consultant et al., 2014; Koj et al., 2019) with CO₂ capture and recovery (Götz et al., 2016).

À cet égard, le PtG est la seule technologie actuellement disponible qui offre la possibilité de stocker de l'énergie de façon saisonnière, en grande quantité, et qui possède aussi le potentiel de décarboner d'autres secteurs tels que la production de chaleur, le transport et certaines applications industrielles (ADEME, 2018; E&E consultant et al., 2014).

Avant d'aller plus loin dans ce travail, le concept de "Power-to-Gas" doit être précisément défini afin de déterminer le champ d'investigation de cette thèse de doctorat. En fait, dans la littérature, le Power-to-Gas peut se référer au Power-to-Hydrogen pour une série d'applications différentes, incluant parfois même le carburant pour la mobilité qui a été au cœur des travaux précédents de notre équipe de recherche au sein du "Laboratoire de Génie Chimique" (De-León Almaraz, 2014). Dans d'autres définitions, le PtG correspond au processus qui relie le réseau électrique au réseau gazier en convertissant l'électricité excédentaire en un gaz compatible via un processus en deux étapes : production d'H₂ par électrolyse de l'eau et conversion de H₂ avec une source externe de CO ou de CO₂ en CH₄ par méthanisation (Götz et al., 2016). Ainsi, l'hydrogène obtenu à partir du PtG peut être principalement :

1. stocké puis retransformé en électricité (Dagdougui, 2012)
2. injecté dans le réseau de gaz naturel (selon les limites du réseau local de gaz naturel, par exemple 6% en France) (E&E consultant et al., 2014).
3. utilisé pour répondre à une partie de la demande du secteur des transports ou de l'industrie (Buttler and Spliethoff, 2018).
4. utilisé dans une seconde étape pour produire du méthane par méthanisation (réaction de Sabatier) (Buttler and Spliethoff, 2018; Götz et al., 2016).

Le méthane (c'est-à-dire le gaz naturel de substitution ou le méthane synthétique) présente certains avantages, car il peut être stocké et injecté sans restrictions techniques dans l'infrastructure de gaz naturel bien développée (Khalilpour, 2018) ou de stockage de gaz, utilisé comme carburant, il peut aussi être facilement utilisé dans des installations industrielles. Malgré ces avantages, le PtG présente certains défis qui doivent être relevés avant qu'il ne puisse jouer un rôle important dans le mix énergétique. Parmi ceux-ci, nous pouvons mentionner que :

- les pertes d'énergie pourraient être importantes tout au long de la chaîne logistique (Thema et al., 2019).
- dans le cas de l'hydrogène, la limite de mélange est actuellement de 10 % (sur la base du volume), la plupart des pays étant en dessous de 2 (Quarton and Samsatli, 2018).
- le méthane pourrait être injecté directement dans le réseau de gaz naturel, mais l'étape de méthanisation implique des coûts supplémentaires (Blanco and Faaij, 2018; E&E consultant et al., 2014).

Pour surmonter ces ambiguïtés et ce manque de précision sémantique, cette thèse promeut plutôt les termes "Power-to-Hydrogen" et "Power-to-Methane". Le terme "PtG" comprendra les deux concepts. Nous mentionnons également leurs chaînes logistiques (SC, en anglais) correspondantes avec les termes explicites HSC (Hydrogen Supply Chain) et MSC (Methane Supply Chain), respectivement pour l'hydrogène et le méthane. Le terme HMSC (Hydrogen and Methane Supply Chain) fera référence à la double production des deux vecteurs. Cela maintient "l'hydrogène" au centre et souligne ainsi sa flexibilité en tant que vecteur d'énergie.

À ce niveau, il convient de souligner que la chaîne logistique d'énergie est plus générique dans sa définition que les concepts de PtG dans le sens où des sources autres que l'électricité peuvent être utilisées pour la production d'hydrogène. Aujourd'hui, l'hydrogène est principalement produit par le reformage du méthane à la vapeur (SMR, « Steam Methane Reforming ») qui est fortement carboné, puisque le gaz naturel est utilisé comme matière première (c'est-à-dire Chaîne gaz-gaz). Sa production avec un minimum d'émissions de gaz à effet de serre (GES), par exemple par électrolyse alimentée par de l'électricité renouvelable ou à partir de bioénergie ou de combustibles fossiles avec capture et stockage du carbone (CSC) (Hydrogen Council, 2020) représente une alternative de grand intérêt. L'ensemble des options est résumé dans la Figure 0.1.

Les services potentiels offerts par les systèmes PtG en général sont conformes à la stratégie de l'UE pour l'intégration du système énergétique (European Commission, 2020a). Les investissements cumulés en hydrogène renouvelable en Europe pourraient atteindre 180 à 470 milliards d'euros d'ici 2050. Cela contribuerait à positionner l'UE en tant que leader mondial dans la fabrication de technologies propres de l'hydrogène, puisque l'on prévoit que l'hydrogène propre pourrait satisfaire 24 % de la demande énergétique mondiale d'ici 2050, avec des ventes annuelles de l'ordre de 630 milliards d'euros (European Commission, 2020b). À cet égard, plusieurs stratégies de transition énergétique des pays européens ont déjà intégré les systèmes PtG dans l'ensemble des technologies pour atteindre leurs objectifs (ADEME, 2018; E&E consultant et al., 2014; Gigler and Weeda, 2018; Thomas et al., 2016), en particulier en Allemagne où la technologie PtG est considérée comme une pierre angulaire de son futur système énergétique (Schiebahn et al., 2015; Varone and Ferrari, 2015). Ces études sont cohérentes avec le rôle majeur que les systèmes PtG devraient jouer à partir de 2030 avec une projection de 80 à 100% de

production d'électricité renouvelable en 2050 (Belderbos et al., 2015). Ils soulignent que la rentabilité des systèmes PtG dépend des circonstances locales et des conditions du marché (Holstein and van den Noort, 2018).

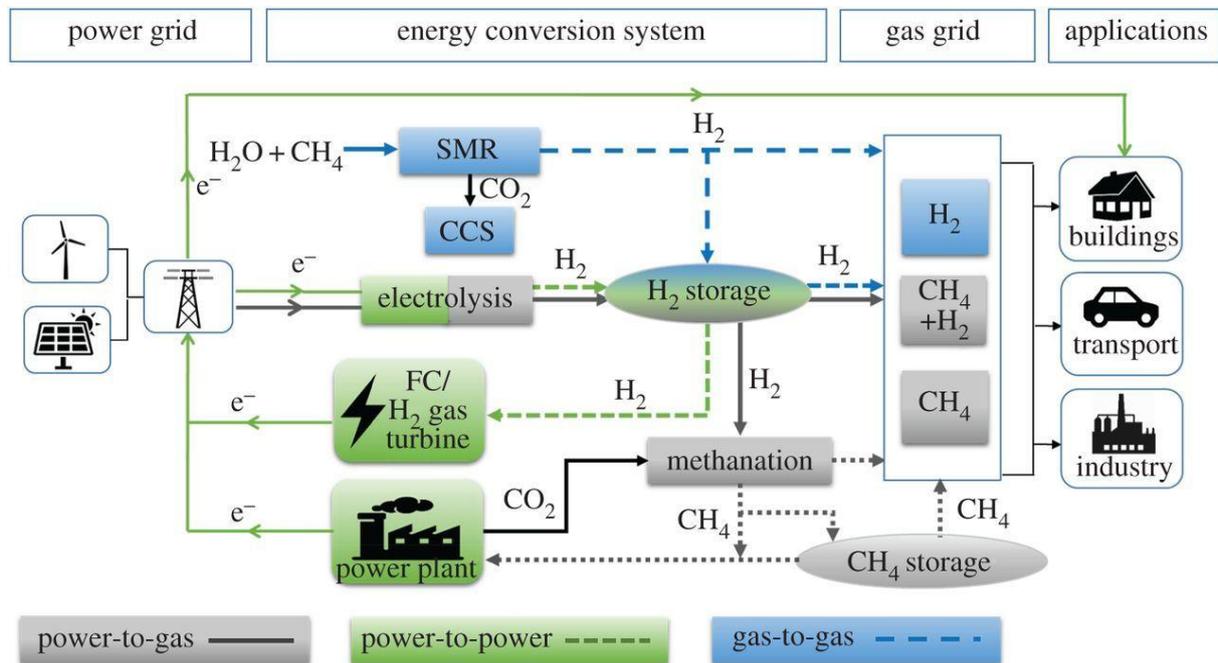


Figure 0.1 Principales voies de conversion de l'énergie (power-to-gas, power-to-power and gas-to-gas) dans un système intégré d'énergie renouvelable (Brandon and Kurban, 2017)

Dans le cas de la France, l'hydrogène renouvelable tel que défini dans la loi française du 18 août 2015 et dans la Stratégie Nationale de Recherche Énergétique (SNRE) (Ministère de l'Économie, des Finances et de la Relance, 2020) et le syn-méthane prendront une place importante dans son futur mix énergétique (ADEME, 2018; E&E consultant et al., 2014; Fuel Cells and Hydrogen Joint Undertaking (FCH JU), 2020). Le réseau français de gaz naturel est le plus long d'Europe et dispose d'importantes interconnexions gazières en raison de ses frontières avec cinq pays européens. En outre, sa capacité de stockage est la troisième en Europe avec 132 TWh (H2V INDUSTRY, n.d.; Ministère de la Transition Écologique et Solidaire, 2018). Huit projets pilotes dédiés aux systèmes PtG ont été mis en œuvre en France de 2000 à 2015 (Thema et al., 2019), et un nouveau pilote PtG sera installé dans le nord de la France, avec une capacité de 610 MW_{el} (H2V INDUSTRY, n.d.).

L'Agence de l'environnement et de la maîtrise de l'énergie (ADEME) ("Agence de la transition écologique," n.d.) a développé une série d'études sur les systèmes PtG dans le contexte français, en tenant compte de la disponibilité des ressources, des applications possibles, des coûts et des limites (ADEME, 2018; E&E consultant et al., 2014). En outre, l'hydrogène est considéré comme un élément essentiel des plans de transition énergétique dans les territoires insulaires et pour le déploiement des

pôles de compétitivité comme un outil efficace pour tirer profit de la croissance des énergies renouvelables. Un exemple phare est le projet MYRTE en Corse, où sont testées les technologies émergentes de l'hydrogène, impliquant des électrolyseurs, des piles à combustible, le stockage de l'hydrogène et des stratégies de couplage avec les sources d'ER (Cristofari et al., 2014).

Au niveau régional, la Région Occitanie a annoncé l'ouverture d'ici 2024 à Francazal à Toulouse un technocampus dédié aux technologies de l'hydrogène en lien étroit avec le projet Hyport qui accueillera le plus grand centre européen de recherche, d'essai et d'innovation technologique dédié à l'hydrogène vert en partenariat avec le CNRS et ses Laboratoires de Recherche (Laboratoire plasma et conversion d'énergie (Laplace), Cirimat, le Laboratoire de Génie Chimique (LGC) et l'Institut de mécanique des fluides de Toulouse (IMFT), l'Université de Toulouse, l'ONERA, des laboratoires de recherche mais aussi des grands groupes comme Safran, Airbus, Liebherr et Vitesco Technologies.

Problématique de recherche et questions de recherche

Au cours des dernières années, l'intérêt scientifique pour les chaînes d'approvisionnement en hydrogène et en méthane (HMSC) s'est progressivement accru, comme le montre la Figure 0.2. Trente articles ont été identifiés de 2005 à 2020, sur la base d'une première recherche dans le Web of Science (WoS) à l'aide des mots clés "renewable hydrogen and methane supply chains". Compte tenu de l'ensemble des technologies, des sources d'énergie et des services concernés par le HMSC, les éléments liés aux systèmes PtG ont été largement abordés dans la littérature (Buttler and Spliethoff, 2018; Götz et al., 2016), en particulier la chaîne logistique d'hydrogène (Dagdougui, 2012; Li et al., 2019). À cet égard, le nombre de publications dont les mots clés sont "Hydrogen Supply Chain" et "Power-to-Gas" est de 720 et 1198, respectivement.

Les modèles de systèmes énergétiques constituent la base de la plupart des scénarios énergétiques quantitatifs. Il existe un grand nombre d'outils de modélisation de tels systèmes qui peuvent être classés de différentes manières, notamment la simulation vs. optimisation, top-down vs. bottom-up, etc. Une revue sur les outils informatiques pour les systèmes énergétiques (Connolly et al., 2010) a identifié 68 outils de modélisation de systèmes énergétiques différents. Suivant la même taxonomie, un examen détaillé et récent de 24 systèmes énergétiques (Lopion et al., 2018) a constaté une tendance claire vers des modèles d'optimisation technico-économiques ascendants afin de répondre aux questions actuelles de la recherche. La tâche la plus difficile pour les modèles de systèmes énergétiques modernes est de saisir de manière intégrée le degré complet de variabilité et de complexité qui existe dans les systèmes énergétiques. Pour que les modèles de systèmes énergétiques fournissent non seulement une représentation précise des systèmes énergétiques, mais aussi qu'ils ne passent pas « à côté » du potentiel des nouvelles technologies telles que celles basées sur l'hydrogène, ils doivent prendre en compte de

manière dynamique le niveau requis en termes temporel, spatial et technologique, (Poncelet et al., 2016) et ainsi que le niveau de détail intersectoriel approprié (Lucchese et al., 2018). *Ce travail de doctorat s'inscrit dans cette perspective pour les systèmes PtG.*

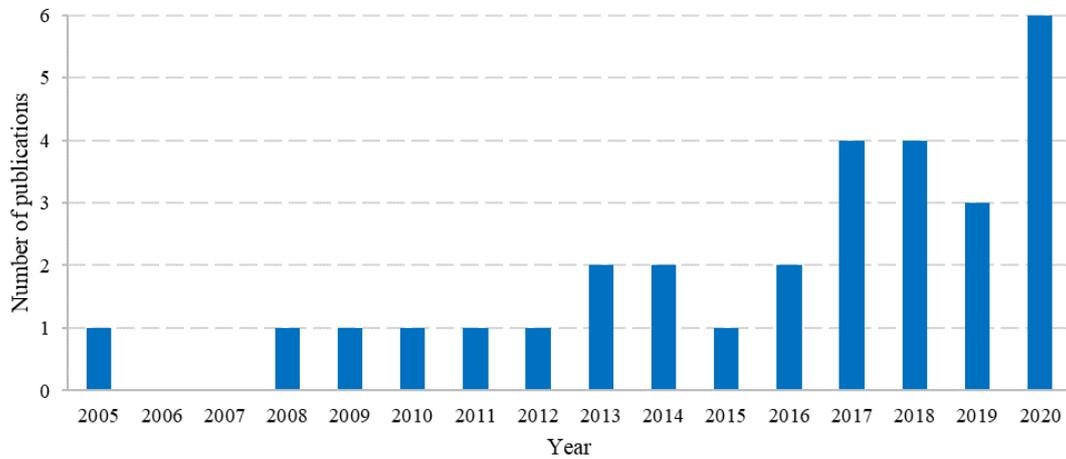


Figure 0.2 Nombre d'articles basés sur une première recherche sur le site web de Science™ (WoS) à l'aide des mots clés "renewable hydrogen and methane supply chains" (5 Décembre 2020)

Un point clé du développement de la chaîne logistique basée sur les systèmes PtG est la démonstration de la faisabilité de son infrastructure alors que de nombreux obstacles techniques, économiques et sociaux doivent être surmontés. Certaines feuilles de route stratégiques ont été publiées sur les potentiels énergétiques de l'hydrogène qui est au cœur des systèmes de PtG à l'échelle européenne, nationale et régionale. Leur principal objectif est d'évaluer certaines questions industrielles, technologiques, environnementales et sociales et d'identifier les principaux obstacles associés à l'économie de l'hydrogène.

La revue de la littérature a montré que de nombreuses recherches ont été menées en considérant les différents usages, les principales briques technologiques, les sources d'énergie et de CO₂ impliquées dans le PtG (Götz et al., 2016, 2014; Lewandowska-Bernat and Desideri, 2018; Li et al., 2019; Rönsch et al., 2016; Wulf et al., 2018). En ce qui concerne les aspects logistiques, la chaîne logistique de l'hydrogène a été abordée dans plusieurs études, comme le soulignent par exemple deux synthèses bibliographiques publiées à huit ans d'intervalle (Dagdougui, 2012; Li et al., 2019). Cependant, seules quelques études ont abordé les aspects logistiques liées au systèmes PtG, en particulier la double production d'hydrogène et de méthane avec les problèmes qui y sont associés, c'est-à-dire la détermination de l'emplacement, de la taille et du nombre d'unités de production, de stockage et de transport, ainsi que des stations de service d'hydrogène.

Des publications scientifiques récentes spécialisées s'accordent sur la nécessité de développer des études systémiques afin de démontrer la faisabilité du secteur, de valider l'intérêt technique et économique de la production et de la récupération de l'hydrogène produit à partir de sources renouvelables. De tels travaux impliquent le développement de modèles basés sur des scénarios économiques pour le déploiement du PtG.

Pour pallier ce manque, l'objectif scientifique de ce travail est de proposer un cadre méthodologique complet pour la conception et le déploiement optimal multi-objectifs des chaînes logistiques de l'hydrogène et du méthane (HMSC), en se concentrant sur les systèmes PtG et leur intégration spatio-temporelle en tenant compte des potentiels régionaux des sources d'énergie renouvelables (SER). Ce cadre conduira à un système d'aide à la décision pour l'analyse des scénarios.

Selon (Fuel Cells and Hydrogen Joint Undertaking, 2019), l'approvisionnement enhydrogène pourrait provenir d'un mélange de sources à très faible teneur en carbone. Bien que la répartition exacte des modes de production puisse varier selon les applications et dépendre de l'évolution des technologies et des coûts, l'électrolyse et le reformage du méthane à la vapeur/le reformage autothermique avec capture et stockage du carbone joueront très probablement un rôle clé. L'électrolyse pourrait fournir le mécanisme de couplage sectoriel requis pour l'intégration des énergies renouvelables et une brique technologique majeure du PtG. Actuellement, des électrolyseurs sont disponibles pour une production à petite échelle (< 1 MW), et des projets de démonstration pour des échelles plus grandes (jusqu'à 10 MW) sont en cours. Le reformage du méthane à la vapeur est la technique de production d'hydrogène à grande échelle la plus courante actuellement et pourrait être décarbonée grâce au captage et à la séquestration du carbone . Les scénarios ne reposant que sur l'une de ces deux voies de production semblent irréalistes et seraient en deçà du déploiement requis. Le couplage des deux technologies doit être étudié pour voir si le reformage à la vapeur peut être considéré comme une technologie de transition nécessaire.

Les principales questions de recherche abordées dans ce travail peuvent être énoncées comme suit.

Question de recherche 1 :

Comment concevoir et intégrer spatialement dans un territoire une chaîne logistique de l'hydrogène et du méthane reposant principalement sur des systèmes "Power-to-Gas" ? Comment pourrait-elle être déployée pour atteindre le niveau de détail temporel, spatial et technologique requis ?

Le HMSC basé sur les systèmes "Power-to-Gas" n'a guère été abordé. Plusieurs aspects tels que le changement de configuration dans le temps et la variation en fonction de différents critères n'ont pas été

tarités à notre connaissance. L'objectif scientifique principal de ce travail de doctorat est de développer un cadre méthodologique pour l'optimisation du HMSC issue des systèmes Power-to-Gas, c'est-à-dire déterminer l'emplacement, la taille et le nombre d'unités de production, de stockage et de transport, ainsi que de stations de ravitaillement en hydrogène.

Question de recherche 2 :

Quels sont les indicateurs clés de performance à utiliser pour la conception des HMSC ? Comment la configuration d'une chaîne logistique d'hydrogène et de méthane changerait-elle en fonction de différents critères de décision ? Comment prendre en compte les aspects multicritères ?

Le processus de production d'énergie et de carburant s'inscrit dans une problématique de développement durable. La conception de chaînes logistiques durables est devenue un sujet de plus en plus important qui suscite aujourd'hui un intérêt croissant de la part des praticiens et des académiques. L'utilisation de mesures appropriées pour déterminer les compromis est un élément essentiel de la prise de décision. Il existe de nombreux indicateurs environnementaux potentiellement concurrents qui pourraient être utilisés pour évaluer l'impact environnemental d'un système énergétique, en suivant les lignes directrices de l'analyse du cycle de vie (Hauschild et al., 2018). Tous ces éléments sont essentiels, mais les mesures des GES sont considérées ici comme pertinentes dans un objectif de réduire les effets sur le changement climatique. L'analyse exergétique en tant qu'outil analyse thermodynamique puissant sera utilisée pour analyser et identifier les inefficacités d'un processus ou d'un système et mettre en évidence le potentiel d'amélioration nécessaire.

Dans le cadre de cette thèse, des méthodes d'optimisation multi-objectifs et des techniques de décision multi-critères seront appliquées pour la sélection de chaînes logistiques énergétiques optimales et durables.

Question de recherche 3 :

Comment le cadre méthodologique pourrait-il être utilisé ? Comment définir des scénarios ?

Comme vu ci-dessus, les concepts de Power-to-Gas ont été étudiés au cours des dernières décennies et offrent un certain nombre de services clés pour décarboner le réseau énergétique. En ce sens, l'étude de ce potentiel dans le contexte français est cruciale pour le développement de ces travaux. En particulier, la méthodologie proposée pourrait être utile pour étudier l'effet des réglementations environnementales (c'est-à-dire la taxe carbone) sur la configuration d'un HMSC.

Question de recherche 4 :

Comment la configuration d'une chaîne logistique d'hydrogène et de méthane changerait-elle en tenant compte de l'interaction avec d'autres technologies ?

Bien que les concepts "Power-to-Gas" offrent plusieurs avantages, ils présentent également un certain nombre de défis à relever par rapport à d'autres types de technologies et de vecteurs énergétiques tels que le reformage à la vapeur et le gaz naturel (notamment en termes de coûts). En ce sens, l'interaction avec d'autres technologies doit être étudiée.

Ce travail de thèse de doctorat présente dans ce manuscrit a été mené au Laboratoire de Génie Chimique, LGC UMR CNRS 5503 INPT/UPS dans le département "Procédés et Systèmes Informatiques" (PSI). Le projet s'inscrit dans le cadre des travaux complémentaires menés au sein du département, en particulier les travaux de thèse de:

- Sofia De León Almaraz, *Multi-objective optimization of a hydrogen supply chain*, 2014.
- Jesus Ochoa Robles, *Multi-objective optimization strategies for design and deployment of hydrogen supply chains*, 2018.

tous deux dédiés à la conception de la chaîne logistique de l'hydrogène. En ce sens, les travaux menés ici visent à apporter une contribution scientifique au domaine de la gestion de la chaîne logistique de l'énergie. Le manuscrit du doctorat est organisé en neuf chapitres. Une brève description du contenu de chacun d'entre eux est présentée ci-dessous :

Le **chapitre 1** a présenté les objectifs de cette recherche.

Le **chapitre 2** explore les principales composantes technologiques d'une HMSC et la diversité des options qui peuvent être utilisées dans sa conception. Une attention particulière sera accordée à la conception et au déploiement du système afin de prendre en compte le niveau de détail technologique et spatial requis.

Le **chapitre 3** décrit l'approche multi-périodes et multi-objectifs, ainsi que les principales méthodes et outils utilisés dans cette étude pour la modélisation, l'optimisation et l'aide à la décision selon différents critères.

Le **chapitre 4** présente l'étude de cas de la région Occitanie en France qui servira de guide pour l'application de la méthodologie développée.

Le **chapitre 5** est consacré à l'élaboration du cadre méthodologique pour la conception et le déploiement de chaînes logistiques de méthane synthétique basées sur des systèmes "Power-to-Gas", grâce à une formulation d'optimisation mono-objectif. L'intégration spatio-temporelle des briques technologiques constituant les chaînes logistiques dans le territoire constitue la base de l'approche de modélisation.

Tout l'hydrogène produit y est utilisé comme matière première pour répondre à la demande en méthane. L'objectif à minimiser est le *coût total annualisé* de l'ensemble du MSC sur l'ensemble de l'horizon étudié. Le coût moyen de l'hydrogène et du méthane (un équivalent LCOE) peut ainsi être calculé. Certaines variables clés de l'optimisation sont le nombre et la taille des unités de production et le transport de l'hydrogène. Ce chapitre amorce les trois chapitres suivants du point de vue méthodologique.

Le **chapitre 6** examine la conception optimale du HMSC avec une demande duale en méthane et hydrogène renouvelable à satisfaire. Dans ce cas, le coût et les impacts environnementaux sont considérés comme des fonctions objectifs.

Le **chapitre 7** étend le HMSC dans lequel les sources d'énergie renouvelables et non renouvelables interagissent. En ce sens, les concepts de PtG, de reformage du méthane à la vapeur et l'importation éventuelle de gaz naturel sont pris en compte pour satisfaire les demandes d'hydrogène et de méthane. En outre, l'utilisation d'une taxe carbone est également considérée comme une mesure réglementaire environnementale, qui peut être un levier pour une utilisation accrue des sources renouvelables. Trois fonctions objectifs seront prises en compte, à savoir le coût, l'impact environnemental et la production totale de méthane renouvelable.

Le **chapitre 8** intègre l'analyse de l'exergie dans l'ensemble des critères de décision. La destruction et la perte d'exergie dans l'ensemble du HMSC sont ensuite évaluées, ce qui permet de mieux utiliser le travail utile disponible. Cette analyse permet de détecter d'éventuels liens avec d'autres systèmes et donc de réduire ces pertes.

Enfin, le **chapitre 9** présente les conclusions générales relatives à la stratégie proposée ainsi que les perspectives des travaux futurs qui découlent de ces résultats.

Ce travail de doctorat a donné lieu aux publications suivantes :

A methodological design framework for Methane Supply Chains based on Power-to-Gas systems: application to the Occitania region in France. Soumise pour publication: Applied Energy (ELSEVIER).

A methodological design framework for hybrid "Power-to-Methane" and "Power-to-Hydrogen" supply chains: application to Occitanie region, France. Publiée dans le 30th European Symposium on

Computer Aided Process Engineering - Computers & Chemical Engineering (ELSEVIER).

Bi-objective optimal design of hydrogen and methane supply chains based on Power-to-Gas systems.
Soumise pour publication: Chemical Engineering Science (ELSEVIER).

A methodological design framework for hydrogen and methane supply chain with special focus on Power-to-Gas systems: application to Occitania region, France. Soumise pour publication: Computers & Chemical Engineering (ELSEVIER).

Cost, environmental and exergy optimization of hydrogen and methane supply chains: application to Occitania region, France. Acceptée pour publication: 31th European Symposium on Computer Aided Process Engineering - Computers & Chemical Engineering (ELSEVIER).

2 DEPLOYMENT AND DESIGN OF HYDROGEN AND METHANE SUPPLY CHAINS BASED ON PTG: HOW TO CAPTURE THE REQUIRED LEVEL OF TECHNOLOGICAL AND SPATIAL DETAIL IN MODELING

ABSTRACT

This chapter presents the main technological components of an HMSC are presented to explore the diversity of options that can be used in the design. Specific focus will be paid to the design and deployment system to capture the required level of, technological and spatial detail needed. The energy system modeling approach will be reviewed to present the main guidelines of the methodological framework for decision-support of PtG systems based on scenario analysis that will be designed in the following chapters.

RÉSUMÉ

Ce chapitre présente les principaux composants technologiques d'une chaîne logistique hydrogène-méthane afin d'explorer la diversité des options qui peuvent être utilisées en conception. Une attention particulière sera accordée à la conception et au déploiement de système afin de prendre en compte le niveau de granularité nécessaire au niveau technologique et spatial. L'approche de modélisation de systèmes énergétiques sera abordée afin de présenter les principales lignes directrices du cadre

méthodologique pour l'aide à la décision, dans le contexte des systèmes PtG, sur la base de l'analyse des scénarios qui seront conçus dans les chapitres suivants.

2.1 Introduction

Power-to-Gas (PtG) systems have received considerable attention given the current energy transition context. They are viewed as a pivotal element for coupling the electric and gas infrastructure, facilitating the integration of renewable forms of energy into well-established and high-performing energy storage and distribution systems. In addition, the technological blocks for their implementation have reached a state of maturity that allows their establishment at a commercial level (Acar and Dincer, 2019; Götz et al., 2014). However, one of the main barriers to PtG deployment is the lack of a hydrogen infrastructure. Given the energy vectors involved, PtG systems can be viewed as a sub-system of hydrogen and methane supply chains, and their role is expected to be crucial in the context of ambitious energy transition objectives. In this regard, the IRENA and the Hydrogen Council indicate that about 6 to 18% of global energy demand can be met through hydrogen (IRENA, 2019). In addition, in the case of scenarios with renewable energy penetrations greater than 80% in the energy grid, synthetic methane can cover about 10% of energy demand.

For this purpose, the deployment of an HMSC involves a set of technological, logistical, and spatial options that need to be analyzed. An example of a generic HMSC can be seen in Figure 2.1. The hydrogen/methane supply chain can be defined as a system of activities from suppliers to customers. These activities, i.e. the so-called echelons are feedstock for hydrogen production, hydrogen transportation and storage, methanation (using an external source of CO₂), and end-uses. Unlike most other fuel infrastructures, hydrogen can be produced either centrally (similar to existing gasoline supply chains) or distributed (as small-scale units that can produce H₂ close to the use point in small quantities) modes at forecourt refueling stations, so that the distribution cost is significantly reduced. In the case of renewable methane, this involves the production of hydrogen through electrolysis up to the injection of methane into the natural gas grid. Besides, for the production of methane, an external source of CO₂ is required, which can come from biogenic (i.e., methanization, gasification...) or industrial (i.e., cement, steel industry...) sources (E&E consultant et al., 2014; Götz et al., 2016). To transform the different raw materials into the final energy vector, multiple options are available. For instance, the solar, wind, and hydraulic energy sources can be used to produce electricity so that hydrogen is then produced by electrolysis. Moreover, biomass, coal, and natural gas can be used to produce hydrogen through biomass gasification, coal gasification, or steam methane reforming (SMR). Depending on the physical form of the hydrogen produced (i.e., liquid or gaseous form), there are various modes of transportation (i.e., tanker-truck, tube trailer, or pipeline), storage, and terminal points. Finally, hydrogen can be used to meet a particular demand (i.e., mainly transport, electricity, industry, and building).

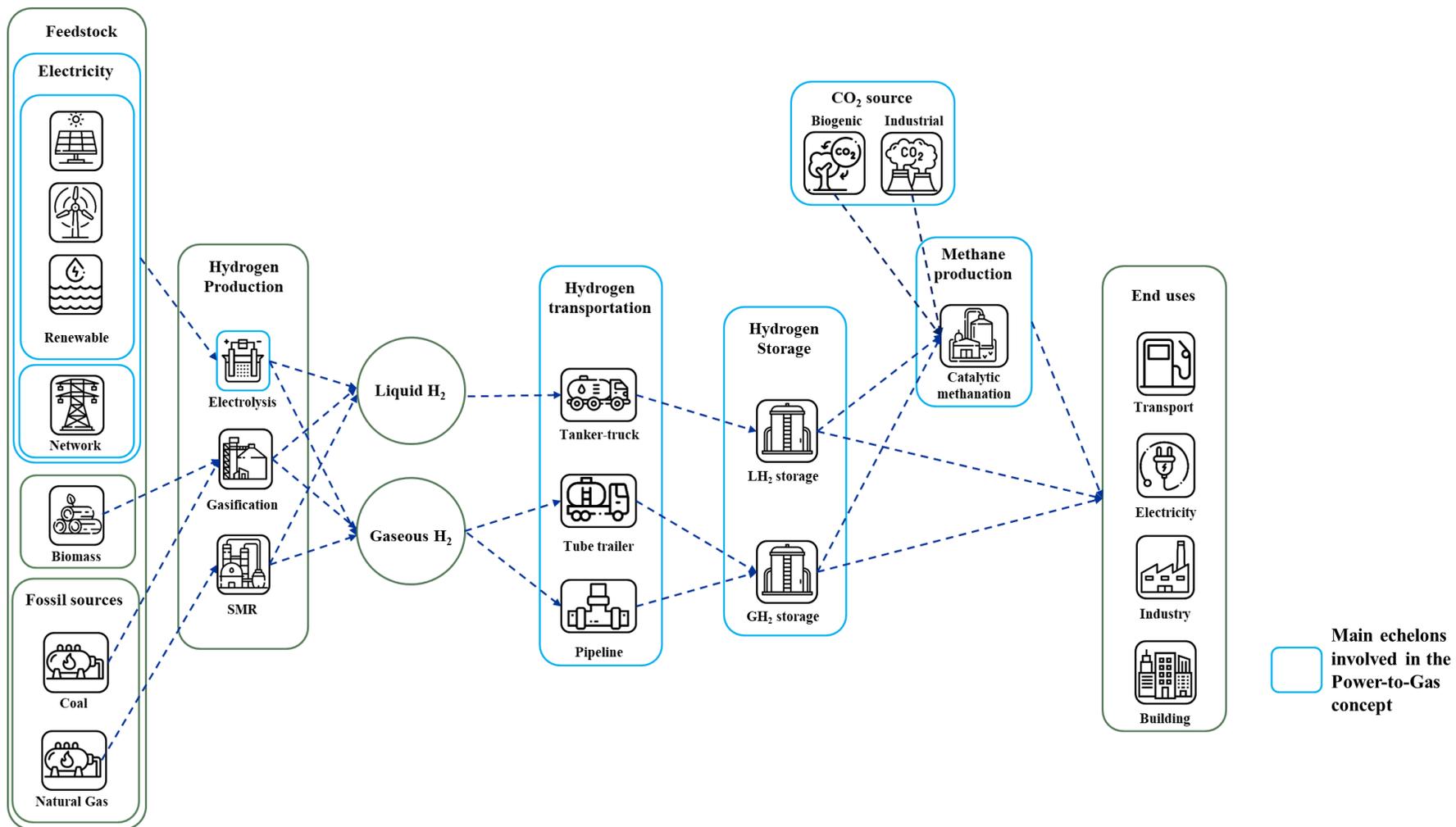


Figure 2.1. General HMSC including feedstock, product, physical form, transport, storage, and end-uses

Renewable energy markets require a development strategy based on complex design, planning, and optimization (Baños et al., 2011; De Meyer et al., 2014). Given the diversity of raw materials, end uses, and markets involved, the corresponding technologies are interconnected. In this regard, process efficiency is another important characteristic. Figure 2.2. illustrates the efficiency losses of described typical Power-to-Power, Power-to-Gas, and Gas-to-Gas processes.

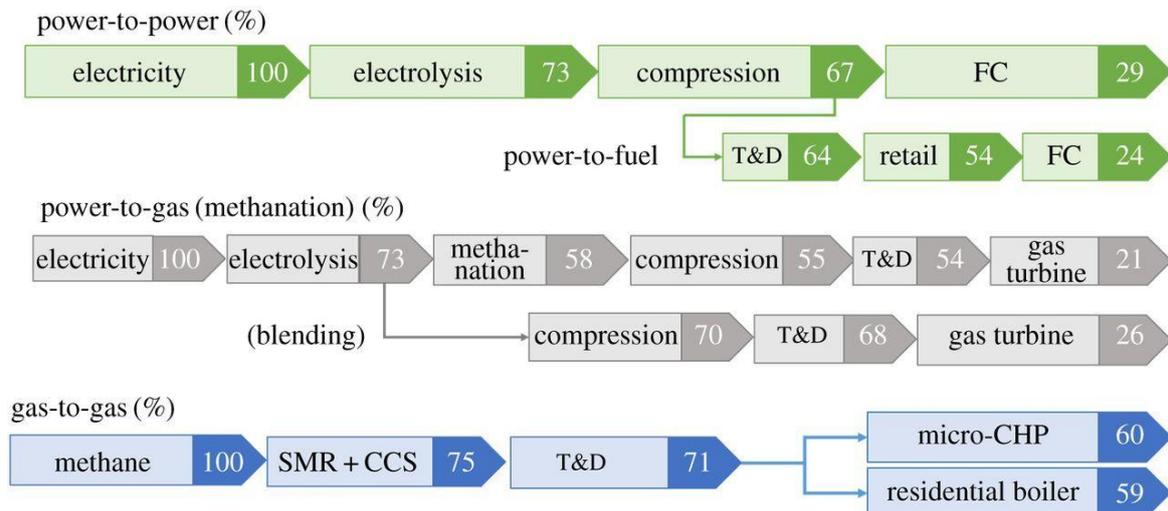


Figure 2.2. The step conversion efficiencies for the hydrogen and methane supply pathways being considered (SMR+CCS efficiency of 75%, transport and distribution (T&D) loss of 5% (distribution only), residential boiler efficiency 84%, and residential micro-CHP fuel cell efficiency 88%) (Brandon and Kurban, 2017)

Consequently, the complexity of HMSC design also depends on these interactions that are also influenced by the territory being analyzed, since the availability and characteristics of raw materials are regionally dependent and therefore affect the location of production units, storage, and the means of transport to be used (Li et al., 2019). In addition, the location of refueling stations for hydrogen also depends on the existing demand (De-León Almaraz, 2014).

All these features make the management of supply chains a complex task mainly due to the large size of the physical supply network and inherent uncertainties and decisions (Papageorgiou, 2009):

- number, size and location of manufacturing sites, storages and distribution centers, and the resources inside them;
- production decision related to plant production planning and scheduling;
- network connectivity (e.g. allocation of suppliers to plants, etc.);
- management of inventory levels and replenishment policies;
- transportation decisions concerning the mode of transportation (e.g. road, rail, etc.) and also sizes of material shipments.

The supply chain management problem can broadly be divided into three main categories:

- (i) strategic phase: supply chain design;

- (ii) tactical phase: supply chain planning and scheduling;
- (iii) operational phase: supply chain control (real-time management).

Only the strategic problem will be tackled in this work. In the next sections, the main components of an HMSC are presented to explore the diversity of design options, with specific focus paid to capture the required level of temporal, spatial, technological detail needed. The energy system modeling approach will be reviewed to present the main guidelines of the methodological framework for decision-support of PtG systems based on scenario analysis that will be the core of the following chapters.

2.2 Main technological bricks involved in hydrogen and methane supply chains

2.2.1 Hydrogen supply

Hydrogen, which is seen as the main product from PtG concepts, is currently produced by the Steam Methane Reforming (SMR) process that satisfies approximately 95% of the total demand for hydrogen. However, the possible production of green hydrogen from water electrolysis with renewable electricity is an opportunity for the process to address a new and large market.

2.2.1.1 Steam methane reforming

Steam methane reforming (SMR) is the most common and cheapest method for hydrogen production, since it has high hydrogen yield efficiency (~74%) and is estimated to produce hydrogen at a cost of around \$1.8kg⁻¹ (Velazquez Abad and Dodds, 2017). The main reaction involved is shown in Eq. 2.1.



In the industrial context, the SMR reaction is carried out in conventional reactors. Several steps are necessary, such as the reduction of carbon monoxide content in the reformat stream by water gas shift reactors, pressure swing adsorption, and further hydrogen separation/purification devices, to obtain a highly pure hydrogen stream (Basile et al., 2015b). A simplified process flow diagram with the main sub-process involved is shown in Figure 2.3. According to (Basile et al., 2015a), when hydrogen needs to be high-grade, a multistep system is used consisting of a conventional reactor (CR), water gas shift (WGS) reactors (high- and low-temperature reactors), and hydrogen separation/purification devices. In the first stage, methane and steam react in the conventional reformer under harsh operating conditions such as 800–1000 °C and 14–20 bar over Ni-based catalysts. Because the CO content is relatively high in the outlet stream of the CR, two reactors in series for WGS are generally required. The first reactor (high-temperature WGS) is packed with chromium-promoted iron oxide catalysts, operating between 350 and 400 °C; the second reactor (low-temperature WGS) is packed with copper-promoted zinc oxide, operating at around 200 °C. Commonly, the outlet stream from the shift reactors contains 86% H₂, 12% CO₂, 0.4% CO, and 1.6% CH₄ on a dry basis (Kirk-Othmer, 2000). Therefore, further steps are mandatory for obtaining high-grade hydrogen. Further techniques can be adopted for

separating/purifying the reformed hydrogen, each of which can be chosen according to hydrogen purity requirements.

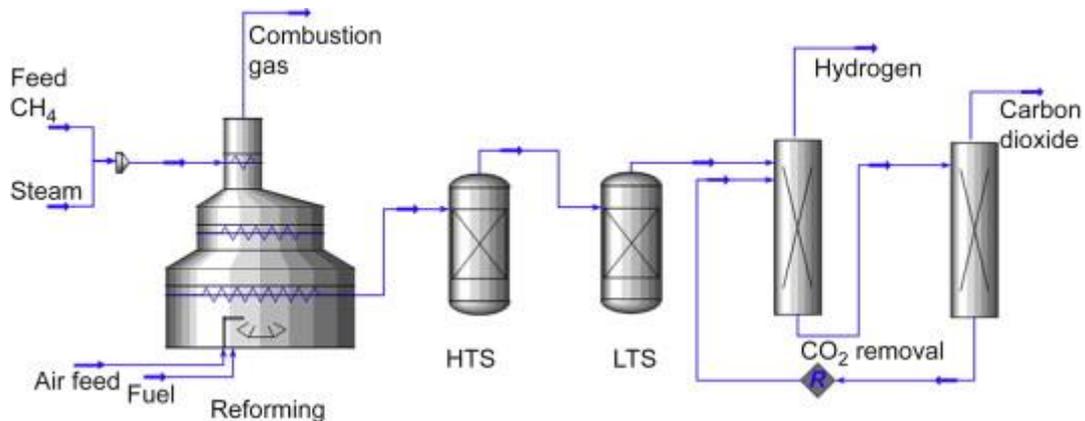


Figure 2.3 Simplified process flow diagram for hydrogen production by steam methane reforming (Basile et al., 2015a)

According to (Speight, 2020), the process has the following advantages: (i) produces 4 mol of hydrogen for each mole of methane consumed, (ii) feedstocks for the process (methane and water are readily available), (iii) the process is adaptable to a wide range of hydrocarbon feedstock, (4) operates at low pressures, less than 450 psi, (v) requires a low steam/carbon ratio (2.5–3), (vi) good utilization of input energy (reaching 93%), (vii) can use catalysts that are stable and resist poisoning, and (viii) acceptable process kinetics. Although SMR is a mature technology, it suffers from significant disadvantages such as mass and heat transfer issues and coke deposition during the reaction (Basile et al., 2015b). Moreover, as the reforming process is not efficient, some of the energy value of the hydrocarbon fuel is lost by conversion to hydrogen but with no tangible environmental benefit, such as a reduction in emission of greenhouse gases (Speight, 2020).

Given the significant CO₂ emissions emanating from this process, a promising option is the integration of carbon capture and storage (CCS). Approximately 90% of the CO₂ emissions can be captured by using CCS technologies. However, its incorporation requires additional energy inputs to scrub, compress, and transport the CO₂, reducing the operating efficiency of the SMR plant by at least 5% (Velazquez Abad and Dodds, 2017).

2.2.1.2 Gasification

Gasification is a technology that has been used from the mid-1800s through the early 1900s to produce gas from coal for heating and lighting purposes. An important moment for this process was during the Second World War when farmers build small gasifiers to convert biomass (i.e., wood) into gas/liquid fuel for their tractors and farming equipment (Speight, 2020). In recent decades, this process has gained increasing interest in hydrogen production.

Gasification is an endothermic process and requires a heat source to promote the reaction. Two main feedstocks are used for this purpose, i.e., biomass and coal. Figure 2.4 shows the main reactions involved in these processes. Both alternatives are described below.

A. Biomass

Gasification is seen as an efficient conversion technology for converting biomass into hydrogen. It is currently in a pilot-scale demonstration or at a commercial stage (Lepage et al., 2021). It can provide an efficient process for hydrogen production that can simultaneously act as a waste disposal system when carried out under appropriate conditions. Other benefits of gasification technology include feedstock flexibility, product flexibility, near-zero emissions, high efficiency, and energy security (Speight, 2020). The conversion takes place generally in a fluidized bed, where biomass (i.e., agriculture waste or wood) is gasified at a temperature between 600–1000 °C (Speight, 2020). Biomass gasification is a promising option that can contribute to building a hydrogen economy but require improvements to produce larger competitive volumes (Lepage et al., 2021).

B. Coal

The conversion of solid coal into a gaseous phase creates opportunities to produce more energy forms than electricity (which is the case in coal combustion systems). Furthermore, it provides the possibility to convert solid fuels into gas with an easier separation of by-products (i.e., CO₂, CO...) (Gnanapragasam et al., 2010). WGS reaction is commercially available for this process, which is generally coupled with membrane separators for producing and separating hydrogen, respectively. Although the main by-product is CO₂, other substances could be present, depending on the upstream processes (Gnanapragasam et al., 2010).

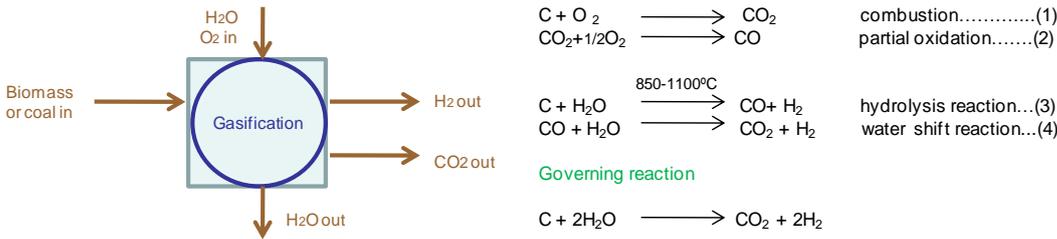


Figure 2.4 Gasification block diagram and governing reaction (de Leon Almaraz, 2014).

2.2.1.3 Water electrolysis

The electrolyzer is the heart of the power-to-gas system. The surplus of electricity from renewable energy sources is used to produce hydrogen via electrolysis and it is the first step of the PtG process Eq. 2.2.



This is required to be efficient, flexible (range of operation and response time) and with a long service life (Götz et al., 2016). Its operation depends on several parameters: temperature, pressure, the electrical resistance of the electrode (conductivity), electrolyte quality (presence of impurities), electrolyte concentration (viscosity), electrolyte circulation, electrode materials (electrical conductivity, chemical resistance), distance between the electrodes, dimensions, and alignment of the electrodes, gas bubbles on the surface of the electrodes, material of the membranes, type of current, among others (Boudellal, 2016). Therefore, its performance significantly affects the overall performance of the chain (Götz et al., 2016). Three concepts of electrolyzer could be used in the short and medium-term commercially production of hydrogen: (1), Alkaline Electrolysis (AEL), (2) Proton Exchange Membrane Electrolysis Cells (PEM), and (3) Solid Oxide Electrolysis Cells (SOEC). Their main characteristics are summarized in Table 2.1. The size of a cell is limited by the capacity of the membrane or the diaphragm to withstand the electric current. Electrolysis cells are therefore piled in stacks that compose the core of an electrolyzer. In addition, an electrolyzer comprises auxiliaries such as a current rectifier, a water demineralization unit, a water pump and a cooling system, a hydrogen purification unit, and instrumentation.

A. Alkaline electrolysis

Alkaline electrolysis is the state-of-the-art technology developed for water electrolysis, and currently the only commercially available power-to-gas application. The main reactions related to this process are shown in Figure 2.5. This is a well-known technology that has been available for decades, which is an advantage for PtG (Kreuter and Hofmann, 1998). It uses an alkaline solution (i.e. sodium hydroxide or potassium hydroxide) as electrolyte material to transfer electrons through hydroxide anions. Depending on the capacity of the electrolyzer and the pressure of the hydrogen delivered, the energy efficiency of the devices varies between 66% and 74% (4.8 - 5.4 kWh_{el}/Nm³H₂). This type of electrolyzers has an operational range between 20 and 100% of the design capacity, and overload operation up to 150% is possible (Götz et al., 2016).

Yet, the electrolytes used are highly corrosive, which implies high maintenance costs and the need to revamp the system every 7 to 12 years. Besides, this type of electrolyzers could be slow due to the inertia of the system, taking between 30 to 60 minutes to restart it following a shutdown. Regarding the intermittent nature of renewable energies, this could be an issue as a rapid response is searched (Götz et al., 2016).

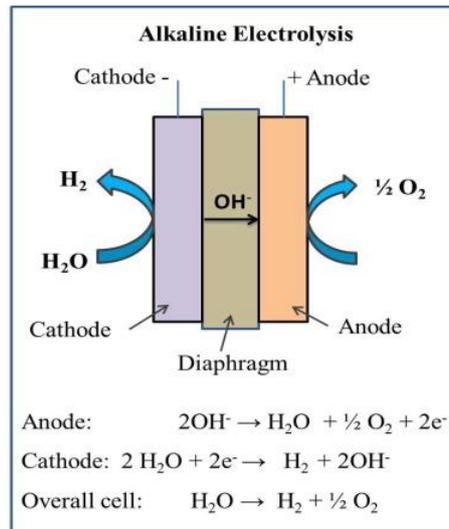


Figure 2.5. Schematic illustration of alkaline water electrolysis (Shiva Kumar and Himabindu, 2019)

B. PEM electrolysis

PEM is a new technology compared to AEL, still in the demonstration phase with only a few experiences at the commercial level (i.e., SILYZER 200) (Hydrogen Council and Hydrogen Council EU, 2017). Figure 2.6 illustrates the main reaction in this electrolysis concept. This technology is based on polymer membranes (Carmo et al., 2013). The efficiency of this technology is similar to AEL technology. The advantages of this type of electrolyzers in PtG systems are very promising: hydrogen with high purity (>99.998%) (Hydrogenics, n.d.), minimal part load (5-10%, compared to 20-40% for alkaline technology), flexibility, and better coupling with intermittent systems thanks to a faster cold start (Götz et al., 2016). However, their life expectancy is limited, mostly because of the membrane that has a fast degradation. Besides, this technology is currently only available on small scales and more expensive than AEL systems (due to the costs for the membrane and the use of a noble metal catalyst) (Gazey et al., 2006; Kreuter and Hofmann, 1998). It is expected to drop to the same level that AEL in the future (up to 500€/kW_{el}) once the production and technology will be fully developed. The major investment cost is related to the membrane, and the use of noble metals, but also the high applied overvoltage (~2 V), especially at high current densities.

Table 2.1. Summary of the key operational parameters of Alkaline, PEM, and Solid Oxide Electrolysis

	AEL	PEM	SOEC
State of development	Commercial (Kreuter and Hofmann, 1998)	Commercial	Laboratory (Carmo et al., 2013)
H₂ production in m³/h (STP, per system)	<760 (Carmo et al., 2013) ≈ 2.7 MW	Up to ≈450 ≈ 1.6 MW	----
Electrolyte	Alkaline solution	Solid polymer membrane (Nafion)	ZrO ₂ ceramic doped with Y ₂ O ₃
Charge carrier	OH-	H ₃ O ⁺ /H ⁺	O ²⁻
Cell temperature in °C	40-90 (Carmo et al., 2013)	40-90 (Carmo et al., 2013)	800-1000 (Laguna-Bercero, 2012)
Cell voltage in V	1.8-2.4 4.5-7	1.8-2.2 4.5-7.5	0.91-1.3
System power consumption (current) in kWh/m³ H₂ (Hs)	4.7-5.4 (Müller-Syring et al., 2013) 5.4-8.2 (Zakeri and Syri, 2015)	5.2-7.1 (Müller-Syring et al., 2013) 4.9-5.2 (Zakeri and Syri, 2015)	----
System power consumption (future) in kWh/m³ H₂ (Hs)	4.3-5.7	4.1-4.8	-----
Cold start time	Minutes-hours (Müller-Syring et al., 2013)(Gahleitner, 2013)	Seconds-minutes (Müller-Syring et al., 2013)	----
Advantages	Available for large plant sizes, cost, lifetime (Ursua et al., 2012)(Bodner et al., 2015)	No corrosive substances, high power densities, high pressure > 100 bar, dynamics (Carmo et al., 2013)(Müller-Syring et al., 2013)	High electrical efficiency, integration of waste heat possible (Reytier et al., n.d.)(Doenitz et al., 1980)
Disadvantages	Low current density, maintenance costs (the system is highly corrosive) (Müller-Syring et al., 2013)	Expensive, fast degradation (Carmo et al., 2013)(Gazey et al., 2006)(Müller-Syring et al., 2013)	Limited long term stability of the cells (Ursua et al., 2012), not suited to fluctuating systems, expensive (Reytier et al., n.d.),
Transient operation	Possible, but leads to problems (Gahleitner, 2013); reduction up to 20% load possible; overload operation possible (Müller-Syring et al., 2013)	Better than AEL (Gahleitner, 2013), dynamic adjustment possible, partial (down to 5%), and overload operation possible (Ulleberg et al., 2010)(Müller-Syring et al., 2013)	Not well suited
Renovations/lifetime (years)	Renovations stack: 8-12 Lifetime: up to 30	Lifetime: 5 (Gahleitner, 2013)	-----

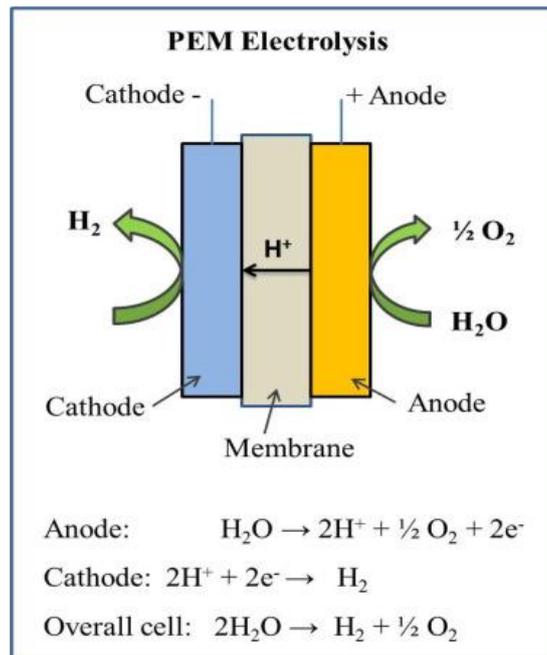
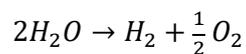
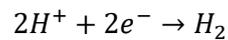
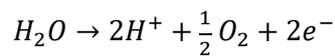


Figure 2.6. Schematic illustration of PEM water electrolysis (Shiva Kumar and Himabindu, 2019)



C. SOEC electrolysis

Solid Oxide Electrolyzer Cells (or high-temperature electrolysis) is the least mature process. The main reactions involved are shown in Figure 2.7. The main interests of the SOEC technology are its higher efficiency and the possible use as a fuel cell in a “reverse” mode. Besides, the low electricity demand is the most significant advantage of SOEC systems. The coupling of a SOEC electrolyzer with an exothermic reactor such as methanation allows heat recovery on the reactor to produce steam for the electrolysis stack, but a constant operation is preferable since ceramics are sensitive to thermal stress so this issue should be widely studied in order to be used in PtG systems (E&E consultant et al., 2014). In particular, the combination with exothermal reactions in PtG and PtL process chains leads to higher overall efficiency (Götz et al., 2016). One of the drawbacks of the system is that they are not stable against fluctuating and intermittent power sources (Götz et al., 2016; Ursua et al., 2012).

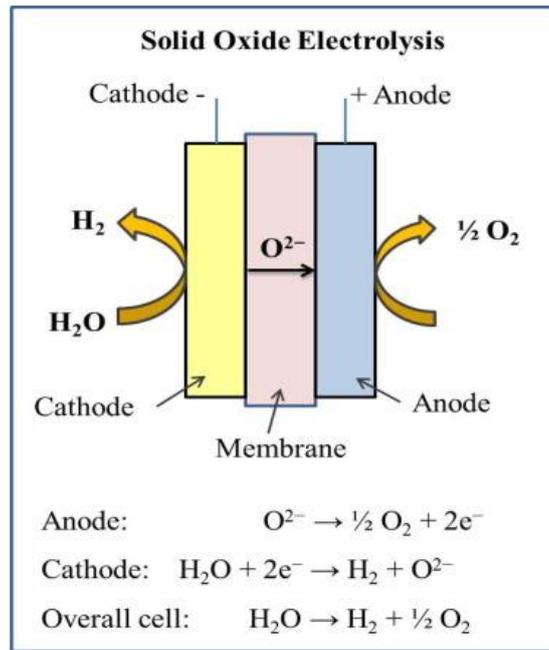


Figure 2.7. Schematic illustration of Solid Oxide electrolysis (Shiva Kumar and Himabindu, 2019).

2.2.2 Methanation for Power-to-Gas

The power-to-gas system offers a second possibility for the hydrogen produced by PtG systems. It can be transformed into methane by using CO₂, through a process of methanation. Paul Sabatier (1854-1941) and Jean-Baptiste Senderens (1856-1937) discovered the methanation reactions in 1902 (Rönsch et al., 2016). The main reaction is shown in Eq. 2.3.



To obtain the best compromise between efficiency and CH₄ obtained, the amount of H₂ transformed varies between 90% and 100%. However, the presence of H₂ in the final product must be adapted to the regulations of each region (E&E consultant et al., 2014).

2.2.2.1 Methanation concepts

The two most promising technological paths at this time are catalytic and biological methanation. The main features of both concepts are indicated in Table 2.1.

2.2.2.1 Catalytic methanation

Catalytic methanation is a thermochemical process operated generally at high temperatures (between 200 and 700 °C) and pressures between 1 and 100 bar (ENEA CONSULTING, 2016; Götz et al., 2016). The principle of the methanation reaction and the typical methanation process chain is shown in Figure 2.8 and Figure 2.9. This is the technology that may be available on the market in the short term because it has been studied for several decades (Götz et al., 2016; Götz and Graf, 2015). The maximum efficiency of the reaction is approximately 80%, without taking into account a later use of waste heat (Schiebahn et al., 2015).

Table 2.2 Summary of the key operational parameters of chemical and biological methanation process

Parameter	Methanation concept					
	Chemical			Biological		
Reactor concept	Fixed-bed	Three-phase methanation (3PH)	Fluidized-bed (FB)	Continuous stirred-tank reactors (CSTR)	Trickle-bed	Fixed-bed
Operation mode	Adiabatic (Rönsch et al., 2016)	Isothermal (Rönsch et al., 2016)		Isothermal (Rönsch et al., 2016)		
Temperature (°C)	200 - 750 (Grond et al., 2013; Kopyscinski, 2010; Sterner, 2009)	300-350 (Götz et al., 2016)	40-700 (Lehner et al., 2014)	20-70 (Götz et al., 2016)		
Pressure (bar)	5-100 (Götz et al., 2016)	<10 (ENEA Consulting, 2016) ≈20 (3PM) (Götz et al., 2016) 2-86 (Lehner et al., 2014)		1 – 10 (Götz et al., 2016)	1 (Burkhardt and Busch, 2013)	1 (Jee et al., 1988)
Maximum production capacity (MW-CH ₄)	<500 (Sudiro and Bertucco, 2010) 1-10 (Suitable capacity for isothermal methanation) (ENEA Consulting, 2016)			≈15 (Electrochaea, 2012; Krassowski, 2012)		
Efficiency (%)	70-85 (Sterner, 2009)(Kopyscinski, 2010)			< 85 (Martin et al., 2013; Nishimura et al., 1992; Seifert et al., 2014)	98 (Burkhardt and Busch, 2013)	34 (Jee et al., 1988)
Heat utilization	Very good (Götz et al., 2016)	Good – Very good (Götz et al., 2016)		Poor (Götz et al., 2016)		
Thermal Stress	High (Rönsch et al., 2016)	Low (Rönsch et al., 2016)		Very low		
Maturity	Commercial (Grond et al., 2013; Rönsch et al., 2016)	Lab scale (Götz et al., 2016)	Pre-commercial (demonstration) stage / Lab scale (Götz et al., 2016)(ENEA Consulting, 2016)	Pre-commercial (demonstration) stage / Lab scale (Grond et al., 2013)	-----	
Process Complexity	High (Rönsch et al., 2016)	Low (Rönsch et al., 2016)	Low (Rönsch et al., 2016)	Low	-----	
Scale	Mid- to large-scale applications (Götz et al., 2016; SBC Energy Institute, 2014)			Small-scale applications (Götz et al., 2016; SBC Energy Institute, 2014)		
Advantages (E&E consultant et al., 2014)	<ul style="list-style-type: none"> - Available commercially and with more than 60 years of development - Recovery of heat at high temperature 			<ul style="list-style-type: none"> - Insensitive to the trace of sulfur - Simple operating principle - Catalyst not required 		
Inconvenient (E&E consultant et al., 2014)	<ul style="list-style-type: none"> - Sensitive to impurities - Difficulties in adapting to the variability of the hydrogen source - High costs and complex operation 			<ul style="list-style-type: none"> - Difficulties for heat recovery - It has not yet been applied at the industrial level 		

A disadvantage of catalytic reactors is the requirement of a high purity in the supplied gas, which then involves additional maintenance and investment costs. In addition, the reaction occurs at high temperatures, which results in greater complexity for its operation and higher costs. This is particularly challenging for adiabatic reactors so that they must be designed as large-scale systems.

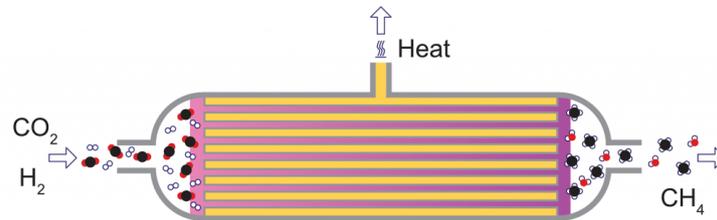


Figure 2.8. Principle of the methanation reaction (Energo, n.d.)

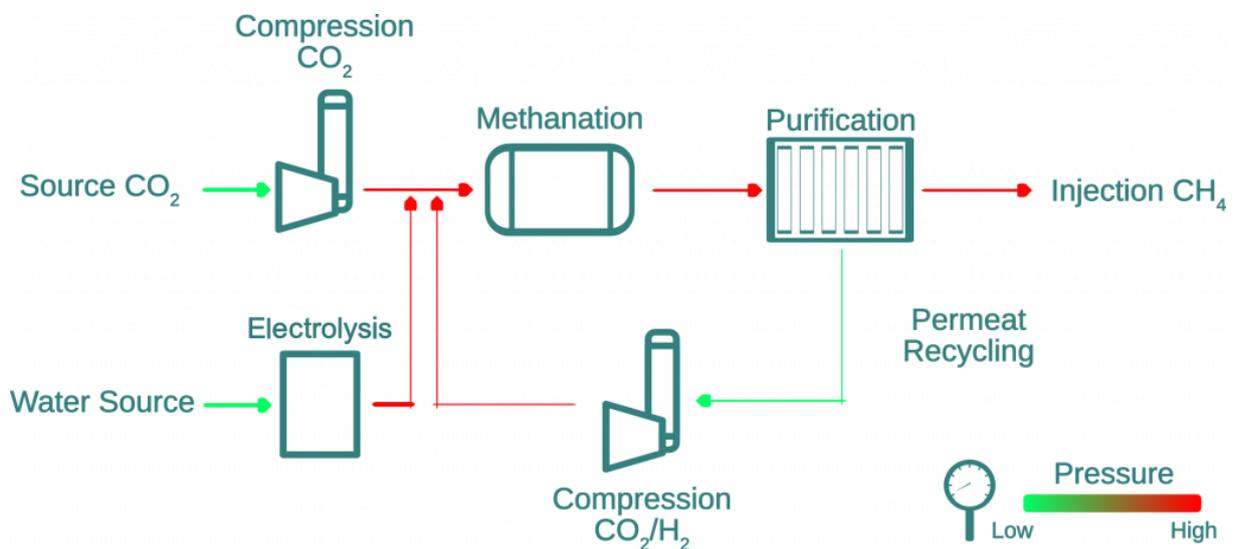


Figure 2.9. Typical methanation process chain (Energo, n.d.)

2.2.2.2 Biological methanation

Biological methanation (BM) produces methane from hydrogen and carbon dioxide using methanogenic microorganisms operating as bio-catalysts. The reaction occurs under anaerobic conditions in an aqueous solution, at atmospheric pressure or under low pressure, between 20 and 70 °C (Götz et al., 2016). In contrast to catalytic methanation, BM has not yet reached a level of commercial maturity. BM has a lower reaction rate due to a lower temperature and a lower volumetric mass transfer coefficient and a high tolerance towards impurities of the input gas (Seifert et al., 2013).

The set of advantages of this concept of methanation is increased when combined with biogas processes: CO₂, which is normally considered as a waste in the production of biomethane (Kougiyas et al., 2017), can be converted into methane by injecting hydrogen into the process. Thus, BM can decrease the production costs of biomethane and improve the properties of the final product. Moreover, this process can also improve the yield concerning the amount of biomass used, thus requiring fewer resources (Lecker et al., 2017).

According to (Lecker et al., 2017), two technical pathways can be used for BM applications. One way is the direct injection of H_2 into an anaerobic digester and therefore the utilization of internally produced CO_2 – the in-situ methanation (Guebitz et al., 2015). The other possibility is the parallel injection of H_2 and CO_2 in a stoichiometric ratio of 4:1 into a reactor – the so-called ex-situ methanation (see Figure 2.10). Despite the advantages offered by these methanisation concepts, some issues still need to be addressed, such as the variability of the yield according to the biomass treated (i.e., cultivated energy crops, sewage sludge...).

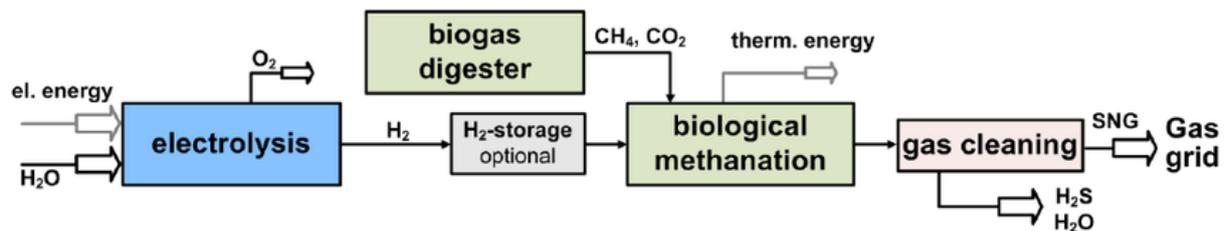


Figure 2.10. Process flow diagram for biological methanation with a separate reactor (Götz et al., 2014)

2.2.3 Storage and transport infrastructure

After being produced, hydrogen has to be transported and properly stored if needed. In the case of methane, it can be injected directly into the natural gas network. According to (Yang and Ogden, 2007), the selection of the infrastructure system depends on the scale and geographic constraints that will also determine the final cost of hydrogen production.

2.2.3.1 Compressed gas storage

Compressed gas storage of hydrogen is the simplest storage solution. However, the main problem with this type of technology is the low storage density, which depends on the storage pressure. Higher storage pressures will necessarily result in higher capital and operating costs (Amos, 1999). The largest operating cost with above-ground gas storage stems from the energy needed for compression itself. The energy requirements would depend on the final pressure. However, a high final storage pressure requires minimal power compared to the initial compression of the gas. The coupling with electrolyzers that produce hydrogen at elevated pressure is a suitable option because the gap with the final storage pressure is reduced and, consequently, the power requirements are lower (E&E consultant et al., 2014).

2.2.3.2 Liquefaction and liquid storage

Liquefaction consists of cooling gas to form a liquid. A typical example is shown in Figure 2.11. This procedure is achieved using a combination of compressors, heat exchangers, expansion engines, and throttle valves to obtain the desired cooling. The most simple liquefaction process is described by the Joule-Thomson or Linde-Hampson expansion cycle (Waele, 2017), in which the gas is compressed at ambient pressure, then cooled in a heat exchanger before passing through a throttle valve where it is carried out an isenthalpic Joule-Thompson expansion producing a liquid.

One of the inconveniences when working with hydrogen is that it warms upon expansion at ambient temperature (contrary to other gases as nitrogen that cools upon expansion) adding complexity to the process as temperature should be below 202 K (inversion temperature) to allow a cooling upon expansion. To make this, liquefaction processes use liquid nitrogen to pre-cool hydrogen. Another major concern in hydrogen liquid storage is minimizing H₂ losses from liquid boil-off¹. Any evaporation will result in a loss of system efficiency, so a very important step to prevent it is to use insulated cryogenic containers specially shaped (normally spherical) to diminish the surface area for heat transfer per volume. All of these characteristics make liquid hydrogen storage a technologically complex issue (De-León Almaraz, 2014; E&E consultant et al., 2014).

The associated costs of liquefaction are higher than those of compression. The highest operating cost for liquefaction is electricity, but small amounts of nitrogen and cooling water are also needed. However, liquid hydrogen storage is foreseen for a scenario of high demand and significant penetration of renewable energy into the grid (De-León Almaraz, 2014).



Figure 2.11. Example of liquid hydrogen tank for space applications (NASA, 2015)

2.2.3.3 Pipeline storage and transport

Blending hydrogen into natural gas is limited, since the natural gas grid has not been designed to withstand the specific properties of hydrogen such as higher permeation and corrosion than natural gas. Limits on hydrogen blending are shown Figure 2.12. It is between 0 and 12% in the EU countries and is expected to increase in the next years until possibly 15 or 20% (Blanco and Faaij, 2018; E&E consultant et al., 2014). Besides, the methanation step produces synthetic methane that is 100% mixable with natural gas, giving the possibility to take advantage of the full capacity of the network without any major modifications. The addition of PtG plants to this system implies the inclusion of injection stations where the gas must be compressed to 40-60 bars in the transmission grid, or to 5 to 10 bars for the distribution grid (ENEA CONSULTING, 2016).

¹ Because liquid hydrogen is stored as a cryogenic liquid that is at its boiling point, any heat transfer to the liquid causes some hydrogen to evaporate. The source of this heat can be, for example, molecular conversion, mixing or pumping energy, radiant, convection, or conducting heating.

For large quantities of hydrogen, pipelines are the cheapest means of transporting hydrogen with high demand. The liquid hydrogen option is the second cheapest means (Penev et al., 2019). The major advantage is that operating costs for pipelines are related to compressor power and maintenance. In addition, storing hydrogen in a pipeline system requires no additional capital expense as the network is already installed and in use. The break-even point between liquid hydrogen and pipeline will vary depending on the distance and quantity of gas being transported (Amos, 1999).

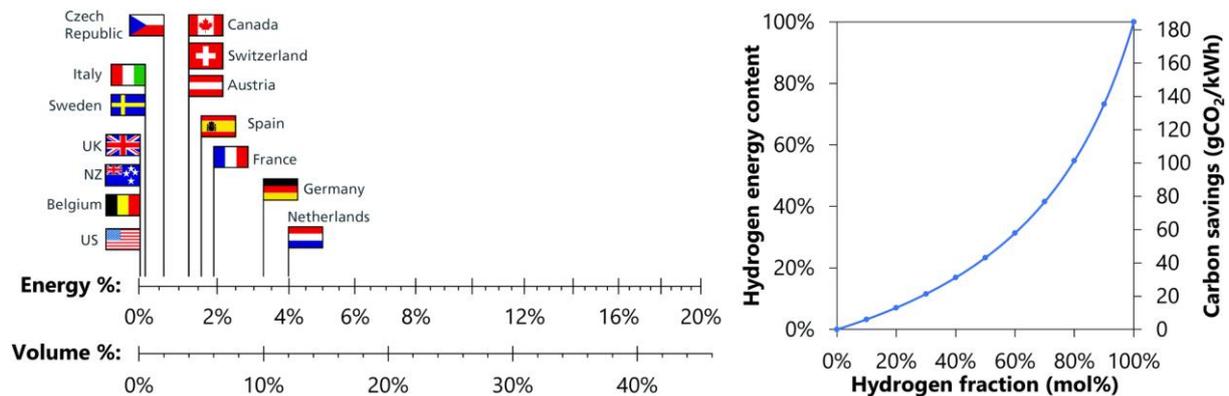


Figure 2.12. Limits on hydrogen blending into national gas grids around the world, and the relationship between energy content, carbon savings, and hydrogen injection mixtures (right) (Staffell et al., 2019).

2.2.3.4 Tube trailer

According to (Reddi et al., 2018), a tube trailer consists of pressure vessels designed to store hydrogen at rated pressure, packaged in a container, and mounted on a trailer to transport the compressed hydrogen gas. The tube trailer can be defined by parameters such as the material used to construct the pressure vessels, the size of the vessels, and how the vessels are installed/packaged on the trailer. This well-established technology (de Leon Almaraz, 2014) could be helpful in the early phases of FCEV introduction, where the infrastructure would have a low-capacity utilization (Reddi et al., 2018). In this regard, this transportation mode is not considered as a long-term delivery solution because they require numerous low-capacity trips to meet the hydrogen demand (Yang and Ogden, 2013).

2.2.3.5 Tanker trucks

Tanker-trucks could be used to transport hydrogen in liquid form. Due to this physical form, they involve a high operating cost due to the electricity needed for liquefaction, but lower capital costs depending on the quantity of hydrogen and the delivery distance (Ball and Wietschel, 2009; Dagdougui et al., 2018). It is an interesting option for medium market penetration (Dagdougui, 2011), since tanker-trucks are more economic than tube trailers: tanker-trucks can hold more hydrogen. According to (Dagdougui et al., 2018), a road tanker that transports high-pressure hydrogen as compressed gas might typically carry 300–400 kg of H₂ and be able to refuel up to about 100 cars. A tanker carrying liquefied hydrogen (LH₂) can carry a much larger inventory, 2.5 to 3.5 tons, and so refuel about 1000 cars (Melaina et al., 2013). Both options, tube trailers, and tanker trucks, are shown in Figure 2.13.

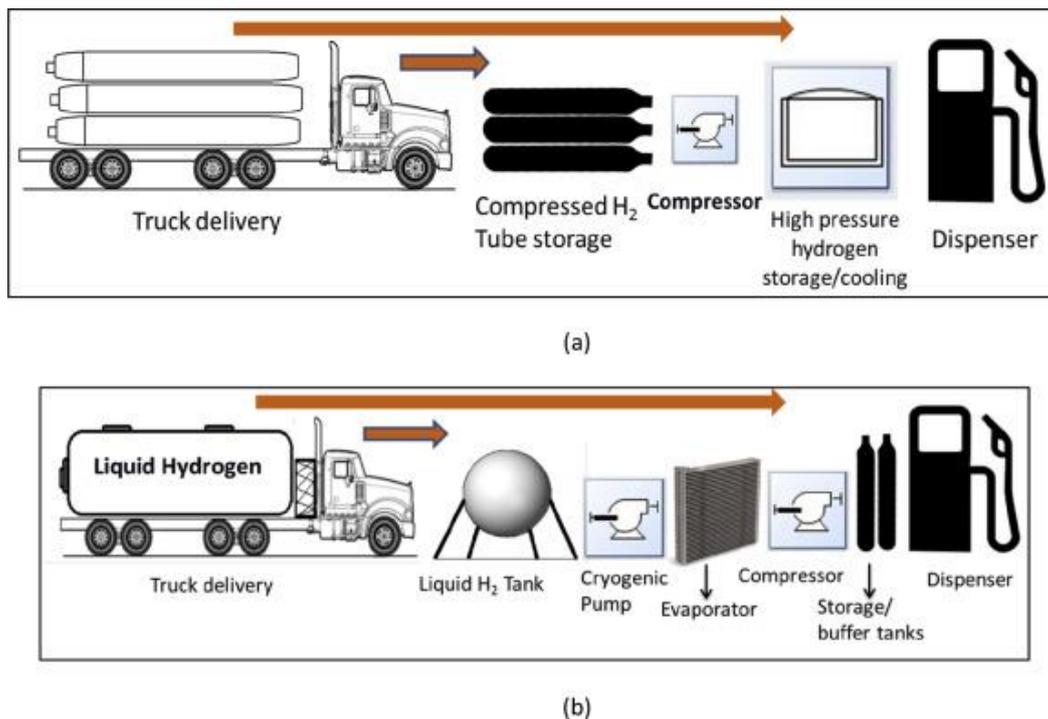


Figure 2.13. Hydrogen transportation options for refueling stations: (a) gaseous hydrogen delivery and (b) liquid hydrogen delivery (Mayyas and Mann, 2019).

2.2.4 End-uses

Several end-uses can be identified for hydrogen and methane, such as industrial uses, electricity production, transport, and buildings. In the case of hydrogen, the transport sector has been identified as one of the most challenging sectors for the reduction of greenhouse gases. Hydrogen appears as one of the possible alternatives to satisfy this demand, since fuel cell electric vehicles (FCEVs) offer high efficiency, high power density, and cold-start capabilities (Pollet et al., 2012; Staffell et al., 2019). The main advantages of FCEV over other alternatives such as battery electric vehicle (BEV) is its autonomy, refueling speed, and lifetime (Staffell et al., 2019).

In the case of synthetic methane, a wide range of uses are identified because no major modifications to current systems are required for its use (Götz et al., 2016), including the residential, industry, transport, and service sectors. To meet these demands, it can be injected directly into the existing natural gas network.

2.2.4.1 Refueling station for hydrogen

Refueling stations represent an end-use for hydrogen and therefore the completion of the hydrogen supply chain in the case of the transport sector. There are two basic types of hydrogen refueling stations: a) Stations where hydrogen is produced off-site and transported to the station; b) stations where hydrogen is produced on-site (Li et al., 2019). The final configuration will then depend on the chosen scheme (see Figure 2.14). Hydrogen can be supplied in liquid or gaseous form. Its establishment does not represent important technological challenges (de Leon Almaraz, 2014) and, in 2019, there were

already around 434 stations in the world, which the majority have been built in Asia and EU (LBST, n.d.).

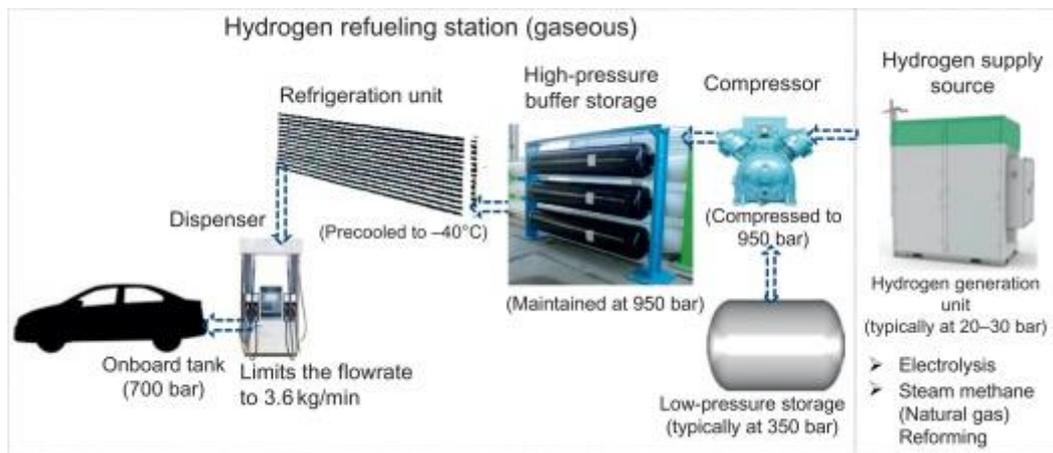


Figure 2.14. Layout of the components of a hydrogen refueling station supplied by an on-site production unit (Reddi et al., 2016).

2.2.5 Feedstock

2.2.5.1 Energy sources

The production of hydrogen and methane from PtG systems requires hydrogen for the production of hydrogen, and then hydrogen and carbon dioxide for the production of methane. However, hydrogen can also be produced through the gasification of coal or biomass, or by SMR using natural gas. A color code nomenclature is becoming commonly used to facilitate discussion: hydrogen produced through SMR / coal gasification (fossil energy sources), gasification or SMR using carbon capture, and electrolysis using renewable electricity sources, is called "grey hydrogen," "blue hydrogen," and "green hydrogen," respectively (IRENA, 2020) (see Figure 2.15).

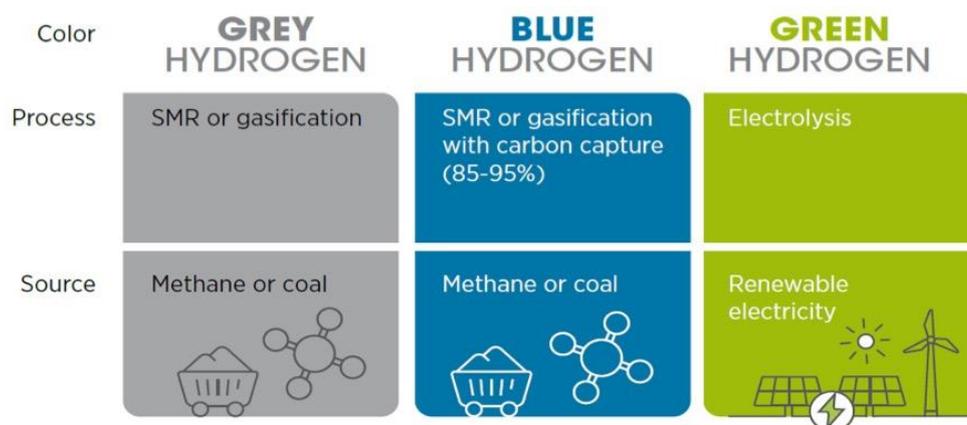


Figure 2.15. Main shades of hydrogen (IRENA, 2020)

2.2.5.2 CO₂ sources

The second raw material for methane production through methanization is carbon dioxide. The CO₂ sources can come from different processes. Four sources can be identified: power plants, industrial

processes, biomass transformation, and direct capture from the air. Ideally, CO₂ should be captured at a low cost and using low energy-intensive processes.

A. CO₂ from industrial plants

Carbon dioxide capture from fossil power plants has been extensively investigated in the context of carbon capture and storage (CCS) (Cuéllar-Franca and Azapagic, 2015). It could be applied for separating CO₂ from the flue gas in industrial or power plants. The most important stationary sources are the steel and cement industry, and power plants using fossil fuels.

For the interest of PtG, it is important to consider that even if the technology is available and the amounts of CO₂ are considerable (or even excessive), the product needs to be transported from the power plant to the methanation plant. Also, the CO₂ is not biogenic, and this should be kept in mind if the objective is to have a renewable PtG network (E&E consultant et al., 2014). Nevertheless, since these emissions in many cases could not be avoided, this could be an interesting solution to improve the industry's carbon footprint (E&E consultant et al., 2014; Koytsoumpa et al., 2018).

B. CO₂ from biomass transformation

Biogas plants can also be used as CO₂ sources. A typical biogas composition is 60% methane (i.e., biomethane) and 40% carbon dioxide (Ademe et al., 2010). Different origins for biomass could be used to produce it, such as municipal solid waste and industrial waste, wastewater treatment sludge, agricultural residues, and energy crops (Sindhu et al., 2019). The upgrading step is essential to remove impurities and undesirable components from biogas, i.e. CO₂, hydrogen sulfides, and water. Specifically considering PtG systems, after removing the trace components, biogas can be directly injected into the methanation reactor improving the efficiency of the process (Collet et al., 2017), as shown in Figure 2.16.

C. CO₂ from air

CCS from the air is a challenging process, due to the concentration of CO₂ in the atmosphere (around 400 ppm). The energy effort needed is about 500-800kJ of primary energy per mole of CO₂ (3000-5000kW_{el}/tCO₂). Despite these issues, there are no restrictions of spatial availability, facilitating the supply chain (E&E consultant et al., 2014).

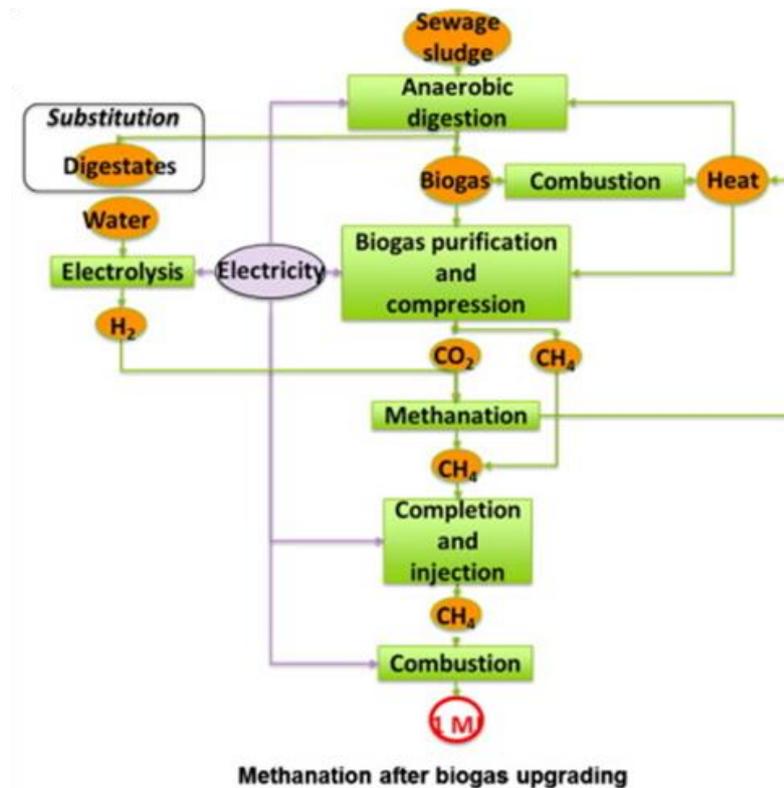


Figure 2.16. Biomethane production with CH_4 synthesis via methanation (Collet et al., 2017)

2.2.6 Conclusions

This assessment on the main echelons of HMSC leads us to conclude that:

- A range of technologies exists for the production of renewable hydrogen and methane, presenting different levels of maturity, advantages, and disadvantages. The production capacity will depend on the technology chosen and production will have to meet a local and/or centralized demand.
- Given the different Technological Readiness Levels (TRL) of hydrogen production technologies (particularly SMC versus electrolysis), a combination of both technologies could be useful to develop the hydrogen market, particularly for its first phase of development. Concerning the production of hydrogen through electrolysis, AEL technology presents the best competitive advantages at present (lower cost and higher maturity- TRL). However, other options such as PEM and SOEC could also play an important role in a hydrogen economy, due to the decrease in costs and higher performance.
- In the case of renewable methane, the addition of methanation as an additional process implies an increase in costs. However, in a scenario of high production of renewable energy and ambitious targets for CO_2 reduction, this solution is interesting to achieve higher levels of decarbonization and flexibility in the energy network. Two technological paths (catalytic and biological) were identified for its implementation. The catalytic route presents a higher level of

maturity, and some pilots demonstrate its viability. However, biological methanation could be interesting if the cost reductions suggested by the experimental scale studies are achieved.

- Hydrogen storage and transport have advantages and disadvantages, which will depend essentially on the penetration of hydrogen in the market and the case study analyzed.
- The variety of raw materials is an important element to consider since it allows the production of hydrogen through different processes. Besides, there are aspects to be addressed as the availability of energy in the territory and its import. The differentiation of these sources (fossil and renewable) is then a key aspect of the design of the HMSC.
- For the production of renewable methane through Power-to-Gas, an external source of CO₂ is required. The main sources can be classified as biogenic, industrial, and directly from the air. The biogenic source offers the best option because it comes from renewable sources and generally presents a higher concentration of CO₂. An industrial source is a second option, which generally includes additional separation costs given the set of gases involved. Obtaining CO₂ from the air has the worst competitive advantage due to the low concentration of CO₂, therefore this option was not considered.
- A set of end uses can be identified (mobility, electricity, buildings, industry...). In the case of hydrogen, the transport sector offers significant competitive advantages over other options such as battery electric vehicles (Staffell et al., 2019). Renewable methane has even more diverse uses due to the reduced adaptation required for its use. Its final use will be diverse because it can be injected directly into the gas network.

This discussion leads to the following selection of technologies, raw materials, and end-uses for HMSC. (see Figure 2.17). In this regard, it can be highlighted that:

- The main technological options, feedstock, and end uses are selected for the application of the PtG concept.
- The liquid physical form of hydrogen has been selected.
- SMR is a mature technology available for large-scale low-cost hydrogen production today and its interaction with PtG systems needs to be studied to facilitate the transition towards the penetration of more renewables. In this sense, fossil and renewable (through electrolysis) energy sources will be involved.
- The HMSC design and deployment will be made at a strategic level.

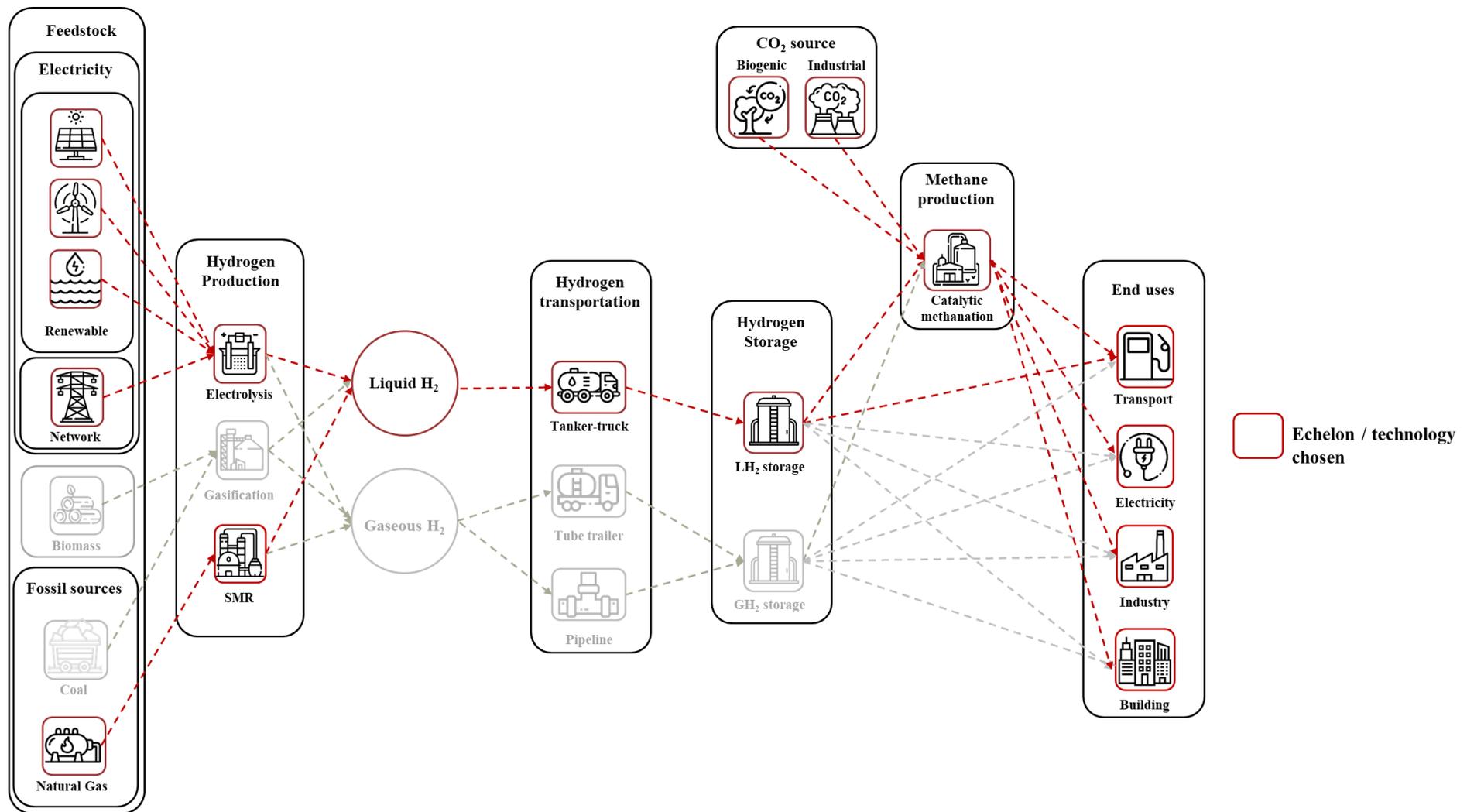


Figure 2.17. Selected technologies and echelons for HMSC modeling

This preselection for the technological components can be summarized in Table 2.3. Each technology associated with its attributes (input and output types and flows; technological parameters, capacity, costs (CAPEX and OPEX), and environmental performances (GHG emissions)) will be the key elements for the energy system modeling and optimization that is the core of this investigation. A first taxonomy of the optimization-based HMSC models is then proposed to develop a technically oriented approach considering the integration of these technologies into a territory, considering the temporal availability and evolution of technologies and sources in conjunction with demand variation over time along the deployment periods.

Table 2.3. Selected technologies for HMSC modeling

Technology	Input	Output
SMR	Natural gas	Liquid H ₂
Electrolysis	Electricity (PV, wind, hydro and network)	Liquid H ₂
Tanker-truck	Liquid H ₂	Liquid H ₂
Liquid H ₂ Storage	Liquid H ₂	Liquid H ₂
Catalytic methanation	H ₂ and CO ₂	Methane

2.3 A first taxonomy of optimization-based HMSC models

Two contrasting energy modeling types have been developed, in order to solve the problem of fulfilling energy demand: the bottom-up engineering approach and the top-down macroeconomic approach (Wene, 1996).

1. The engineering approach is to develop bottom-up models with specific emphasis on **technical aspects of the energy system** and its future development. Energy demand is typically provided exogenously, and the models analyze how the given energy demand should be fulfilled generally using mono-objective optimization based on a cost criterion.
2. The economic approach is to build top-down models that describe the whole economy, with a specific focus on the potential substitution of different production factors to optimize social welfare. These models **are generally not technically-oriented**. The interplay between energy and other production factors to create economic growth is captured in production functions, and opportunities to make changes in fuel mixes are described by elasticities of substitution. Another key parameter is the autonomous energy efficiency improvement, which allows the improvement in production due to assumed technical improvements (Murphy et al., 2007).

The two approaches differ in their identification of the relevant system, and may therefore produce different guidance for policy-makers. This work is based on a bottom-up approach. According to (Agnolucci and Mcdowall, 2013; Li et al., 2019), three categories for HMSC can be listed regarding the objective that is targeted:

2.3.1 *Energy system optimization models (ESOM)*

Generally formulated as an LP or MILP problem, the objective of such models is to find the energy system that satisfies the entire demand for services at the minimum cost. Other objectives may also be included, such as CO₂ emissions and other environmental impact indicators, oil imports, or a combination of them. Typically, the entire supply chain (SC) is addressed, along with various technologies and distribution modes. Technologies at the end of the chain (i.e., natural gas vehicles, electric vehicles, FCEV...) compete with each other to satisfy a requested service (i.e., mobility needs), thus both the demand and supply of energy carriers (i.e., electricity, methane, hydrogen...) are optimized (Agnolucci and Mcdowall, 2013; Li et al., 2019). This represents the main advantage of using these types of models. One of the most common frameworks is the MARKAL LP model (Rath-Nagel and Stocks, 1982), developed in the 1970s, and its evolution, the Integrated MARKAL-EFOM System (TIMES) (Loulou and Goldstein, 2005; Loulou and Labriet, 2008). They have been applied at different geographical levels (global, regional, and national). In Europe, many countries have developed their own national TIMES model (Pfenninger et al., 2014). Both models have been used to address the HSC and MSC in different countries and regions (Agnolucci and Mcdowall, 2013; Blanco et al., 2018a, 2018b; Li et al., 2019). In addition to the models mentioned above, there is a range of open-source or commercial multi-energy modeling tools that can be used for HMSC optimization, based on LP, dynamic programming (DP), and MILP formulations (Klemm and Vennemann, 2021). Similar models have been also employed as an approach to HSC and MSC modeling, such as operational models based on MILP (Vandewalle et al., 2015), and economic dispatch models (He et al., 2018; Wang et al., 2019).

2.3.2 *Geographically explicit optimization models (GEOM)*

These models focus on infrastructure deployment, considering the entire SC. They are usually taken as case studies at regional and national scales. They differ from ESOMs since they can take into account continuous, binary, and integer variables to address location, size, and number of units to be installed, flow direction, selection of production technologies, storage, and transport modes (Eskandarpour et al., 2015; Li et al., 2019). Therefore, GEOMs are generally based on MILP formulations.

Two reference models have been identified for HSC. (Almansoori and Shah, 2006) developed a pioneering model, taking Great Britain as a case study. It is a demand-driven, steady-state model that integrates the main stages of HSC: production, storage, and transport. This model was then improved by taking into account aspects such as resource availability, demand variation over time, demand uncertainty, environmental impacts, and the progressive deployment of the chain by periods (Dayhim et al., 2014; De-León Almaraz et al., 2015; Han et al., 2013; Moreno-Benito et al., 2017; Nunes et al., 2015; Sabio et al., 2012). The work developed in (Samsatli and Samsatli, 2018) proposes a MILP model that can simultaneously determine the design and operation of any integrated multi-vector energy networks comprising technologies for conversion, storage, and transport. The model takes into account the spatial distribution and temporal variability of system properties (such as demands and availability of resources) and determines the spatial structure of the integrated networks. It embeds a multi-objective

optimization problem, which comprises the trade-offs between minimization of cost, maximization of profit, minimization of CO₂ emissions, maximization of energy production, or different combinations of these, considering Great Britain as the region of interest. Some works propose the extension of the hydrogen SC to other energy vectors such as methanol and ammonia. For example, the study conducted in (Ogumerem et al., 2019) shows that the use of these two carriers could be economically viable and complementary to hydrogen, due to a higher energy density per volume.

For methane supply chains, two studies are particularly interesting. The work developed in (Moeller et al., 2014) involves an NLP formulation to analyze feasible scenarios with a focus on the 2020s and 2030s in the Berlin-Brandenburg region, Germany. They were numerically evaluated in hourly time steps using a cost optimization approach. In (Clegg and Mancarella, 2015), a dispatch model is used to focus on the integration of electricity and gas networks, and the impact of PtG systems applied to Great Britain as a case study. Technical, environmental, and economic operational aspects of PtG are considered.

Other GEOMs have been developed, considering the interactions between HSC and MSC. (Mesfun et al., 2017) used the “BeWhere” model based on MILP formulation, with the Alpine Region as a case study. The impact of the intermittent renewable energy sources and the way how the surplus of electricity can be recovered through PtG and PtL systems is examined. The objective function to minimize is the total cost of the complete supply chain, including the cost of CO₂ emissions. (Bramstoft et al., 2020) investigate the role of renewable gas and liquid fuel production pathways in various future system configurations, with a fossil-independent Danish energy system by 2050 as a case study. This modeling framework explores synergies between renewable gas and renewable liquid fuel production, and the power and district heating networks. Two strategies, i.e., Balmorel and OptiFlow are used to obtain the optimal SC. Balmorel optimizes simultaneously the economic dispatch and investments, by minimizing the total costs for satisfying the district heating and power demands, using a perfect foresight approach within the year of optimization. OptiFlow optimizes the location, size, and operation of conversion plants, depending on e.g. costs of transporting local biomass resources, their seasonal availability, and the district heating demand.

2.3.3 *Location or partial optimization models (LoPOM)*

The deployment of a transport network and refueling stations for the supply of hydrogen at low cost and safety are two of the challenges that are repeatedly highlighted so that hydrogen can play an appropriate role in the energy mix (Ball and Weeda, 2015; *The Hydrogen Economy - Opportunities, costs, barriers, and R&D needs*, 2005). Since it is expected that PtM concepts will be deployed easier once HSC will be established (E&E consultant et al., 2014), it also influences the deployment of MSC. Several models have been developed to optimize networks of hydrogen refueling stations mainly in small geographical spaces (cities or states), where the location of service stations is treated with different approaches: the “Flow-Refueling Location Model” used for studying strategies for initial infrastructure deployment in Florida (Kuby and Lim, 2005), the “Fuel-travel-back” approach applied to Southern California (Lin et

al., 2008), a model for planning a network of refueling stations of a given company within a competitive framework used in the Northern Italy (Bersani et al., 2009), and the life cost approach (Chaoming He et al., 2017; Sun et al., 2017).

- *Choice of the framework for the optimization-based HMSC models*

Table 2.4 shows the main categories of optimization-based HMSC models. ESOMs offer a global vision of an energy system involving all possible technologies that can satisfy a set of services but do not include all the aspects related to logistical issues (e.g, number, size, and location of units). LoPOMs only address a part of the SC. In this regard, GEOMs are the most suitable for the design and deployment of SC, as they encompass geographical and logistical aspects of the entire SC (i.e., number of plants to be installed, their location, transport, and distribution network). For this work, the approach chosen is GEOM oriented, due to the reasons aforementioned with a MILP formulation: the involved constraints will be explained in the following chapters.

Table 2.4. Three main types of HMSC models

	Research object		Spatial scale				Main formulation			Main interest	
	E	P	I	N	R	U	LP	NLP	MILP		MINLP
ESOM	•		•	•	•		•		•		Balance of the entire energy system / identification of the part attributed to a set of technologies / strategy aspects
GEOM	•			•	•				•		Strategy and logistic aspects / identification of the part attributed to a technology
LoPOM		•					•		•	•	Logistic, tactical and/or operational aspects

E: Entire HMSC; P: Partial HMSC; I: International; N: National; R: Regional; U: Urban

2.4 Conclusion

In this chapter, a description of the main technological options involved in HMSC was presented. Based on the bibliography consulted, a preselection of the technologies to be considered for modeling was carried out based on the following elements.

- Emphasis on power-to-gas systems, including raw materials and major end-uses.
- The possible use of SMR process, given its current high market penetration, in order to facilitate the implementation of the energy transition.
- Scenario with high penetration of renewable energies.

Because the interest of this work is to address decision-making at the strategic/logistical level and the entire HMSC, a GEOM model will be developed. In this regard, the optimization model must indicate the hypotheses adopted for each use case, due to the different end uses that both energy vectors can have and the interactions between the technologies involved.

Given the variability of demand, availability of raw materials, the learning rate of each technology as well as the economic and environmental aspects, the selection made so far needs to be consolidated considering the temporal and multi-objective aspects. In addition, the specific selection of technological options needs to be in line with the case study even if the methodology has the ambition to be generic to be replicated in other geographical contexts. These aspects will be addressed in chapters 3 and 4.

3 METHODS AND TOOLS FOR MULTI-PERIOD, MULTI-CRITERIA AND MULTI-ECHELON HMSC DESIGN

ABSTRACT

This chapter presents the methods and tools used in this work for Hydrogen and Methane Supply Chains (HMSC) design. The main aspects for the solution of the mono and multi-objective optimization problems are thus presented: problem formulation, optimization framework, and tools for multi-criteria decision making (MCDM) steps. This chapter should be seen as a complement to the understanding of the models and methodological approach used in the following chapters.

RÉSUMÉ

Ce chapitre présente les méthodes et les outils utilisés dans ce travail pour la conception des chaînes logistiques de l'hydrogène et de méthane (HMSC). Les points clés en vue de la résolution des problèmes d'optimisation mono et multi-objectifs sont ainsi présentés : formulation du problème, cadre d'optimisation et outils pour les phases de prise de décision multi-critères (MCDM). Ce chapitre doit être considéré comme un complément à la compréhension des modèles et de l'approche méthodologique utilisées dans les chapitres suivants.

Acronyms

AUGMECOM	Augmented ε -constraint method
ELECTRE	Elimination and Choice Translating Reality
GAMS	General Algebraic Modeling System
HMSC	Hydrogen and Methane Supply Chain
HSC	Hydrogen Supply Chain
LP	Linear Programming
MCDM	Multi-criteria Decision Making
MILP	Mixed Integer Linear Programming
MINLP	Mixed-Integer Nonlinear Programming
MSC	Methane Supply Chain
M-TOPSIS	Modified TOPSIS
NLP	Nonlinear Programming
TOPSIS	Technique for Order Preference by Similarly to Ideal Solution
RHS	Right-hand-side

3.1 Introduction

Optimization techniques have been widely used in engineering in diverse applications, at very different scales, and with various objectives (Boix et al., 2015; Cui et al., 2017; Guillén et al., 2005). This includes the design and assessment of energy systems, which are generally addressed using mathematical models that encompass their key features: costs and benefits, technical performance, thermodynamic indicators, flows, environmental effects, and/or resource requirements (land, water, and other natural resources) (Kanbur et al., 2019; Rajesh et al., 2001; Rath-Nagel and Stocks, 1982). In this regard, the problem formulations can be linear (Linear Programming, LP; or MILP) or nonlinear (Nonlinear Programming, NLP; or Mixed-Integer Nonlinear Programming, MINLP). Several studies have been employed to address hydrogen supply chains (HSC), methane supply chains (MSC), and HMSC with optimization strategies (Agnolucci and Mcdowall, 2013; Blanco and Faaij, 2018; Li et al., 2019).

This chapter is a summary of the main approaches used and the choices made in this work to develop the methodological framework for HMSC design and address the case studies that will be studied in the following chapters. This analysis is based on previous reviews (Agnolucci and Mcdowall, 2013; Blanco and Faaij, 2018; Li et al., 2019), and on the expertise acquired in the *Laboratoire de Génie Chimique* (LGC) (De-León Almaraz, 2014). This chapter can also be seen as a complement to the understanding of the models and methodological approach used in the following chapters.

The chapter is divided into 5 sections. The categories of the models are described in section 2, following this introduction, in which particular attention is devoted to the geographically explicit optimization models (GEOM). In section 3, the optimization framework for GEOM is addressed. The decisions and

planning time horizon, performance measures, and solution methods will be presented. Section 4 is devoted to the decision aid methods for multi-criteria decision making. Finally, the main methods and tools chosen will be justified.

3.2 Optimization framework for GEOM

The development of the optimization framework for HMSC involves considering the following key items.

3.2.1 Decisions and planning time horizon

The HMSC design problem involves strategic-level decisions since these refer to the location, number, and size of facilities, the transport mode to be used, and the geographical customer areas to serve (Li et al., 2019). These decisions influence the deployment of HMSC in the medium and long term (Klibi et al., 2010). This means that when designing an HMSC, no tactical or operational decisions are explicitly taken into account, which is carried out in the short term (Mula et al., 2010; Yue et al., 2014).

3.2.2 Strategic decisions

According to the previous classification by (Li et al., 2019), the main strategic decision variables in the HMSC can include: feedstock (type, availability), production (number, location, production rate, type, physical form, size), transportation (flow rate, type, number, existence, direction), storage (number, location, inventory, type, size), refueling station (location, number, type, size), emission reduction (amount reduced, type). In this regard, facility location is a key function (Melo et al., 2009), that is highly dependent on the geographic discretization chosen. Furthermore, according to (Melo et al., 2009), feedstock can significantly influence the supply chain configuration. These aspects have been addressed in (De-León Almaraz et al., 2015), considering the type of energy sources used for hydrogen production. However, other issues such as the interaction with the CO₂ feedstock have proven to be critical to the HMSC (Sunny et al., 2020), and have insufficiently been addressed. Another key aspect to consider is the refueling station problems, due to its high contribution to the capital cost (Li et al., 2019).

3.2.3 Multi-period approach

Since some of the technologies associated with HMSC have not yet reached a maximum level of maturity (Götz et al., 2016, 2014), and the demand satisfied by these energy carriers is expected to grow in the coming years (ADEME, 2016a; E&E consultant et al., 2014), the HMSC evolution capacity should be considered. This is generally implemented through a multi-period approach, involving the variation of some parameters in given periods (Melo et al., 2009) as illustrated in Figure xxx. This means that demand variation and the learning rate of technologies have to be taken into account (Böhm et al., 2020). Fine-grained modeling can also include shorter periods (month-step patterns for instance) to

consider feedstock and demand variability (Kim and Kim, 2016). A generic example is shown in Figure 3.1.

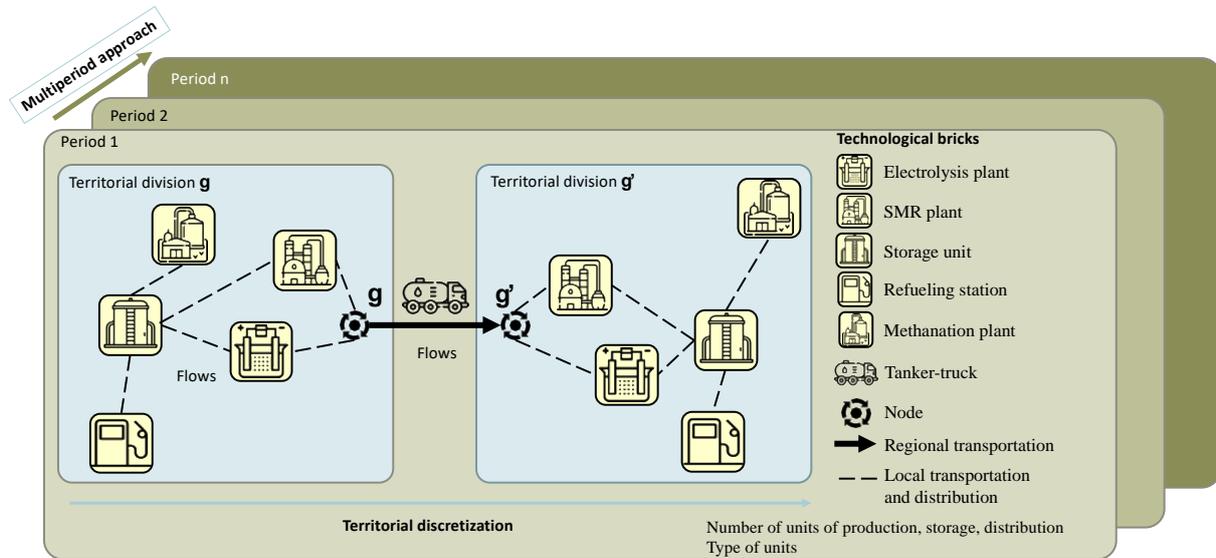


Figure 3.1. Generic HMSC modeling with a multi-period approach

3.2.4 Performance measures

3.2.4.1 Cost

Cost is the most widely used objective function to measure the performance of an HMSC (Blanco and Faaij, 2018; Li et al., 2019). This function is generally stated using an approach similar to (Almansoori and Shah, 2006), where investment and operating costs are included. Other researches included several aspects such as the cost of land (Han et al., 2012; Kim et al., 2008; Kim and Kim, 2016), environmental aspects (Bramstoft et al., 2020; Mesfun et al., 2017; Samsatli et al., 2016), and the discount rate (Carrera Guilarte and Azzaro-Pantel, 2020). Some improvements that could be made in this regard are the incorporation of profit maximization (Samsatli et al., 2016), and the calculation of the Levelized cost of energy (LCOE) for hydrogen and methane (Carrera Guilarte and Azzaro-Pantel, 2020).

3.2.4.2 Environmental impacts

Environmental impacts are measured using a life cycle assessment (LCA) approach, taking into account the global warming potential (GWP) indicator based on GHG emissions (Carrera Guilarte and Azzaro-Pantel, 2020; De-León Almaraz et al., 2015; Han et al., 2013) or several Life Cycle Assessment (LCA) indicators (Guillén-Gosálbez et al., 2009; Sabio et al., 2012). However, even though the most used indicator is GWP, the impact of PtG systems can be important in other categories. For example, (Blanco et al., 2020) indicate that PtM can have an impact up to 10 times larger for impact categories other than climate change.

3.2.4.3 Safety

Although there is no clear consensus on how to analyze the risks associated with hydrogen and methane production, storage, and transport, several studies suggest that this strategic aspect should be addressed.

It is particularly important for the HSC, because hydrogen can react dangerously (i.e., its explosion can generate fatal accidents) (Kim and Moon, 2008; Li et al., 2019). (Kim and Moon, 2008) proposed a pioneering model to address this issue. However, there is no standard methodology to address these aspects (Govindan et al., 2017), especially when linked to other energy vectors (i.e., methane).

3.2.4.4 Exergy

Another aspect to be addressed is the destruction and loss of exergy throughout the HMSC. While energy is conserved (first law of thermodynamics), exergy is destroyed when a process is irreversible (second law of thermodynamics) (Querol et al., 2013). Exergy can also be lost due to by-products. Hence, exergy analysis provides insight into the performance of the HMSC that would elude purely first-law analysis. The overall exergy efficiency of the HMSC can vary dramatically depending on the feedstock and the technologies involved (Khosravi et al., 2018). Thus, the estimation of destroyed and lost exergy is essential to propose energy improvements and compare options. The reported studies that have carried out an exergy analysis on the processes linked to the HMSC technology components have shown that new processes with higher yields can be obtained (Bozoglan et al., 2012; Calderón et al., 2011; Hajjaji et al., 2012; Obara, 2019; Obara and Li, 2020; Toonssen et al., 2008).

3.2.5 Multi-objective approach

Most of the approaches used to model a SC involving hydrogen or methane state from a mono-objective approach generally involve a cost function as an objective function. However, cost is related to only one pillar of sustainability. Even if carbon pricing can be used to represent the hidden cost of greenhouse gas pollution and could be included in a cost function, it does not necessarily imply the achievement of a solution with minimum CO₂-eq emissions. Other criteria involved in a more comprehensive approach need to be included in appropriate multi-objective solution approaches in order to address sustainability correctly.

3.2.6 Integration in the GEOM model

Only Mesfun et al. (Mesfun et al., 2017) and Bramstoft et al. (Bramstoft et al., 2020), are listed as GEOMs that satisfy simultaneously several fuel demands through PtG concepts. The evolution of the supply chain over time, the installation of refueling stations, and exergy analysis are not covered in either of the studies. This work contributes to bridging these gaps. Table 3.1 shows the main characteristics of (Mesfun et al., 2017), (Bramstoft et al., 2020), and this work.

Table 3.1. Strategic decisions in HMSC models

Articles	Feed.	Prod.	Transp.	Stor.	H ₂ Ref. Stat.	Units			Dep. per p.	Cost O.	Env. O.	Ex O.
						Loc.	Size	Tech.				
(Mesfun et al., 2017)	•	•		•		•	•	•		•	•	
(Bramstoft et al., 2020)	•	•	•	•		•	•	•		•	•	
This study	•	•	•	•	•	•	•	•	•	•	•	•

Feed.: Feedstock; Prod.: Hydrogen and methane production; Transp.: Hydrogen transportation; Loc.: Location; Tech.: Technology; Dep. in time: Development of the supply chain in time; Cost O.: Costs included in the objective function; Env. O.: Environmental effects included in the objective function. Ex O.: Exergy analysis included in the objective function.

According to the preselection model conducted in chapter 2, the design methodology will have to encompass all these elements:

- Modeling of HMSC through a GEOM optimization approach, with emphasis on strategic and deployment aspects.
- Particular attention is paid to power-to-gas systems, their raw materials, and end-uses. In addition, the use of fossil fuels, particularly for hydrogen production via SMR is taken will also be included in the modeling options.
- A multi-period approach will be deployed to cover aspects such as demand variability, resource availability, and technological maturity.
- The HMSC configuration has to be designed considering cost criterion, environmental impact, and exergy analysis.

3.3 Solution methods

3.3.1 Mono-objective problems

According to (Bierlaire 2006), the mono-objective approach can be stated as in Eq. 3.1.

$$\text{Min } f(x) \tag{Eq. 3.1}$$

st

$$g(x) \leq 0$$

$$h(x) = 0$$

where f is a function $R^{n1} \times [0,1]^{n2}$ in R , $g(x) \in \mathfrak{R}^m$, $h(x) \in \mathfrak{R}^p$, $x \in S$

In the abovementioned formulation, $f(x)$ is the objective function; vectors $g(x)$ and $h(x)$ are respectively m inequality constraints and p equality constraints, and x is an element of S . The set of restrictions defines the domain where the optimal solution is found, in which continuous and discrete variables can be involved.

According to (Li et al., 2019; Melo et al., 2009), SC optimization problem solving can be classified into two categories: problems solved with a predetermined software (either commercial or non-commercial), and problems solved with an adapted algorithm. Within each of these two classifications, solutions can be of the "exact" or "heuristic" type. This distinction can be seen in Figure 3.2. Most of the solution approaches found in the literature are solved with a "general solver". The GAMS environment ("GAMS - Cutting Edge Modeling," n.d.) is one of the most used for this purpose. Although GAMS includes a large number of well-known solvers for the solution of MILP, the mono-objective problems related to SC are generally solved through the efficient CPLEX solver ("IBM CPLEX Optimizer," n.d.), (Klibi et al., 2010). However, when the number of periods and potential locations is relatively high, the number of integer and binary variables can increase considerably. In this sense, hybrid methods adapted for HMSC optimization could provide quasi-optimal solutions using less computational effort (Copado-méndez et al., 2013).

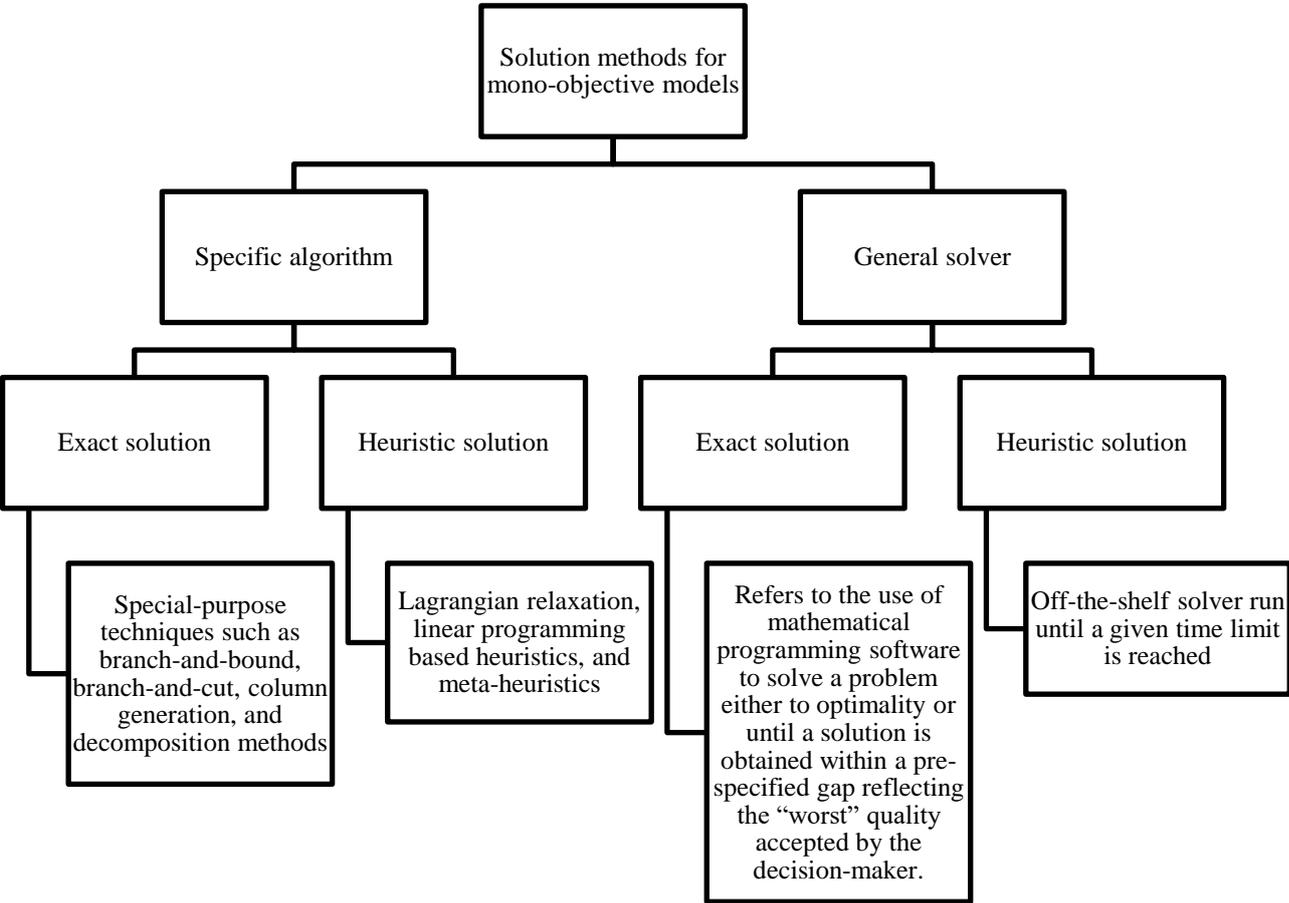


Figure 3.2. Solution methods for mono-objective models (modified from (Li et al., 2019))

- Criteria to be considered

The mono-objective problems considered in this work involve the Total Annual Cost (TAC), the environmental impact through the Global Warming Potential (GWP), or the Total Exergy Lost and Destroyed (TELD) as the optimization criteria. The problem formulation uses the GAMS environment with the CPLEX solver.

3.3.2 Multi-objective problems

Multi-objective optimization is carried out when two or more objective functions are of interest and there is an antagonistic conflict between several criteria. The general formulation of a multi-objective optimization problem is shown in Eq. 3.2, where f_i with $i = 1, k$ is a function of $R^{n1} \times [0,1]^{n2}$ in $R, g(x) \in R^m, h(x) \in R^p$ and x is an element of S .

$$\text{Min} [f_1(x), f_2(x), \dots, f_{1k}(x)] \quad \text{Eq. 3.2}$$

st

$$g(x) \leq 0$$

$$h(x) = 0$$

$$x \in S$$

The approaches used to solve multi-objective problems can be classified into a priori, a posteriori, and hybrid models as shown in Figure 3.3.

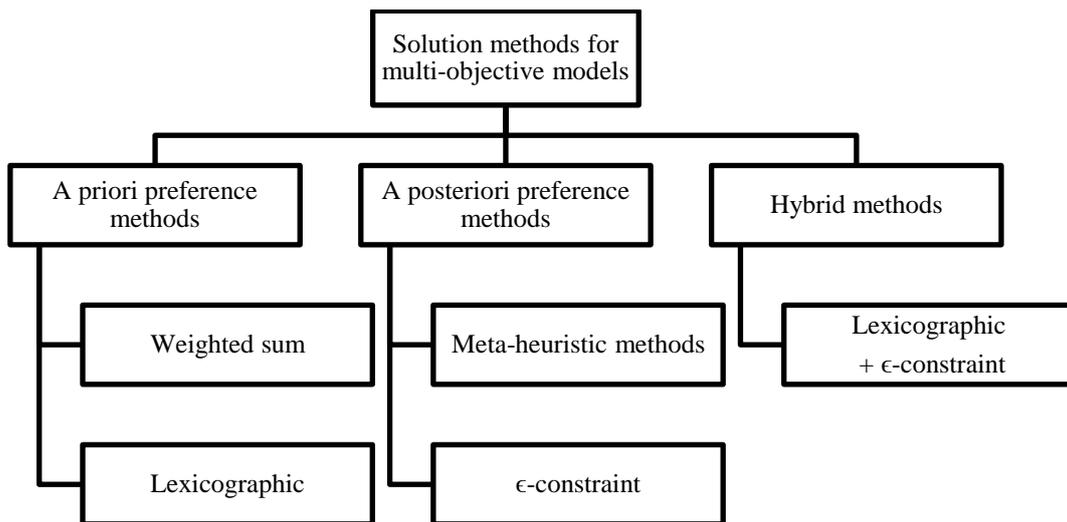


Figure 3.3 Solution methods for multi-objective models (modified from (Li et al., 2019))

In this case, a set of optimal solutions is obtained. It is represented through the Pareto front (or efficient, non-dominated, non-inferior), which are the solutions that cannot be improved in one objective function without degrading their performance in at least one of the rest (Mavrotas, 2009). The Pareto front is illustrated in Figure 3.4. This property can be mathematically expressed as follows: A feasible solution $x^* \in S$ is called efficient or Pareto optimal if there is no other $x \in S$ such that $f(x^*) < f(x)$ and $f(x) \neq f(x)$ (Khorram et al. 2010).

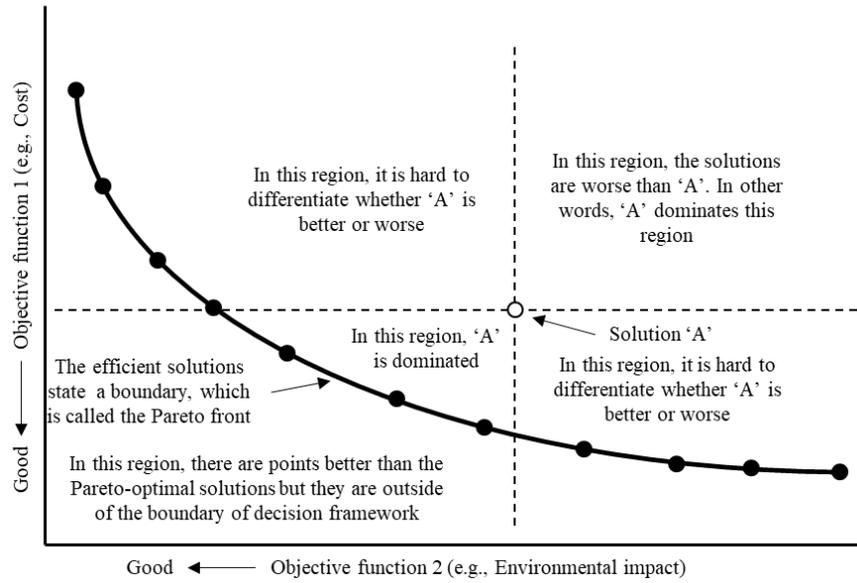


Figure 3.4. Illustration of Pareto optimality in two-objective optimization (modified from (Xin, 2013))

3.3.2.1 ϵ -constraint method formulation

The literature indicates that for HSC chains the most used approach is the ϵ -constraint (Li et al., 2019). The advantage of this method is that leads to a set of Pareto- optimal solutions that can then be used to make decisions. The ϵ -constraint method is based on the minimization of one objective function while the others are considered as restrictions within a certain allowed level, usually called nadir and utopia (or ideal) solutions. These two points are assumed to be the worst and best solution, respectively (Miettinen, 1998). The problem can be formulated as shown in Eq. 3.3.

$$\begin{aligned}
 & \text{Min } f_1(x) && \text{Eq. 3.3} \\
 & \text{st} \\
 & f_2(x) \leq \epsilon_2 \\
 & f_3(x) \leq \epsilon_3 \\
 & \dots \\
 & f_p(x) \leq \epsilon_p \\
 & x \in S
 \end{aligned}$$

By parametrical variation in the right-hand-side (RHS) of the objective functions (ϵ_i), the efficient solutions of the problem are obtained (Mavrotas, 2009). Despite its easy implementation, three major difficulties are generally highlighted (González-Rodríguez, 2020):

- Estimation of the range of objective functions over the efficient set.
- Guarantee of efficiency of the obtained solutions.
- Increased solution time for problems with more than two objectives.

3.3.2.2 Hybrid methods: augmented ϵ -constraint method

To address the abovementioned issues, the ϵ -constraint method can be combined with a priori method (i.e., lexicographic), thus allowing only efficient solutions to be obtained. They are considered as “hybrid methods”. An example of this approach is the augmented ϵ -constraint method (AUGMECON) (Mavrotas, 2009), which allows obtaining strong optimal solutions and avoids redundant iterations, compared to the conventional ϵ -constraint method, and other methods as the “weighted sum approach” (Costa-Carrapiço et al., 2020).

The major difference between the conventional ϵ -constraint method and the AUGMECON involves the calculation of the payoff table. In the conventional method, the widely used approach is to find these values with the upper and lower values (results of the mono-objective optimizations for each objective function). Although the nadir point is calculated with the maximum values of this table, this can lead to non-optimal solutions. To deal with this problem, AUGMECON uses the lexicographical optimization for each objective function to obtain the payoff table with only optimal solutions. This implies that the solutions above these values are excluded. Lexicographic optimization is applied as follows:

- The first objective function (of higher priority) is optimized so that $\max(f_1) = z_1^*$.
- Then the second objective function is optimized by adding the constraint $f_1 = z_1^*$, to keep the optimal value of the first optimization.
- Assuming that $\max(f_2) = z_2^*$ was obtained, the third objective function is optimized by adding the two constraints $f_1 = z_1^*$ and $f_2 = z_2^*$ to keep the previous optimal values.
- The process continues until the whole set of the objective function is treated.

Another aspect addressed by (Mavrotas, 2009) is the guarantee that the solutions obtained are efficient. If an optimal alternative solution can be obtained, then the solutions found are “weakly” efficient. The solutions are efficient only if the constraints associated with the objective functions are interconnected. To address this aspect, the method incorporates slack variables that are used in a second term with a lower priority (according to the lexicographic optimization), forcing the program to generate only efficient solutions. In addition, the inequalities are transformed into equations incorporating the slack variables. The new formulation is indicated in Eq. 3.4, where *eps* is an adequately small number (i.e., 10^{-3} to 10^{-6}).

$$\text{Min} \left(f(x) + \text{eps} \times (s_2/r_2 + s_3/r_3 + \dots + s_p/r_p) \right) \quad \text{Eq. 3.4}$$

st

$$f_2(x) - s_2 = \varepsilon_2$$

$$f_3(x) - s_3 = \varepsilon_3$$

...

$$f_p(x) - s_p = \varepsilon_p$$

$$x \in S \text{ and } s_i \in R$$

An alternative is the meta-heuristic approaches, which are rarely used for this type of problem compared to the deterministic approach. However, they represent an interesting branch of development to be addressed. For example, (Cantú et al. 2020) have developed a hybrid algorithm based on meta-heuristic methods for HSC that reproduces accurately the Pareto front while reducing drastically (up to 20 times) the CPU time with respect to the MILP solver (CPLEX) (Cantú et al., 2020). This type of approach can be extended to HMSC, to address non-linear aspects as well as multiple periods, and a large number of patterns in the network.

- Choices about mono and multi-objective approaches

The mono and multi-objective problems are solved in the GAMS environment. The literature review allowed identifying the ε -constraint method approach as the most suitable to solve the multi-objective problems. To address the weaknesses of the conventional approach previously mentioned (determination of the Nadir point, the guarantee of finding efficient solutions...), a hybrid (lexicographic+ ε -constraint) method was chosen, i.e., AUGMECON.

3.4 Multi-criteria decision making (MCDM): decision aid methods

The set of solutions encompassed in the Pareto front is based on a compromise between the different decision criteria. In this regard, a multi-criteria decision analysis method can be used to choose a solution, considering the order relation between various elements. Two methods and their respective variants are the most common for multi-criteria decision making: ELECTRE and TOPSIS (De-León Almaraz, 2014).

3.4.1 ELECTRE

The Elimination and Choice Translating Reality (ELECTRE) method can be used to select which alternative is preferable, incomparable, or indifferent by pairwise comparison of the alternatives under each criterion. It utilizes outranking relations or priority functions for ranking the alternatives in terms of priorities among the criteria (Peng et al., 2015). This method is sometimes unable to identify the most preferred alternative, only producing a core of leading alternatives. It is especially convenient when

there are decision problems that involve a few criteria with a large number of alternatives saving much time (De-León Almaraz, 2014).

3.4.2 TOPSIS

The TOPSIS (Technique for Order Preference by Similarly to Ideal Solution) (Hwang and Yoon, 1981; Yoon, 1987) is a popular approach to multiple criteria decision making (MCDM) and has been widely used in the engineering literature (Wang and Elhag, 2006). The fundament of this method is that the selected alternative should have the shortest distance from the Positive Ideal Solution (PIS) and the farthest distance from the Negative Ideal Solution (NIS) in a geometrical sense (De-León Almaraz, 2014; Wang and Elhag, 2006). Its implementation requires a simple mathematical form, and adaptability to SC management (Behzadian et al., 2012). In this method, a decision matrix is evaluated, involving “m” alternatives (i.e., efficient solutions), and “n” criteria (i.e., TAC, GWP, TELD...). (see Figure 3.5). According to (González-Rodríguez, 2020), the steps involved can be summarized as follow:

- Step 1: To build the decision matrix that shows “m” efficient solutions “i” evaluated by “n” criteria “j”.

		<i>j=1</i>	<i>j=2</i>	<i>j=3</i>	...	<i>j=n</i>
		C1	C2	C3		Cn
<i>i=1</i>	A1	<i>Xij</i>	<i>Xij</i>	<i>Xij</i>	...	<i>Xij</i>
<i>i=2</i>	A2	<i>Xij</i>	<i>Xij</i>	<i>Xij</i>	...	<i>Xij</i>
<i>i=3</i>	A3	<i>Xij</i>	<i>Xij</i>	<i>Xij</i>	...	<i>Xij</i>
⋮	⋮	⋮	⋮	⋮	⋮	⋮
<i>i=m</i>	<i>Am</i>	<i>Xij</i>	<i>Xij</i>	<i>Xij</i>	...	<i>Xij</i>

Figure 3.5. Generic decision matrix for application of the TOPSIS method (from (González-Rodríguez, 2020))

- Step 2: Each value in the decision matrix is transformed into a normalized, non-dimensional value as is shown in Eq. 3.5, where *a_{ij}* stands for the normalized values.

$$a_{ij} = \frac{x_{ij}}{\sqrt{\sum_{j=1}^n x_{ij}^2}} \tag{Eq. 3.5}$$

- Step 3: The decision-makers assign weights to each criterion, thus allowing the objectives to be ranked. The matrix values are multiplied according to the applicable weight. It is shown in Eq. 3.6, where *w_i* stands for the weight assigned.

$$v_{ij} = w_i \cdot a_{ij} \tag{Eq. 3.6}$$

- Step 4. The positive and negative ideal solutions from matrix A are determined. The positive ideal solution (A^+) is the group of weighted normalized criteria values, which indicates the best solution that can be reached. The negative ideal solution (A^-) is a group of weighted normalized criteria values, which represents the negative ideal criteria values. This is shown in Eq. 3.7 and Eq. 3.8.

$$A^+ = \{v_{1+}, v_{2+}, \dots, v_{n+}\} \quad \text{Eq. 3.7}$$

$$A^- = \{v_{1-}, v_{2-}, \dots, v_{n-}\} \quad \text{Eq. 3.8}$$

Thereafter, the distances to the ideal solution S_{i+} and to the negative ideal solution S_{i-} are calculated for each efficient solution using the Euclidean distance. It is shown in Eq. 3.9 and Eq. 3.10, where $v_{j+} = \max_i v_{ij}$ and $v_{j-} = \min_i v_{ij}$.

$$S_{i+} = \sqrt{\sum_{j=1}^n (v_{j+} - v_{ij})^2} \text{ for } i = 1, 2, \dots, m, \quad \text{Eq. 3.9}$$

$$S_{i-} = \sqrt{\sum_{j=1}^n (v_{j-} - v_{ij})^2}, \text{ for } i = 1, 2, \dots, m, \quad \text{Eq. 3.10}$$

Thus, a rank of alternatives is created in decreasing order according to the value of C_i . The best alternative is the one that has the TOPSIS coefficient C_i nearest to 0. It is shown in Eq. 3.11.

$$C_i = \frac{S_{i-}}{(S_{i+} + S_{i-})} \quad \text{Eq. 3.11}$$

3.4.3 M-TOPSIS

One of the problems related to TOPSIS is that it can cause a phenomenon known as rank reversal (García-Cascales and Lamata, 2012). In this phenomenon, the alternative order of preference changes when an alternative is added or removed from the decision problem. To deal with this problem, the M-TOPSIS variant (Ren et al., 2007) has been proposed. To address this issue, the M-TOPSIS sets the optimized ideal reference point ($\min(S_{i+}), \max(S_{i-})$), and then the distance from each alternative to that point is calculated (Eq. 3.12). The best alternative corresponds to a coefficient C_i^M nearest to 0.

$$C_i^M = \sqrt{(S_{i+} - \min(S_{i+}))^2 + (S_{i-} - \max(S_{i-}))^2} \quad \text{Eq. 3.12}$$

- Choices about the MCDM method

Supply Chain Management and logistics are considered the most popular topic in TOPSIS approach applications, due to their simple application and adaptability to this type of problem (Behzadian et al., 2012). A variant of the conventional TOPSIS method, the modified-TOPSIS (m-TOPSIS), is used in this work, since it allows avoiding rank reversals and evaluation failure that often occurs in the conventional TOPSIS approach (Ren et al., 2007).

3.5 Conclusion

In this chapter, the approach used to implement the solution of the optimization problems is described. The basis of the models and tools are described and supplemented in the following chapters. The main choices made are listed below:

- A. The set of models used can be categorized as GEOM with MILP formulations. The decision variables include: feedstock, production, transportation, storage, and refueling station. A multi-period approach is considered. Three objective functions are included: cost, environmental impact, and exergy. The model formulation is carried out in the GAMS environment
- B. Mono-objective optimizations will be solved through CPLEX,
- C. Multi-objective optimizations will be tackled through the AUGMECON framework
- D. The M-TOPSIS technique will be selected as an MCDM method.

4 CASE STUDY FOR THE HMSC OPTIMIZATION

ABSTRACT

The case study of Occitania for HMSC deployment serves as an illustration of the proposed methodology. The Region is on the cutting edge of developing green hydrogen with its own Green Hydrogen Plan (FuelCellsWorks, 2020). The collection of techno-economic data to be used in the optimization model, the territorial characteristics, and the available feedstock in the Occitania Region are presented. This chapter can be viewed as a supplement to the next chapters, which will develop the methodological optimization for HMSC design.

RÉSUMÉ

L'étude de cas de l'Occitanie pour le déploiement du HMSC sert d'illustration de la méthodologie proposée. Cette région est à l'avant-garde du développement de l'hydrogène vert avec son propre plan (FuelCellsWorks, 2020). La collecte de données technico-économiques à utiliser dans le modèle d'optimisation, les caractéristiques territoriales et les matières premières disponibles dans la région de l'Occitanie sont présentées. Ce chapitre peut être considéré comme un complément aux chapitres suivants, qui développeront la méthodologie d'optimisation pour la conception du HMSC.

4.1 Introduction

The profound transformation that the energy system must undergo to achieve the objectives of greenhouse gas emission reduction has led to the development of different roadmaps, particularly in the European context (European Commission, 2020a; Fuel Cells and Hydrogen Joint Undertaking, 2019). In France, ADEME has carried out a set of studies with energy-climate scenarios (ADEME, 2018, 2016b; E&E consultant et al., 2014). In this sense, the development of energy vectors such as hydrogen and syn-methane, along with the incorporation of Power-to-Gas (PtG) systems, has been seen as a promising solution to substitute the fossil fuels currently used.

The French primary energy mix is composed of 16% natural gas, which represents 505 TWh (Ministère de la Transition Écologique et Solidaire, 2018). Natural gas consumption relies mainly on imports due to the depletion of the French fields in Lacq since 2013 (ADEME, 2018; E&E consultant et al., 2014). There is still a marginal production of gas in the north of France (247 GWh in 2015, versus 2 TWh in 2000). The French natural gas network has 16 underground gas deposits with a total capacity of 132 TWh. This allows great flexibility, balancing the relatively constant gas import during the year against a highly variable consumption according to the period of the year.

Green gas production is an emerging sector. Biomethane from methanization is a new source of gas, producing a relatively low amount of 82 GWh in 2015 and 215 GWh in 2016 (ADEME, 2018). One of the main advantages of these emerging energy vectors is the existence of a well-established infrastructure for the storage and transport of natural gas, which facilitates the development of this market. ADEME indicates that 100% of the gas consumed in France could potentially be renewable by 2050 (ADEME, 2018). ADEME's study analyses a decarbonized gas network in 2050 with a total capacity of 460 TWh. Three main technologies are involved, i.e. 30 % methanization, 40% pyrogasification, and 30% PtM. Although all three options are seen as alternatives to accelerate the energy transition in France, PtG requires specific conditions. This technology could be used in the case of significant penetration of renewable energies in the electricity network, i.e. solar or wind sources, to balance the production and demand of electricity. Other studies made on a European scale and taking into account the whole energy system, indicate that the role of these systems could be important only in scenarios where there is an ambitious reduction of greenhouse gases (Blanco et al., 2018b).

To apply the methodology framework proposed in this research, the Occitania region is chosen as a case study. It is located in the south of France. It corresponds to 13.2% of the French metropolitan territory and is the fifth most populated region in France (Région Occitanie / Pyrénées-Méditerranée, 2017, n.d.).

4.2 Energy transition in the Occitania region

The energy transition in the Occitania region has been framed in a set of international, national, and regional agreements: the Paris Agreement (2015), the energy transition law for growth (2015) (“Energy transition | Gouvernement.fr,” n.d.), the Climate Plan of the Ministry of Energy and Solidarity Transition (2017) (“France’s Climate Plan | Gouvernement.fr,” n.d.) and the REPOS (Positive Energy Region, in French) scenario for the Occitania region (Région Occitanie / and Pyrénées-Méditerranée, 2017). In line with this, the ADEME and the Council of the Region have established the following priorities: a 40% decrease in energy consumption and a threefold increase in the production of renewable energies. Besides, the authorities have set as objective to substitute all-oil for the mobility of people and goods involves developing public transportation, with new services and an offer of vehicles powered either by electricity (mainly in urban areas) or using gas from renewable sources (hydrogen, methane, and other fuels from renewable sources). Currently, the demand for natural gas in the Occitania region represents approximately 4% (20 TWh) of France's total demand (Report, 2017). Renewable energy production in the region is 28 TWh, of which 13.6 TWh is electricity (Région Occitanie/Pyrénées-Méditerranée, 2018). By 2050, the following renewable electricity production is expected in the region to be met by 23 TWh of solar, 25.5 TWh of wind (taking into account both onshore and offshore), and 10 TWh of hydro (Région Occitanie / and Pyrénées-Méditerranée, 2017). This study adopts the ADEME and REPOS Occitania scenarios as the main forecasts and assumptions to propose an HMSC.

To perform the supply chain optimization, preliminary work must be carried out, depending on the case study. In this section, three topics are addressed, i.e., the collection of techno-economic data, geographic division, and the estimate of energy sources and demands.

4.3 Collection of techno-economic data

In general, the main sources used for the collection of the technical-economic data are reports, statistics, and journal articles. Since there is no historical information on the hydrogen and methane supply chains produced from PtG according to current climate strategies and objectives, their estimate is based on forecasts and the state of maturity of each technological cluster.

The techno-economic data for each echelon is shown in Table 4.1. Alkaline water electrolysis, SMR, and catalytic reaction (methanation) are the selected technologies for hydrogen and methane production, according to their high Technology Readiness Levels (TRL) (De-León Almaraz et al., 2015; Götz et al., 2016, 2014). The capacity for each technology was selected according to the data consulted. The CAPEX was estimated by using a classical exponential rule, as expressed in Eq. 4.1, where C_b stands for the equipment cost at the scale S_b (size, capacity, nominal power) of the component, and C_a and S_a represent the costs and scale of the known reference component, respectively. In this expression, f is the

scale factor applied to a given technology (Böhm et al., 2020). In this work, an 0.9 exponent was used, according to (Böhm et al., 2020).

$$C_b = C_a \left(\frac{S_b}{S_a} \right)^f \quad \text{Eq. 4.1}$$

Tanker-trucks were chosen as the transport option, as they are the most suitable for short distances (Abdin et al., 2020). For hydrogen storage, the use of liquid storage was selected because of its high efficiency in treating large amounts of hydrogen and its economic advantages over gaseous storage (Abdalla et al., 2018; De-León Almaraz et al., 2015). No plant is assumed to have been installed before 2035. The plant capacity installed between 2035 and 2045 should be sufficient to meet the increase in demand between 2045 and 2050, which means that no plant will be installed in 2050. To calculate the present value of future investments, a discount rate of 5.25% was considered (ADEME, 2018).

Table 4.2 shows the costs of feedstock and carbon price. For the cost of electricity, a differentiation was established for each source. This cost also varies according to the period analyzed. The cost of CO₂ sources represents the average value, involving its capture, storage, and transport. Yet, the associated costs can vary considerably according to the source, transport and storage needs, and the number of resources used (Oei et al., 2014). Besides, the minimum and maximum values of the carbon price are indicated. This parameter is incorporated to take into account environmental regulations, which are essential for the development of technological options based on renewable energies such as PtG.

Table 4.3 shows the values of the GHG emissions for the energy and CO₂ sources. For the environmental impact analysis, a comprehensive LCA was not carried out. Only the GHG emissions impact was computed, as the main indicator to assess the effects on climate change (Collet et al., 2017; Reiter and Lindorfer, 2015). Furthermore, the sources of electricity used for electrolysis and the CO₂ sources for methanation have been identified as the most sensitive parameters for the environmental impact of PtG systems (Blanco et al., 2020).

Table 4.1. Main techno-economic assumptions of the case study

Process	Type	Year	Capacity	Cost		References
				CAPEX	OPEX	
Electrolysis ¹	Alkaline	2035	1.53, 7.7, 13.8, and 20 MWe	500 €/kWe	5% of CAPEX per year + electricity cost	(Almansoori and Shah, 2009; De-León Almaraz et al., 2015; E&E consultant et al., 2014; Götz et al., 2016; Lehner et al., 2014)
		2050		300 €/kWe		
SMR			2.8 MW _{H2}	1.05 M€	5% of CAPEX per year + natural gas cost	(De-León Almaraz et al., 2015; Li et al., 2020)
			4.8 MW _{H2}	1.7 M€		
			7 MW _{H2}	2.28 M€		
H ₂ Transport ²	Tanker-truck	-----	117 MWh _{H2} /trip	500 k€	Driver wage (DW): 12.67 €/h; Fuel economy (FE): 2.5 km/liter; Fuel price (FP): 1.06 €/liter; General expenses (GE): 2,610 €/year; Load and unload time (LUT): 2 h; Maintenance expenses (ME): 0.11 €/km; Average speed (SP)=66.8 km/h; Availability of transportation (TMA): 6570 h/year; Weight of transportation (w): 40 ton	(Almansoori and Shah, 2009; De-León Almaraz et al., 2015)
Hydrogen storage	Tank	-----	15 MWh _{H2}	0.7 M€	1.92 €/ MWh _{H2}	(Almansoori and Shah, 2009; De-León Almaraz et al., 2015)
			317 MWh _{H2}	4.350 M€	0.96 €/ MWh _{H2}	
			5,000 MWh _{H2}	28.700 M€	0.30 €/ MWh _{H2}	
			18,000 MWh _{H2}	106 M€	0.15 €/ MWh _{H2}	
Refueling stations		2035	2.3; 3.8; and 4.6 MWh _{H2}	1300 €/kW _{H2}	10% of CAPEX	(De-León Almaraz et al., 2015; Li et al., 2020)
		2050		960 €/kW _{H2}		
Methanation	Catalytic	-----	5 – 100 MW _{CH4}	400 €/kW _{CH4}	5% of CAPEX per year + CO ₂ cost	(Collet et al., 2017; E&E consultant et al., 2014; Götz et al., 2016; Lehner et al., 2014)
CH ₄ Injection		-----			6.55 €/MWh	(Collet et al., 2017)

¹ The reference capacity is shown. See the Database for more details.

² The operating costs were calculated according to the parameters indicated in Eq. 5.39 to 5.43.

Table 4.2. Primary energy, CO₂ cost, and carbon price

Item	Type	Cost				References
		Year				
		2035	2040	2045	2050	
Energy source [€/MWh]	Natural gas	40	40	40	40	(Eurostat, 2020)
	Solar	45	42.3	41	38.3	(ADEME, 2016c; IRENA, 2018b, 2018c, 2016; Lazard, 2019)
	Wind	39.4	39	38.8	38.5	
	Hydro	24.5	24.3	24.2	24	
	Network	120	120	120	120	
CO ₂ source [€/tonCO ₂]	Methanation /gasification			42		(ADEME, 2018; E&E consultant et al., 2014)
	Others			85		
Carbon price [€/tonCO ₂]	Low			50		(Li et al., 2020)
	High			270		

Table 4.3. GWP for electricity and CO₂ sources

Source	Type	GHG emissions	References
Energy [gCO ₂ -eq/kWh]	Natural gas	202	(Collet et al., 2017)
	Solar	48	(ADEME, 2016b; Kriegler and Janssens-maenhout, 2014; Schlömer et al., 2014)
	Wind	12	
	Hydro	12	
	Network	23-27	
CO ₂ [gCO ₂ -eq/kg CO ₂]	From grid	278	(Collet et al., 2017; E&E consultant et al., 2014; Reiter and Lindorfer, 2015)
	Import	442	

4.4 Geographical division

As indicated in Chapters 2 and 3, GEOM formulations address relatively large areas (countries or regions), which are divided into sub-regions (grids or nodes). Each may have an associated demand or capacity to produce hydrogen or methane, which may be limited to the available feedstock. This explains why the decision variables will involve the location of the different production, storage, and transport units, and the flow between the grids involved in the proposed optimization framework that will be developed in the following chapters. One of the aspects to be taken into account is the flow of resources and products, and for this purpose, the appropriate transport network for these sub-regions will be determined.

Two major trends are observed in the literature in this regard. A subdivision, taking the United Kingdom as a case study is proposed in (Almansoori and Shah, 2012, 2009, 2006). Other studies propose an administrative segmentation to obtain more realistic divisions, particularly concerning the transport network and available local statistics (population, demands, etc.). The latter is the main approach adopted in the previous studies (De-León Almaraz et al., 2015; Johnson and Ogden, 2012; Murthy Konda et al., 2011; Parker et al., 2010).

For this work, the region of Occitania (*Région Occitanie* in French) has been divided into sub-regions, which correspond to the different departments of the region (13 departments) (see Figure 4.1), thus clearly identifying the hydrogen transport routes. Furthermore, specific information on feedstock and population density are available.

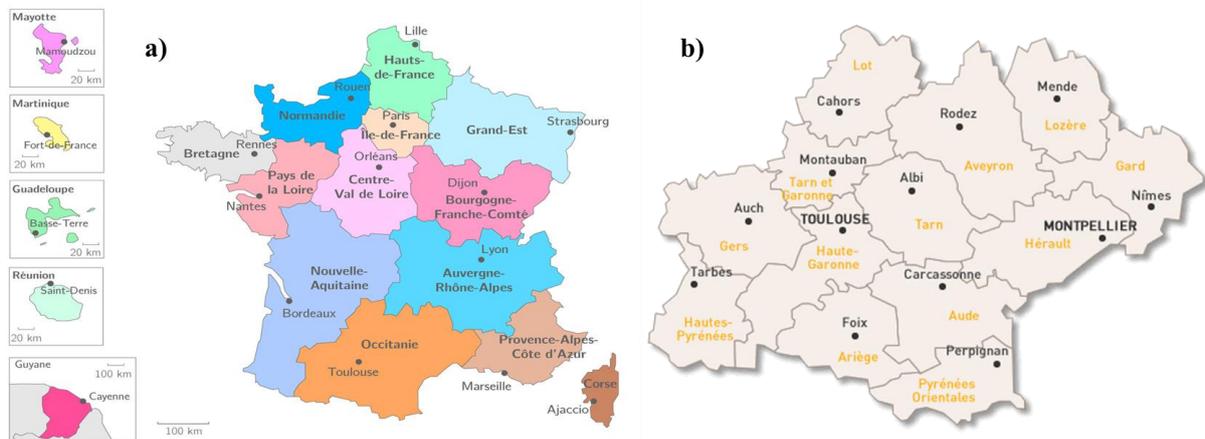


Figure 4.1. a) Administrative regions in France; b) Departments in the Occitania region

4.5 Demand estimate

Determining the hydrogen and methane demand to be met is a key step because the demand profile directly influences the configuration of the HMSC. Since most of the studies carried out focus on hydrogen demand for the transport sector, this depends mainly on the penetration that the vehicles will have, the population density, and its characteristics (urban or rural). (Li et al., 2019) identifies three ways to identify hydrogen demand: socio-economic studies (Melendez and Milbrandt, 2008), the assumption of an S-shaped curve as with other emerging technologies (Agnolucci and Mcdowall, 2013),

and coupling the energy system model with an HSCN model following a MARKAL/TIMES model (Agnolucci and Mcdowall, 2013).

Two known demands for hydrogen and methane must be met depending on the availability of resources and their possible importation. The demand for hydrogen is indicated for each grid, while the demand for methane is fixed for the territory as a whole. The evolution of the demands is shown in Figure 4.2. The demand for hydrogen is related to the transport sector. The hypotheses and assumptions used to calculate the penetration of FCEV in the transport sector were taken from the REPOS scenario (Région Occitanie / Pyrénées-Méditerranée, 2017) and (De-León Almaraz et al., 2015). The demand for methane was estimated based on the ADEME, and the REPOS scenario (ADEME, 2016a; Région Occitanie / Pyrénées-Méditerranée, 2017). It includes the residential, industry, transport, and service sectors.

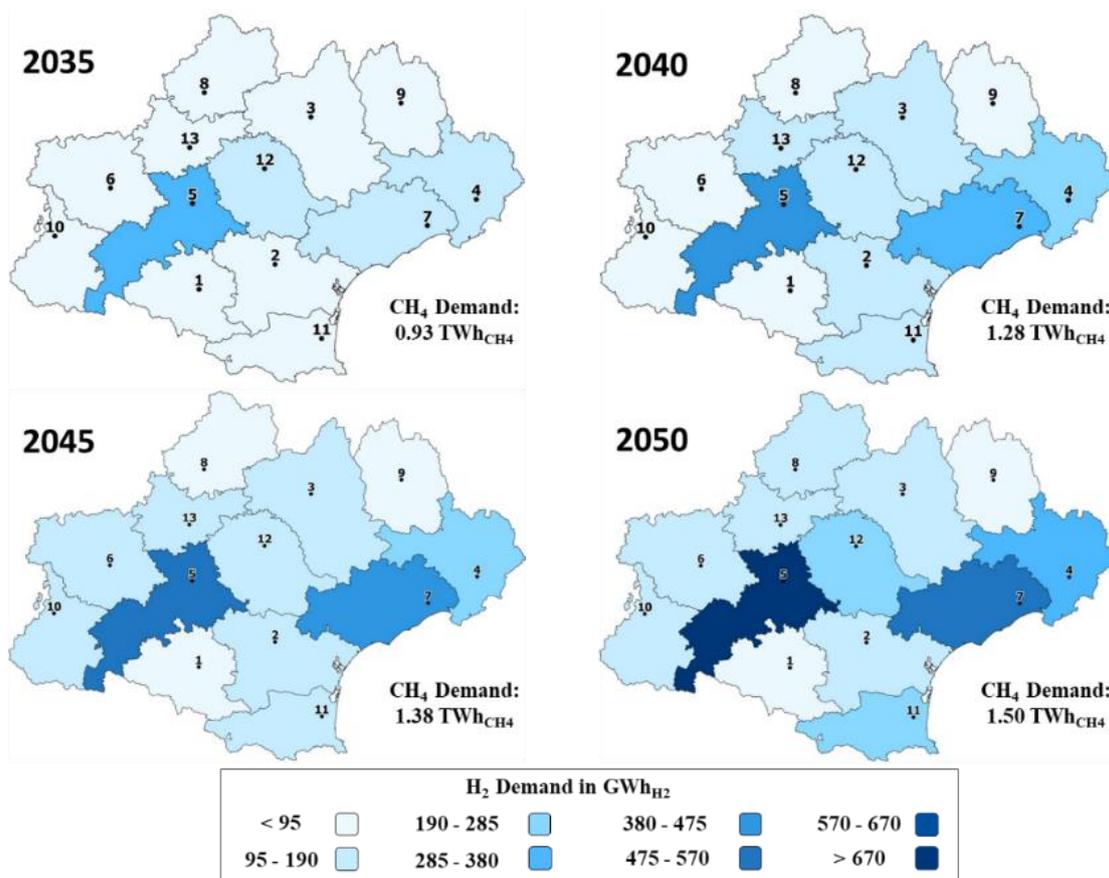


Figure 4.2. Hydrogen and methane demand in the period 2035-2050

4.6 Feedstock estimate

The estimation of the available resources for the production of hydrogen and methane through Power-to-Gas systems is key, since this influences the viability of the HMSC, conditioning aspects such as LCOE, CO₂ emissions, location of the units, and hydrogen transport network. The available primary energy and CO₂ sources on the territory are described below. They correspond to the available feedstock in the territory to meet the expected demands.

4.6.1.1 Energy sources

For electricity, the solar source represents 58-60%, the wind source 38-39%, and the water source 1-2% of the total energy available for PtG between 2035 and 2050 (BRU et al., 2019; Région Occitanie / and Pyrénées-Méditerranée, 2017). Currently, the production of renewable energy in the region is 28 TWh, with electricity accounting for 13.6 TWh of this total (Région Occitanie/Pyrénées-Méditerranée, 2018). Electricity from the national power grid can be used as a back-up. All the natural gas needed to satisfy hydrogen and methane demands through fossil sources must be imported, since there are no oil/gas natural fields in the territory. The available energy in the territory is shown in Figure 4.3 and Figure 4.4.

4.6.1.2 CO₂ sources

The main sources of CO₂ for this study are from the processes of methanization and gasification of biomass, according to the resources that can be provided for methanation (ADEME, 2018; E&E consultant et al., 2014). The total available CO₂ amount could satisfy at most 79% of the methane demand, according to Sabatier reaction (see Eq. 2.3). Its distribution in the territory is shown in Figure 4.5. Industrial CO₂ sources can be used when required.

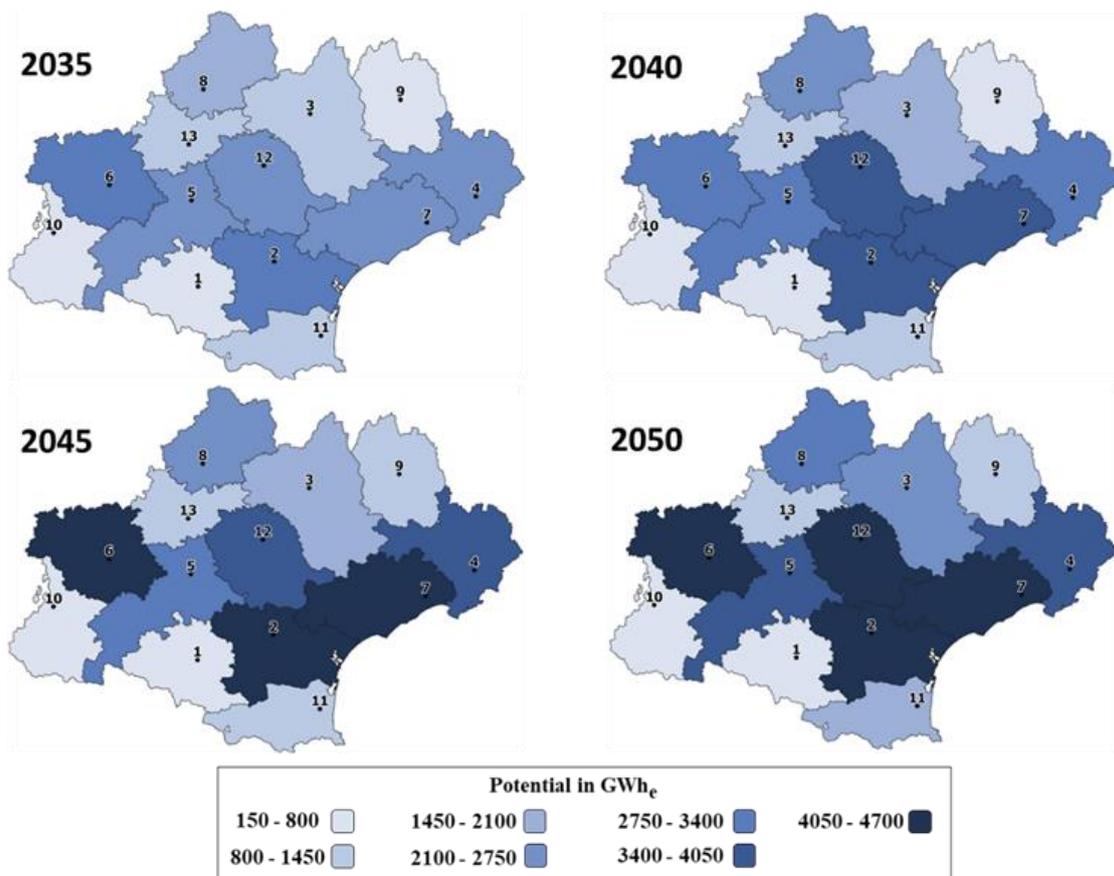


Figure 4.3. Regional electricity availability in 2035-2050

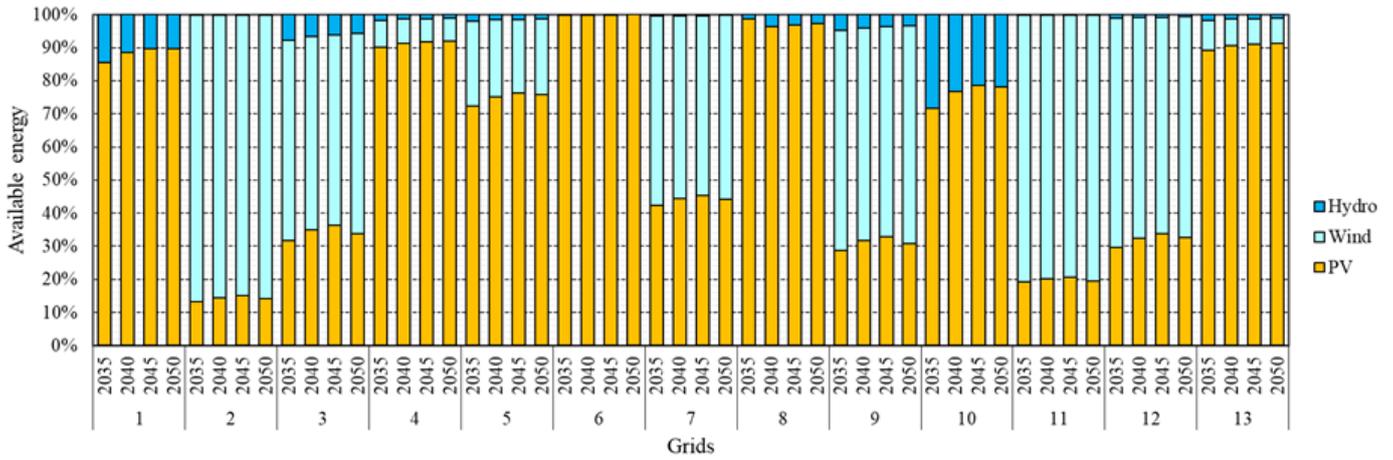


Figure 4.4. Available energy in each grid by source

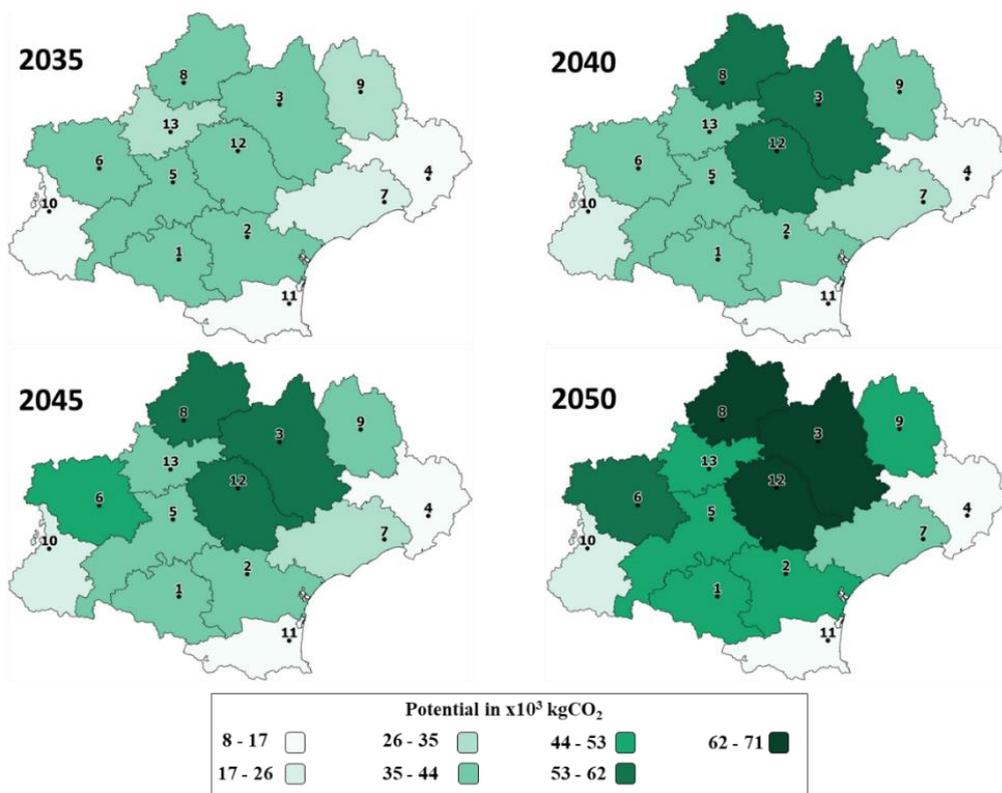


Figure 4.5. Regional CO₂ availability for methane production in 2035-2050

4.7 Conclusion

The case study that will serve as an illustration of the proposed methodology for HMSC design was developed in this chapter through the collection of techno-economic data to be used in the optimization model, the territorial characteristics, and the available feedstock in the Occitania region. The data and HMSC configuration presented in this chapter can be viewed as a supplement to the next chapters, which will address the following aspects:

- Chapter 5: HMSC to satisfy a demand for methane (i.e., MSC), in which all the hydrogen produced will be used as a raw material for methane production through methanation.
- Chapter 6: HMSC to satisfy a demand for methane and hydrogen, only using the PtG concept.
- Chapter 7 and 8: HMSC to meet the demand for methane and hydrogen through renewable and fossil energy sources.

5 AN OPTIMIZATION FRAMEWORK FOR POWER-TO-METHANE SYSTEM DESIGN AND DEPLOYMENT

ABSTRACT

This chapter focuses on the design of methane supply chains based on Power-to-Gas systems, where all the hydrogen produced is used as a feedstock to meet the demand for methane. This work focuses on the deployment and design of an MSC, using an optimization formulation based on Mixed-Integer Linear Programming (MILP) implemented in the General Algebraic Modeling System (GAMS) environment with a multi-period horizon (2035-2050). The objective to minimize is the Total Annual Cost (TAC) of the entire MSC throughout the total horizon studied. Key optimization variables are the number and size of unit plants and hydrogen transport. The methodology is applied to the case study of the Occitania region in France. The parameters that have been chosen were taken from the scientific literature review and some reports released by French authorities. Two scenarios are analyzed considering low and high demands for methane.

RÉSUMÉ

Ce chapitre est centré sur la conception de chaînes logistiques de méthane basées sur des systèmes Power-to-Gas, où tout l'hydrogène produit est utilisé comme matière première pour répondre à la

demande de méthane. Ce travail se concentre sur le déploiement et la conception d'une MSC, en utilisant une formulation d'optimisation basée sur la programmation linéaire mixte en nombres entiers (MILP) mise en œuvre dans l'environnement du *General Algebraic Modeling System* (GAMS) avec un horizon multi-période (2035-2050). L'objectif à minimiser est le coût annuel total (TAC) de l'ensemble du MSC tout au long de l'horizon étudié. Des variables d'optimisation clés sont le nombre et la taille des unités de production et le transport de l'hydrogène. La méthodologie est appliquée à l'étude de cas de la région Occitanie en France. Les paramètres qui ont été choisis sont issus de la littérature scientifique et de certains rapports institutionnels français. Deux scénarios sont analysés en considérant une demande faible et élevée de méthane.

Abbreviations

ADEME	Agence de l'environnement et de la maîtrise de l'énergie
CO2SC	Cost of carbon dioxide source
ESCh	Cost of electricity for hydrogen production
FCCg	Facility capital cost of methane
FOCg	Facility operating cost of methane
FCCh	Facility capital cost of hydrogen
FOCh	Facility operating cost of hydrogen
FCCs	Storage capital cost of hydrogen
FOCs	Storage operating cost of hydrogen
GAMS	General Algebraic Modeling System
GWP	Global warming potential
HHV	High Heating Value
HSC	Hydrogen supply chain
LCA	Life Cycle Assessment
LCOE	Levelized cost of energy
LH₂	Liquid hydrogen
MILP	Mixed Integer Linear Programming
MSC	Methane supply chain
PtG	Power-to-Gas
PtH	Power-to-Hydrogen
PtM	Power-to-Methane
SMR	Steam methane reforming
TCCh	Transport capital cost for hydrogen
TOCh	Transport operating cost for hydrogen

5.1 Introduction

Considerable attention has been focused on the design of Power-to-Hydrogen systems in the dedicated literature for several years (Li et al., 2019) since hydrogen can be subsequently used as an energy carrier

or as a reactant for further compounds, such as methane. In this chapter, methanation is considered for the production of synthetic methane due to the additional environmental benefit of the CO₂ reuse: via a methanation step, hydrogen can be converted with CO₂ to produce synthetic methane and water (Sabatier reaction), which facilitates the use of the existing gas network (E&E consultant et al., 2014; Götz et al., 2016).

This chapter targets exclusively the design of methane supply chains based on Power-to-Gas systems, where all the hydrogen produced is used as a feedstock to meet the demand for methane².

The technological aspects and techno-economic analysis of the processes involved in Power-to-Methane systems have been highlighted in several works (Götz et al., 2016). The catalytic and biological reactors are the most advantageous pathways for methane production (Rönsch et al., 2016). Although the catalytic route is available commercially, the biological route is expected to be more profitable (Lecker et al., 2017). Such process conversions have a typical efficiency of 65–75% for Power-to-Hydrogen (PtH) and 75% for hydrogen to methane (High Heating Value, HHV) (Götz et al., 2016). This Power-to-Methane (PtM) pathway is explored in what follows, considering hydrogen as an intermediate product to produce methane.

As previously mentioned, PtG is the only technology currently available that offers the possibility of storing energy seasonally, in large quantities, and also the potential of decarbonizing other sectors such as heat production, transport, and some industrial applications (ADEME, 2018; E&E consultant et al., 2014). Besides, PtG allows greater flexibility in energy systems and acts as a link between electric power and natural gas networks. This is one of the pillars of the EU Strategy for Energy System Integration (European Commission, 2020a), which means that the system is planned and operated as a whole, linking different energy carriers, infrastructures, and end-uses, thus increasing the system efficiency and providing more flexibility to achieve larger emissions cuts. Several countries have conducted some studies to include these systems within their energy transition roadmaps (ADEME, 2018; E&E consultant et al., 2014; Gigler and Weeda, 2018; Judd and Pinchbeck, 2013; Thomas et al., 2016), in particular in Germany where PtG technology is seen as a possible cornerstone of the future energy system (Schiebahn et al., 2015; Varone and Ferrari, 2015). These studies are consistent with the major role that PtG systems are expected to play from 2030 with a projection of 80 to 100% renewable electricity production in 2050 (Belderbos et al., 2015). They highlight that the profitability of PtG depends on local circumstances and market conditions (Holstein and van den Noort, 2018).

² This work will be submitted for publication in *Computers and Chemical Engineering* from ELSEVIER

To include PtG into energy systems, technical demonstration and systems integration are of major importance (Wulf et al., 2018). The integration of PtG systems with other technologies or their role within the energy network has been widely addressed (Al Rafea et al., 2017; Nastasi et al., 2017; Qadrdan et al., 2015; Vandewalle et al., 2015). Within these possible interactions, the coupling of biomass transformation (i.e., methanization or gasification) and PtG systems has been particularly addressed, since it allows the increase of the efficiency of both processes through the recovery of by-products (Collet et al., 2017; Frigo and Spazzafumo, 2018; Rosenfeld et al., 2020).

It must be yet emphasized that PtG systems face some challenges.

1. Electrolysis is an essential step in the development of PtG systems. In the first stage of its application, hydrogen is expected to be used as the main product (Power-to-Hydrogen) and then methane as a secondary one (ADEME, 2018; E&E consultant et al., 2014). This requires hydrogen injection into existing gas networks or the development of a new dedicated infrastructure for hydrogen. For existing networks, the blending limit is currently up to 10% (volume basis) with most of the countries below 2% (Quarton and Samsatli, 2018). Increasing the presence of hydrogen in the natural gas grid leads to different values of the Wobbe Index that can affect the operation of downstream equipment, lower energy density, and higher pressure drops (need for relocating recompression stations with a too high content) (Melaina et al., 2013). This implies hydrogen embrittlement for high-pressure pipelines that would require a change in the pipeline integrity system and closer monitoring (Dodds and Demoullin, 2013; Dolci et al., 2019). Although the transport of hydrogen through pipelines could be economically competitive in the medium or long term by modifying the natural gas network or adding inhibitor admixture (Cerniauskas et al., 2020), methane can be stored and injected without technical restrictions into the natural gas infrastructure already in place (Khalilpour, 2018). Therefore, in terms of the use of existing well-developed infrastructure, methanation is preferable. This issue will thus be specifically addressed in this chapter.
2. The cost of methane produced by PtG systems is another challenge. Some indicators suggest that methane would not be competitive with natural gas or other alternatives such as biomethane. The cost of industrial natural gas in Europe is approximately 33 €/MWh (Eurostat, 2020). Biomethane produced in France is assessed from 94 to 122 €/MWh in 2018 and could decrease to 66-82 €/MWh in 2030 (ENEA, 2018). In contrast, the cost of methane from PtG is about 101-160 €/MWh in 2020 and could reach 95 to 150 €/MWh in 2030 (E&E consultant et al., 2014). This is mainly due to the cost of electricity. It can reach a minimum value of about 22-45 €/MWh with a zero electricity cost with 2000-3000 operating hours (Vandewalle et al., 2015). With few operating hours and non-zero electricity costs, this system is not profitable and is challenged by other options.

However, PtM systems offer advantages despite their high cost. They provide a bidirectional interaction between the natural and electricity network, improving the flexibility of both markets and reduces operational risks (Khalilpour, 2018), which are considered as drivers for their deployment.

A literature review has shown that numerous researches have been carried out taking into account different issues of PtG, where possible applications, main technological blocks, energy sources, and CO₂ sources are highlighted (Götz et al., 2016, 2014; Lewandowska-Bernat and Desideri, 2018; Li et al., 2019; Rönsch et al., 2016; Wulf et al., 2018). In this regard, most investigations have been developed to study the hydrogen supply chain based on PtG systems (Li et al., 2019). Few studies have considered methane as the main product (Götz et al., 2016) and the logistical aspects of gas production from these systems are not sufficiently addressed in which the location, size, and the number of production, storage, and transport units have to be determined.

The scientific issue of this chapter is thus to contribute to the development of a methodological framework for the deployment and design of PtG systems, involving Methane Supply Chains (MSC) from an optimization viewpoint.

This chapter is divided into five sections. The problem definition and the mathematical model are described in Sections 2 and 3, respectively, following this introduction section. Section 4 shows the results obtained are analyzed for two demand profiles and a sensitivity assessment is also conducted based on three parameters: the cost of electricity to produce hydrogen, hydrogen storage capacity, and the cost of CO₂. Finally, the conclusions and perspectives are presented in Section 5.

5.2 Problem definition

From the literature review performed in Chapter 3, it can be highlighted that the optimization problem is mainly addressed with a MILP formulation to minimize costs. Most studies emphasize the role that these systems can play according to certain scenarios (i.e., the flexibility of the electricity network, linking the electricity and gas network, meeting estimated demands under low CO₂ emission scenarios, renewable electricity penetration in the energy grid, among others). The models used are: a) equilibrium models, which assumes perfect competition, and economic rationality (i.e, Balmorel (Bramstoft et al., 2020) and TIMES (Blanco et al., 2018b)), b) geographic explicit cost optimization models (Mesfun et al., 2017), and c) bi-level models and economic dispatch formulation (Chuan He et al., 2017; He et al., 2018; Li et al., 2016), with limited attention to logistical aspects (i.e., numbers, size, rate and location of production facilities, storage, and transport units).

This study proposes a contribution to bridging this gap, taking as a starting point the previous work dedicated to hydrogen supply chains carried out by Almaraz et al. (De-León Almaraz et al., 2015). Its

scientific objective is to contribute to the development of a comprehensive optimization model that can determine the design and operation of an MSC. It is mainly based on the conversion of renewable energies and CO₂ through PtG systems such that the Total Annual Cost (TAC) is minimized. The main contributions are:

- Addressing various aspects of the MSC: primary resources, production units, transport, and storage.
- Embedding the possibility of using locally produced electricity (i.e. solar, wind, and water) and importing electricity from the national network.
- Considering the evolution of the logistics chain over time according to different scenarios.
- Determining the size, location, and the number of units involved in the supply chain.

The main research questions tackled in this chapter are:

- Where hydrogen and methane production plants, storage units should be located to meet a demand for methane in specific periods by minimizing costs?
- Is it necessary to use hydrogen transport between grids? If so, where will the hydrogen be transported from?
- What will be the production of each grid?
- How does the availability of CO₂ and electricity from renewable sources affect the selection of grids for hydrogen and methane production?

The model presented has as limitations the satisfaction of a given demand for methane through MSC in a steady state. Furthermore, the seasonal aspects of electricity production from renewable energy during the year, the CO₂ supply chain management, and interaction with other technologies and demands (i.e., Steam Methane Reforming SMS, Methanization) are beyond the scope of this research. The HMSC supply chain that will be studied in this chapter can be visualized in Figure 5.1.

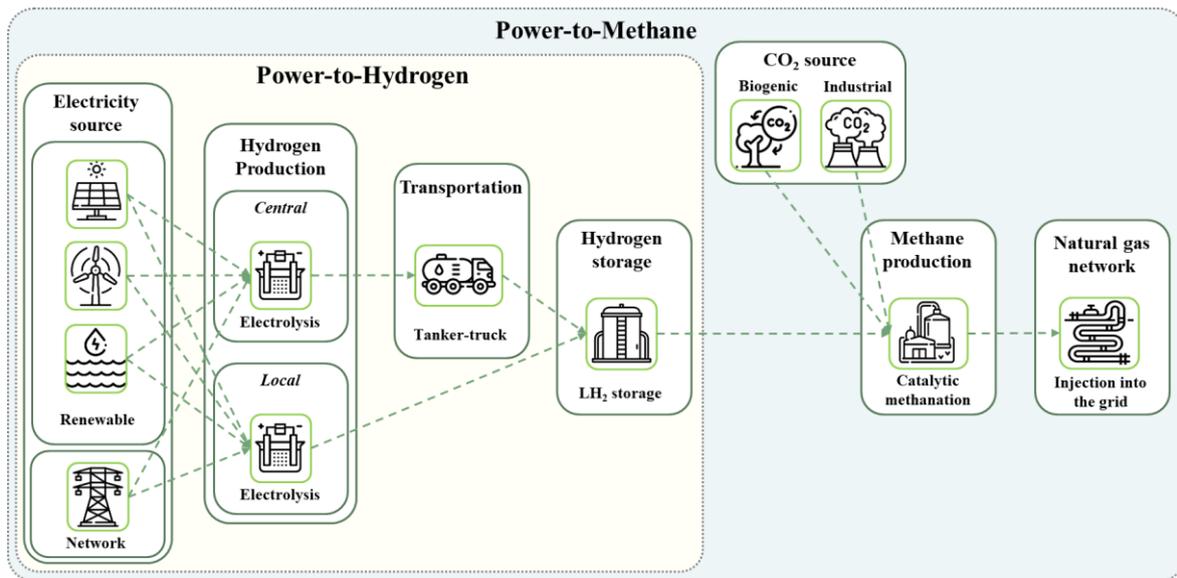


Figure 5.1. HMSC supply chain based for the case study

The problem formulation for the design and operation of an MSC is based on the guidelines proposed by De-León Almaraz et al. (De-León Almaraz et al., 2015) for HSC considering a 3-tier approach is visualized in Figure 5.2. The model is formulated with the following assumptions:

- The territory to be studied is divided into grids.
- In each grid, production, and storage plants of hydrogen and methane can be installed.
- Hydrogen transport between grids is possible.
- The methane demand is satisfied only through Power-to-Gas systems.
- All the hydrogen produced is used to meet methane demand.
- All the gas produced is injected directly into the gas network.
- There is limited availability of primary energy and CO₂ sources. However, it is possible to import primary resources if necessary.
- Transport may exist only between grids. Transport within the grid is not modeled.
- A multi-period formulation is involved.

The decision variables considered are the following:

- Number, size, capacity, production rate, and location of hydrogen and methane production and storage units.
- The number of transport units and the flow of hydrogen transported between grids.
- Specification of the sources and quantity of electricity and CO₂ consumed.
- Costs related to each section of the supply chain.

So as to:

- Minimize TAC.

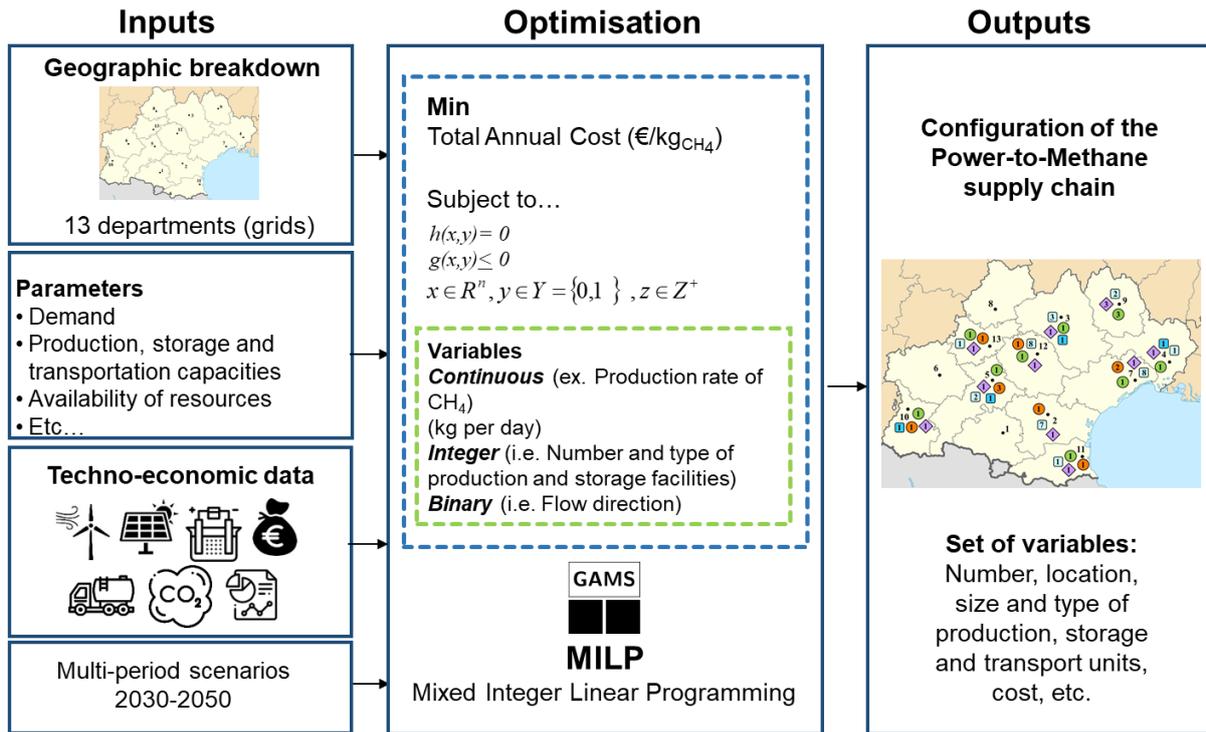


Figure 5.2. Methodology framework for the HMSC (Chapter 4)

5.3 Mathematical model

Sets

c	CO ₂ source
e	energy source
g	grid
i	final product
j	size of the production facilities
l	type of hydrogen transportation modes
p	type of hydrogen production technology
q	type of methane production technology
s	type of hydrogen storage facility
t	time period of the planning horizon

Parameters

MD_{ig}	Total demand for methane in period t in form i in grid g (kg per year)
r	Discount rate

Sources

EA_{egt}	Initial average availability of primary energy source e in grid g during time period t (MWh per year)
CA_{cgt}	Initial average availability of CO ₂ source c in grid g during time period t (Kg per year)
UEC_{et}	Unit electricity cost for source e produced in period t (€ per kg)

UCC_{ct}	Unit production cost for product i produced by plant type p (€ per kg)
<i>Production data</i>	
$HNPO_{pijg}$	Initial number of plants of type p for product form i in size j in grid g
$HPCapmax_{pij}$	Maximum production capacity of electrolyzer type p for product form i and size j (kg per year)
$HPCapmin_{pij}$	Minimum production capacity of electrolyzer type p for product form i and size j (kg per year)
$HSCapmax_{sit}$	Maximum storage capacity of storage type s for product form i (kg per year)
$HSCapmin_{sit}$	Minimum storage capacity of storage type s for product form i (kg per year)
$MNPO_{qijg}$	Initial number of plants of type q for product form i in size j in grid g
$MPCapmax_{qij}$	Maximum production capacity of catalytic reactor type q for product form i (kg per year)
$MPCapmin_{qij}$	Minimum production capacity of catalytic reactor type q for product form i (kg per year)
PCC_{pi}	Capital cost of establishing electrolyser type p producing product form i (€)
SCC_{si}	Capital cost of establishing storage type s storing product form i (€)
SSF	Safety stock factor of primary source within a grid (%)
δ_{cqi}	Rate of utilization of primary CO ₂ source c by plant type q and size j (Kg-CO ₂ per Kg-H ₂)
γ_{epj}	Rate of utilization of primary energy source e by plant type p and size j (MWh per Kg H ₂)
ϕ_{H2}	Rate of utilization of hydrogen for methane production (Kg H ₂ per Kg CH ₄)
<i>Storage data</i>	
$HUSC_{si}$	Unit storage cost for hydrogen at storage type s in form i (€ per kg-year)
β	Factor of hydrogen storage
$NHSO_{sig}$	Initial number of storage facilities of type s for product form i in grid g
<i>Transportation data</i>	
$AD_{gg'}$	Average delivery distance between g and g' by transportation l (km per trip)
DW_l	Driver wage of transportation mode l (€ per hour)
FE_l	Fuel economy of transportation mode l (km per liter)
FP_l	Fuel price of transportation mode l (€ per liter)
GE_l	General expenses of transportation mode l (€ per year)
LUT_l	Load and unload time of product for transportation mode l (hours per trip)
ME_l	Maintenance expenses of transportation mode l (€ per km)
$Q_{max_{il}}$	Maximum flow rate of product form i by transportation mode l (kg per year)
$Q_{min_{il}}$	Minimum flow rate of product form i by transportation mode l (kg per year)
SP_l	Average speed of transportation mode l (km per hour)
$TCap_{il}$	Capacity of transportation mode l transporting product form i (kg per trip)
TMA_l	Availability of transportation mode l (hours per year)
TMC_{il}	Cost of establishing transportation mode l for product form i (€)
W_l	Weight of transportation mode l (tons)

Variables

Continuous variables

C_{cgt}	Average availability of CO ₂ source c in grid g during time period t (Kg per year)
CSC_t	CO ₂ source cost during time period t (€)
E_{egt}	Average availability of primary energy source e in grid g during time period t (MWh per year)
ESC_t	Primary energy source cost during time period t (€)
FC_t	Fuel cost in period t (€ per year)
GC_t	General cost in period t (€ per year)
HD^I_{igt}	Imported demand of product form i to grid g in period t (kg per year)
HD^L_{igt}	Demand for product i in grid g in period t satisfied by local production (kg per year)
HD^T_{igt}	Total demand for hydrogen in form i in grid g in period t (kg per year)
$HFCC_t$	Facility capital cost of hydrogen production in period t (€)
$HFOC_t$	Facility operating cost of hydrogen production in period t (€ per year)
HPR_{pijgt}	Production rate in plant type p produced in form i and size j in grid g in period t (kg per year)
HP^T_{igt}	Total production rate of hydrogen in form i in grid g in period t (kg per year)
HS^T_{igt}	Total average inventory of hydrogen in form i in grid g in period t (kg/year)
$HTCC_t$	Hydrogen transportation capital cost in period t (€)
$HTOC_t$	Hydrogen transportation operating cost in period t (€ per year)
$IPCS_{cgt}$	Imported CO ₂ from source c in grid g in period t
$IPES_{egt}$	Imported electricity from source e in grid g in period t
LC_t	Labour cost in period t (€ per year)
MC_t	Maintenance cost in period t (€ per year)
$MFCC$	Facility capital cost of methane production in period t (€)
$MFOC_t$	Facility operating cost of methane production in period t (€ per year)
MPR_{qijgt}	Production rate in plant type q produced in form i and size j in grid g in period t (kg per year)
MP^T_{igt}	Total production rate of methane in form i in grid g in period t (kg per year)
$MTAC$	Total annual cost of the network in the whole period (€)
$MTAC_t$	Total annual cost of the network in period t (€ per year)
$Q_{ilgg't}$	Flow rate of product i by transportation mode l between g and g' (kg per year)
$SFCC_t$	Facility capital cost of hydrogen storage in period t (€)
$SFOC_t$	Facility operating cost of hydrogen storage in period t (€ per year)
$V_{ilgg't}$	Artificial variable with values between 0 and 1
<i>Integer variables</i>	
HIP_{pijgt}	Number of installed plants of hydrogen production type p producing in form i and size j in grid g in period t
HIS_{sijgt}	Number of installed plants of hydrogen storage type s producing in form i and size j in grid g in period t
HNP_{pijgt}	Number of plants of hydrogen production type p producing product form i and size j in grid g in period t
MIP_{qijgt}	Number of installed plants of methane production type q producing in form i and size j in grid g in period t

MNP_{qijgt}	Number of plants of methane production type q producing product form i and size j in grid g in period t
NHS_{sigt}	Number of hydrogen storage facilities of type s for product form i in grid g in period t
$NTU_{ilgg't}$	Number of transport units between g and g' in period t
<i>Binary variables</i>	
$X_{ilgg't}$	1 when the product form i is to be transported from grids g to g' in period t
Y_{igt}	1 if product form i is to be exported from grid g or 0 otherwise in period t
Z_{igt}	1 if product form i is to be imported from grid g or 0 otherwise in period t

The formulation aims to meet a demand of methane “i”, considering: the potential of each grid “g”, the availability of electricity source “e” and CO₂ “c”, production type “p”, storage option “s”, transport option “l”, for each period “t”. The model involves a steady-state balance of mass and energy, with the possibility of the import of primary resources. The fundamental constraint is the resource balance, which considers all possible flows of each resource into or out of each grid (import or export). Only the key points are discussed below.

5.3.1 Demand constraints

5.3.1.1 Methane demand

The total demand for methane (MD_{it}^T , kg/year) (see Eq. 5.1) is met by the production of methane from each grid (MPR_{igt}^T , kg/year).

$$MD_{it}^T = \sum_g MP_{igt}^T \quad \forall i, t \quad \text{Eq. 5.1}$$

5.3.1.2 Hydrogen demand

Methane production (MP_{igt}^T , kg/year) requires hydrogen production (HD_{igt}^T , kg/year) according to the Sabatier reaction (see Eq. 2.3). It is shown in Eq. 5.2. The hydrogen stoichiometric coefficient (φ_{h_2}) is multiplied by a safety factor (SSF = 5%) that takes into account a small inventory of hydrogen.

$$HD_{igt}^T = \varphi_{h_2} SSF \sum MP_{igt}^T \quad \forall i, g, t \quad \text{Eq. 5.2}$$

The total demand for hydrogen (HD_{igt}^T , kg/year) is met by the local production due to the facilities established within a grid (HD_{igt}^L , kg/year), or by importing from other neighboring grids (HD_{igt}^I , kg/year). Furthermore, the demand for hydrogen in grid g satisfied by importation is calculated by taking into account the flow of hydrogen from transportation modes ($Q_{ilg'gt}$, kg/year). The impossibility of exporting and importing hydrogen from the same grid is stated as a restriction ($g \neq g'$) expressed in Eq. 5.3, Eq. 5.4 and Eq. 5.5.

$$HD_{igt}^L \leq HPR_{igt}^T \quad \forall i, g, t \quad \text{Eq. 5.3}$$

$$HD_{igt}^I = \sum_{l, g'} Q_{ilg'gt} \quad \forall i, g, t; g \neq g' \quad \text{Eq. 5.4}$$

$$HD_{igt}^T = HD_{igt}^L + HD_{igt}^I \quad \forall i, g, t \quad \text{Eq. 5.5}$$

5.3.2 Production facilities constraints

The total production of methane (MP_{igt}^T , kg/year) is equal to the total production of each methanation plant (MPR_{qijgt} , kg/year) (See Eq. 5.6). This production will be restricted according to the capacity of the methanation plants installed (See Eq. 5.7 and Eq. 5.8).

$$MP_{igt}^T = \sum_{pj} MPR_{qijgt} \quad \forall i, g, t \quad \text{Eq. 5.6}$$

$$\sum_{pj} MPCap_{qij}^{\min} MNP_{qijgt} \leq MP_{igt}^T \leq \sum_{pj} MPCap_{qij}^{\max} MNP_{qijgt} \quad \forall i, g, t \quad \text{Eq. 5.7}$$

$$MPCap_{qij}^{\min} MNP_{qijgt} \leq MPR_{qijgt} \leq MPCap_{qij}^{\max} MNP_{qijgt} \quad \forall q, i, j, g, t \quad \text{Eq. 5.8}$$

A mass balance is carried out on each grid (see in Eq. 5.9) to determine the total production of hydrogen in each grid (HP_{igt}^T , kg/year). The imported ($Q_{ilg'gt}$, kg/year) and exported ($Q_{ilg'g't}$, kg/year) hydrogen and the total demand (HD_{igt}^T , kg/year) are considered. Then the total hydrogen produced is equal to the total hydrogen produced by each plant installed in the same grid (HPR_{pigt} , kg/year) with a limitation related to the production capacity of each plant (see Eq. 5.10 to Eq. 5.12).

$$HP_{igt}^T = \sum_{l, g'} (Q_{ilg'gt} - Q_{ilg'g't}) + HD_{igt}^T \quad \forall i, g, t \quad \text{Eq. 5.9}$$

$$HP_{igt}^T = \sum_{pj} HPR_{pigt} \quad \forall i, g, t \quad \text{Eq. 5.10}$$

$$\sum_{pj} HPCap_{prij}^{\min} HNP_{prijgt} \leq HP_{igt}^T \leq \sum_{pj} HPCap_{prij}^{\max} HNP_{prijgt} \quad \forall i, g, t \quad \text{Eq. 5.11}$$

$$HPCap_{prij}^{\min} HNP_{prijgt} \leq HPR_{pigt} \leq HPCap_{prij}^{\max} HNP_{prijgt} \quad \forall p, i, j, g, t \quad \text{Eq. 5.12}$$

5.3.3 Transportation constraints

The flow of hydrogen from one grid to another ($Q_{ilg'g't}$, kg/year) can also occur if necessary. For this purpose, a minimum (Q_{il}^{\min} , kg/year) and maximum (Q_{il}^{\max} , kg/year) flow capacities are introduced, which are associated with the transport capacities of the chosen option as expressed in Eq. 5.13.

$$Q_{il}^{\min} X_{ilg_g't} \leq Q_{ilg_g't} \leq Q_{il}^{\max} X_{ilg_g't} \quad \forall i, l, g, g', t; \quad g \neq g' \quad \text{Eq. 5.13}$$

The flows between different grids can only occur in one direction, as expressed in (Eq. 5.14) as a restriction that a grid cannot import and export at the same time:

$$X_{ilg_g't} + X_{ilg'gt} \leq 1 \quad \forall i, l, g, g', t; \quad g \neq g' \quad \text{Eq. 5.14}$$

A particular network can only import products from neighboring networks or export products to other networks, but can never import or export both at the same time. It is expressed in Eq. 5.15 to 5.17.

$$Y_{igt} \geq X_{ilg_g't} \quad \forall i, l, g, g', t; \quad g \neq g' \quad \text{Eq. 5.15}$$

$$Z_{igt} \geq X_{ilg'gt} \quad \forall i, l, g, g', t; \quad g \neq g' \quad \text{Eq. 5.16}$$

$$Y_{igt} + Z_{igt} \leq 1 \quad \forall i, g, t \quad \text{Eq. 5.17}$$

5.3.4 Storage facilities constraints

The total inventory of hydrogen in grid g in the period t (HS_{igt}^T , kg/year) is equal to a function of the corresponding demand (HD_{igt}^T , kg/year) multiplied by the factor of hydrogen storage (β) (see Eq. 5.18).

The parameter (β) is introduced to cover uncertainties in both supply and demand as well as plant interruptions.

$$HS_{igt}^T = \beta HD_{igt}^T \quad \forall i, g, t \quad \text{Eq. 5.18}$$

This is restricted to the minimum and maximum installed storage capacity (Eq. 5.19).

$$\sum_s HSCap_{si}^{\min} NHS_{sigt} \leq HS_{igt}^T \leq \sum_s HSCap_{sit}^{\max} NHS_{sigt} \quad \forall i, g, t \quad \text{Eq. 5.19}$$

In addition to the above constraints, it is added that all variables must be positive or equal to zero.

Based on the technological options available, the HMSC used in this chapter can be visualized in Figure 5.3. Each grid (g or g') represents a department in the Occitania region. They enclose different hydrogen plants, storage units, and methane plants, and refueling stations. The transportation of hydrogen is used to transport the hydrogen from g to g' , if necessary. Local (intra-regional) hydrogen distribution and transportation will not be considered.

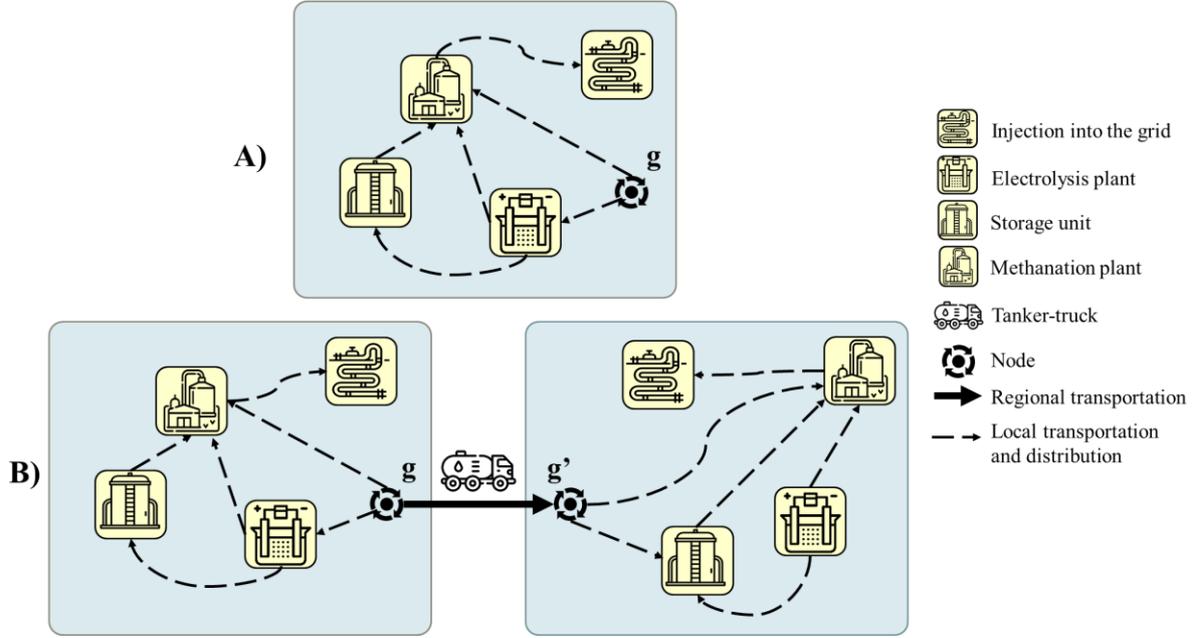


Figure 5.3. Possible grids configurations: A) without transportation and B) with transportation

5.3.5 Determination of the number of units

5.3.5.1 Number of plants

The total number of plants installed during each period for methane production (NMP_{qijgt}) and hydrogen storage and production (NHP_{pijgt}) is determined from Eq. 5.20 to 5.22. The number of plants installed at the beginning is indicated as parameters ($MNPO_{qijg}$, $NHSO_{sig}$, $MNPO_{qijg}$). Hydrogen plants are not installed if renewable energy is not produced within the grid (see Eq. 5.23). Methane plants are not installed if carbon dioxide sources are not available in the grid (see Eq. 5.24).

$$NMP_{qijgt} = NMP_{qijgt-1} + MIP_{qijgt} \quad \forall q, i, j, g, t \neq 1 \quad \text{Eq. 5.20}$$

$$NHP_{pijgt} = NHP_{pijgt-1} + HIP_{pijgt} \quad \forall p, i, j, g, t \neq 1 \quad \text{Eq. 5.21}$$

$$NHS_{sijgt} = NHS_{sijgt-1} + HIS_{sijgt} \quad \forall s, i, j, g, t \neq 1 \quad \text{Eq. 5.22}$$

$$HIP_{p(RES)ijgt} = 0 \quad \text{if } EA_{e(RES)gt} = 0 \quad \forall p(RES), i, j, g, t \quad \text{Eq. 5.23}$$

$$MIP_{qijgt} = 0 \quad \text{if } CA_{cgt} = 0 \quad \forall q, i, j, g, t \quad \text{Eq. 5.24}$$

5.3.5.2 Number of transport units

The number of transport units ($NTU_{ilg'g't}$) is estimated from Eq. 5.25. The continuous variable ($V_{ilg'g't}$) (see Eq. 5.27 and 5.27) represents the partial use of a transport unit.

$$NTU_{ilg'g't} = \left(\frac{Q_{ilg'g't}}{TMA_l TCap_{il}} \left(\frac{2AD_{gg'}}{SP_l} + LUT_l \right) \right) + V_{ilg'g't} \quad \forall i, l, g, g', t \quad g \neq g' \quad \text{Eq. 5.25}$$

$$V_{ilg'g't} \geq 0 \quad \forall i, l, g, g', t; \quad g \neq g' \quad \text{Eq. 5.26}$$

$$V_{ilg' t} \leq X_{ilg' t} \quad \forall i, l, g, g', t \quad g \neq g' \quad \text{Eq. 5.27}$$

5.3.6 Energy constraint

Eq. 5.28 indicates the energy available to produce hydrogen (E_{egt} , GWh/year). This depends on the energy available over the territory (EA_{egt} , GWh/year), the energy imported from the grid ($IPES_{egt}$, GWh/year), and the energy consumed to produce hydrogen ($SSF \sum_{pji} \gamma_{epj} HPR_{pijgt}$, GWh/year). A rate of utilization of primary energy source e by plant type “ p ” and size “ j ” (γ_{epj} , Gwh/kg) is also introduced.

$$E_{egt} = EA_{egt} + IPES_{egt} - SSF \sum_{pji} \gamma_{epj} HPR_{pijgt} \quad \forall e, t, g; g \neq g' \quad \text{Eq. 5.28}$$

5.3.7 CO₂ source constraint

The CO₂ available amount (C_{cgt} , kg-CO₂) to produce methane is calculated in Eq. 5.29. It is composed of three terms the availability of CO₂ sources on the territory, the imported CO₂, and the consumed CO₂ during methane production, from left to right, respectively. In the last term, the amount of CO₂ needed to produce 1 kg of methane (δ_{cpj} , kg-CO₂/kg-CH₄) and a safety stock factor (SSF equal to 5%) for storing a small inventory of CO₂ is involved.

$$C_{cgt} = CA_{cgt} + IPCS_{cgt} - SSF \sum_{qji} \delta_{cpj} MPR_{qijgt} \quad \forall c, t, g; g \neq g' \quad \text{Eq. 5.29}$$

5.3.8 Cost objective

5.3.8.1 Facility capital cost

The facility capital cost (FCC _{t} , €/year) involves the Capital Cost for the installation of a (PCC_{xijt} , €) as well as the Number of Facilities installed in each period (IP_{xijgt} , year⁻¹). The hydrogen, storage, and methane facility capital costs are shown in Eq. 5.30 to 5.32, respectively.

$$HFCC_t = \sum_{pij} HPCC_{pij} HIP_{pijgt} \quad \forall t \quad \text{Eq. 5.30}$$

$$SFCC_t = \sum_{sij} SPCC_{sij} SIP_{sijgt} \quad \forall t \quad \text{Eq. 5.31}$$

$$MFCC_t = \sum_{qij} MPCC_{qij} MIP_{qijgt} \quad \forall t \quad \text{Eq. 5.32}$$

5.3.8.2 Transportation capital cost

The transportation capital cost (HTCC _{t} , €/year) is determined by Eq. 5.33. It involves the number of transport units ($NTU_{ilg' t}$, year⁻¹) and the cost of establishing a transportation mode “ l ” (TMC_{il} , €).

$$HTCC_t = \sum_{i,l,g,g'} NTU_{ilg,g'} TMC_{il} \quad \forall t \quad \text{Eq. 5.33}$$

5.3.8.3 Facility operating cost

The operational cost of hydrogen production ($HFOC_t$, €/year), storage hydrogen ($SFOC_t$, €/year) and methane ($MFOC_t$, €/year), the cost of energy (ESC_t , €/year), and the cost of CO₂ (CSC_t , €/year) are calculated in Eq. 5.34 to 5.38. The Unit Cost of Energy source “e” (UEC_{et}) and Unit Cost of CO₂ source “c” (UCC_{ct}) are involved.

$$HFOC_t = \sum_{pig} (HFOC_{pit} HPR_{pig}) + ESC_t \quad \forall t \quad \text{Eq. 5.34}$$

$$SFOC_t = \sum_{sig} HUSC_{si} HS_{igt}^T \quad \forall t \quad \text{Eq. 5.35}$$

$$MFOC_t = \sum_{qijg} MFOC_{qij} MPR_{qijgt} + CO2SC_t \quad \forall t \quad \text{Eq. 5.36}$$

$$ESC_t = \sum_{eg} UEC_{et} \left(IPES_{egt} + SSF \sum_{pji} \gamma_{epj} HPR_{pji} \right) \quad \forall t \quad \text{Eq. 5.37}$$

$$CSC_t = \sum_{cg} UCC_{ct} \left(IPCS_{cgt} + \sum_{pji} \delta_{cuj} MPR_{pji} \right) \quad \forall t \quad \text{Eq. 5.38}$$

5.3.8.4 Transportation operating cost

The operating cost of the hydrogen transport ($HTOC_t$, €/year) is calculated in Eq. 5.44 as the sum of the annual fuel cost (FC_t , €/year), annual labor cost (LC_t , €/year), maintenance cost (MC_t , €/year), general cost (GC_t , €/year) (see Eq. 5.45 to 5.48).

$$HTOC_t = FC_t + LC_t + MC_t + GC_t \quad \forall t \quad \text{Eq. 5.39}$$

$$FC_t = \sum_{i,l,g,g'} FP_l \left(\frac{2AD_{gg'} Q_{ilg,g'}}{FE_l TCap_{il}} \right) \quad \forall t \quad \text{Eq. 5.40}$$

$$LC_t = \sum_{i,l,g,g'} DW_l \left(\frac{Q_{ilg,g'}}{TCap_{il}} \left(\frac{2AD_{gg'}}{SP_l} + LUT_l \right) \right) \quad \forall t \quad \text{Eq. 5.41}$$

$$MC_t = \sum_{i,l,g,g'} ME_l \left(\frac{2AD_{gg'} Q_{ilg,g'}}{TCap_{il}} \right) \quad \forall t \quad \text{Eq. 5.42}$$

$$GC_t = \sum_{i,l,g,g'} GE_l \left(\frac{Q_{ilg,g'}}{TMA_l TCap_{il}} \left(\frac{2AD_{gg'}}{SP_l} + LUT_l \right) \right) \quad \forall t \quad \text{Eq. 5.43}$$

5.3.8.5 Objective function

The objective function is the Total Annual Cost of the MSC ($MTAC$, €), which involves the total annual cost of the methane supply chain in each period ($MTAC_t$, €/year) and a discount rate (r). It is shown in Eq. 5.49 to 5.39.

$$MTAC_t = MFCC_t + HFCC_t + SFCC_t + HTCC_t + MFOC_t + HFOC_t + SFOC_t + HTOC_t \quad \forall t \quad \text{Eq. 5.44}$$

$$MTAC = \sum_t \frac{MTAC_t}{(1+r)^t} \quad \text{Eq. 5.45}$$

5.3.8.6 Levelized Cost of Energy

The Levelized Cost of Energy ($LCOE$, €/MWh) for methane is calculated in Eq. 5.40, which represents the average net present cost of methane for the set of plants over the studied period (2035-2050). It is calculated as the ratio between all the costs of the supply chain over the period divided by a discounted sum of the actual methane amounts delivered (Lai and McCulloch, 2017). The LCOE then represents the average revenue per unit of methane generated that would be required to recover the costs of building and operating a generating plant during an assumed financial life and duty cycle (EIA, 2020).

$$LCOE = \frac{\left(\sum_t \frac{MTDC_t}{(1+r)^t} \right)}{\left(\sum_t \frac{MD_t^T}{(1+r)^t} \right)} \quad \text{Eq. 5.46}$$

The superstructure of the proposed model is shown in Figure 5.4, which summarizes the set of equations and inequalities described. Each grid (g or g') represents a different area. For this case study, they represent each of the departments within the Occitania Region. They enclose different types (p) and sizes (j) of hydrogen plants, different types (s) and sizes (j) of storage technologies, and also different types (q) and sizes (j) of methane plants. The hydrogen plants will be located according to the availability of resources, either energy (e) for hydrogen production or CO_2 (c) for methanation, while a hydrogen storage unit will only be installed if demand for hydrogen is present. The transportation of hydrogen (l) is used to transport the hydrogen from g to g' if it is necessary. It is worth noting that in the case where the entire demand can be met with hydrogen, transport is not required ($g=g'$).

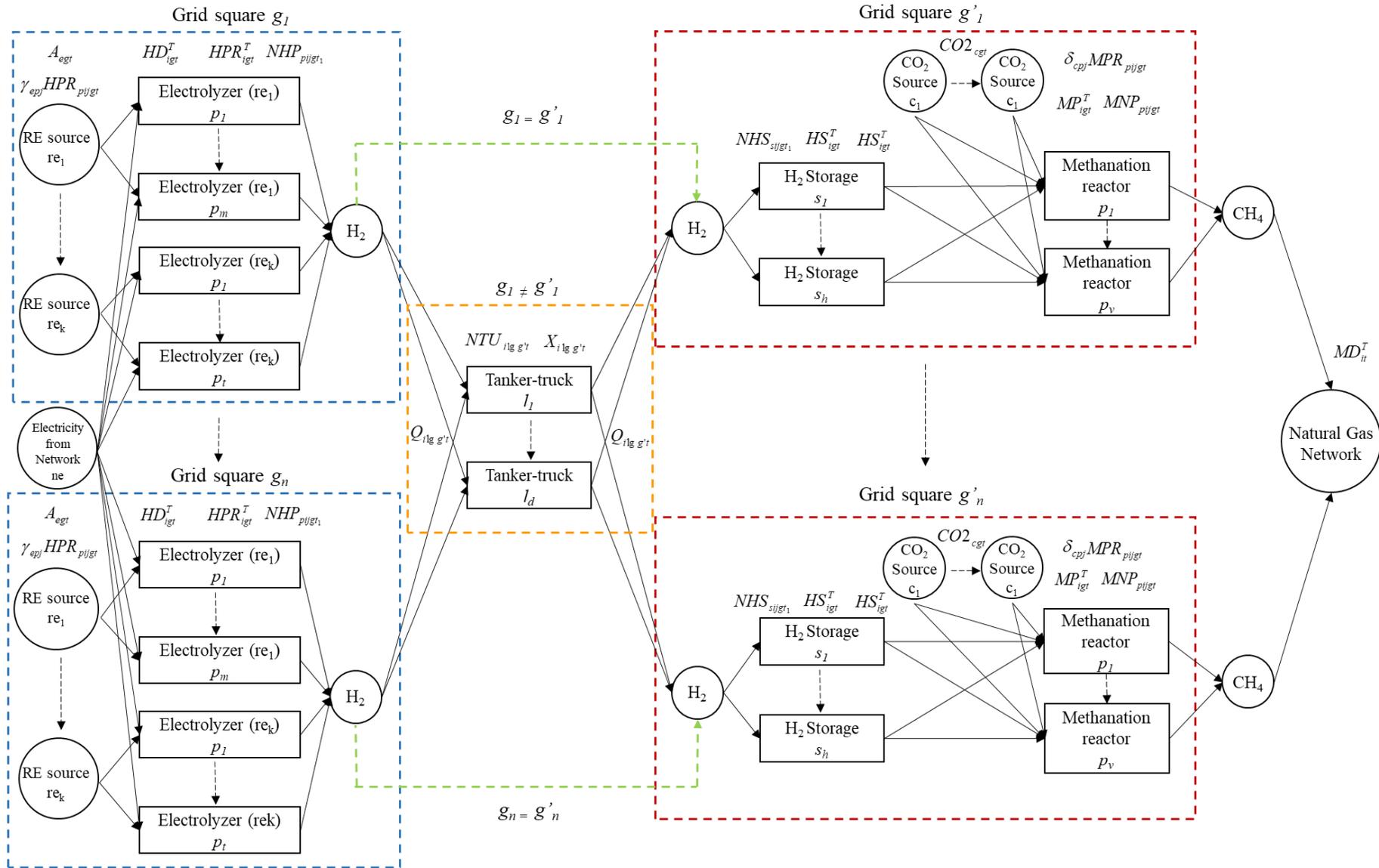


Figure 5.4. Superstructure of the multi-period model for the Methane Supply Chain (MSC)

5.4 Results and discussion

A mono-objective optimization was carried out to obtain the optimal MSC. Two scenarios with low and high demand were studied.

5.4.1 Analysis of scenarios

The decision/operating variables that have been calculated from the optimization run are shown in Table 5.1. In Scenario 1, the totality of the methanation plants is installed in the first two periods, as well as most hydrogen storage units. An increase in demand in Scenario 2 implies several plants and storage units to be installed in the third period. Since there are sufficient resources for hydrogen and methane production in each grid, regional hydrogen imports are not necessary. However, transportation could be key if a greater diversity of end uses for the produced hydrogen is considered (this will be observed in the following chapters).

The total network cost is directly influenced by four parameters: the capital cost of hydrogen plants and storage facilities, electricity, and CO₂ cost. Their contributions can be up to 43%, 15%, 74%, and 14%. Although an increase in demand implies additional capital and operating costs in the MSC, it only represents a 2% increment in the LCOE. This slight variation in the cost of methane can be attributed to the assumption of a constant average cost of electricity. Considering the fluctuating cost of electricity according to the amount of electricity consumed would change this result (Vandewalle et al., 2015).

The detailed configurations of the MSC in each period are displayed in Figure 5.5 and Figure 5.6 and. Facilities are installed progressively from 2035 to 2050. It is observed that the MSC obtained is decentralized. The number of units installed for hydrogen and methane production, in addition to hydrogen storage units, is shown in each grid. The electrolyzers are installed in the first instance to produce hydrogen from the available electricity in the territory. None of the electrolyzers is installed to be directly connected to the national network.

Table 5.1. Mono-objective optimization results of methane supply chain

Year	Scenario 1 : low demand				Scenario 2 : high demand			
	2035	2040	2045	2050	2035	2040	2045	2050
Demand (TWh)	2.296	3.143	3.405	3.67	6.304	8.631	9.35	10.13
Number of hydrogen production facilities	33	47	57	57	91	126	155	155
Number of storage facilities	4	6	8	8	11	15	17	17
Number of methane production facilities	4	6	6	6	10	14	16	16
Number of transport units	0	0	0	0	0	0	0	0
Capital cost (M€)								
Hydrogen plants facilities	149.4	54.45	24.97	0.000	409.4	149.7	68.74	0.000
Hydrogen storage facilities	53.28	20.63	1.404	0.000	66.38	20.56	2.536	0.000
Methane plant facilities	26.55	10.28	0.000	0.000	20.19	17.95	14.76	0.000
Hydrogen transport	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Operating cost (M€)								
Hydrogen plant facilities	7.373	6.537	5.376	3.286	20.19	17.95	14.76	9.023
Electricity cost	84.75	88.68	73.98	61.05	243.9	251.4	207.3	169.6
Hydrogen storage facilities	8.516	9.027	7.572	6.352	23.39	24.79	20.79	17.44
Methane plants facilities	1.327	1.542	1.194	0.924	3.319	3.597	3.012	2.332
CO ₂ cost	15.71	15.44	12.98	10.53	45.68	47.31	39.57	32.78
Hydrogen transport	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Total network cost (M€)	346.9	206.6	127.4	82.14	958.7	556.5	374.7	231.1
Obj: Total Annual Cost (TAC) (M€)	763				2121			
LCOE (€/MWh)	138.1				140.5			

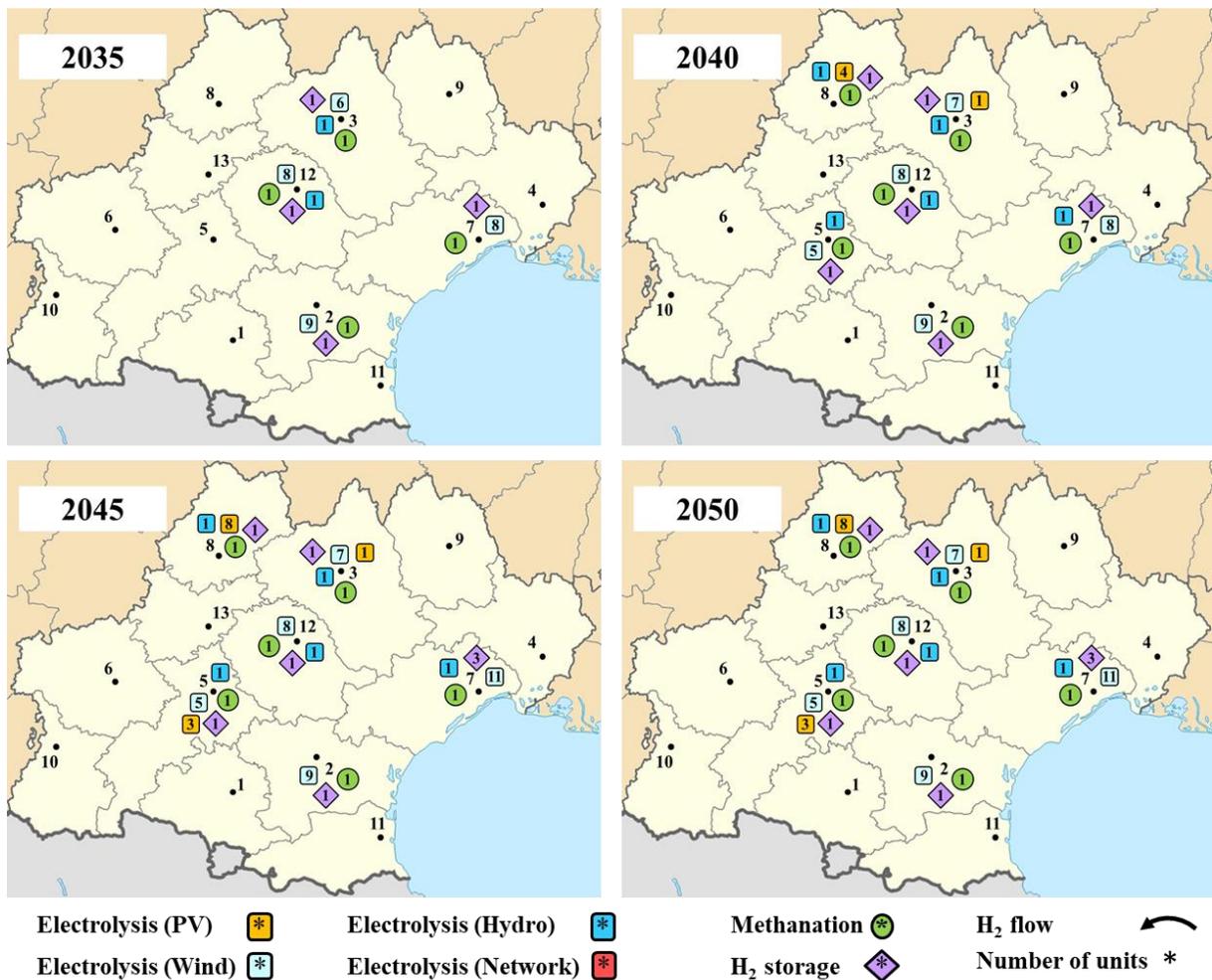


Figure 5.5. Methane Supply Chain Structure for low demand

Figure 5.7 shows the production of methane in each grid for both scenarios. Methane production is equally distributed among the producing grids for Scenario 1. However, in Scenario 2, grids 2, 7, and 12 satisfy 23%, 23%, and 12% of the total demand for methane, respectively. These grids have a high potential for methane production, due to the energy available for PtG, as shown in Figure 4.4. Since the cost of electricity is the parameter with the highest influence on the production of hydrogen and therefore of methane, the availability of electricity from renewable energy sources is dominant over the availability of CO₂.

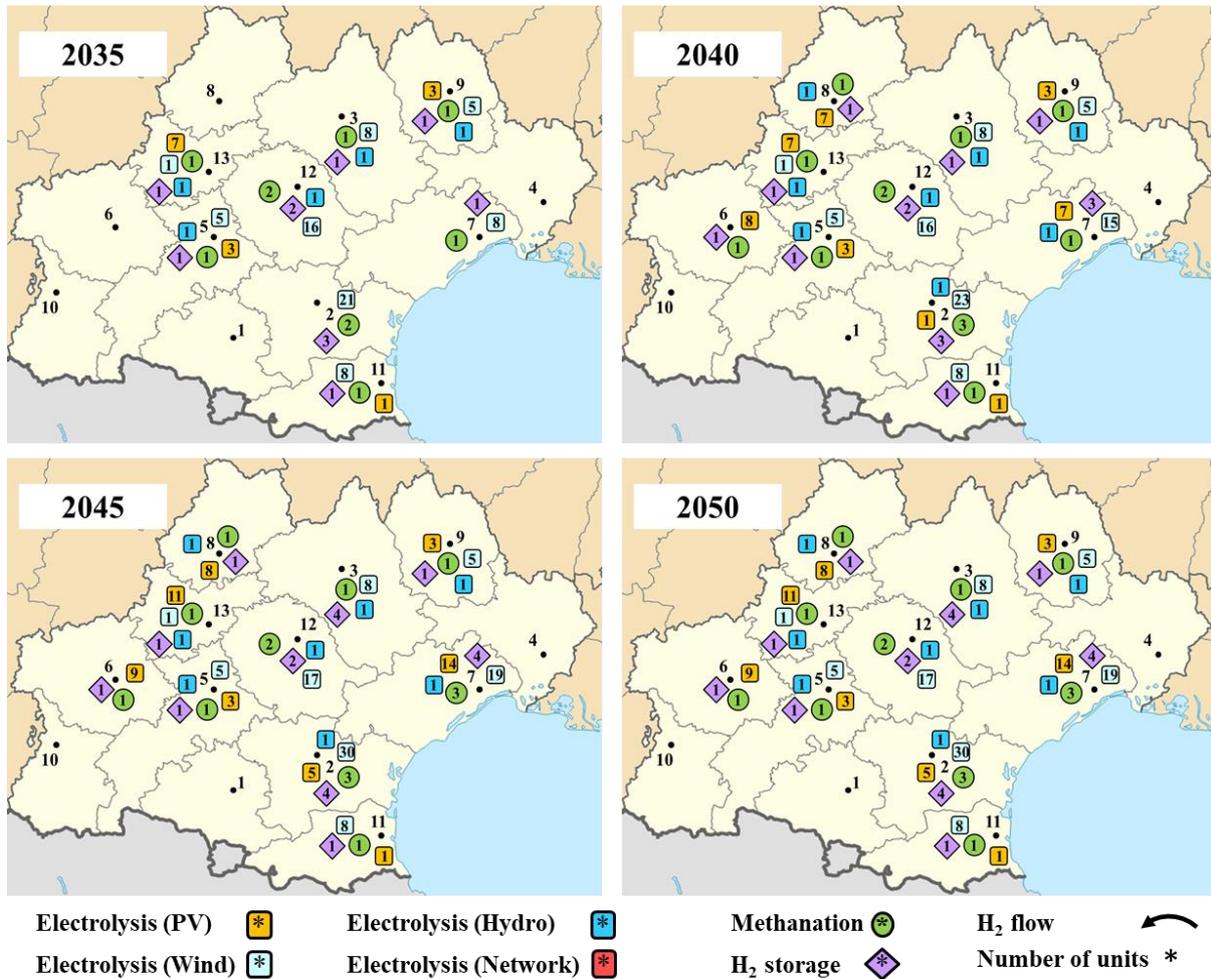


Figure 5.6. Methane Supply Chain Structure for high demand

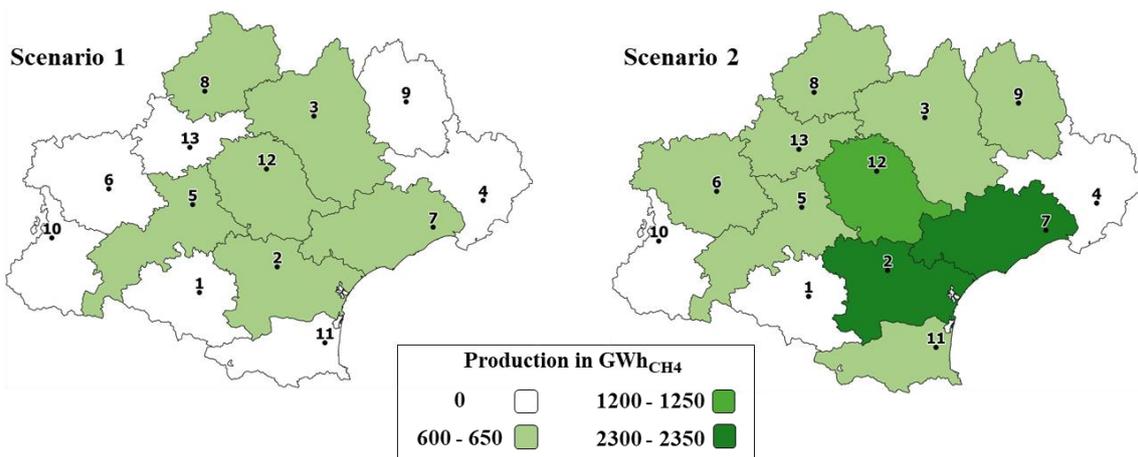


Figure 5.7. Production of methane in each grid (2050)

Figure 5.8 shows the available electricity and the electricity consumption to produce hydrogen in each grid and both scenarios. The major source of energy is wind followed by PV. It can be seen that wind energy source is chosen in priority, due to its lower cost compared to the solar source (see Table 4.2). However, these results could change drastically if the seasonal variation of both sources in the region,

and the costs associated with them are considered. On the one hand, low-cost solar electricity will be usually produced in large quantities during the summer. This implies a more stable and predictable production of hydrogen (Belderbos et al., 2015). On the other hand, the seasonal variation of solar and wind sources has a direct impact on the cost. It could considerably increase the cost of hydrogen and then the cost of methane if the main energy sources are solar and wind.

Some considerations could significantly change these results. According to some scenarios and researches, Power-to-Gas systems will initially be used to meet hydrogen demands (ADEME, 2018; E&E consultant et al., 2014; Lehner et al., 2014). The use of available electricity to meet hydrogen and methane demands will eventually influence the configuration of the MSC since electrolysis is a common stage for both supply chains. This implies that in grids (such as 4 and 5), where energy is available and not used by the MSC, hydrogen can be produced to meet a specific demand (e.g. transport). This can also potentially increase the interaction between grids (De-León Almaraz et al., 2015).

Besides, the results have also shown that the availability of CO₂ is a key parameter. Figure 5.9 shows the CO₂ consumed to produce methane. It can be observed that an import of 28 and 56% of CO₂ is required in scenarios 1 and 2, respectively, to meet the demand for methane at a minimum cost. However, not all the available CO₂ is used. The total CO₂ amount available in the territory is not used because the methane plants are installed in the same grid where hydrogen is produced. Only 57% of CO₂ is consumed from the grids in Scenario 1, up to 85% in Scenario 2. The optimization of the use of CO₂ in each network can directly influence the results since the CO₂ amounts from processes as methanation and gasification are diffuse sources (ADEME, 2018; E&E consultant et al., 2014). Taking into account more precisely the origin of the CO₂ source, location, distribution, and storage could imply a variation in its cost and therefore in the configuration of the MSC (Jarvis and Samsatli, 2018; Oei et al., 2014).

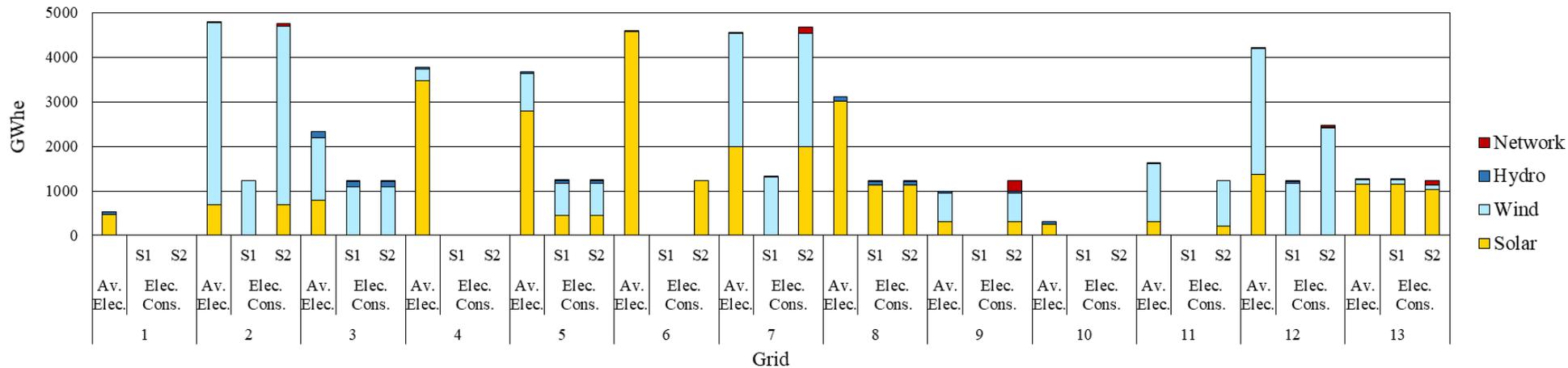


Figure 5.8. Available energy and energy consumption (by sources) in each grid (2050)

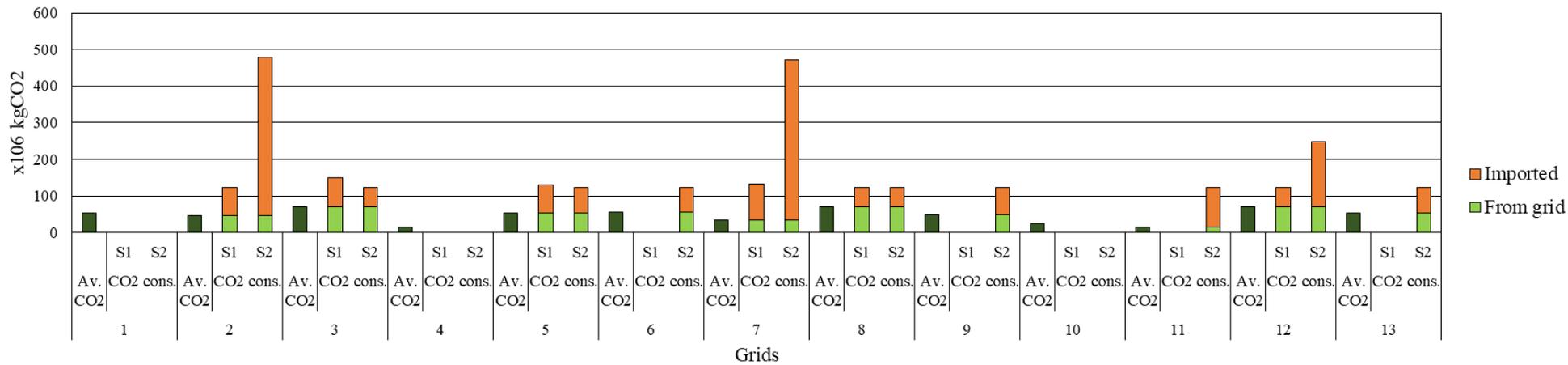


Figure 5.9. Available CO₂ and CO₂ consumption (from the grid and imported) in each grid (2050)

5.4.2 Sensitivity analysis

Table 5.2. Parameters for sensitivity analysis

Case	Demand for methane in 2050 (TWh)	Electricity cost (MWh)	CO ₂ cost (€/kg)	Factor of hydrogen storage (β)
1) Baseline	3.67	Values in Table 4.1	Values in Table 4.1	2
2) High demand	10.13			2
3) Constant electricity cost	3.67	25 €/MWh	Biogenic CO ₂ in grid = 95 €/kg, Industrial CO ₂ = 119 €/kg	2
4) High cost of CO ₂	3.67	Values in Table 4.1		2
5) Increase in hydrogen storage	3.67	Values in Table 4.1	Values in Table 4.1	3

Figure 5.10 shows the LCOE of methane for each case (Table 5.2). The values obtained are between 109 (Scenario 3) and 146 €/MWh (Scenario 5). These costs are similar to those proposed in other studies (ADEME, 2018; Collet et al., 2017; E&E consultant et al., 2014; Götz et al., 2016). A key parameter is the cost of electricity used to produce hydrogen, ranging between 46 and 57% of the total cost of methane. Based on the results obtained, the average cost of electricity varies between 39 and 43 €/MWh for scenarios 1, 2, 4, and 5. The cost of CO₂ is the second most influential parameter representing between 10 and 15% of the LCOE. The electrolysis and hydrogen storage capital costs are also significant, contributing to a range between 7 and 10, and 10 and 14% of LCOE, respectively. Some reductions in LCOE could be achieved by the use of residual heat from methanation or the recovery of O₂ from the electrolysis process (Götz et al., 2016; Vandewalle et al., 2015).

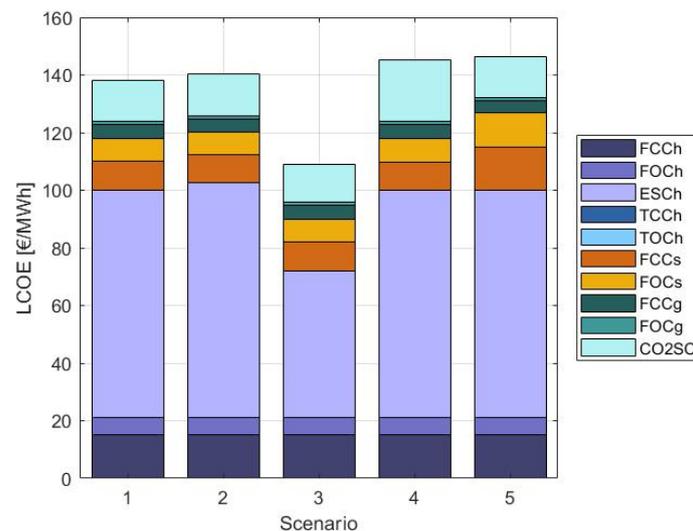


Figure 5.10. LCOE of Methane for: 1) Low demand, 2) High demand, 3) Low cost of electricity, 4) High cost of CO₂ and 5) High capacity of hydrogen storage. The LCOE is constituted by Facility Capital (FCCh) and Operating (FOCh) Cost of Hydrogen, Cost of Electricity (ESCh), Transportation Capital (TCCh) and operating cost (TOCh), Storage Capital (FCCs) and Operating (FOCs) Cost of Hydrogen, Facility Capital (FCCg) and operating (FOCg) of Methane and Cost of CO₂ source (CO2SC).

5.5 Conclusions

A general methodology for the design and deployment of an MSC based on PtG systems was developed and used for a base case study devoted to the Occitania region in France. The main technological blocks associated with the MSC were considered by minimizing the Total Annual Cost (TAC). The model provides the configurations of the supply chain that are implemented for the period studied. The supply chains obtained for two different demands were decentralized so that the use of transport between the grids is not necessary. The results show where methane and hydrogen production units can be located to meet the demand for methane, considering the potential of CO₂ and electricity from renewable sources available on the territory. The grids with the highest potential were identified, as well as the CO₂ and electricity consumption of each, according to the source. The costs associated with each of the blocks in the chain were calculated, which allowed the LCOE for the entire period to be estimated. A sensitivity analysis for key parameters (i.e., electricity and CO₂ cost and hydrogen storage capacity) was carried out to estimate its variation.

The model obtained could be useful for policy-makers, as it allows them to identify the locations where measures are required to promote the development of this type of technology. A more comprehensive methodological framework could be achieved by carrying out a multi-objective optimization, taking into account environmental impacts (i.e. Greenhouse gas emissions). Furthermore, the interaction with other technologies and energy carriers (Bramstoft et al., 2020), the optimization of CO₂ used considering other sources, and the recovery of by-products (i.e. oxygen or heat) (Götz et al., 2016; Vandewalle et al., 2015) is an interesting perspective that can significantly change the configuration of the supply chain. Both aspects are now explored in the following chapter.

6 BI-OBJECTIVE OPTIMAL DESIGN OF PTH AND PTM SUPPLY CHAINS

ABSTRACT

This chapter focuses on the development of the methodological design framework for Hydrogen and Methane Supply Chains (HMSC) based on Power-to-Gas (PtG) systems, with a specific demand for hydrogen for electromobility in addition to the hydrogen demand required as a feedstock to produce methane from the methanation process. As in chapter 4, the case study is the Occitania region, France. The problem formulation is based on a Mixed Integer Linear Programming (MILP) approach with augmented epsilon-constraint implemented in the GAMS environment according to a multi-period approach (2035-2050). Hydrogen demand was determined based on the expected use of fuel cell electric vehicles (FCEVs) whereas the demand for renewable methane has been estimated from the scenarios developed by the local authorities. The objectives to be minimized simultaneously are the Total Annual Cost (TAC) and the Global Warming Potential (GWP) related to the whole HMSC over the entire period studied. A modified Technique for Order of Preference by Similarity to Ideal Solution (m-TOPSIS) has been implemented to select a good compromise solution from the Pareto front. The optimization variables involve the number and size of production and storage units, the number of tanker-trucks as well as the flows of imported/exported hydrogen from one grid to another one. The Levelized Cost of Energy (LCOE) and the Greenhouse gas emission for hydrogen and methane are also computed. Finally, a sensitivity assessment is carried out to study the influence of three parameters, i.e., discount rate, cost of electricity, and cost of CO₂ on LCOE for hydrogen and methane.

RÉSUMÉ

Ce chapitre est dédié au développement du cadre de conception méthodologique pour les chaînes logistiques de l'hydrogène et du méthane (HMSC) basées sur les systèmes Power-to-Gas (PtG), avec une demande spécifique en hydrogène pour l'électromobilité en plus de la demande d'hydrogène nécessaire comme matière première pour produire du méthane à partir du processus de méthanation. Comme dans le chapitre 4, l'étude de cas porte sur la région Occitanie, en France. La formulation du problème est basée sur une approche de programmation linéaire mixte en nombres entiers (MILP) avec la méthode « Augmented epsilon-constraint, » mise en œuvre dans l'environnement GAMS selon une approche multi-période (2035-2050). La demande d'hydrogène a été déterminée sur la base d'une prévision de la demande en véhicules électriques à pile à combustible (FCEV), tandis que la demande de méthane renouvelable a été estimée à partir de scénarios élaborés par schémas régionaux de développement. Les objectifs à minimiser simultanément sont le coût annuel total (TAC) et les émissions de gaz à effet de serre (GHG) liés à l'ensemble des FCEV sur toute la période étudiée. La méthode « modified Technique for Order of Preference by Similarity to Ideal Solution » (m-TOPSIS) a été mise en œuvre pour sélectionner une solution de compromis sur le front de Pareto. Les variables d'optimisation concernent le nombre et la taille des unités de production et de stockage, le nombre de camions-citernes ainsi que les flux d'hydrogène importés/exportés d'un réseau à l'autre. Le coût de l'énergie (LCOE) et les émissions de gaz à effet de serre pour l'hydrogène et le méthane sont également calculés. Enfin, une analyse de sensibilité est réalisée pour étudier l'influence de trois paramètres, à savoir le taux d'actualisation, le coût de l'électricité et le coût du CO₂ sur le LCOE de l'hydrogène et du méthane.

Nomenclature

Abbreviations

GAMS	General Algebraic Modeling System
GHG	Greenhouse gas emissions
GWP	Global warming potential
HSC	Hydrogen supply chain
LCOE	Levelized cost of energy
MSC	Methane supply chain
MILP	Mixed Integer Linear Programming
O&M costs	Operation and Maintenance cost
RES	Renewable energy sources
SMR	Steam methane reforming
TAC	Total Annual Cost

6.1 Introduction

The elements involving the energy supply chains linked to PtG systems have been extensively addressed in the literature (Buttler and Spliethoff, 2018; Götz et al., 2016), especially the hydrogen supply chain (Dagdougui, 2012; Li et al., 2019). However, only a few studies have addressed the logistical aspects involved in the dual production of hydrogen and methane with their associated supply chain issues. In this regard, the scientific objective of this chapter is to extend the methodological framework presented in Chapter 4³. The optimization formulation based on Mixed-Integer Linear Programming (MILP) implemented in the General Algebraic Modeling System (GAMS) environment (“GAMS - Cutting Edge Modeling,” n.d.) with a multi-period horizon (2035-2050) is then extended for this purpose. The Occitania region in France serves as a case study. In the light of climate change mitigation and the resulting urge to decrease greenhouse gas (GHG) emissions, this chapter highlights hydrogen as an important component for future energy systems because its energetic use does not lead to direct CO₂ emissions. Additionally, hydrogen as an energy carrier shows a broad range of existing and potential use cases, e.g., within the electricity, transport, industrial, and heating sector.

Two objective functions will be involved in the optimization framework, i.e., the Total Annual Cost (TAC) and the Greenhouse Gas emissions (GHG). These functions are calculated for the entire HMSC throughout the total horizon studied.

As explained in the previous chapter, the costs associated with the HMSC and the consequent LCOE implies that PtG systems can be viewed as prohibitive as compared to other options. For this reason, the consideration of the reduction in GHG by satisfying the same service is key to its implementation in the energy system. The Levelized Cost of Energy (LCOE) for hydrogen and methane produced by PtG systems is between 64-74 €/MWh (2.1-2.5 €/kgH₂) and 95 to 150 €/MWh in 2030 (E&E consultant et al., 2014), respectively whereas other options are more economically advantageous, i.e., hydrogen from Steam Methane Reforming (SMR) (48-64 €/MWh (IEA - International Energy Agency, 2019)), or methane from biomass transformation (66-82 €/MWh in 2030 (ENEA, 2018)) and natural gas (33-37 €/MWh (Eurostat, 2020)). By considering the associated CO₂ equivalent emissions and the break-even point for certain applications, hydrogen produced by PtG systems could be a viable option, with a possible application in the transport sector. The current cost of hydrogen at German service stations is 9.5 €/kgH₂ (285 €/MWh) (Welder et al., 2018), a competitive value compared to the LCOE for hydrogen from PtG indicated above. A drastic decrease in electricity costs and operating hours over 4000 h per year allows reaching minimum values of about 22-45 €/MWh (Vandewalle et al., 2015). However, these

³ This work has been submitted for publication in *Chemical Engineering Science* from ELSEVIER

costs can rise to 9.60-49.38 €/kgH₂ depending on demand, the technology employed and environmental measures applied (carbon tax) (Li et al., 2020). In addition, the GWP for petroleum-derived diesel and SMR are 355 kgCO₂-eq/MWh and 327 kgCO₂-eq/MWh (Reiter and Lindorfer, 2015), respectively. The cost for hydrogen from PtG reaches 18-90 kgCO₂-eq/MWh when produced from wind and solar sources (Reiter and Lindorfer, 2015). In the case of methane from PtG, it could be 22-104 kgCO₂-eq/MWh when produced from the wind energy source, and 108-191 kgCO₂-eq/MWh with solar as an energy source. The ambitious environmental targets set by the European Union promote the use of energy carriers such as hydrogen and methane to satisfy energy services with low CO₂ emissions. However, using electrical energy from fossil sources in PtG systems degrade this environmental advantage: the GWP for hydrogen and methane are 850 and 994-1076 kgCO₂-eq/MWh, respectively, when the electricity used comes from the European energy mix (Reiter and Lindorfer, 2015).

This chapter addresses the mono and bi-objective optimization of HMSC, taking into account cost and environmental impact as objectives (i.e., TAC and GHG). Figure 6.1 summarizes the HMSC to be analyzed. The main contributions of this chapter are:

- Addressing various aspects of the HMSC: primary resources, production units, hydrogen transportation, storage, and refueling stations.
- Embedding the possibility of using locally produced electricity (i.e. solar, wind, and water) and importing electricity from the national network.
- Considering the evolution of the supply over time according to different criteria (total annual cost, GWP, and both of them).
- Determining the size, the location, and the number of units involved in the supply chain (production, storage, transport, and refueling stations).

The main research questions tackled in this chapter are:

- Where hydrogen and methane production plants, storage units should be located to meet a demand for hydrogen and methane in specific periods by minimizing costs and/or environmental impacts?
- Is it necessary to transport hydrogen between grids? If so, where will the hydrogen be transported from?
- What will be the production in each grid?
- How does the availability of CO₂ and electricity from renewable sources impact the selection of grids for hydrogen and methane production?

The model has as limitations that the demand for both vectors is external and not optimized at the same time given, for example, the competition of several technologies to satisfy the same service. Furthermore, it is a steady-state model, which does not address the intermittent characteristics of the energy sources (i.e., solar and wind). Furthermore, the electricity and CO₂ supply chains, i.e. aspects such as installation, transport, and storage of units for electricity and CO₂, are beyond the scope of this work.

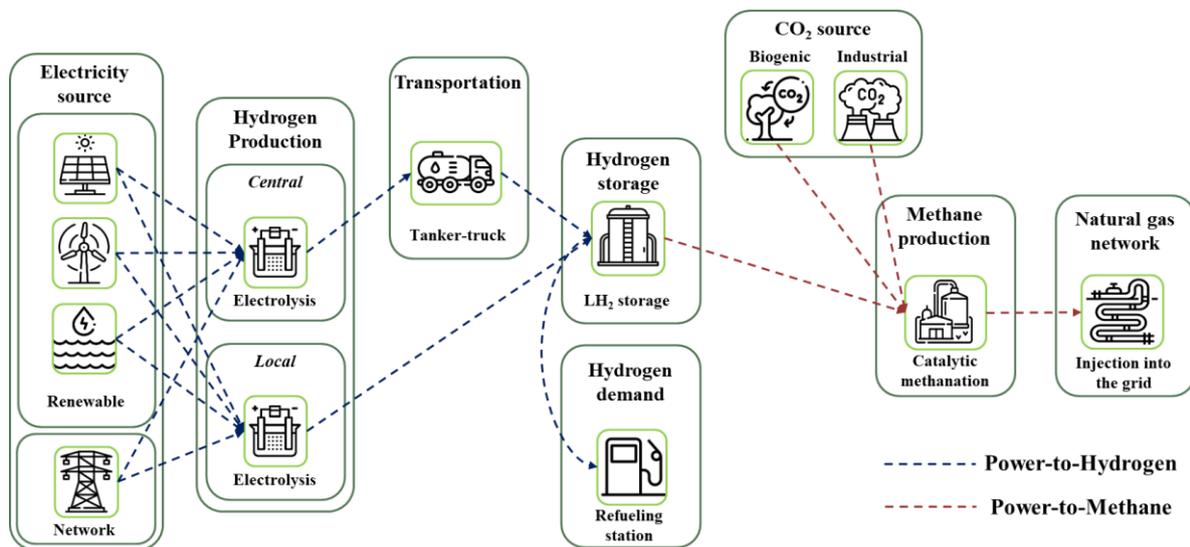


Figure 6.1. HMSC supply chain based on PtG concepts

The chapter is divided into six sections following this introduction, which sets the context and its scientific objective. Section 2 is dedicated to the problem formulation. The mathematical model and solution strategy are discussed in Sections 3 and 4, respectively. Finally, the main results (i.e., mono and multi-objective optimizations, and decision variables) are analyzed in Section 5, followed by the conclusions in Section 6.

6.2 Problem definition

The optimization model is similar to the prior model developed. However, the satisfaction of the demand for hydrogen and the inclusion of environmental impact as an objective function requires some modifications. From a macroscopic viewpoint, it can be summarized as follows:

Given:

- Spatio-temporal demands for *hydrogen* and methane
- Spatio-temporal availability of primary energy and CO₂ sources.
- Characteristics of each technology (i.e., Capital costs, O&M costs, Capacities, CO₂-eq emissions...)

Assuming:

- A division of the territory into grids.
- Only PtG systems are used to satisfy the methane and *hydrogen* demands.
- Direct injection of the methane produced into the natural gas network.
- Limited availability of primary energy and CO₂ sources.
- Possibility of importing primary resources.
- Possibility of transporting hydrogen only between grids by tanker-trucks.

Determine:

- The location, number, capacity, and rate of production, storage, transport, and refueling stations of hydrogen and methane.
- Transport flows between the grids.
- The specification of the sources, electricity quantity, and CO₂ consumed.
- Costs and CO₂-eq emissions related to each section of the supply chain.

Subject to:

- The satisfaction of the demands for hydrogen and methane.
- Conservation of mass and energy and other physical laws.

Objective:

- Minimize Total Annual Cost.
- *Minimize Global Warming Potential.*
- Combination of both through ϵ -constraint method.

The methodology framework used in this chapter is summarized in Figure 6.2.

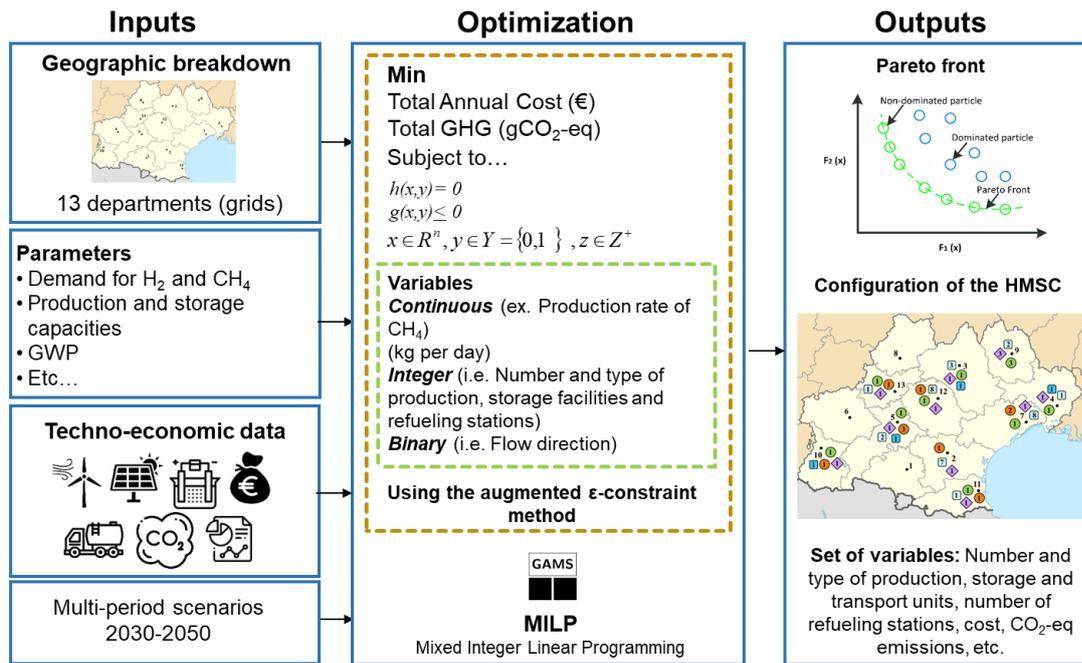


Figure 6.2. Methodology framework for the HMSC

6.3 Mathematical model

The main sets, parameters, variables added to the model described in chapter 4 are shown below, in addition to the main modifications made to the restrictions.

Sets

f types of refueling station

Parameters

Demand

HD_{igt} Hydrogen demand for the final product i in grid g in period t (kg per year)

Production data

FCC_{pjt} Capital cost of establishing refueling station type f and size j in period t (€)

$FNPO_{fjg}$ Initial number of refueling 1 stations of type f and size j in grid g

$HGWP_{ip}$ Global warming potential for hydrogen production for the final product i using plant type p (gCO₂-eq per kg H₂)

$MGWP_{iq}$ Global warming potential for methane production for the final product i using plant type q (gCO₂-eq per kg CH₄)

Storage data

$SGWP_{is}$ Global warming potential for hydrogen storage for the final product i establishing storage type s (gCO₂-eq per kg H₂)

Transportation data

$TGWP_l$ Global warming potential for hydrogen transport establishing transportation mode l (gCO₂-eq per ton-km)

Variables

Continuous variables

CH_4Cost_{it}	Cost of methane for final product i in period t (€ per MWh)
CH_4GWP_{it}	Global warming potential for methane ($i=CH_4$) in period t (gCO ₂ -eq per MWh)
CH_4LCOE_i	Levelized cost of energy for methane (€ per MWh)
GHG	Total global warming potential for the whole supply chain (gCO ₂ -eq)
GHG_t	Global warming potential for the whole supply chain in period t (gCO ₂ -eq per year)
H_2Cost_{it}	Cost of hydrogen for the final product i in period t (€ per MWh)
H_2GWP_{it}	Global warming potential for hydrogen having i as the final product in period t (gCO ₂ -eq per MWh)
H_2LCOE_i	Levelized cost of energy for hydrogen (€ per MWh)
HD_{igt}^T	Total hydrogen demand for the final product i in grid g in period t (kg per year)
$HGHG_t$	Global warming potential for hydrogen production in period t (gCO ₂ -eq per year)
$SGHG_t$	Global warming potential for hydrogen storage in period t (gCO ₂ -eq per year)
$TGHG_t$	Global warming potential for hydrogen transport in period t (gCO ₂ -eq per year)
XP_{ipjgt}	Allocation fraction for hydrogen production considering the final product i , type of plant p and size j in grid g in period t
XQ_{ilt}	Allocation fraction for hydrogen transport considering the final product i , transportation mode l in period t
XS_{isjgt}	Allocation fraction for hydrogen storage considering the final product i , type of storage s and size j in grid g in period t

Integer variables

HIF_{fjgt}	Number of fuel station installed type f and size j in grid g in period t
NFS_{fjgt}	Number of Fuel Stations type f and size j in grid g in period t

The model formulation involves a set of equalities (mainly mass balances) and inequalities that take into account the possible flows between the grids. It aims to meet a demand for final product “ i ” (hydrogen and methane), considering the potential of each grid “ g ”. The availability of primary resources “ e ” (electricity) and “ c ” (CO₂), hydrogen production type “ p ”, storage option “ s ”, transport mode “ l ”, methane production type “ q ” and refueling station “ f ” for each period “ t ” are considered. The model includes modifications to meet both methane and hydrogen demands. The set of equations and constraints are described below.

The ensemble of equations shown in the previous chapter was modified to include the new sets. In addition, modifications were made to include the hydrogen demand for the transportation sector. The main modifications and the new equations considered are shown below.

6.3.1 Hydrogen demand

The demand for hydrogen to satisfy the methane production (MPR_{igt}^T , kg/year) as feedstock and the demand for hydrogen as a final product (for mobility usage in this case) (HD_{igt} , kg/year) imply a total demand for hydrogen (HD_{igt}^T , kg/year). Based on Sabatier reaction (Eq. 2.3) the hydrogen demand to meet the methane production is calculated using the hydrogen stoichiometric coefficient (φ_{h_2}) and a safety factor (SSF = 5%) for storing a small inventory of hydrogen. It is shown in Eq. 6.1.

$$HD_{igt}^T = \varphi_{h_2}SSF \sum MPR_{igt}^T + HD_{igt} \quad \forall i, g, t \quad \text{Eq. 6.1}$$

6.3.2 Refueling station constraints

The total supply of hydrogen from the refueling stations (FSR_{igt}^T , kg/year) is calculated by using the hydrogen supply of each refueling station (FSR_{fijgt} , kg/year) (See Eq. 6.2). It is restricted according to the capacity of the refueling stations installed (See Eq. 6.3).

$$FSR_{igt}^T = \sum_{fj} FSR_{qijgt} \quad \forall i, g, t \quad \text{Eq. 6.2}$$

$$FSCap_{fj}^{\min} NFS_{fjgt} \leq \sum_i FSR_{fijgt} \leq FSCap_{fj}^{\max} NFS_{fjgt} \quad \forall q, j, g, t \quad \text{Eq. 6.3}$$

6.3.3 Number of refueling stations

The total number of refueling stations installed during each period (NFS_{ifjgt}) is determined from Eq. 6.4. The numbers of units installed at t equal to 0 are indicated as parameters ($NFS0_{ifjg}$).

$$NFS_{ifjgt} = NFS_{ifjgt-1} + HIF_{ifjgt} \quad \forall i, f, j, g, t \neq 1 \quad \text{Eq. 6.4}$$

6.3.4 Refueling station capital cost

The facility capital cost for the refueling stations ($FFCC_t$, €/year) involves the capital cost for the installation ($FFCC_{fjt}$, €) as well as the number of facilities installed in each period (HIF_{fjgt} , year⁻¹). It is shown in Eq. 6.5.

$$FFCC_t = \sum_{fjg} FCC_{fjt} HIF_{fjgt} \quad \forall t \quad \text{Eq. 6.5}$$

6.3.5 Environmental objective function and indicators

The Greenhouse Gas emissions indicator (GHG) is used to assess the environmental impact of the supply chain. Although an exhaustive LCA was not performed, its methodological guidelines were followed to calculate GWP. The GHG_t related to the hydrogen (HGHG_t, gCO₂-eq per year) and methane (MGHG_t, gCO₂-eq per year) production, hydrogen storage (SGHG_t, gCO₂-eq per year), and hydrogen transport

($TGHG_t$, gCO₂-eq per year) is calculated through Eq. 6.6 to 6.10. Finally, the function GHG is obtained as expressed in Eq. 6.11.

$$HGHG_t = \sum_{p|jg} (HPR_{p|jgt} HGWP_{ip}) \quad \text{Eq. 6.6}$$

$$SGHG_t = \sum_{ig} (HS_{igt}^T SGWP_{is}) \quad \text{Eq. 6.7}$$

$$TGHG_t = \sum_{i|lg} \left(\frac{2AD_{lg} \cdot Q_{i|lg}}{TCap_i} \right) TGWP_i \quad \text{Eq. 6.8}$$

$$MGHG_t = \sum_{q|jg} (MPR_{q|jgt} MGWP_{iq}) \quad \text{Eq. 6.9}$$

$$GHG_t = HGHG_t + SGHG_t + TGHG_t + MGHG_t \quad \text{Eq. 6.10}$$

$$GHG = \sum_t GHG_t \quad \text{Eq. 6.11}$$

6.3.6 Physical allocation for cost, and environmental indicators

A physical allocation procedure is used to compute the cost and environmental indicators, since physical relationships can be established. Other relationships between the products are not necessary in this case, because it involves the same product (da Silva et al., 2017). They were calculated once the solution of the set of variables is obtained. For the common steps shared by hydrogen and methane production (production, storage, and transport of hydrogen), the allocation is performed according to the amount of hydrogen used respectively for both final products. This means that the allocation parameter is based on mass fraction for the production ($XP_{ip|jgt}$, kg/kg), storage ($XS_{is|jgt}$, kg/kg), and transport (XQ_{ilt} , kg/kg) of hydrogen as defined in Eq. 6.12 to 6.14.

As an example, it is shown how the investment ($HFCC_{it}$, €/year) and the operating cost ($HFOC_{it}$, €/year) for hydrogen production associated with each final product are calculated (see Eq. 6.14 and 6.16). Similarly, the other costs involved in the TAC for each final product (TAC_{it} , €/year) (Eq. 6.17) are computed. Then, the TAC for methane and hydrogen ($MTAC_{it}$ and $HTAC_{it}$), were computed (see Eq. 6.18 and 6.19), taking into account their contribution to the TAC.

$$XP_{ip|jgt} = \left(\frac{PR_{p|jgt}}{\sum_i PR_{p|jgt}} \right) \quad \forall i, p, j, g, t \quad \text{Eq. 6.12}$$

$$XS_{is|jgt} = \left(\frac{HS_{is|jgt}}{\sum_i HS_{is|jgt}} \right) \quad \forall i, s, j, g, t \quad \text{Eq. 6.13}$$

$$XQ_{ilt} = \left(\frac{\sum_{g,g'} Q_{i l g g' t}}{\sum_{i,g,g'} Q_{i l g g' t}} \right) \forall i, l, t \quad \text{Eq. 6.14}$$

$$HFCC_{it} = \sum_{pjg} XP_{ipjgt} HCC_{pj} HIP_{pjgt} \quad \forall t \quad \text{Eq. 6.15}$$

$$HFOC_{it} = \sum_{pjg} XP_{ipjgt} (HUPC_{pit} HPR_{pjgt}) + \sum_{ejg} XP_{ipjgt} UEC_{et} \left(IPES_{eigt} + SSF \sum_{pj} \gamma_{epj} HPR_{pjgt} \right) \quad \forall i, t \quad \text{Eq. 6.16}$$

$$TAC_{it} = MFCC_{it} + HFCC_{it} + SFCC_{it} + HTCC_{it} + FFCC_{it} + MFOC_{it} + HFOC_{it} + SFOC_{it} + HTOC_{it} + FFOC_{it} \quad \forall i, t \quad \text{Eq. 6.17}$$

$$MTAC_{it} = TAC_{it} - FFCC_{it} - FFOC_{it} \quad \forall t, i = CH_4 \quad \text{Eq. 6.18}$$

$$HTAC_{it} = TAC_{it} - MFCC_{it} - MFOC_{it} \quad \forall t, i \quad \text{Eq. 6.19}$$

The Levelized Cost of Energy for methane (CH_4LCOE , €/MWh) and hydrogen (H_2LCOE , €/MWh) are calculated through Eq. 6.20 and 6.21, respectively.

$$CH_4LCOE = \left(\frac{\sum_t \frac{MTAC_{it}}{(1+r)^t}}{\sum_t \frac{MD_{it}^T}{(1+r)^t}} \right) \quad \text{Eq. 6.20}$$

$$H_2LCOE = \left(\frac{\sum_t \frac{HTAC_{it}}{(1+r)^t}}{\sum_t \frac{HD_{it}}{(1+r)^t}} \right) \quad \text{Eq. 6.21}$$

Another cost indicator was defined to determine the cost of methane (CH_4Cost_{it} , €/MWh) and hydrogen (H_2Cost_{it} , €/MWh) in each period (Eq. 6.22 and 6.23, respectively). In contrast to the LCOE which represents the average cost for the entire period (2035 to 2050), the cost indicator indicated here represents the average cost for each of the years studied. Similar to the previous case, the Total Annual Cost was reformulated (TAC_{it}) and the allocation fractions for hydrogen production, storage, and transport were employed.

$$CH_4Cost_{it} = \frac{TAC_{it}}{MD_{it}^T} \quad \text{Eq. 6.22}$$

$$H_2Cost_{it} = \frac{TAC_{it}}{HD_{it}} \quad \forall i, t \quad \text{Eq. 6.23}$$

The GWP associated with each energy vector (CH_4 and H_2 GWP, gCO₂-eq per MWh) was estimated through Eq. 6.25 and 6.25 respectively, using the same physical allocation as above.

$$CH_4GWP_t = \frac{GWP_t}{MD_t^T} \quad \forall t \quad \text{Eq. 6.24}$$

$$H_2GWP_{it} = \frac{GWP_{it}}{HD_{it}} \quad \forall i, t \quad \text{Eq. 6.25}$$

The superstructure of the HMSC is shown in Figure 6.3 and the main variables involved are illustrated.

6.4 Solution strategy

The model involves a MILP formulation. The objective functions TAC and GHG are used to carry out a mono-objective optimization for each function. Both optimizations are solved through CPLEX 12 in the GAMS environment. Subsequently, a multi-objective optimization with these two objective functions is performed. The Pareto front was obtained for this purpose through the augmented ε -constraint method (AUGMECON) (Mavrotas, 2009), which allows obtaining strong optimal solutions and avoids redundant iterations, compared to the conventional ε -constraint method (Miettinen, 1998) and other methods as the “weighted sum approach” (Costa-Carrapiço et al., 2020). The AUGMECON strategy has been implemented in GAMS. Given that all Pareto solutions are optimal, a multi-criteria decision analysis method has been used to choose a solution. The modified-TOPSIS (m-TOPSIS) is used in this work. It allows avoiding rank reversals and evaluation failure that often occurs in the original TOPSIS (Ren et al., 2007).

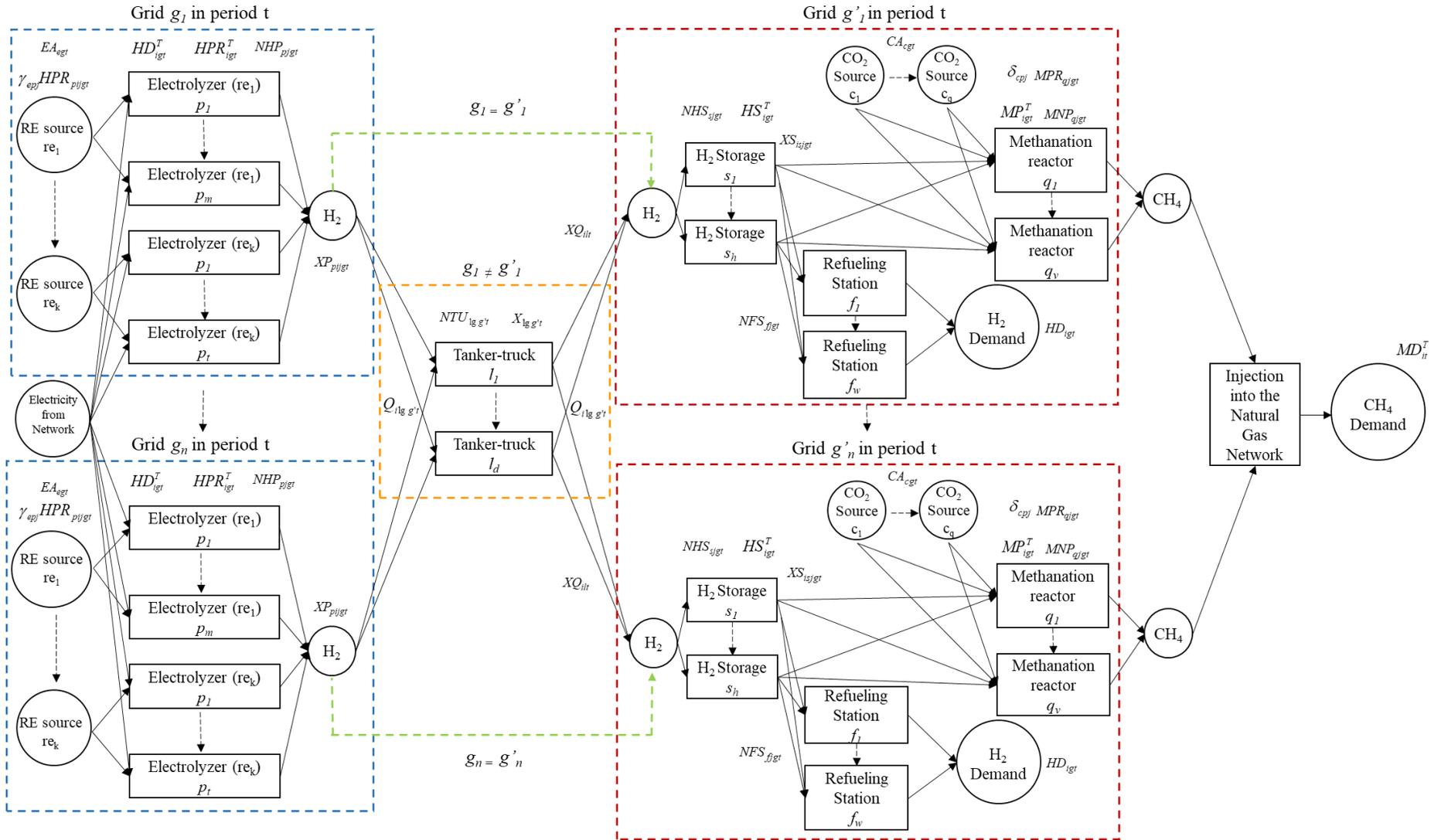


Figure 6.3. Superstructure of the multi-period model for the HMSC

6.5 Results and discussions

6.5.1 Mono-objective optimizations

Two mono-objective optimizations were carried out for the TAC (“min TAC”) and GHG (“min GWP”) functions. The values of the main decision variables are shown in Table 6.1. Although only a 4% difference in TAC between the “min TAC” and “min GWP” solution is achieved, a 20% GHG variation is observed. The production and storage units are installed progressively. However, the number of refueling stations installed in each period is the same for each solution, since the demand to be satisfied remains constant and their effects on CO₂ emissions are considered negligible. There is an important installation of plants to use renewable energy resources with lower emissions and the use of biogenic CO₂ in the “min GWP” solution. The difference between the total investment costs for the “min TAC” and “min GWP” solutions is 19 %. Most of the investment costs are involved in the first period. The investment costs in 2035 represent 67% of the TAC of the same year for the "min TAC" solution, while for the "min GWP" solution it involves 71%. In terms of operating costs, the cost of electricity for hydrogen production lies between 20 and 70% of the total cost for each period. The costs of hydrogen storage could be as high as 11%, which may vary according to the demand or availability of low-cost energy. The cost of CO₂ stands between 2 and 3%. CO₂ emissions between 2035 and 2050 increase between 40% ("min GWP") and 46% ("min TAC"). The “min TAC” solution corresponds to the minimum values of the LCOE and the “min GWP” solution to its maximum value.

Although the difference of the LCOE between both solutions is only 1.5 %, the configurations of the HMSC are different for each solution. The detailed configurations of the HMSC in each period are shown in Figure 6.4 and Figure 6.5. It is observed that the supply chains obtained are decentralized. The number of units installed for hydrogen and methane production, in addition to hydrogen storage, transport units, and refueling stations, can be shown in each grid. All of the electrolyzers operate with the energy source that is available in the territory, i.e., none of the electrolyzers is installed to use only electricity from the national grid. Only the configuration obtained for “min TAC” proposed the installation of hydrogen production plants using PV electricity source. The electricity issued from solar source can be competitive with those of the wind source. However, the GWP associated with the solar source is higher than those from wind or hydro sources, and therefore the "min GWP" solution does not propose its use (see Table 4). In addition, the configurations of the “min GWP” and “TOPSIS” solutions are more decentralized. This implies the use of more hydrogen transport units and the exploitation of wind and hydraulic energy sources with lower CO₂-eq emissions than the PV source. The configuration of the HMSC tends to be decentralized because both the energy and CO₂ sources used are diffuse. This type of configuration can be observed in previous studies (Almansoori and Shah, 2006; Bramstoft et al., 2020; Mesfun et al., 2017; Samsatli and Samsatli, 2018). In addition, a decrease in hydrogen and methane cost indicators in each period due to fewer installations in the following years is observed for

both solutions. The cost indicators of both solutions differ significantly in 2035. As for hydrogen, there is a 16% increase in the cost indicator. A difference of 13% is observed for methane.

Table 6.2 allows comparing the LCOE and GWP obtained for hydrogen and methane in previous investigations with those computed in this study. Only studies of hydrogen and methane supply chains approached from GEOMs and taking France as a case study were chosen. On the one hand, for the minimization of TAC, the LCOE of hydrogen in this study is higher than those shown in (De-León Almaraz et al., 2015). This is because in (De-León Almaraz et al., 2015) SMC is involved as a technology option. However, the GWP is 8 times higher. Finally, in (Li et al., 2020) there is the highest LCOE since the horizon studied is 2030 and environmental costs are also taken into account. The minimization of GWP corresponds in both cases to an exhaustive use of PtG systems. However, the wider use of solar sources in (De-León Almaraz et al., 2015) implies an increase of the LCOE and GWP. there is a 37% difference between the LCOE for hydrogen when comparing (De-León Almaraz et al., 2015) and this study. Besides, the GWP for hydrogen in (De-León Almaraz et al., 2015) is twice the value in this study. In the case of methane, only TAC minimization can be compared.

6.5.2 *Multi-objective optimization and comparison*

The Pareto front obtained can be visualized in Figure 6.6 from which two TOPSIS solutions can be obtained:

- « TOPSIS solution »: m-TOPSIS applied with TAC and GWP as criteria.
- « TOPSIS solution' »: m-TOPSIS applied with TAC, GWP, H_2COST_{it} , CH_4COST_{it} , H_2GWP_{it} , CH_4GWP_{it} in 2035 as criteria.

The values used to apply the m-TOPSIS method are shown in Table 6.3. Several comments can be highlighted from these two solutions. The TOPSIS solution' allows reducing by 14% the total GWP, with a 1% increase for TAC, and 5 and 6% of the cost indicators of hydrogen and methane, respectively, compared to the "min TAC" solution. The final choice will depend on the viewpoint of the decision-maker. In this study, the « TOPSIS solution » is analyzed, suggesting two solutions that can be achieved according to the decision criteria of decision-makers.

The main values of the decision variables for the TOPSIS solution are shown in Table 6.1. In addition, the configuration of the HSCH for this solution is shown in Figure 6.7. As with the mono-objective optimization solutions, it is a decentralized HMSC, with a higher presence of hydrogen transport compared to the "min TAC" solution. The main characteristics and computational effort of the solutions obtained are shown in Table 6.4. The solution of the model with the TAC function implies a greater complexity and therefore a longer calculation time, compared to the "min GWP" solution.

Table 6.1. Main results for the solutions obtained through the proposed methodology

Year	min TAC				min GWP				TOPSIS solution			
	2035	2040	2045	2050	2035	2040	2045	2050	2035	2040	2045	2050
Number of facilities												
Hydrogen production	52	72	91	91	52	68	87	87	51	67	85	85
Hydrogen storage	18	24	30	30	15	17	20	20	16	19	21	21
Refueling station	31	55	93	93	31	55	93	93	31	55	93	93
Methane production	6	7	13	13	33	37	52	52	24	33	35	35
Hydrogen transport	5	4	1	5	12	14	17	19	9	15	15	22
Capital cost (M€)												
Hydrogen plants facilities	103.1	40.67	25.41	0.000	109.5	41.69	25.05	0.000	106.1	35.35	26.15	0.000
Hydrogen storage facilities	138.0	26.88	20.81	0.000	177.2	20.63	10.79	0.000	156.6	22.19	9.195	0.000
Refueling stations facilities	63.20	14.79	2.560	0.000	63.20	14.79	2.560	0.000	63.20	14.79	2.560	0.000
Methane plants facilities	39.82	5.139	3.339	0.000	64.89	4.313	7.318	0.000	50.83	10.89	4.436	0.000
Transportation modes	1.160	0.719	0.139	0.539	3.017	2.516	2.405	1.939	2.089	2.695	2.087	2.370
Operating cost (M€)												
Hydrogen plants facilities	10.07	9.149	8.027	4.907	10.73	9.660	8.369	5.116	10.41	9.044	8.011	4.897
Electricity cost	112.6	121.2	106.7	90.76	111.5	119.9	105.5	90.53	111.9	120.3	105.8	90.84
Hydrogen storage facilities	15.80	17.44	15.94	14.08	15.80	17.44	15.94	14.08	15.80	17.44	15.94	14.08
Methane plants facilities	1.991	1.799	1.560	1.208	3.244	2.728	2.478	1.919	2.541	2.512	2.172	1.682
Refueling stations facilities	6.320	5.674	3.070	5.131	6.320	5.674	3.070	5.131	6.320	5.674	3.070	5.131
CO ₂ cost	14.52	15.27	12.23	10.01	11.25	12.44	10.31	8.149	12.31	12.65	10.43	8.688
Injection cost	6.982	7.401	6.207	5.208	6.982	7.401	6.207	5.208	6.982	7.401	6.207	5.208
Transportation modes	0.448	0.253	0.027	0.169	0.862	0.893	0.815	0.774	0.796	1.039	0.842	0.901
Total network cost (M€)	514.1	266.4	206.0	132.0	584.5	260.1	200.9	132.8	545.9	262.0	197.0	133.8
GWP (GgCO ₂)	283.5	405.0	475.3	534.9	249.8	350.8	387.7	418.4	258.4	354.4	390.0	430.8
Obj: Total Annual Cost (M€)		1118				1178				1139		
Obj: GWP (GgCO₂)		1699				1407				1434		
LCOE – Hydrogen (€/MWh)		137				139				138		
LCOE – Methane (€/MWh)		132				133				132		

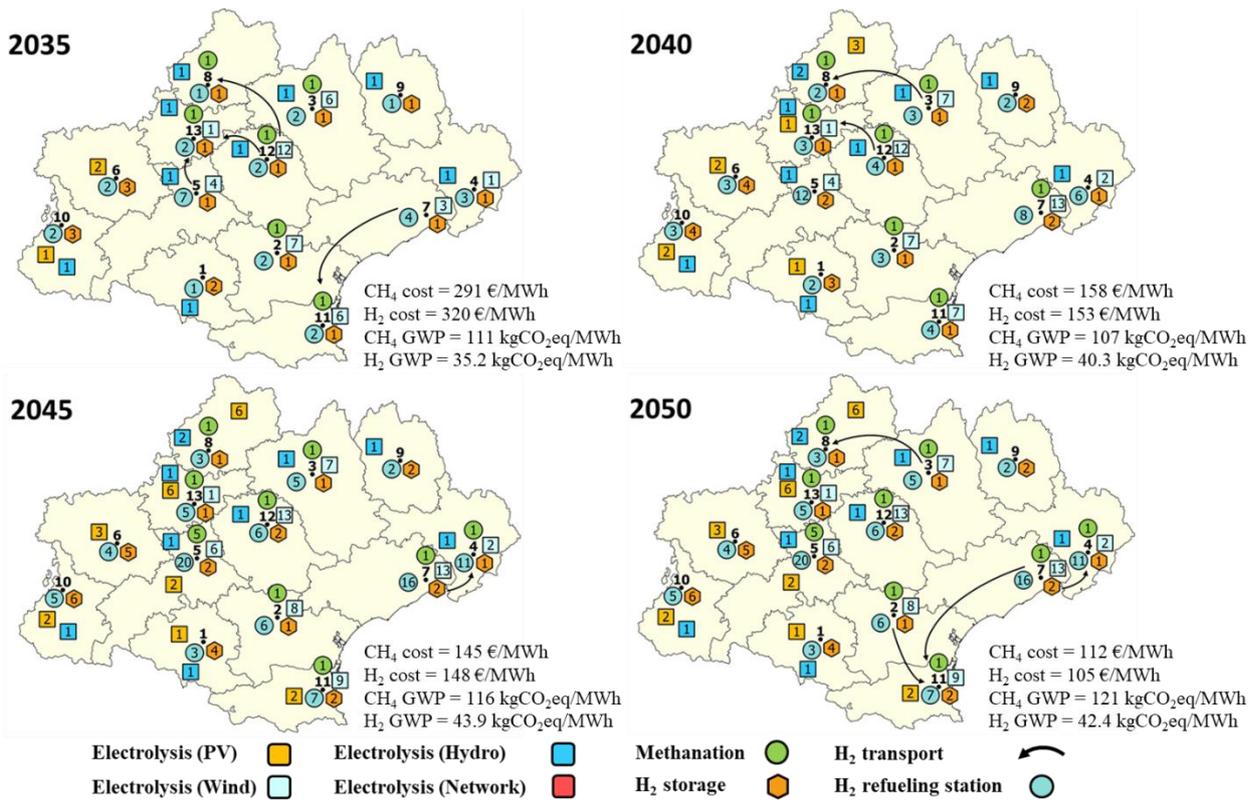


Figure 6.4. Supply chain configuration for min TAC

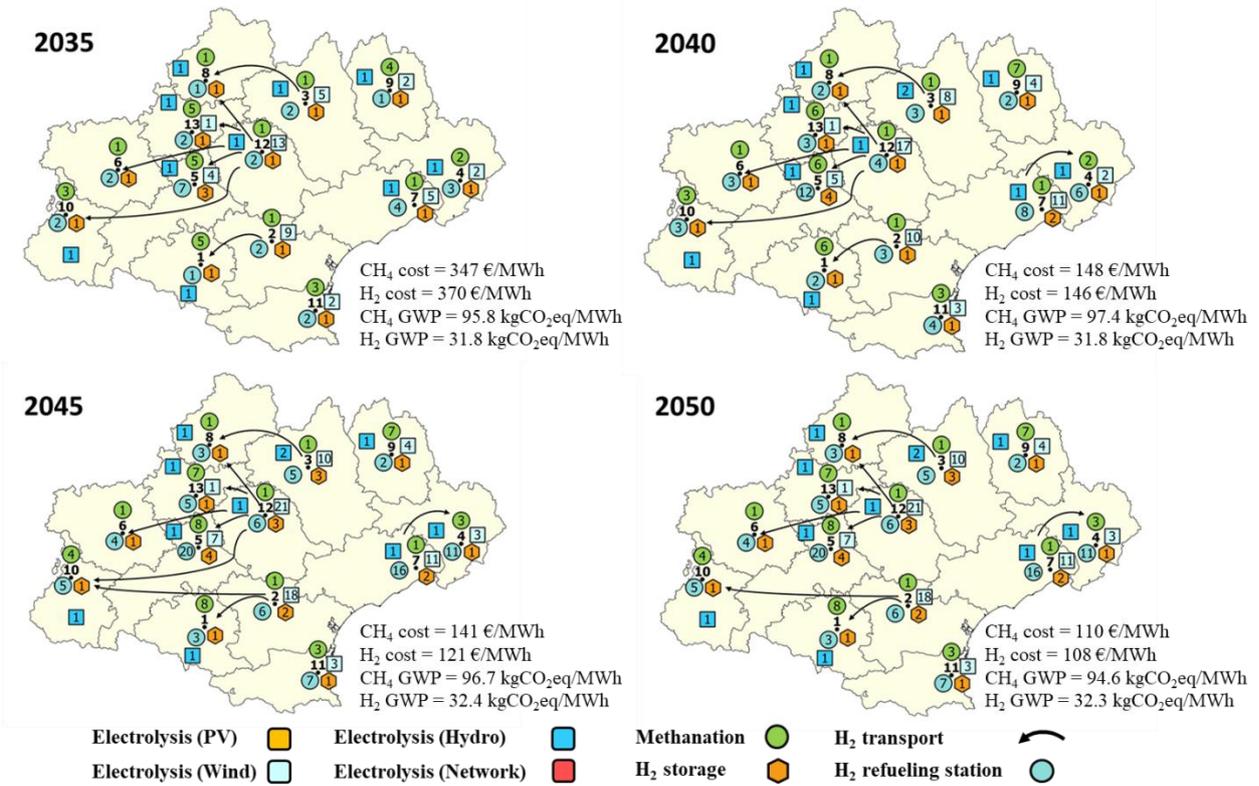


Figure 6.5. Supply chain configuration for min GWP

Table 6.2. LCOE and GWP for hydrogen and methane

Study	Period	Case of study	Objective function							
			TAC				GWP			
			LCOE [€/MWh]		GWP [gCO ₂ eq/MWh]		LCOE [€/MWh]		GWP [gCO ₂ eq/MWh]	
			H ₂	CH ₄	H ₂	CH ₄	H ₂	CH ₄	H ₂	CH ₄
(De-León Almaraz et al., 2015)	2020-2050	France	102	---	330	---	222	---	66	---
(Li et al., 2020)	2030	Franche-Comté	288 to 1482	---	---	---	---	---	---	---
This study	2035-2050	Occitanie	137	132	41	115	139	133	32	96

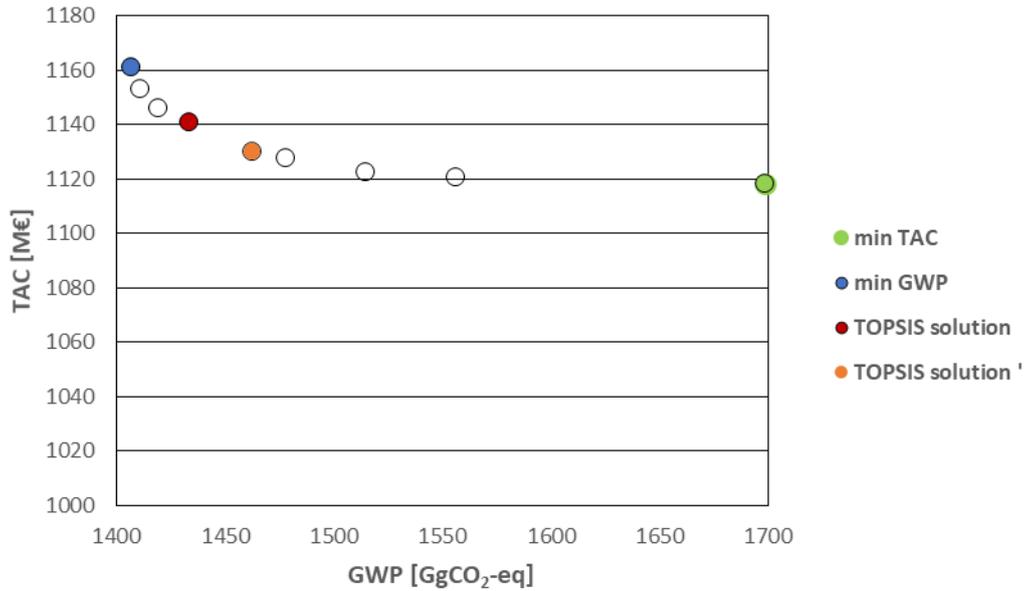


Figure 6.6. Pareto front obtained through the AUGMECON method. « TOPSIS solution » and « TOPSIS solution' »

Table 6.3. Values used to apply the m-TOPSIS method

Solutions	TAC [M€]	Total GWP [GgCO ₂]	Year: 2035			
			CH ₄ Cost [€/kWh]	CH ₄ GWP [gCO ₂ /kWh]	H ₂ Cost [€/kWh]	H ₂ Cost [gCO ₂ /kWh]
Min GWP	1162	1407	347	95.8	370	31.8
	1153	1411	336	96.1	366	31.8
	1146	1419	334	96.9	366	31.5
TOPSIS	1141	1433	316	99.5	348	31.9

Alternative	1130	1462	308	103	336	31.9
	1128	1478	311	102	340	31.9
	1123	1515	301	105	332	31.7
	1121	1556	290	109	319	33.7
Min TAC	1118	1699	291	111	320	35.2

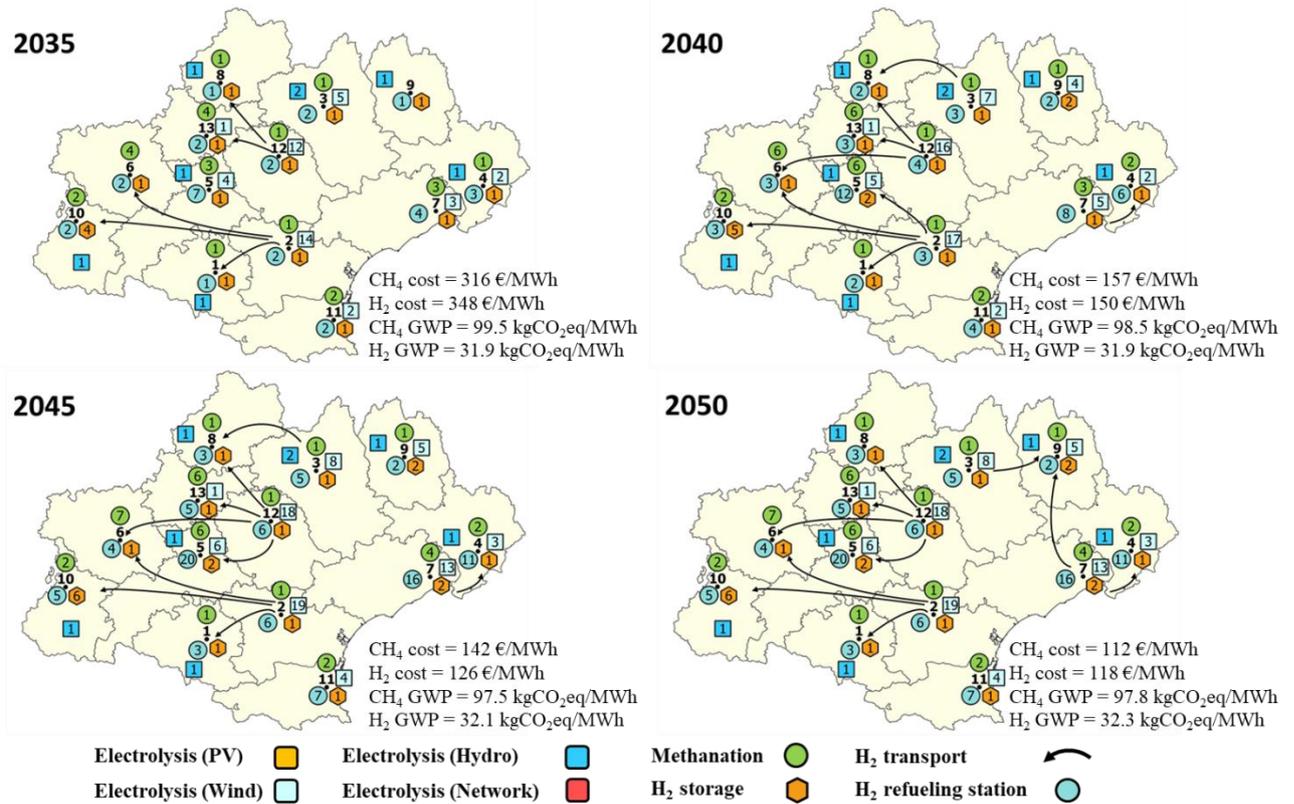


Figure 6.7. Supply chain configuration for TOPSIS solution

Table 6.4. Computational results for the optimization

Continuous variables	Binary variables	Constraints	CPU (s)			GAP		
			TAC	GWP	TOPSIS	TAC	GWP	TOPSIS
52,893	23,628	385,319	5,500	427	5,500	0.006	0.001	0.007

A color code with various intensities for different ranges has been used to visualize (Figure 6.8) the production of methane (PtM), hydrogen for methane production (PtH_{CH₄}), and transport (PtH_{TRANSP}) in each grid for the three solutions analyzed. The grids with the largest amount of low-cost available energy match the most hydrogen-producing grids, i.e. grids like 2, 7, 12 (see Figure 4.3). Conversely, those with the fewest available energy sources produce the least amount of hydrogen., i.e. 6 and 8. For methane production, the “min GWP” and “TOPSIS” solutions reflect methane production almost all over the territory, using mainly the available biogenic CO₂ sources and with less environmental impact compared

to imported CO₂ from other sources (see Figure 4.5). The TAC solution shows no methane production in grids 1, 6, 9, and 10 with a decrease in investment costs.

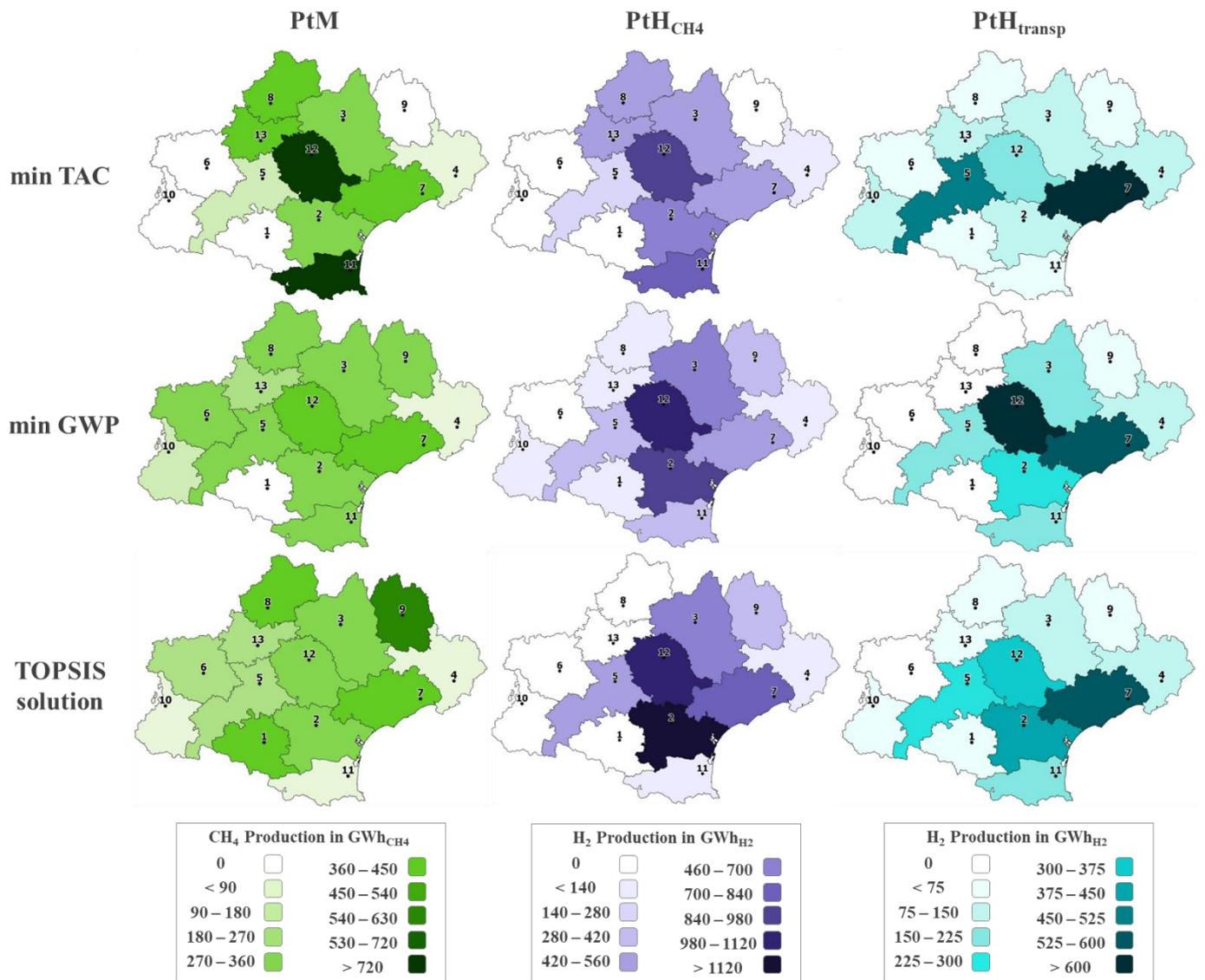


Figure 6.8. Production of methane (PtM), hydrogen for PtM (PtH_{CH_4}), and hydrogen for transportation (PtH_{trans}) in each grid (2050)

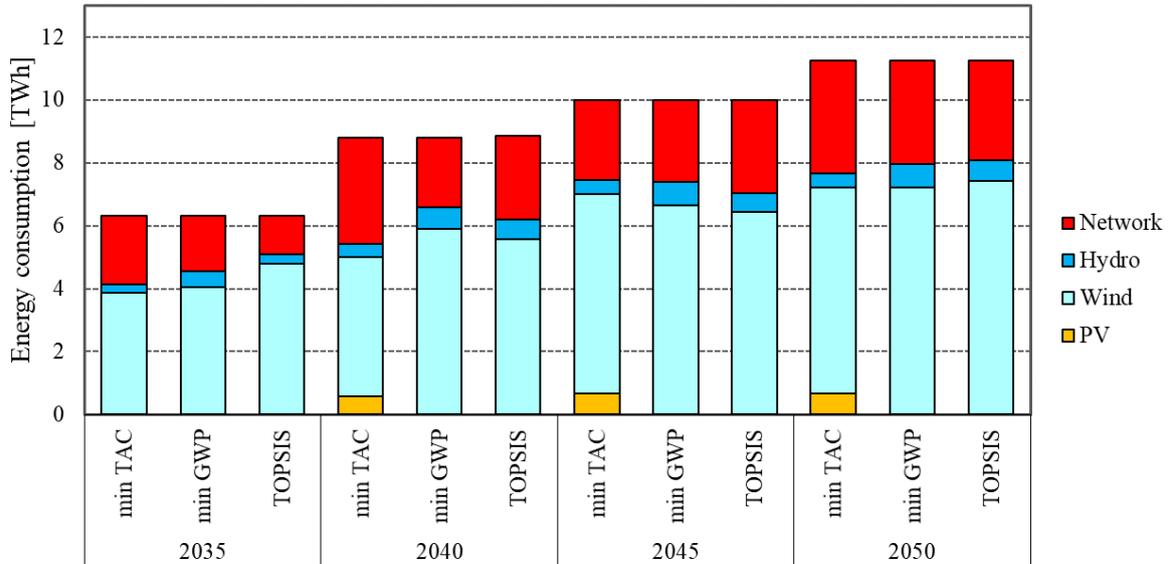


Figure 6.9. Electricity consumption for min TAC, min GWP, and TOPSIS solution

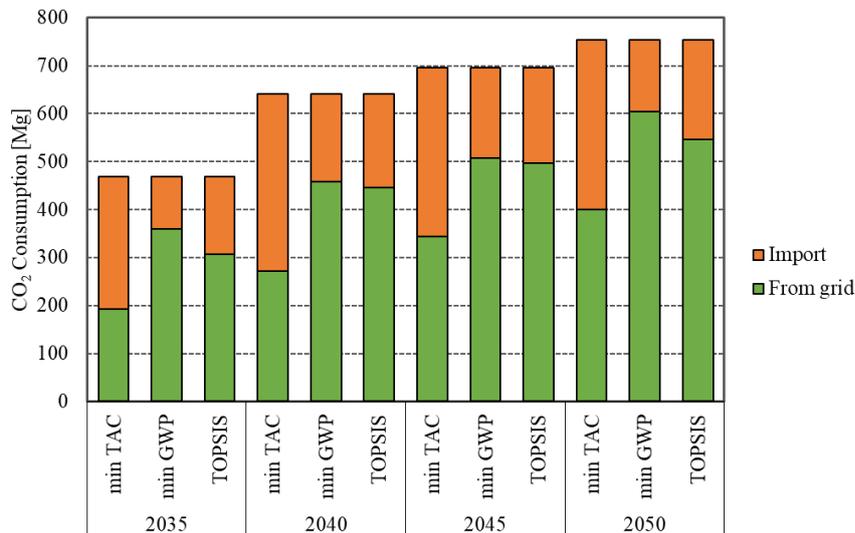


Figure 6.10. CO₂ consumption for min TAC, min GWP, and TOPSIS solution

Figure 6.9 shows the electricity consumption for the production of hydrogen. The major source is wind power followed by the use of the grid. Wind energy is chosen as a priority in the three solutions because of its low cost and GWP. In addition, the energy from the network is used to supply energy to the electrolyzers already installed from wind and hydraulic sources. A part of the energy satisfied in the “min TAC” solution comes from the PV source. However, this only represents a maximum of 7% of the total energy consumption.

Figure 6.10 shows the CO₂ consumption to produce methane. The total available CO₂ could satisfy at most 79% of methane production. Nevertheless, the imported CO₂ in the “min TAC” solution is up to 58% while in the “min GWP” solution it is up to 23%. This significant import of CO₂ in the “min TAC”

solution is due to the prioritization of the use of low-cost energy sources, and the reduction of hydrogen and methanation plants against the use of the available CO₂.

Figure 6.11 shows the GWP for hydrogen (PtH_{transp}), which value corresponds to 32-44 gCO₂-eq/MWh_{H₂}. The electricity consumption represents between 63 and 74% of CO₂-eq emissions, in which the highest value corresponds to “min TAC”. Figure 6.12 shows the GWP for methane (PtM), which lies between 95 and 121 gCO₂-eq/MWh_{CH₄}. The methanation process with CO₂ consumption represents 61-71% of CO₂-eq emissions. Besides, the difference in equivalent emissions between the “min TAC” and “min GWP” solutions can be up to 20%. Since energy use and CO₂ sources have the greatest impact on hydrogen and methane production, the results obtained are close to the values indicated for PtG (Wind), since most of the energy consumed comes from wind sources.

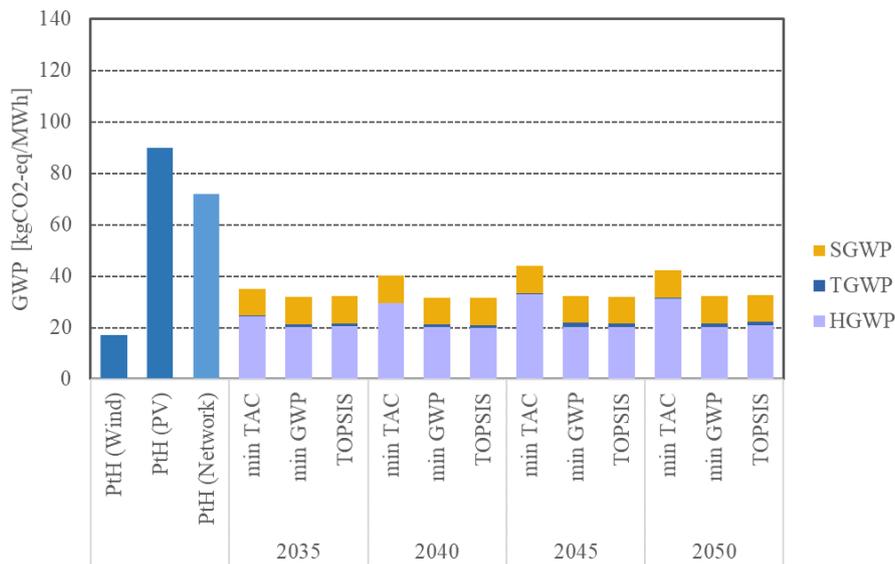


Figure 6.11. GWP of hydrogen associated with each solution per year, compared to PtH (Wind): Hydrogen produced from wind energy source (Collet et al., 2017); PtH (PV): Hydrogen produced from solar source (Collet et al., 2017); and PtH (Network): Hydrogen produced from French electricity network (Reiter and Lindorfer, 2015)

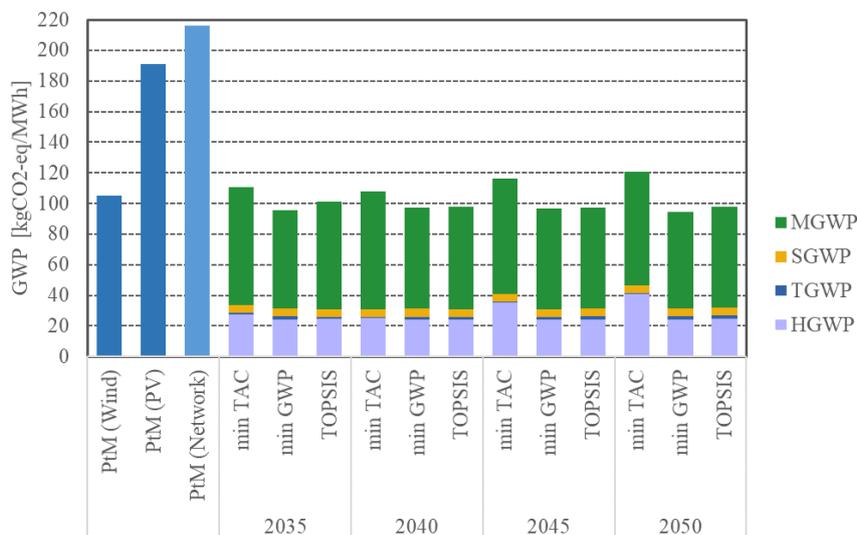


Figure 6.12. GWP of methane associated with each solution per year, compared to PtM (Wind): Methane produced from wind energy source (Collet et al., 2017); PtM (PV): Methane produced from solar source (Collet et al., 2017); and PtM (Network): Methane produced from French electricity network (Reiter and Lindorfer, 2015)

6.5.3 Sensitivity analysis

Table 6.5. Parameters for sensitivity analysis

Parameter	Values
Baseline	r = 5.25% Electricity and CO ₂ in Table 4.2
Discount rate (r)	0; 2.0; 7.5; 10 %
Electricity cost and CO ₂ cost	Variation of ±30%

Figure 6.13 and Figure 6.14 show the LCOE for hydrogen (PtH_{transp}) and methane (PtM), respectively. In both cases, the sensitivity to three parameters was studied: discount rate, electricity cost, and CO₂ cost (see Table 6.5). The cost of electricity has a significant impact on the LCOE. The LCOE for hydrogen lies between 119 and 159 €/MWh_{H₂}, while the LCOE for Methane between 108 and 156 €/MWh_{CH₄}. These values are similar to those indicated in the ADEME reports (ADEME, 2018; E&E consultant et al., 2014). Nevertheless, as indicated above, a solution that takes into account the variation in electricity costs and the demands for methane and hydrogen over the year could have a significant impact on this indicator (Götz et al., 2016; Vandewalle et al., 2015). As for the variation of the discount rate, the LCOE increases mainly due to its influence on investment costs. It is between 128-150 €/MWh_{H₂} for hydrogen and 126-139 €/MWh_{CH₄} for methane. Although in the HMSC the production of hydrogen and methane are connected through the process of electrolysis, a variation in the cost of CO₂ does not necessarily imply a significant variation in the LCOE for hydrogen. For methane, a ±30% variation in the CO₂ implies a ±3% variation in the LCOE. In addition, the difference between the LCOE for the “min TAC” and “min GWP” solutions is negligible compared to the variation of the parameters previously addressed.

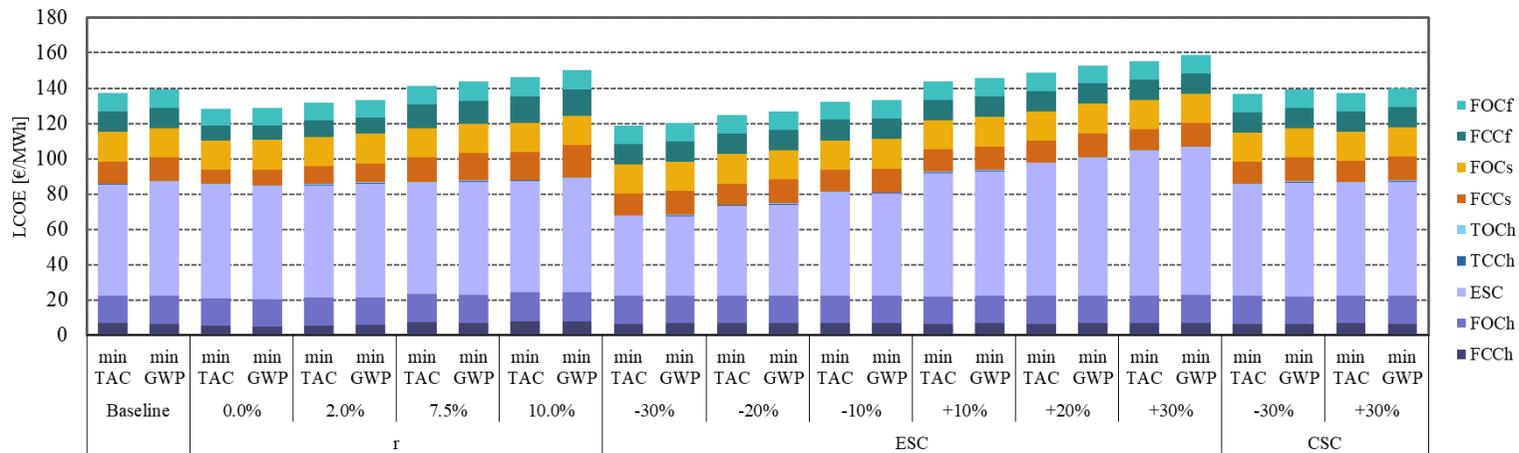


Figure 6.13. LCOE for methane according to discount rate (r), electricity cost (ESC), and CO_2 cost (CSC). The LCOE is constituted by Facility Capital ($FCCh$) and Operating ($FOCh$) Cost of Hydrogen, Cost of Electricity (ESC), Transportation Capital ($TCCh$) and operating cost ($TOCh$), Storage Capital ($FCCs$), and Operating ($FOCs$) Cost of Hydrogen, Facility Capital ($FCCf$) and operating ($FOCf$) of Refueling station.

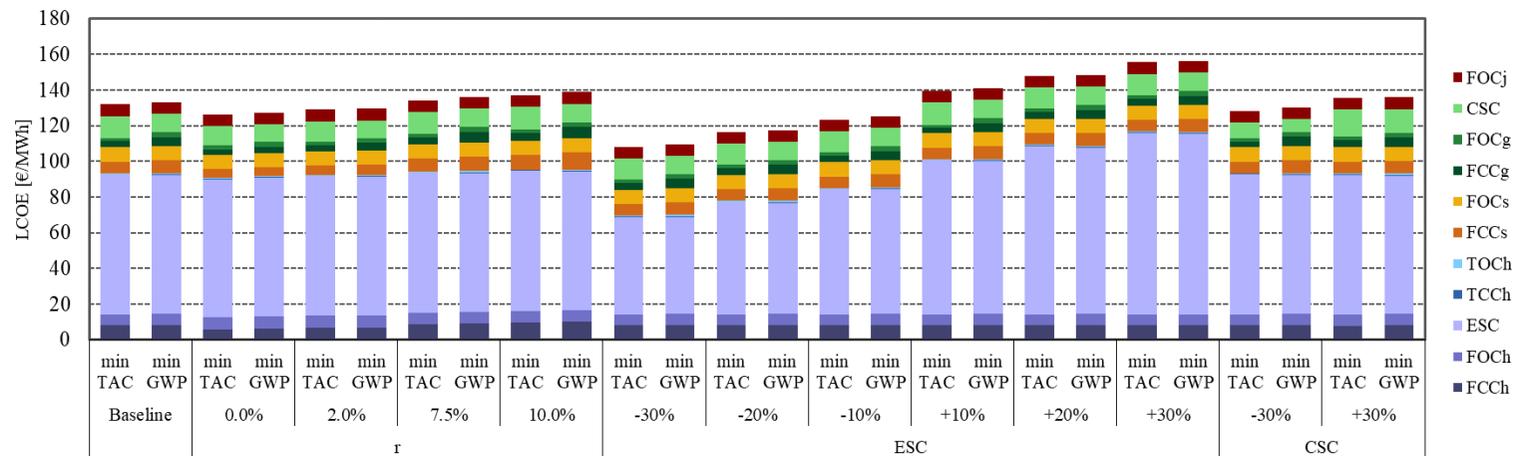


Figure 6.14. LCOE for methane according to discount rate (r), electricity cost (ESC), and CO_2 cost (CSC). The LCOE is constituted by Facility Capital ($FCCh$) and Operating ($FOCh$) Cost of Hydrogen, Cost of Electricity (ESC), Transportation Capital ($TCCh$) and operating cost ($TOCh$), Storage Capital ($FCCs$), and Operating ($FOCs$) Cost of Hydrogen, Facility Capital ($FCCg$) and operating ($FOCg$) of Methane, Cost of CO_2 source (CSC) and the cost of injection into the natural gas network ($FOCj$).

6.6 Conclusions

In this chapter, a general methodology for the design and deployment of an HMSC based on PtG systems was developed, taking the region of Occitania, France, as a case study. The methodology is based on an optimization strategy that takes into account two families of criteria, i.e. cost and environmental impact, based on a multi-period, multi-source MILP formulation. Special attention was given to distinguish the use of hydrogen to meet a demand for methane and hydrogen for the transport sector, to calculate economic and environmental indicators. To this end, three physical allocation factors were defined for the production, transport, and storage of hydrogen. A mono-objective optimization was applied to obtain two solutions: the minimization of TAC ("Min TAC") and the minimization of GHG ("min GWP"). To obtain the optimal solution set, the Pareto front is computed through the AUGMECON method. The m-TOPSIS is used to choose the optimal solutions according to the chosen criteria (cost and GWP indicators). The values of the main decision variables and the temporal configuration of each solution on the territory, the production of hydrogen and methane on each grid, and the consumption of CO₂ and electricity according to their sources can thus be obtained from the optimization framework. A sensitivity assessment was carried out to study the influence of three parameters: discount rate, cost of electricity, and cost of CO₂ on the LCOE for hydrogen and methane.

From the results obtained, it can be seen that the configuration of the HMSC changes significantly according to the decision criteria. Besides, since the wind energy source has a low cost and associated low CO₂ emissions compared to the PV source, this is the favored energy in both solutions and directly influences the configuration of the supply chain. The GHG of the methanation process has an important weight in the CO₂ emissions and not in the cost, so the use of almost all the CO₂ available in the territory was proposed for the "min GWP" solution. However, it implies an increase in investment costs.

The AUGMECON strategy combined with m-TOPSIS proved to be useful to obtain and analyze different optimal solutions, according to the chosen criteria. The results obtained were compared with some previous studies carried out in France and the effect of the hybridization of the supply chain, i.e. satisfaction of simultaneous demands of hydrogen and methane has been studied. It indicates a decrease in the cost of methane. The LCOE obtained for both energy vectors shows that only hydrogen can be economically competitive according to the literature review. Other drivers must be explored in the optimization with additional decision criteria that may boost methane production.

The sensitivity analysis shows that the cost of electricity and the discount rate are key parameters. An extension could be achieved by considering the influence of energy source, demand variability, the use of alternative other technologies, and the CO₂ supply chain.

7 BEYOND RENEWABLE POWER-TO-GAS: COMPETITION WITH FOSSIL SOURCES

ABSTRACT

This chapter focuses on the design of Hydrogen and Methane Supply Chains (HMSC), focusing on Power-to-Hydrogen (PtH) and Power-to-Methane (PtM) concepts, and their interactions with other technologies, and energy vectors (i.e., Steam Methane Reforming – SMR, and natural gas). The methodological framework developed is based on a Mixed Integer Linear Programming (MILP) approach with augmented ϵ -constraint implemented in the GAMS environment according to a multi-period approach (2035-2050). Several available energy sources (wind, PV, hydro, national network, and natural gas) for hydrogen production through electrolysis and SMR are included. Methane demand can be met by methanation or importing natural gas. For this reason, particular attention will be paid to differentiate between fossil and renewable sources. The objective to be minimized in the mono-optimization approach is the total annual cost considering the externality of greenhouse gas emissions through the carbon price for the whole HMSC over the entire period studied. The multi-objective optimization includes as objectives the total annual cost, greenhouse gas emissions, and the total methane production from methanation. A modified Technique for Order of Preference by Similarity to Ideal Solution (m-TOPSIS) has been implemented to select a good compromise solution from the Pareto front. The optimization variables involve the number and size of production and storage units, the number of tanker-trucks for hydrogen transport as well as the flows of imported/exported hydrogen from one grid to another one. The methodology is applied to the case study of Occitania (France). The Levelized Cost of Energy (LCOE), and the greenhouse gas emissions for each energy carrier are also

computed. Finally, a sensitivity assessment is carried out to study the influence of three parameters on the LCOE (i.e., discount rate, cost of electricity, and carbon price).

RÉSUMÉ

Ce chapitre est consacré à la conception des chaînes logistiques de l'hydrogène et de méthane (HMSC), en se concentrant sur les concepts Power-to-Hydrogen (PtH) et Power-to-Methane (PtM), et leurs interactions avec d'autres technologies et vecteurs énergétiques (c'est-à-dire le reformage du méthane à la vapeur - SMR, et le gaz naturel). Le cadre méthodologique développé est basé sur une approche de programmation linéaire mixte en nombres entiers (MILP) avec la méthode « augmented ϵ -constraint », mise en œuvre dans l'environnement GAMS selon une approche multi-période (2035-2050). Plusieurs sources d'énergie disponibles (éolienne, photovoltaïque, hydraulique, réseau national et gaz naturel) pour la production d'hydrogène par électrolyse et par SMR sont incluses. La demande de méthane peut être satisfaite par méthanation ou l'importation de gaz naturel. Pour cette raison, une attention particulière sera accordée à la différenciation des sources fossiles et renouvelables. L'objectif à minimiser dans l'approche mono-objectif est le coût annuel total en considérant l'externalité des émissions de gaz à effet de serre par le biais du prix du carbone pour l'ensemble du HMSC sur toute la période étudiée. L'optimisation multi-objectifs inclut comme objectifs le coût annuel total, les émissions de gaz à effet de serre et la production totale de méthane par méthanisation. Le « modified Technique for Order of Preference by Similarity to Ideal Solution » (m-TOPSIS) a été mise en œuvre pour sélectionner une bonne solution de compromis sur le front de Pareto. Les variables d'optimisation concernent le nombre et la taille des unités de production et de stockage, le nombre de camions-citernes pour le transport de l'hydrogène ainsi que les flux d'hydrogène importés/exportés d'un réseau à l'autre. La méthodologie est appliquée à l'étude de cas de l'Occitanie (France). Le coût de l'énergie (LCOE) et les émissions de gaz à effet de serre pour chaque vecteur d'énergie sont également calculés. Enfin, une étude de sensibilité est réalisée vis-à-vis de l'influence de trois paramètres sur le LCOE (à savoir le taux d'actualisation, le coût de l'électricité et le prix du carbone).

Abbreviations

CCS	Carbon Capture and Storage
CP	Carbon price
FCEV	Fuel cell electric vehicle
GHG	Greenhouse gas
GWP	Global warming potential
MILP	Mixed Integer Linear Programming
SMR	Steam Methane Reforming
TAC	Total Annual Cost

7.1 Introduction and context

A pre-requisite for increased hydrogen use, in the context of climate change mitigation efforts, is hydrogen provision with zero or at least low GHG emissions. The most prominent technological bricks involved in hydrogen production are steam methane reforming (SMR) combined with Carbon Capture and Storage (CCS) and water electrolysis powered by electricity from renewable sources of energy. Thus far, both processes play only a minor role in global hydrogen production. Only 12 large-scale CCS projects are in progress globally (Kapetaki and Scowcroft, 2017) of which two are related to hydrogen (Global CCS Institute, 2018). Furthermore, hydrogen from water electrolysis covers less than 1% of the global hydrogen demand (IEA, 2020). One of the objectives of this chapter is to demonstrate if SMR from natural gas, which has the highest H/C ratio of 25% among all hydrocarbons, is prompted to be the short-term solution.

In chapters 4 and 5, two models have been presented that address different aspects of HMSC based on Power-to-Gas (PtG) systems. This chapter will thus be devoted to studying the interaction with technologies such as SMR that can be useful to accelerate the transition⁴. Figure 7.1 shows the HMSC studied in this chapter.

A correlated aspect that will be studied within the scope of this chapter is the influence of environmental regulations on the configuration of the HMSC. Carbon price, as one of the main environmental measures in the focus of policies that seek to drive the energy transition, has been incorporated into the model. It must be highlighted that it is viewed as an essential driver to promote the penetration of renewable energy sources in the energy network (Li et al., 2020; Milani et al., 2020) because it penalizes the technologies with higher GHG emissions. For this study, the interest of its incorporation is to evaluate the “PtG degree” in supply chains and trace the resulting grey and green hydrogen as well as synthetic renewable methane as opposed to natural gas import if necessary to satisfy the demand.

The main contributions of this chapter are to address various aspects of the HMSC (i.e, primary resources, production units, hydrogen transportation, storage, and refueling stations), taking into account renewable and fossil sources. The evolution of the HMSC over time according to different criteria (TAC, GHG, and synthetic methane production), and the incorporation of a carbon price will be investigated. The HMSC addressed in this chapter is illustrated in Figure 7.1.

The chapter is paper is divided into five sections following this introduction which sets the context and its scientific objective. Section 2 is dedicated to the problem formulation and the mathematical model.

⁴ This work has been submitted for publication in *Computers & Chemical Engineering* from ELSEVIER

The solution strategy is discussed in Section 3. Finally, the main results (i.e., mono and multi-objective optimizations, and decision variables) are analyzed in Section 4, followed by the conclusions in Section 5.

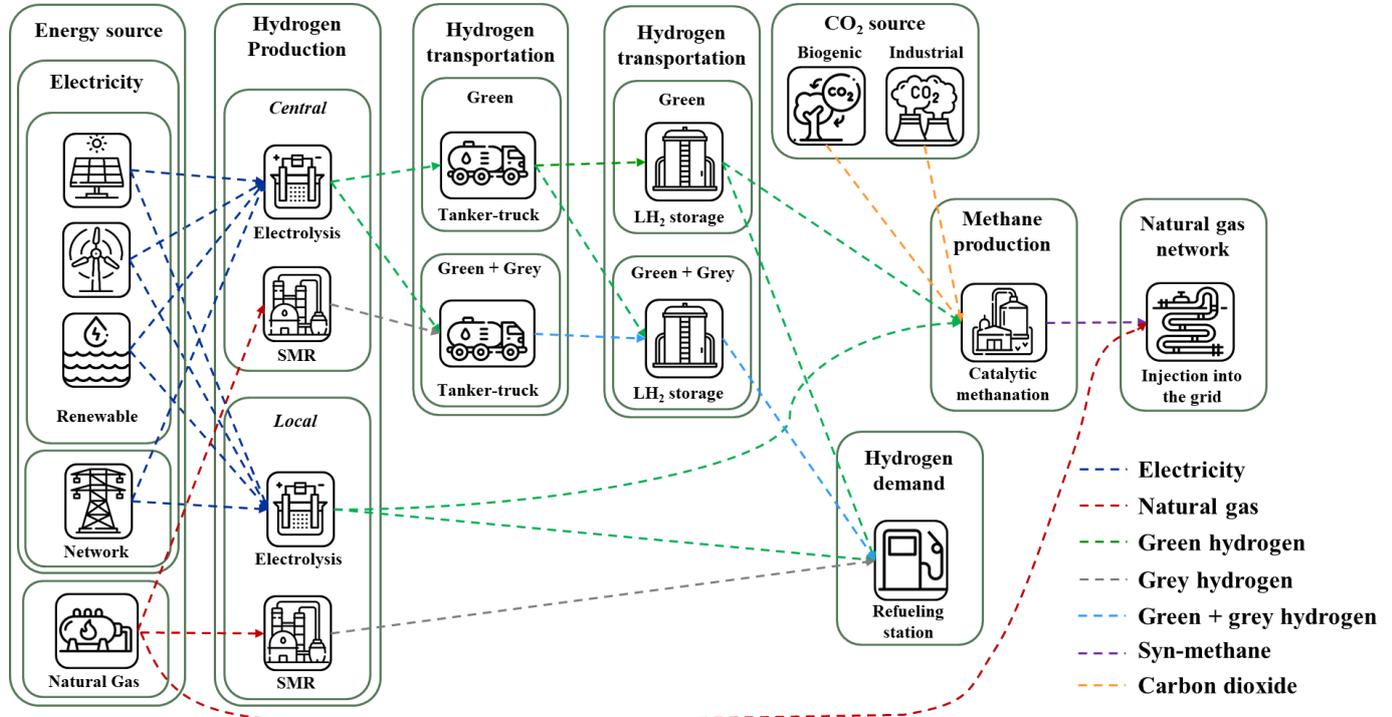


Figure 7.1. HMSC studied in this chapter

7.2 Methodology and formulation of the HSC problem

7.2.1 Problem definition

The proposed new model based on what was presented in the previous chapters can solve problems posed in the following terms:

Given:

- Spatio-temporal demands for hydrogen and methane
- Spatio-temporal availability of primary renewable and *fossil* energies, and CO₂ sources.
- Characteristics of each technology (i.e., Capital costs, O&M costs, Capacities, CO₂-eq emissions...)

Assuming:

- A division of the territory into grids.
- *SMR*, *PtG*, and *natural gas* are used to satisfy the methane and hydrogen demands.
- Direct injection of the methane produced into the natural gas network.
- Limited availability of primary energy and CO₂ sources.
- Possibility of importing primary resources.

- Possibility of transporting hydrogen only between grids by tanker-trucks.
- *The carbon price is constant throughout the period studied.*

Determine:

- The location, number, capacity, and rate of production, storage, transport, and refueling stations of hydrogen and methane.
- Transport flows between the grids.
- The specification of the sources, energy quantity, and CO₂ consumed.
- Costs and CO₂-eq emissions related to each section of the supply chain.

Subject to:

- The satisfaction of the demands for hydrogen and methane.
- Conservation of mass and energy and other physical laws.

Objective:

- Minimize Total Annual Cost.
- Minimize Greenhouse Emissions (GWP).
- *Maximize Total Methane Production.*
- Combination of them through ϵ -constraint method.

The methodology framework adapted to this chapter is shown in Figure 7.2.

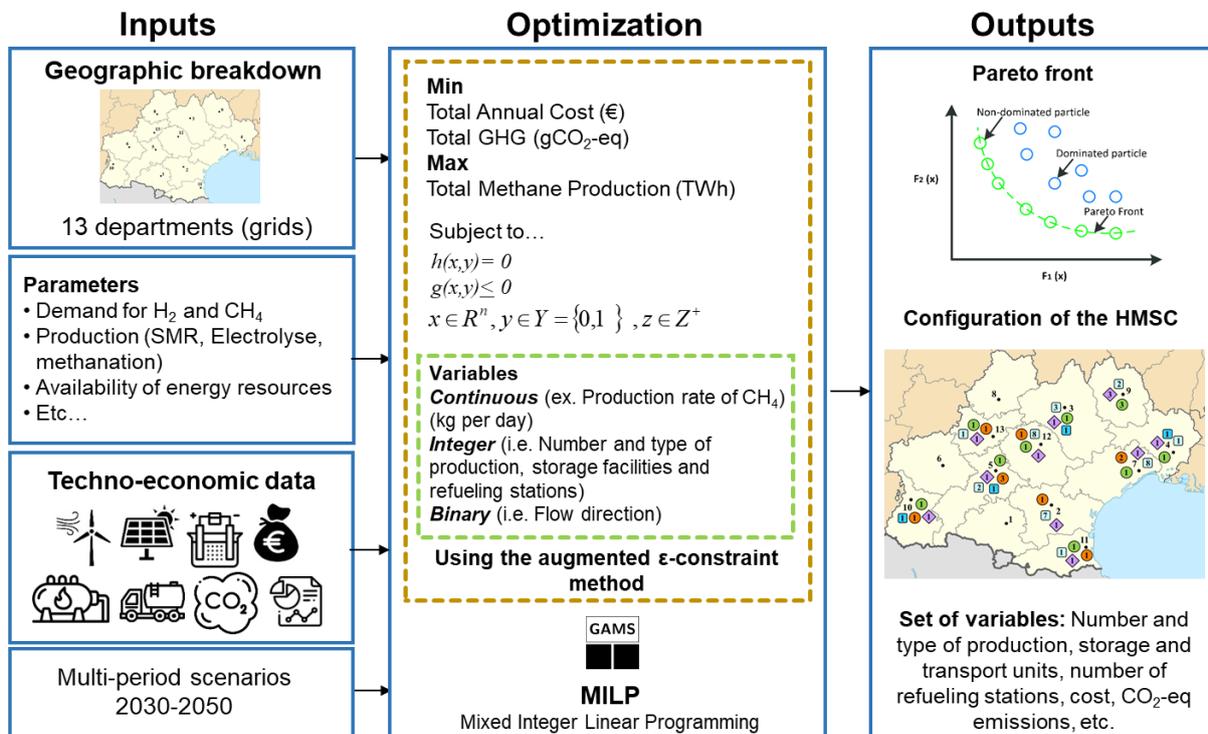


Figure 7.2. Methodology framework for the HMSC

The previous model was extended to include the possible production of hydrogen through SMR, and the satisfaction of methane demand through natural gas. It aims to meet a demand for final product “i” (hydrogen and methane), through fossil or renewable energy sources “b”, and considering the potentialities of each grid “g”. The availability of primary resources “e” (electricity and natural gas) and “c” (CO₂), hydrogen production type “p”, storage option “s”, transport mode “l”, methane production type “q”, and refueling station “f” for each period “t” with years “d” are considered. The model includes modifications to meet hydrogen and methane demands in a differentiated mode when fossil and renewable energy sources are used simultaneously. Thus, the set of equations used in Chapter 5 were modified and adapted to the HMSC studied in this chapter, since only PtG systems are considered to meet methane and hydrogen demands. The main modification in the sets, parameters, variables, equations, and constraints are described below.

Sets

b Type of energy source (fossil or renewable)

Parameters

CP Carbon price

UCIM_t Cost of import natural gas in period *t*

Sources

CA_{cg_t} Initial average availability of CO₂ source *c* in grid *g* during period *t* (Kg per year)

EA_{eg_t} Initial average availability of primary energy source *e* in grid *g* during period *t* (MWh per year)

UCC_{ct} Unit CO₂ cost for source *c* in period *t* (€ per kg)

UEC_{et} Unit electricity cost for source *e* produced in period *t* (€ per kg)

Variables

AEC_t Annual emission cost in period *t*

CIM_t Cost of imported methane in period *t*

IM_{ibt} Import methane for final product *i*, from source *b*, in period *t*.

TMP Total methane production

7.2.2 Methane demand

The total demand for methane (MD_{it}^T , kg/year) (see Eq. 7.1) is met by the methane demand satisfied locally (MD_{ibt}^L , kg/year) (see Eq. 7.2), and the import of natural gas (IM_{ibt} , kg/year). A constraint was added to indicate that importing methane only satisfies the demand for methane as a final product.

$$MD_{it}^T = \sum_{gb} (MD_{ibt}^L + IM_{ibt}) \forall i, t \quad \text{Eq. 7.1}$$

$$MD_{ibt}^L = \sum_g MPR_{ibgt}^T \forall i, b, t \quad \text{Eq. 7.2}$$

7.2.3 Distinction between renewable and fossil energy sources

The production of hydrogen through electrolysis is an intermediate process to satisfy both the hydrogen demand of the transport sector and the production of renewable methane. For this reason, a distinction must be made between hydrogen produced through SMR and electrolysis. Hydrogen destined to satisfy the demand of the transport sector can be stored and transported in the same units, without considering the process used for its production. However, the hydrogen used to produce methane through methanation must be produced entirely by electrolysis. Therefore, hydrogen storage and transport units should be designed for this purpose. The set "b" was used to distinguish between renewable and fossil energy sources.

7.2.4 Cost objective function

The cost objective function is the Total Annual Cost (TAC , €) (Eq. 7.6), which involves the total annual cost of the HMSC in each period (TAC_t , €/year) (Eq. 7.3), was modified to include the cost of the natural gas imported to meet part of the methane demand (CIM_t , €/year) (Eq. 7.4) and the annual emission cost (AEC_t , €/year) (Eq. 7.5).

$$TAC_t = \sum_d \frac{MFCC_t + HFCC_t + SFCC_t + HTCC_t + FFCC_t}{(1+r)^{(nc_t)}} + \sum_d \frac{MFOC_t + HFOC_t + SFOC_t + HTOC_t + FFOC_t + CIM_t + AEC_t}{(1+r)^{(na_t)}} \quad \forall t \quad \text{Eq. 7.3}$$

$$CIM_t = UCIM_t IM_{ibt} \quad \text{Eq. 7.4}$$

$$AEC_t = (CP)(GHG_t) \quad \text{Eq. 7.5}$$

$$TAC = \sum_t TAC_t \quad \text{Eq. 7.6}$$

7.2.5 Environmental objective function

The Global Warming Potential (GWP) is taken as the environmental impact of the supply chain. The GWP_t related to the import of methane ($IMGWP^T_t$, gCO₂-eq per year) (Eq. 7.7) was included in this function.

$$IMGWP^T_t = \sum_{ib} (IM_{ibt} IMGWP_b) \quad \text{Eq. 7.7}$$

7.2.6 Methane production objective function

The total production (TMP , kgCH₄) of renewable methane over the entire study period was defined as an objective function. This corresponds to the total methane produced in the territory through methanation (see Eq. 7.8).

$$TMP = \sum_{ibgt} (MPR_{ibgt}^T) \quad \text{Eq. 7.8}$$

The superstructure of the supply chain is shown in Figure 7.3. Compared to the previous chapter, the possibility of using fossil energy sources (natural gas) and SMR processes was incorporated. A fundamental aspect is the distinction of energy sources to avoid inconsistencies such as the production of methane through hydrogen produced by SMR processes. This involves the hydrogen transport and storage units.

7.3 Solution strategy

The model involves a MILP formulation. The objective function TAC is chosen to carry out a mono-objective optimization for the low and high carbon price. Both optimizations are solved through CPLEX 12 in the GAMS environment. Subsequently, a multi-objective optimization with these three objective functions (TAC, GWP, and TMP) is performed. The Pareto front was obtained for this purpose through the augmented ε -constraint method (AUGMECON) (Mavrotas, 2009), which allows obtaining strong optimal solutions. This method avoids redundant iterations, compared to the conventional ε -constraint method (Miettinen, 1998), and other methods as the “weighted sum approach” (Costa-Carrapiço et al., 2020). With the optimal solution set obtained, the modified-TOPSIS (m-TOPSIS) was applied to choose a compromise solution. It is a variant of the Technique for Order of Preference by Similarity to Ideal Solution (TOPSIS) (Hwang and Yoon, 1981; Yoon, 1987).

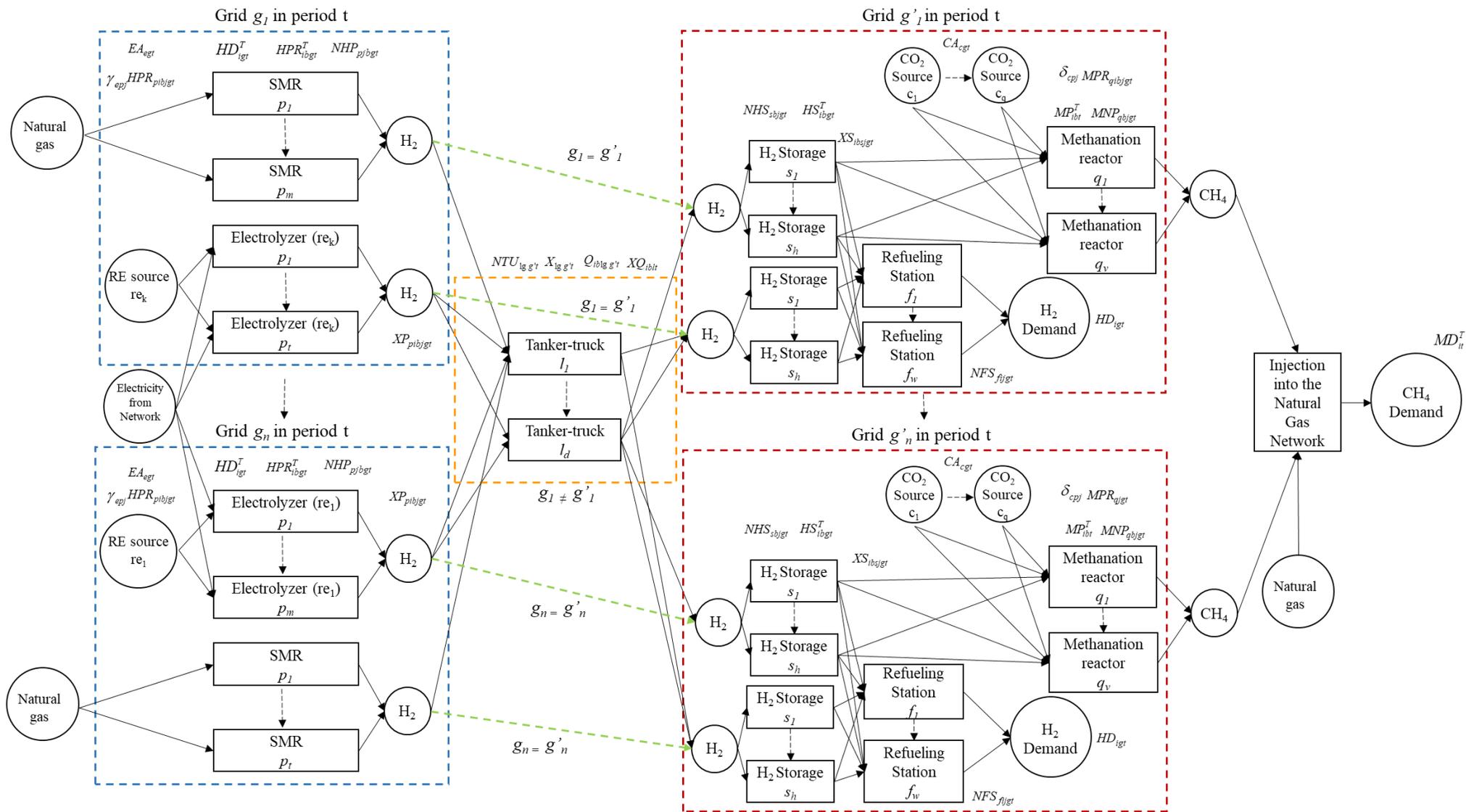


Figure 7.3. Superstructure of the multi-period model for the HMSC

7.4 Results and discussion

7.4.1 *Mono-objective optimizations*

Two mono-objective optimizations were carried out for the TAC to evaluate the impact of low and high CP in the HMSC configurations. The values of the main decision variables are shown in Table 7.1. The high CP solution involves a 31% increase in TAC compared to the low CP solution. However, a 34% reduction in GHG is achieved. The set of units are installed progressively throughout the period studied. In both cases, the same number of refueling stations are installed, because their contribution to the GHG is negligible. For the low CP solution, the number of installed methane plants is zero, and only one plant is installed for the high CP solution, due to the high costs associated with the production of synthetic methane compared to natural gas.

The costs for each stage of the HMSC indicate that the investment costs for the hydrogen plants and the refueling station, the costs of the energy sources, and the CP are determining parameters in the TAC. The investment cost of production units and refueling stations can be as high as 16% and 17%, respectively, compared to the total cost in 2035-2039. It is due to the high investment in this period. The energy source costs contribute between 32 and 54% for the low CP solution, and 25 to 34% for the high CP solution. As for the CP, it can be as high as 14% and 40%, for the low and high CP solutions, respectively.

The configurations of the HMSC for low and high CP in each period are shown in Figure 7.4 and Figure 7.5. For both cases, it can be seen that decentralized HMSC is obtained. The production of hydrogen from electrolysis meets a significant proportion of the demand in both solutions, ranging from 48 to 75% in the low CP solution, and from 81 to 100% for the high CP solution. The lower costs and GWP of the wind source prioritize its use over the solar source (see Table 4.3). The hydraulic energy source is present in both solutions, however, in a minor quantity due to its reduced availability. In both solutions, the demand for methane is met with natural gas. Only one methane plant is installed, satisfying less than 1% of the demand. The structure of both HMSC is decentralized. However, the high CP solution implies the increased use of electrolyzers, and therefore a further decentralization to use the available energy sources. Similar configurations have been obtained in previous studies (Bramstoft et al., 2020; Mesfun et al., 2017; Quarton and Samsatli, 2018). However, the inclusion of other modes of transport at different scales (i.e., injection into the gas network, and installation of pipelines for the exclusive use of hydrogen), could modify the configuration of the network proposed (Moreno-Benito et al., 2017). A decrease in hydrogen cost indicators in each period is observed, mainly due to a reduction in investment costs (see Table 7.1). The GWP of hydrogen decreases by up to 52% in the high CP solution compared to the low CP solution.

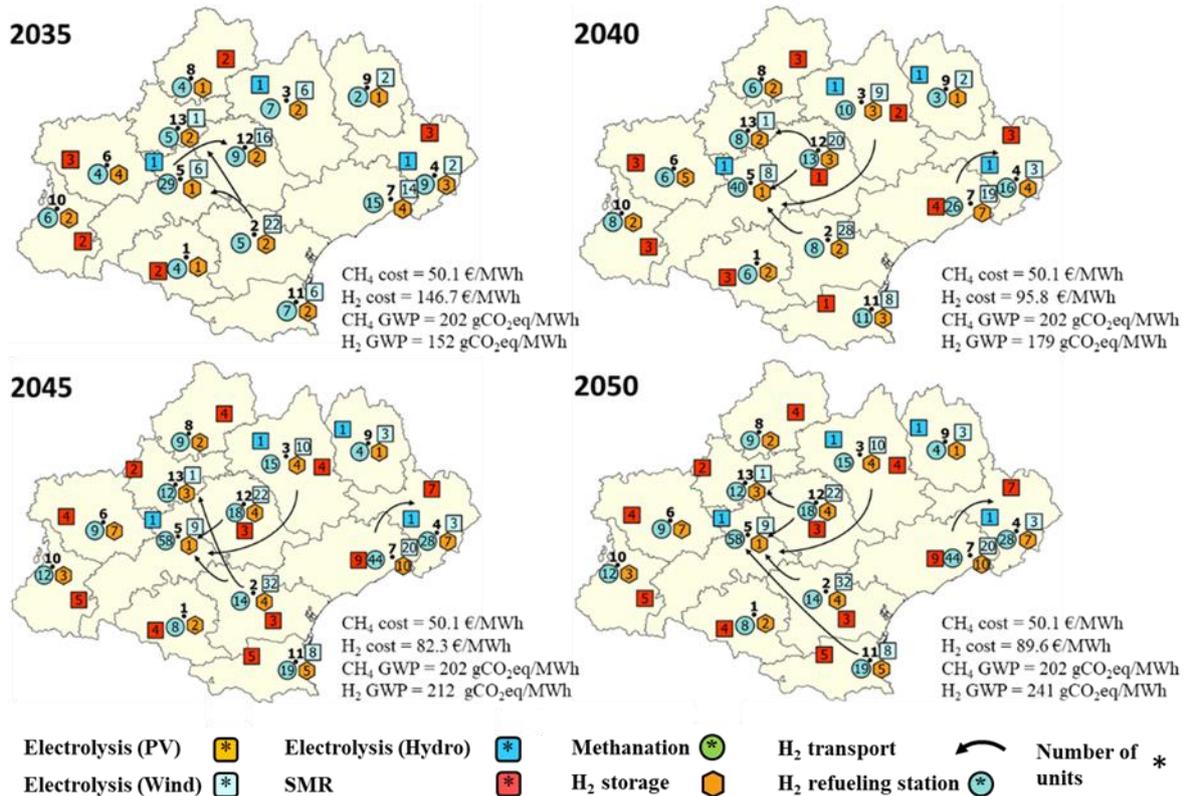


Figure 7.4. Supply chain configuration for low CP

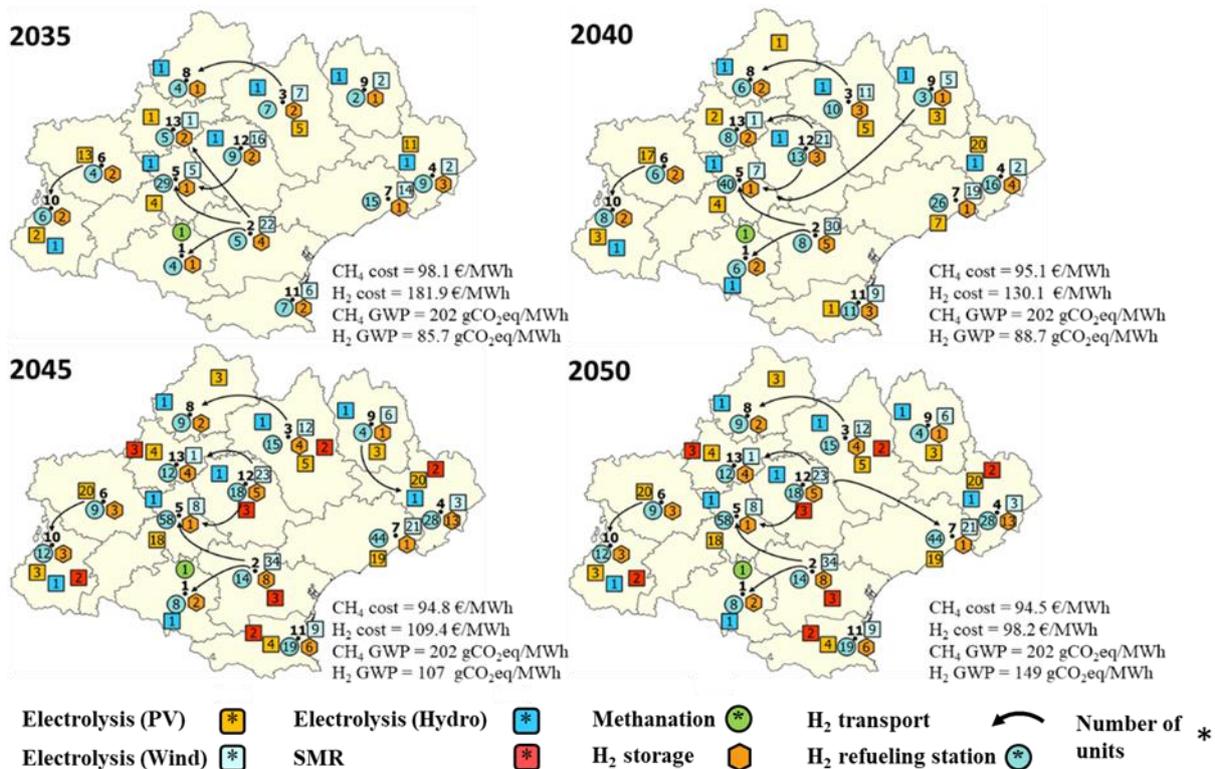


Figure 7.5. Supply chain configuration for high CP

7.4.2 *Multi-objective optimization and comparison*

A multi-objective optimization was carried out to study a scenario in which minimizing TAC, GHG, and maximizing the use of the syn-methane (TMP) are aimed. The Pareto front (Figure 7.6) allowed finding the compromise solutions through the m-TOPSIS. A solution in which the weighting is identical for the three criteria indicated above is considered in this study. The solution obtained corresponds to an HMSC in which the entire demand for hydrogen and methane is met through PtG systems. It implies an increase in the TAC of 59 and 21% with respect to the low and high CP solutions. Regarding the GHG objective function, it decreases by 48 and 21%, compared to the two solutions analyzed above.

The values of the main decision variables for the TOPSIS solution are shown in Table 7.1. Besides, the configuration of the HMSC for this solution is shown in Figure 7.7. The TOPSIS solution implies an increase in the decentralization of hydrogen production due to the satisfaction of methane demand through methanation units. Thus, the number of electrolysis units increases by 161 and 261 compared to the low and high CP solutions, respectively. Although a low GWP is achieved by meeting the demand for methane through PtG systems (a decrease of up to 51%), its cost indicator can be between 4.7 and 3.4 times superior to natural gas, compared to the previous solutions.

The main characteristics and computational effort of the proposed solutions are shown in Table 7.2. The CPU time limit is reached for the three solutions (20h), due to the number and characteristics of the variables involved (continuous, integer, and binary).

Table 7.1. Main results for the solutions obtained through the proposed methodology

Year	Low CP				High CP				TOPSIS			
	2035	2040	2045	2050	2035	2040	2045	2050	2035	2040	2045	2050
Number of facilities												
Hydrogen production	90	125	162	162	118	176	242	242	191	280	403	403
Hydrogen storage	27	37	53	53	24	31	53	53	25	33	42	42
Refueling station	106	161	250	250	106	161	250	250	106	161	250	250
Methane production	0	0	0	0	1	1	1	1	22	27	27	27
Hydrogen transport	4	7	8	9	7	10	9	11	5	6	10	14
Capital cost (M€)												
Hydrogen plants facilities	137	45.0	26.4	0.00	190	92.4	65.7	0.00	309	143	142	0.00
Hydrogen storage facilities	60.7	15.6	17.3	0.00	66.0	10.9	16.5	0.00	84.4	12.5	5.81	0.00
Refueling stations facilities	152	33.9	5.99	0.00	152	33.9	5.99	0.00	152	33.9	5.99	0.00
Methane plants facilities	0.00	0.00	0.00	0.00	0.93	0.00	0.00	0.00	26.1	3.59	5.99	0.00
Transportation modes	0.93	1.26	1.11	0.97	1.62	1.80	1.25	1.18	1.16	1.08	1.39	1.51
Operating cost (M€)												
Hydrogen plants facilities	56.7	49.4	41.4	5.82	81.9	79.2	73.4	10.0	133	127	129	17.4
Energy cost (hydrogen)	290	322	309	61.8	293	305	270	51.2	463	459	374	61.8
Hydrogen storage facilities	15.3	17.7	17.6	3.59	15.4	17.7	17.6	3.59	23.1	26.0	24.6	4.88
Methane plants facilities	0.00	0.00	0.00	0.00	0.21	0.16	0.13	0.02	5.91	5.39	4.17	0.71
Refueling stations facilities	68.7	48.7	12.0	12.1	68.7	48.7	12.0	12.1	68.7	48.7	12.0	12.1
CO ₂ cost	0.00	0.00	0.00	0.00	0.16	0.10	0.08	0.01	17.2	20.6	17.3	3.55
Injection cost	0.00	0.00	0.00	0.00	0.12	0.07	0.06	0.01	12.9	13.6	11.4	2.12
Transportation modes	1.73	2.01	1.80	0.39	2.19	2.53	1.87	0.43	1.72	1.75	2.10	0.50
Natural gas (Methane demand)	78.4	83.1	69.7	12.9	77.7	82.7	69.4	12.9	0.00	0.00	0.00	0.00
Carbon price	54.6	68.5	73.6	16.2	212	240	248	60.8	171	199	196	39.5
Total network cost (M€)	916	687	576	114	1160	915	782	152	1469	1095	926	144
Total CO ₂ eq. emissions (TgCO ₂)	2.08	3.37	4.67	1.51	1.50	2.18	2.92	1.04	1.20	1.81	2.31	0.68
Total Annual Cost (M€)	2292				3011				3633			
Total GWP (10⁹ kg CO₂-eq)	11.63				7.64				6.00			

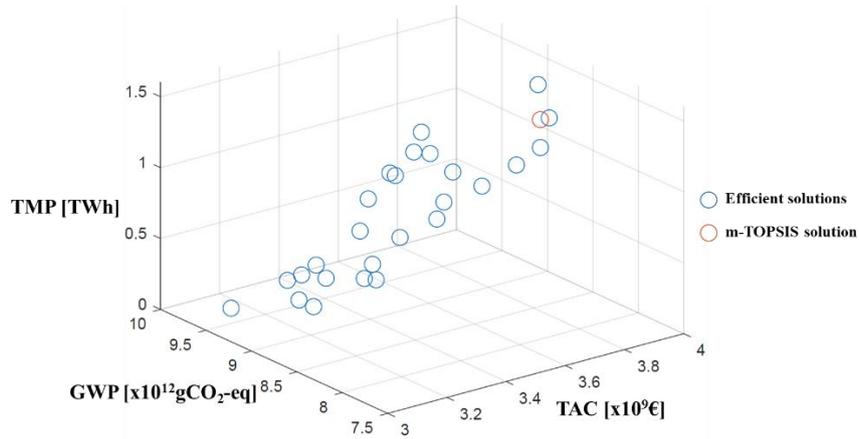


Figure 7.6. Pareto front obtained through the AUGMECON method and TOPSIS solution

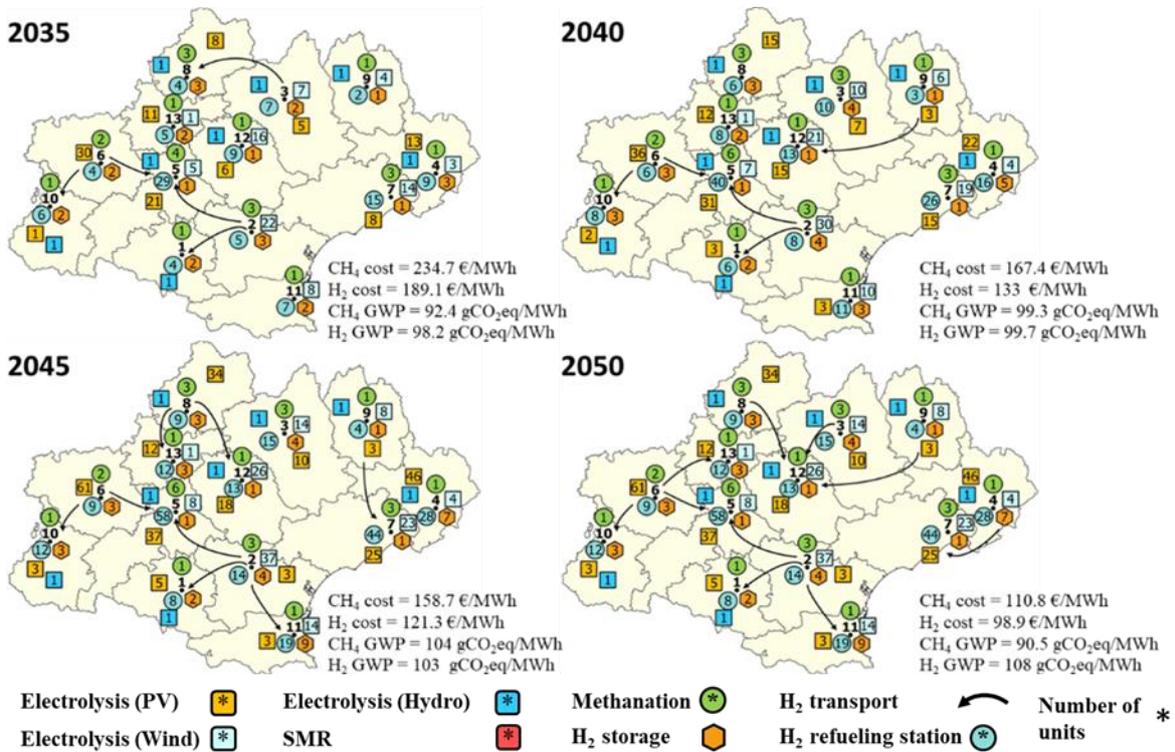


Figure 7.7. Supply chain configuration for TOPSIS solution

Table 7.2. Computational results for the optimization

Continuous variables	Binary variables	Constraints	CPU (h)			GAP		
			Low CP	High CP	TOPSIS	Low CP	High CP	TOPSIS
98,172	32,873	814,564	20	20	20	0.006	0.006	0.008

The hydrogen production in each grid for the three solutions is visualized in Figure 7.8. It can be said on the one hand that the grids with the largest amount of energy available (i.e., 2, 5, 7, 12) (see Figure 4.3) at low cost are those that produce the most hydrogen. On the other hand, grids with less energy

availability (i.e., 1, 9, 10, and 13) have lower hydrogen production. Hydrogen production through the SMR (grey hydrogen) process is present mainly in the grids where a part of the demand is met by electrolysis (green hydrogen), particularly for the low CP solution. This allows a decrease in costs to meet the demand for hydrogen in the transport sector, since a differentiation between renewable and fossil sources for storage, transport, and refueling stations is not required. For methane production, the TOPSIS solution reflects methane production all over the territory. Grid 12 concentrates 51% of methane production through methanation. Although a plant with such a capacity is technically feasible, a more detailed analysis of the logistical aspects with a finer level of territorial discretization is necessary. This is because the sources of CO₂ (mainly from the methanization and gasification process) are diffuse, and its storage would influence the configuration of the MSC.

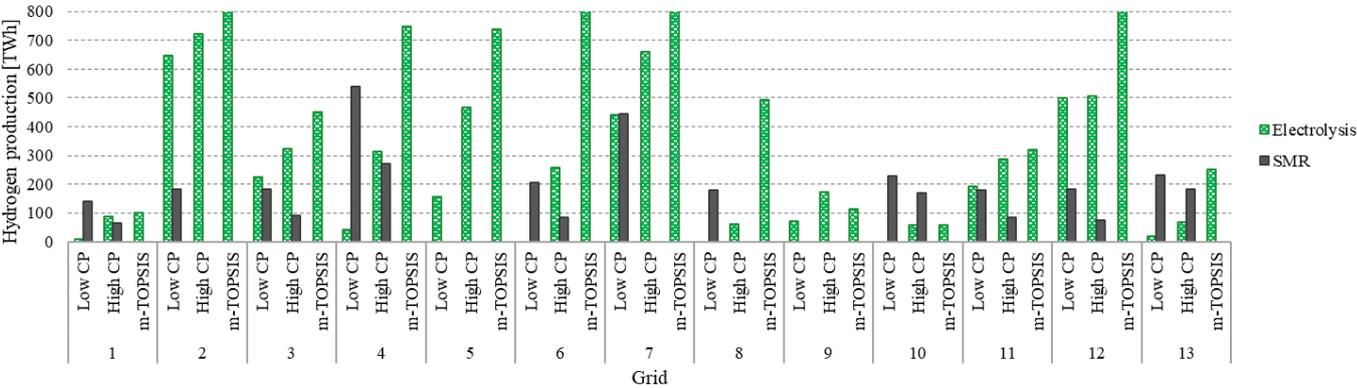


Figure 7.8. Production of hydrogen in each grid (2050)

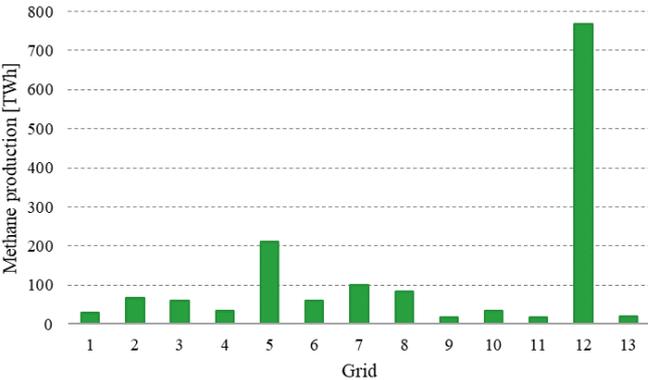


Figure 7.9. Production of methane in each grid for TOPSIS Solution (2050)

Figure 7.10 shows the energy consumption for hydrogen production. The major source is wind power for the low and high CP solutions, with 60 and 64% of the energy consumed, respectively. This is due to its low cost and GWP, compared to the solar and natural gas sources. For the low CP solution, the second source used is the natural gas source. It represents 38% of the energy consumed, with important exploitation in 2050, outperforming the wind source. For the high CP solution, the second source used is solar, representing 30% of the energy used. Natural gas consumption stands at 3.7% of energy

demand. For the TOPSIS solution, the solar and wind sources represent 52.7 and 45.7% of the total energy supplied, respectively.

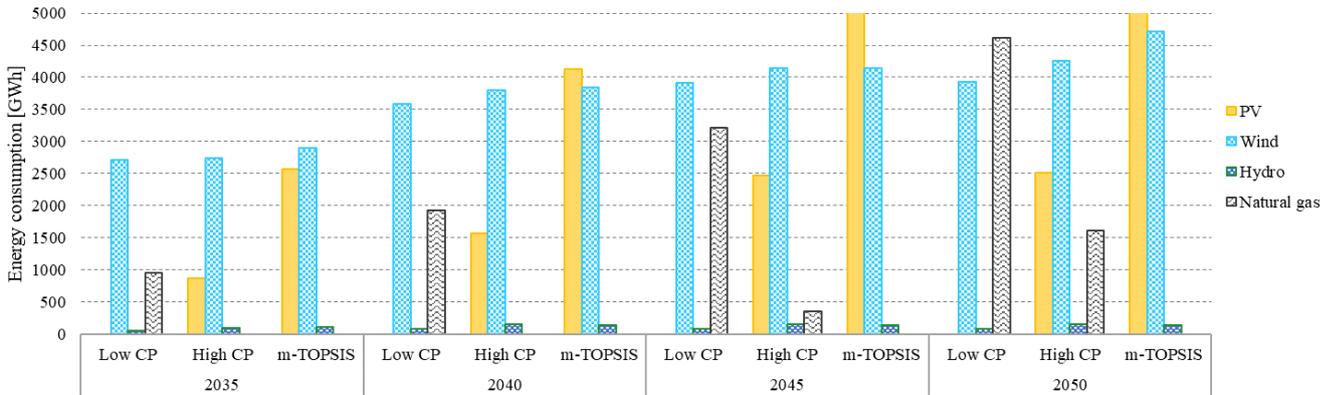


Figure 7.10. Electricity consumption for low and high CP, and m-TOPSIS solutions

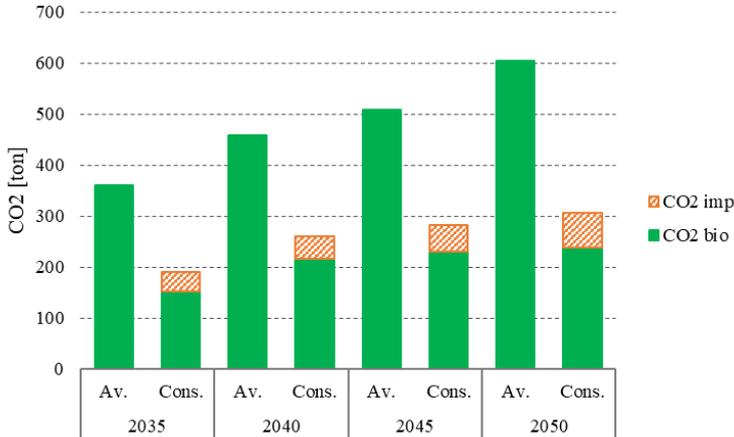


Figure 7.11. CO₂ consumption for Topsis solution

Figure 7.11 shows the CO₂ consumption to produce syn-methane in the TOPSIS solution. Between 78 and 82% of the CO₂ consumed comes from biogenic sources (from methanization and gasification) available in the territory. A part of the CO₂ comes from industrial processes although biogenic sources are available in other grids, thus avoiding the installation of additional methanation plants.

7.4.3 Sensitivity analysis

Table 7.3. Parameters for sensitivity analysis

Parameter	Values
Baseline	r = 5.25%
Discount rate (r)	Electricity cost and CP (high) in Table 4.2 0; 2.0; 7.5; 10 %

Electricity cost
Carbon price

Variation of $\pm 30\%$
0; 50; 120; 200; 270 €/ton_{CO2}

Figure 7.12 shows the LCOE for hydrogen (PtH) and methane (PtM). In both cases, the sensitivity to three parameters was studied: discount rate (r), energy source cost (ESC), and carbon price (CP) (see Table 7.3) which impacts are not linear. The LCOE for hydrogen lies between 129 and 177 €/MWh_{H2}, while the LCOE for methane between 156 and 192 €/MWh_{CH4}. Similar values have been indicated in other studies (ADEME, 2018; E&E consultant et al., 2014; Götz et al., 2016). The cost of ESC has the most significant impact on the LCOE for both vectors, standing at 29-42% for hydrogen and 30-46% for methane. Since one of the main functions of PtG systems is to recover the overproduction of electricity in a context of high penetration of renewable energies, more comprehensive research should be carried out, taking into account the variability of costs of each energy source. This fact has already been highlighted in other studies (Götz et al., 2016; Vandewalle et al., 2015). A discount rate of zero implies a decrease in the LCOE for hydrogen and methane of 40 and 30%, respectively. While the maximum discount rate of 10% represents a 20 and 30% increase. This variation is mainly reflected in capital costs. The CP represents between 5-27 and 7-25 € of the LCOE for hydrogen and methane, respectively. Despite an increase in LCOE for both vectors, only a high carbon price could drive a renewable HMSC.

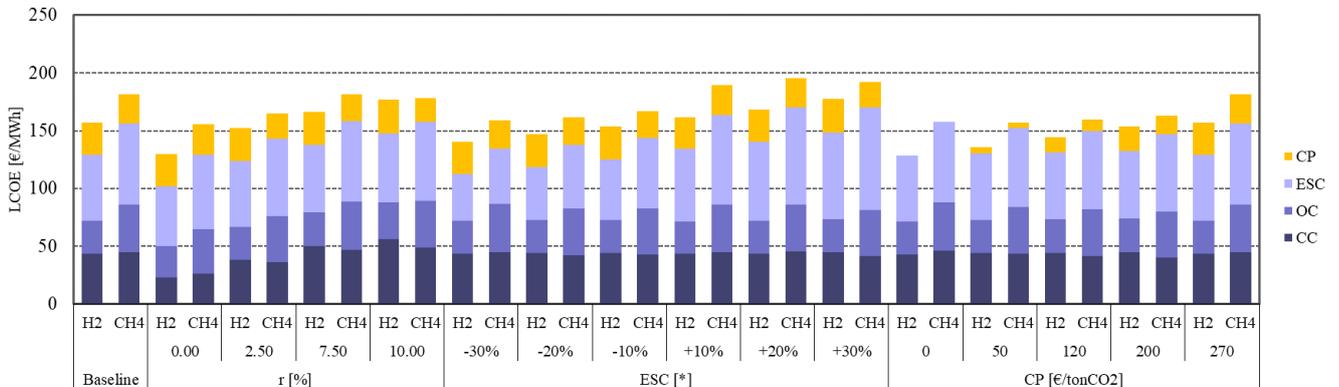


Figure 7.12. LCOE for methane according to discount rate (r), energy source cost (ESC), and Carbon Price (CP). The LCOE is constituted by Capital Costs (CC), Operating Costs (OC) ESC, and CP

In order to set the results in perspective, Table 7.4 allows comparing the LCOE and GWP obtained for hydrogen and methane in previous research with those calculated in this study. In this sense, the prior results obtained by the authors, and those reported in other studies were considered. The common point between these studies is that they all address HSC and/or without MSC with a GEOM approach in the French context. A study conducted by French institutions is also reported for comparison purposes (E&E consultant et al., 2014), as a reference roadmap. The LCOE values for hydrogen in this study is higher than those shown in (De-León Almaraz et al., 2015), which can be explained by the fact that CP is not

taken into account, and the main technology obtained by minimizing TAC is SMR, which is also reflected in the GWP. In addition, it is observed that the solution minimizing GHG emissions obtained in (De-León Almaraz et al., 2015) involves an LCOE for hydrogen outside the range obtained in this study, even superior to that obtained for the TOPSIS solution (100% renewable sources). It must be yet emphasized that in (De-León Almaraz et al., 2015), Occitania (72 724 km²; 5 845 102 inhabitants) (2017) is 1.6 times larger in surface area than the former Midi-Pyrénées region and almost twice as populated. Midi-Pyrénées involves only eight of the thirteen grids indicated in this study: 1, 3, 5, 6, 8, 10, 12, and 13. In the HMSC configuration obtained, the interactions between the grids belonging to the Midi-Pyrénées region and the external grids are observed. In particular, the energy available in grid 2 is used for the production of hydrogen and subsequently can satisfy the demand in the neighboring grids through its export using tanker-trucks.

A second study was carried out by (Li et al., 2020) with a mono-period approach (2030). A higher LCOE is observed for hydrogen, which could decrease if an extension of the horizon is made towards 2050. This could lead the LCOE closer to values similar to those obtained in this study. When this study is compared with (E&E consultant et al., 2014), a difference in costs is observed: (E&E consultant et al., 2014) considers other technologies for the production of hydrogen (i.e., PEM and SOEC) and renewable methane (biological reactors).

Table 7.4. LCOE and GWP for hydrogen and methane

Study	Period	Case of study	Objective function							
			TAC				GWP			
			LCOE [€/MWh]		GWP [gCO ₂ eq/MWh]		LCOE [€/MWh]		GWP [gCO ₂ eq/MWh]	
			H ₂	CH ₄	H ₂	CH ₄	H ₂	CH ₄	H ₂	CH ₄
(De-León Almaraz et al., 2015)	2020-2050	Midi-Pyrenees	102	--	330	--	222	--	66	--
(Li et al., 2020)	2030	Franche-Comté	288 to 1482	--	--	--	--	--	--	--
This study	2035-2050	Occitania	107 to 177	50 to 181	101 to 240	98 to 201	--	--	--	--
(E&E consultant et al., 2014)	2030-2050	France	56-86	92-160						

7.5 Conclusion

A general methodology for the design and deployment of an HMSC was developed and applied to the Occitania region (France) as a case study. The methodology is based on an optimization strategy with a multi-period, multi-source MILP formulation that allows minimizing the Total Annual Cost or obtaining an optimal solution taking into account three criteria: total annual cost (TAC), greenhouse gas emissions (GHG), and total renewable methane production (TMP). The distinction between the demands to be satisfied and energy sources (renewable and fossil) was incorporated, which allowed calculating the cost, and GHG emissions indicators according to allocation criteria. A mono-objective optimization was implemented to obtain two solutions with a low and high carbon price (CP). To obtain the optimal

solution set, the Pareto front is found through the AUGMECON method. The m-TOPSIS method is used to choose the optimal solutions according to the chosen criteria. The three solutions were analyzed through the decision variables and the configuration of the HMSC. Hydrogen and methane production (TOPSIS solution) were analyzed, as well as the energy and CO₂ sources used. A sensitivity analysis led to the study of three key parameters on the LCOE for hydrogen and methane: discount rate, cost of electricity, and CP.

The results show that the HMSC changes significantly depending on the CP applied. The configuration of the HMSC implies a nearly total satisfaction of the hydrogen demand for the transport sector through electrolysis (green hydrogen), with a high CP. Synthetic methane from renewable sources needs other drivers to support its implementation, such as the consideration of a significant reduction of GHG, and the decarbonization of the gas network. In this sense, the TOPSIS solution shows that methane and hydrogen produced for the HMSC configuration stem only from renewable sources when TAC, GHG, and TMP are considered simultaneously. From the sensitivity analysis performed, it can be deduced that only hydrogen produced by electrolysis is competitive with SMR under the parameters and scenario analyzed. In the case of synthetic methane, the available resources can meet the demand through PtG, and even if synthetic methane for grid injection is thus far from competitiveness with natural gas, PtG technologies have the most potential to decarbonize the fossil economy and achieve a circular economy via CO₂ valorization. Besides, only the production of “grey” and “green” hydrogen was taken into account in this research. A perspective to be considered in future work could be the incorporation of “blue” hydrogen production with CCU systems so that the contribution of the SMR process in a low carbon economy scenario could be studied.

8 COST, ENVIRONMENTAL, AND EXERGY OPTIMIZATION OF HYDROGEN AND METHANE SUPPLY CHAINS

ABSTRACT

Very few studies have been carried out on the exergy analysis of a PtG system based on hydrogen and methane. The scientific objective of this short chapter is to propose a first insight to build a comprehensive optimization model that can determine the design and deployment of an HMSC also considering the total exergy lost and destroyed (TELD) as objective functions to be minimized in addition to the cost and environmental criteria considered in the previous chapters. The aim is thus to quantify the loss and destruction of exergy to identify the potential of valorization. The results show that a significant part of the exergy is lost throughout the HMSC (up to 202 TWh). Not surprisingly, the highest losses are observed in energy production due to low exergy yields for renewable energy sources compared to fossil sources. Hence, in the current state of the model presented, minimization of energy losses favors the use of fossil energy sources (i.e., natural gas). The coupling of this PtG ecosystem with other energy systems is necessary to reduce the exergy losses (i.e., copying with the cement industry or with methanization processes for heat recovery). In addition, the recovery of other by-products such as oxygen could be interesting.

RÉSUMÉ

Très peu d'études ont été réalisées sur l'analyse de l'énergie d'un système PtG basé sur l'hydrogène et le méthane. L'objectif scientifique de ce court chapitre est de proposer un modèle d'optimisation complet qui puisse déterminer la conception et le déploiement d'un HMSC en considérant également l'exergie totale perdue et détruite (TELD) comme des fonctions objectives à minimiser en plus des critères de coût et d'environnement considérés dans les chapitres précédents. La démarche vise donc à quantifier la perte et la destruction de l'énergie afin d'identifier le potentiel de valorisation. Les résultats montrent qu'une partie importante de l'exergie est perdue dans le HMSC (jusqu'à 202 TWh), le couplage de cet écosystème PtG avec d'autres systèmes énergétiques est nécessaire pour réduire les pertes d'exergie (c'est-à-dire par exemple le couplage avec l'industrie du ciment ou avec les procédés de méthanisation pour la récupération de chaleur). En outre, la récupération d'autres sous-produits tels que l'oxygène pourrait être intéressante.

8.1 Introduction

In the previous chapters, the HMSC design has been tackled using an optimization methodology that mainly considers costs and environmental impact as objective functions. Since HMSC involves the interaction of different technologies with different energy performances, exergy analysis is an interesting aspect to be addressed. While energy is conserved (first law of thermodynamics), exergy is destroyed when a process is irreversible (second law of thermodynamics) (Querol et al., 2013). Hence, exergy analysis provides insight into the performance of the HMSC that would elude purely first-law analysis. The overall exergy efficiency of the HMSC can vary dramatically depending on the feedstock and technologies involved (Khosravi et al., 2018). Thus, the estimation of destroyed and lost exergy is essential to propose energy improvements and compare options.

During the past several decades, many researchers have carried out exergy analyses especially for power generation and cogeneration (Dincer et al., 2014). Exergy analysis has proven to be useful to identify losses, possible valorization, and better system integration. Thus, methodologies have emerged that integrate economic, environmental, and thermodynamic criteria (Abusoglu and Kanoglu, 2009; Kumar, 2017). In addition, new conceptual frameworks taking into account territorial particularities and negotiation mechanisms have been proposed in the context of circular economy and sustainability for industrial parks, taking into account exergy analysis (Chahla and Zoughaib, 2019). To the best of our knowledge, very few studies have been carried out on the exergy analysis of a PtG system based on hydrogen and methane. The scientific objective of this short chapter is to propose a comprehensive optimization model that can determine the design and deployment of an HMSC considering Total Annual Cost (TAC), Greenhouse gas (GHG) emissions, and total exergy lost and destroyed (TELD) as

objective functions to be minimized⁵. The typical features of the HMSC (i.e., primary resources, production units, hydrogen transportation, storage, and refueling stations) are addressed, considering the evolution of its configuration over time according to these different criteria. As in the previous chapters, Figure 8.1 shows the main aspects of the methodology framework applied for the multi-objective optimization of the HMSC.

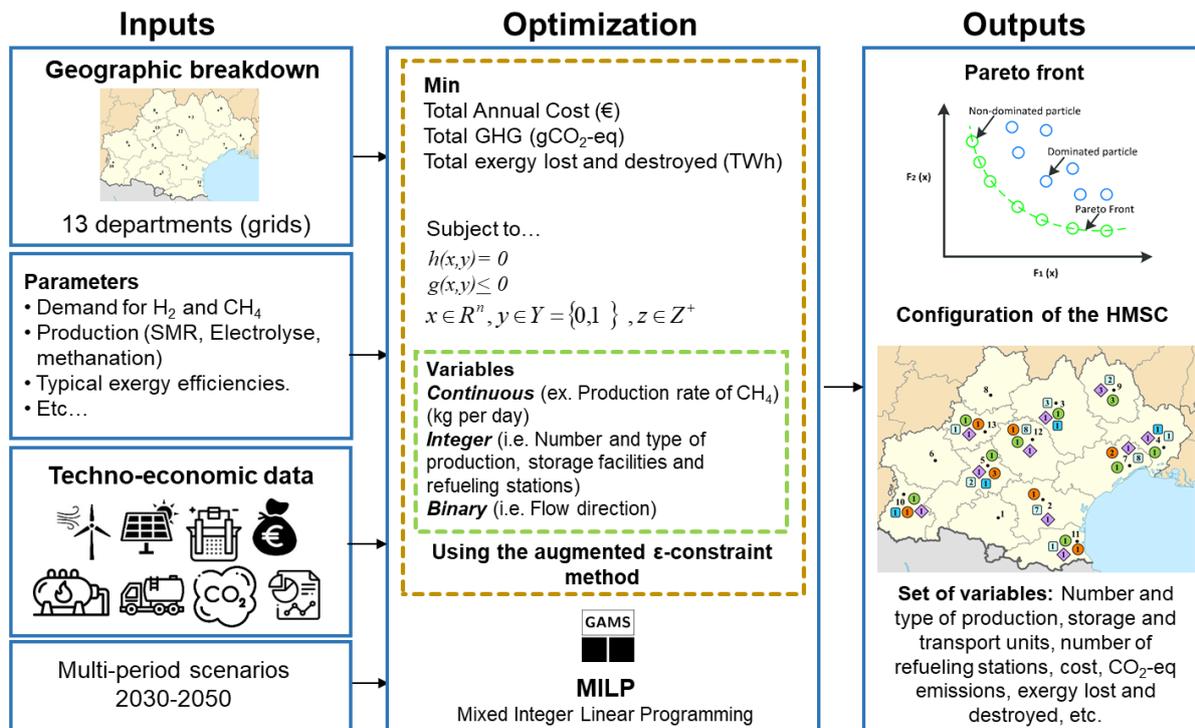


Figure 8.1. Methodology framework for HMSC

The chapter is divided into six sections following this introduction, which sets the context and its scientific objective. Section 2 is dedicated to the definition of exergy and the main equation to take into account. The methodology and formulation of the HSC problem are discussed in Section 3. Finally, the main results (i.e., mono and multi-objective optimizations and decision variables) are analyzed in Section 4, followed by the conclusions in Section 5.

8.2 Exergy definition

Exergy is a thermodynamic property. The exergy of a portion of matter is equal to the maximum useful work obtainable when taken from its given state to the thermodynamic equilibrium with the environment (Kotas, 2012). Such a final state of equilibrium is known as the dead state (Querol et al., 2013). While energy is conserved (first law of thermodynamics), exergy is destroyed when a process is irreversible

⁵ This work was accepted for publication in the 31st European Symposium on Computer-Aided Process Engineering (ESCAPE-31)

(second law of thermodynamics). Exergy can also be lost due to by-products. Hence, exergy analysis provides insight into the performance of the HMSC that would elude purely first-law analysis. The overall exergy efficiency of the HMSC can vary dramatically depending on the feedstock and the technologies involved (Khosravi et al., 2018). Thus, the estimation of destroyed and lost exergy is essential to propose energy improvements and compare options. The total exergy can be calculated by the sum of the chemical and physical exergy, since the kinetic and potential exergy can be neglected in most cases.

The physical exergy (b_{ph}) is the maximum useful work obtained by passing the unit of mass of a substance of the generic state (T,p) to the environmental (T_o,p_o) state through purely physical processes. For this calculation, the variation of enthalpy and entropy are taken into account (Eq. 8.1). Besides, the chemical exergy can be defined as the maximum useful energy, which would be attained by passing from the environmental state to the dead state through a chemical reaction. A general method for obtaining the chemical exergy of a substance X is shown Eq. 8.2, which involves the chemical exergy of each element ($b_{ch,i}(x_i)$) and the free energy of formation of X ($g_{fo}(X)$).

$$b_{ph} = [h(T, p) - h(T_o, p_o)] - T_o [s(T, p) - s(T_o, p_o)] \quad \text{Eq. 8.1}$$

$$b_{ch}(X) = \sum n_i b_{ch,i}(x_i) + g_{fo}(X) \quad \text{Eq. 8.2}$$

A simplified exergy analysis was carried out taking into account the information available in the literature. A synthetic example of a process or equipment is shown in Figure 8.2. Exergy is either lost through waste (L) or destroyed due to irreversibilities (D). Exergy efficiency (η) can be defined as the ratio between the total exergy contained in the resources “R” (TER) and the total exergy of the product “P” (TEP), which is shown in Eq. 8.3. Similarly, a coefficient (φ) can be defined by the relationship between lost/destroyed exergy (TEDL) and the TER (See Eq. 8.4). A correlation between the two parameters can be established through Eq. 8.5.

$$\eta = \frac{TEP}{TER} \quad \text{Eq. 8.3}$$

$$\varphi = \frac{TEDL}{TER} \quad \text{Eq. 8.4}$$

$$\varphi = 1 - \eta \quad \text{Eq. 8.5}$$

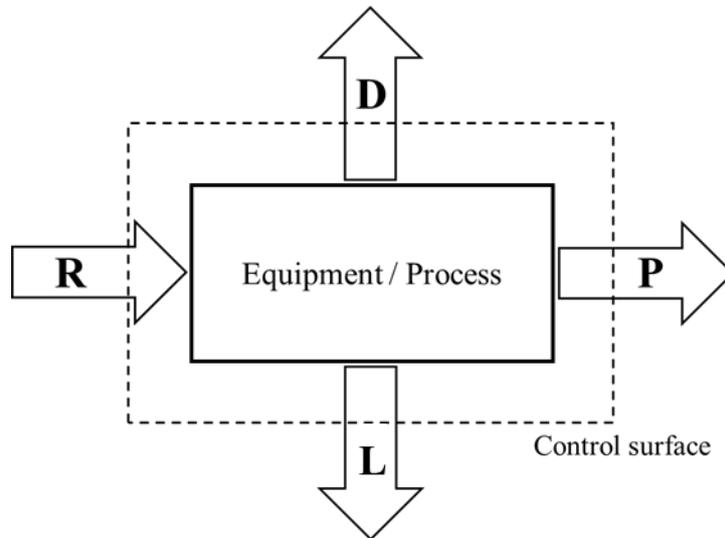


Figure 8.2. General case for equipment or process involved in the HMSC, involving exergy as resource (R), product (P), and exergy lost as waste (L) and due to irreversibilities (D)

8.3 Methodology and formulation of the HSC problem

The problem formulation can be expressed as in Chapter 5, incorporating the typical exergy efficiencies as parameters. The following objectives to be minimized have been considered either separately or with a combination through ϵ -constraint method:

- Total Annual Cost.
- GHG emissions.
- Total Exergy Loss and Destruction.

Sets

b Type of energy source (fossil or renewable)

Parameters

CP Carbon price

$UCIM_t$ Cost of import natural gas in period t

Variables

$HELD_t$ Lost/destroyed exergy related to hydrogen production

$SELD_t$ Lost/destroyed exergy related to hydrogen storage

$TrELD_t$ Lost/destroyed exergy related to hydrogen transport

$MELD_t$ Lost/destroyed exergy related to methane production

$IMELD_t$ Lost/destroyed exergy related to natural gas import

$TELD$ Total Lost/destroyed exergy for the entire supply chain for the period studied

8.3.1 Exergy objective function

The Total Exergy Lost and Destroyed (TELD) is taken as the exergy objective function (Eq. 8.6). The exergy destroyed and lost in each process was calculated considering the typical exergetic yield of each stage (Khosravi et al., 2018; Safari and Dincer, 2018). The lost/destroyed exergy related to hydrogen ($HELD_t$, MWh/year), and methane ($MELD_t$, MWh/year) production, hydrogen storage ($SELD_t$,

MWh/year), hydrogen transport ($TrELD_t$, MWh/year), and methane import ($IMELD_t$, MWh/year) are involved as follows:

$$TELD = \sum_t (HELD_t + SELD_t + TrELD_t + MELD_t + IMELD_t) \quad \forall t \quad Eq. 8.6$$

Table 8.1 shows the typical values for the efficiency of the main equipment and processes involved in the HMSC studied.

Table 8.1. Typical exergy yield for the main processes involved in the HMSC

	Typical exergy efficiency (%)	References
PV	11	(Bayrak et al., 2017; Khosravi et al., 2018)
Wind turbine	30	(Khosravi et al., 2018)
Hydro turbine	75	(Nazari-Heris and Mohammadi-Ivatloo, 2017)
Natural gas production	65	(Voldsund, 2014)
SMR	62	(Simpson and Lutz, 2007)
Electrolyzer	56	(Safari and Dincer, 2018)
Refueling station (storage)	95	(Khosravi et al., 2018)
Methanation reactor	87.2	(Safari and Dincer, 2018)
Hydrogen storage	95	(Khosravi et al., 2018)
Aqueous-MEA capture (CO ₂ sources)	46.1	(Wiesberg et al., 2019)

8.4 Results and discussion

The optimal values of the decision variables from the mono-objective approach for each objective function (TAC, TGHG, and TELD), and the TOPSIS solution chosen are shown in Table 8.2. It can thus be observed that The analysis of the three mono-objective optimizations shows a drastic change in the configuration of the HMSC. Since a significant part of the exergy is lost throughout the HMSC (up to 202 TWh), The production, storage, and transport units are installed progressively over the periods. The minimization of exergy implies no transport of hydrogen, due to the significant amount of exergy destroyed. The LCOE for hydrogen is between 130-285 €/MWh, which corresponds to the range of acceptable values to be economically viable (Hydrogen Europe, 2018; Welder et al., 2018). However, only the “min TGHG” and TOPSIS solutions propose methane production from methanation, where an important reduction of CO₂-eq emissions is imposed. For “min TAC”, the methane demand is met through natural gas, since its cost is two times lower than the one of methane produced from methanation. In the case of “min TELD”, likewise, the demand for methane is satisfied with the import of natural gas, because its production presents a lower loss and destruction of exergy.

The configuration of the HMSC for the TOPSIS solution is shown in Figure 8.3. Due to the capacity factor associated with each energy source, the HMSC configuration is decentralized. Under the applied scenario and decision criteria, the total hydrogen demand is fully satisfied by electrolysis while only 24% of the methane demand is provided by methanation (the other portion is satisfied by natural gas

imports). The cost indicator decreases progressively for hydrogen up to 47%. The cost indicator for methane varies between 96.8 and 119 euros/MWh, because approximately 86% of the demand is satisfied through natural gas. The GHG for both vectors corresponds to the range of values reported from the literature (see Figure 6.11 and Figure 6.12), and varies according to the energy sources used in each period. The exergy lost and destroyed over the period varies between 84 and 89%, mainly due to the exergetic yield associated with energy production (i.e., 11 and 30% for PV and wind power plants, respectively). The main source of electricity used is solar followed by wind sources. The main characteristics and computational effort of the solution found by the optimization model are summarized in Table 8.3.

Table 8.2. Optimization results of the HMSC

Solutions	min TAC				min TGHG				min TELD				TOPSIS							
	2035	2040	2045	2050	2035	2040	2045	2050	2035	2040	2045	2050	2035	2040	2045	2050				
Year																				
Number of H ₂ production facilities	119	181	246	246	196	283	402	402	96	98	133	133	126	190	300	300				
Number of H ₂ storage facilities	28	42	57	57	24	32	46	46	58	58	64	64	22	28	42	42				
Number of methane production facilities	0	0	0	0	13	15	16	16	0	0	0	0	3	4	14	14				
Number of H ₂ transport units	7	8	10	14	4	4	5	6	0	0	0	0	7	8	6	8				
Number of refueling station	61	92	137	137	61	92	137	137	61	92	137	137	61	92	137	137				
CH ₄ LCOE (€/MWh)		94.4					190					94.4					106			
H ₂ LCOE (€/MWh)		142					144					208					141			
Objective functions																				
TAC (M€)		3158					3930					4256					3303			
TGHG (x10 ¹² gCO ₂ -eq)		2.71					1.99					6.67					2.16			
TELD (TWh)		108					202					23					97			

When considering TELD, the results diverge from those presented in the previous chapter. For the proposed case study, a minimization of exergy losses and destruction favors the use of SMR technologies. However, these results could change if possible interactions between PtG systems with other processes are considered (i.e., methanization, steel and cement industries, etc.) and the recovery of by-products (oxygen). In this sense, the results obtained open a new perspective towards the improvement of the model and its extension towards the possible interactions necessary to increase the exergy efficiency of the HMSC.

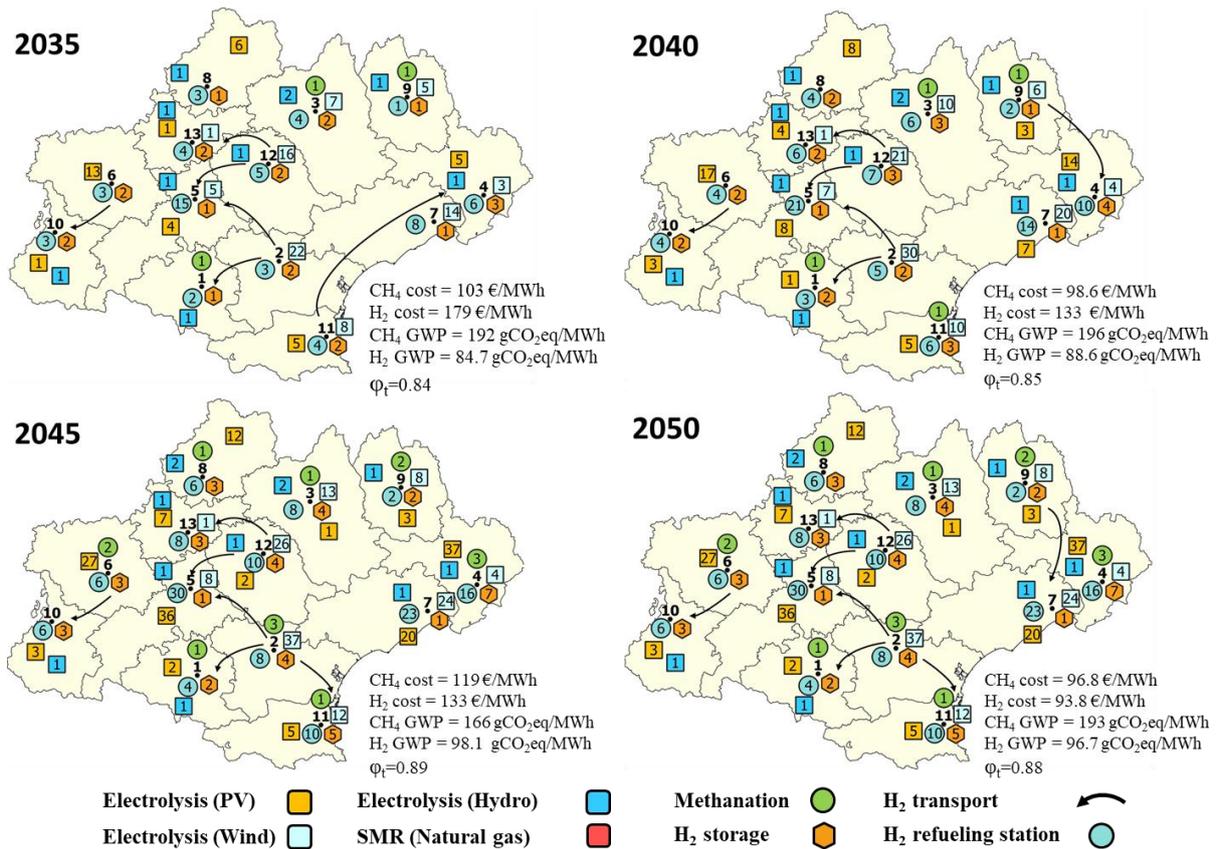


Figure 8.3. Network structure of the HMSC for TOPSIS solution

Table 8.3. Computational results for the optimization

			min TDC		min TGHG		min TELD		TOPSIS	
Single variables	Discrete variables	Single equations	CPU (s)	GAP	CPU (s)	GAP	CPU (s)	GAP	CPU (s)	GAP
98,245	23,816	814,640	7200	0.007	8	0.000	25	0.000	7200	0.008

8.5 Conclusions

In this chapter, the methodology developed in the previous chapters was enriched by incorporating the TELD as one of the objective functions. This first model serves to open a new perspective for more detailed and in-depth work to consider exergy analysis within HMSC optimization. An analysis of the three mono-objective optimizations of TAC, GHG, and TELD shows a considerable change in the configuration of the HMSC. Since a significant part of the exergy is lost throughout the HMSC (up to 202 TWh), the coupling of this PtG ecosystem with other energy systems is necessary to reduce the exergy losses (i.e., copying with the cement industry or with methanization processes for heat recovery). In addition, the recovery of other by-products such as oxygen could be interesting.

The methodology, approach, and results found can be useful to enrich the decision-making process, especially to favor the deployment of specific technologies. The model is general enough to incorporate other criteria. The methodological framework paves the way for further extensions to other types of processes and energy vectors, thus creating a synergetic effect with other energy systems, thus exploring the scale-up potential of increase in efficiency and decrease in costs.

9 GENERAL CONCLUSIONS AND PERSPECTIVES

9.1 Conclusions

Renewable gas has a major role to play in the future energy system and Power-to-gas (PtG) is the key technology to meet that demand as it provides the solution to produce the required large amounts of green molecules hydrogen (H₂) or methane (CH₄). The strategy of several countries and regions for energy transition has incorporated these systems to achieve the objective of deep decarbonization of the energy sector. In this sense, the strategy to be adopted for the implementation of PtG concepts can vary significantly according to the territory to be considered and has to be adapted to different scenarios and to interact with various technologies. The development of a methodology for the design of such systems and the deployment of their corresponding infrastructures was the core of this Ph.D.

Since the energy systems are becoming more technologically diverse, spatially distributed, and temporally variable, this work proposed a methodological framework for the spatial, temporal, and multi-objective optimization of energy supply chains based on PtG concepts.

The Region of Occitania was considered as a case study but the approach developed is generic enough to be replicated to other territories and technologies. A time horizon between 2035-2050 was considered as the deployment period with a 5-year increment. Several energy sources for hydrogen production were

identified (solar, wind, hydro, natural gas, and imported electricity through the power grid), as well as CO₂ sources (biogenic and industrial) for methane production through methanation. Special attention was paid to the satisfaction of hydrogen and methane demands, taking into account both fossil and renewable energy sources. Mono and multi-objective optimizations were carried out with several objective functions considered either in isolation or in combination according to the studied scenario: cost, environmental impact, total production of renewable methane, and exergy lost and destroyed.

The literature review has emphasized that only a few studies addressed the logistical aspects involved in PtG chains, in particular the dual production of hydrogen and methane with their associated issues, i.e. determining the location, size, and the number of production, storage, and transport units, and refueling stations. The proposed framework enlarged the scope of previous works implemented in our research group. The management of the duality hydrogen/methane complexifies significantly the supply chain modeling by increasing the number of constraints and criteria from a mathematical viewpoint. The MILP problem was solved using the CPLEX solver for mono-objective optimization in the GAMS environment. In the case of multi-objective optimization, the epsilon-constraint method was applied to obtain the set of optimal solutions (Pareto front). The m-TOPSIS method was then used to choose the solution that represents the best compromise. The quantitative information provided by the optimization model then allows studying the deployment of the HMSC for a planned transition scenario. The main contributions from a modeling viewpoint can be summarized as follows:

- The extension of HSC to an HMSC, incorporating the demand for renewable methane through methanation.
- The incorporation of other objective functions such as total methane production and exergy destruction and loss.
- The reformulation of the Total Annual Cost objective function to consider discount rate, which then allowed the LCOE to be calculated.
- The differentiation of energy sources leads to an additional set of constraints to meet the requirements of both hydrogen and methane demands.

The main echelons of the HMSC were described in a dedicated chapter from a process and technological viewpoint. Emphasis was placed on PtG concepts (i.e., green hydrogen and renewable methane production) and put into perspective by incorporating fossil alternatives such as SMR and natural gas, taking into account the environmental regulation (i.e., carbon pricing). The use of the selected technologies was also justified, given their technological maturity or suitability for the case study.

One of the main challenges that Power-to-Gas systems must overcome is their economic viability concerning other options, due to the significant investment and feedstock costs (particularly electricity) they involve. Yet these technologies are also a key enabler for decarbonization. The values of LCOE

and GHG for hydrogen and methane from PtG systems, according to the case study, are shown in Table 9.1.

Table 9.1. LCOE and GHG for hydrogen and methane from PtG systems

Energy vector	LCOE [€/MWh]	GHG [gCO ₂ -eq/kWh]
Hydrogen	107-177	32-41
Methane	133-181	96-139

In the proposed methodology, an exergy analysis allowed identifying the energy destruction (irreversibilities) and loss (by-products). In the current state of the model, the consideration of the exergy loss along the HMSC as an optimization criterion favors the use of fossil fuels. It suggests that the system should be further enlarged by interacting with other systems (i.e., methanization, and steel and cement industry) or valorizing some by-products (i.e., oxygen), to favor the use of renewable sources.

The configuration of the HMSC changes considerably according to the objective function. On the one hand, TAC minimization generally implies a centralized HMSC, due to the economy of scale. On the other hand, GHG minimization favors decentralization with maximum use of raw materials with low carbon emissions.

Regarding the interaction between fossil and renewable energy sources, it must be highlighted that green hydrogen requires strict environmental regulations (i.e., high carbon price) to substitute gray hydrogen. However, for a lower carbon price, the role of both options is complementary and allows to satisfy the estimated demand. For renewable methane, ambitious GHG reduction targets are required to make it play a significant role. Its contribution can be measured not only in terms of economic viability but also in terms of additional services of flexibility to the network and contribution to the achievement of a circular economy by recovering the CO₂ emitted by other processes. The flexibility contribution of PtG needs to be assessed through a novel flexibility metric.

9.2 Lessons learned

In general, it can be said that multi-objective problems are more complex, compared to mono-objective ones with a higher computational cost. In this study, the spatial and multi-period aspects make the problem more complex. A four-period problem using 13 grids as geographical divisions allowed to obtain an interesting first solution for strategic planning in an investment stage. In this sense, an increase in problem complexity would be more realistic but also require major computational efforts.

The proposed methodology provides an operational and efficient solution to the guidelines highlighted in the roadmaps proposed by ADEME and local institutions, which show the final objectives to be achieved without indicating the logistical aspects linked to the deployment of HMSC in the territory. The framework has been designed with genericity purpose to be adapted to other regions and can be modified to embed other technologies, indicators, and criteria. However, it is important to note that the parameters involved require assumptions that are beyond the scope of the methodology. Validation by a set of experts is interesting to improve the solutions that will be obtained with the model.

9.3 Perspectives

Finally, several perspectives can be suggested to improve the proposed methodological framework:

- Gaseous and liquid physical forms of hydrogen for transportation, other production options, transport, and storage modes can be considered to extend the model. This perspective is being addressed by the LGC research team in particular for a better granularity level for storage options. The case of an insular territory (Corsica) with its specific constraints is now under study (Cristofari et al., 2014).
- In this work, the concepts "green hydrogen" and "grey hydrogen" were incorporated into the HMSC. However, an interesting option would be the incorporation of "blue hydrogen". This implies the use of carbon capture and storage (CCS) and thus the reduction of GHG emissions associated with the SMR process. The development of production facilities with CCS is identified as a key requirement to produce low-carbon low-cost hydrogen. However, the development of a credible standard is needed to demonstrate that the hydrogen produced is sufficiently sustainable. The model could then provide the basis for the allocation of Green Methane/Hydrogen Certification Scheme.
- A hydrogen demand for the transport sector (FCEV) and a demand for methane that is injected directly into the natural gas network have been used. Yet, hydrogen has the potential to be used in various applications in the conversion and final consumption in the electricity, gas, transportation (i.e., ships and aircraft), residential and industrial sectors that could be further considered.
- In this research, only a global variation of the carbon price was considered, so a more detailed analysis of the different regulatory mechanisms and their influence on the HMSC configuration and indicators could be an interesting perspective to explore.
- Another interesting perspective could be to extend the HMSC model to an Energy Supply Chain based on Power-to-X concepts. Finally, hydrogen is central to several sector-coupling options, including power-to-gas (for the gas grid), power-to-heat, power-to-liquids, and power-to-ammonia, thus enhancing the opportunity for energy transfer between sectors, as this can unlock potential for cost and resource efficiency savings (i.e., minimization of exergy loss and

destruction). An example of the possible options for the extension of the model is presented in Figure 9.1.

- Another aspect to be addressed is the variability of demand and costs for energy, CO₂, etc. More detailed work on this subject could significantly enrich the results obtained. The results obtained through HMSC optimization have shown that the cost of electricity significantly influences the final cost of hydrogen. In this sense, the seasonal variation of renewable energy sources from a multi-period aspect (i.e., month-step discretization) may be an interesting aspect to address.

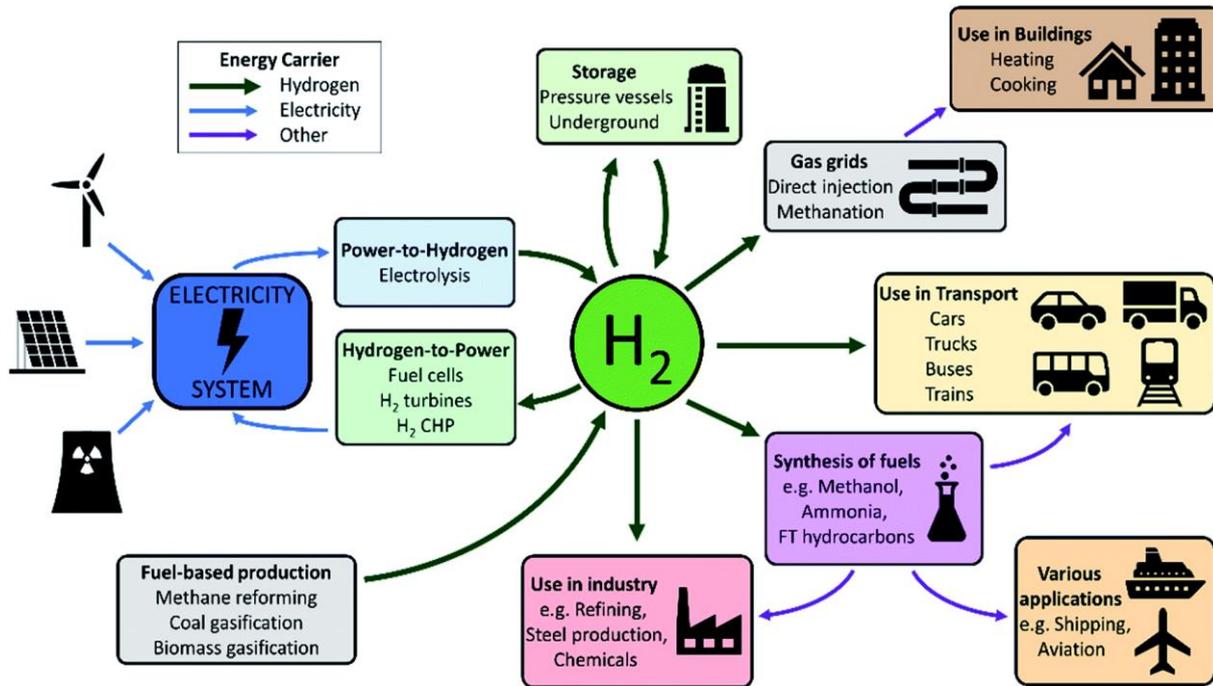


Figure 9.1. Energy supply chain involving Power-to-X concepts (Quarton et al., 2020)

- Integration of process models and supply chain models. It should be mentioned that in the field of operational research and Process Systems Engineering, a very large amount of work has been undertaken to address both the infrastructure network design problem and the optimization of established networks. However, in most of the work to date, the potential benefit of including more detail in the manufacturing process has not been established. Since there is huge uncertainty regarding the efficiency of each PtG process option in its mature state, at this stage of development these uncertainties should be captured in the global network model, to direct the research in a way that most benefits the global network. A schematic example for Multi-scale modeling for holistic energy supply chains is shown in Figure 9.2

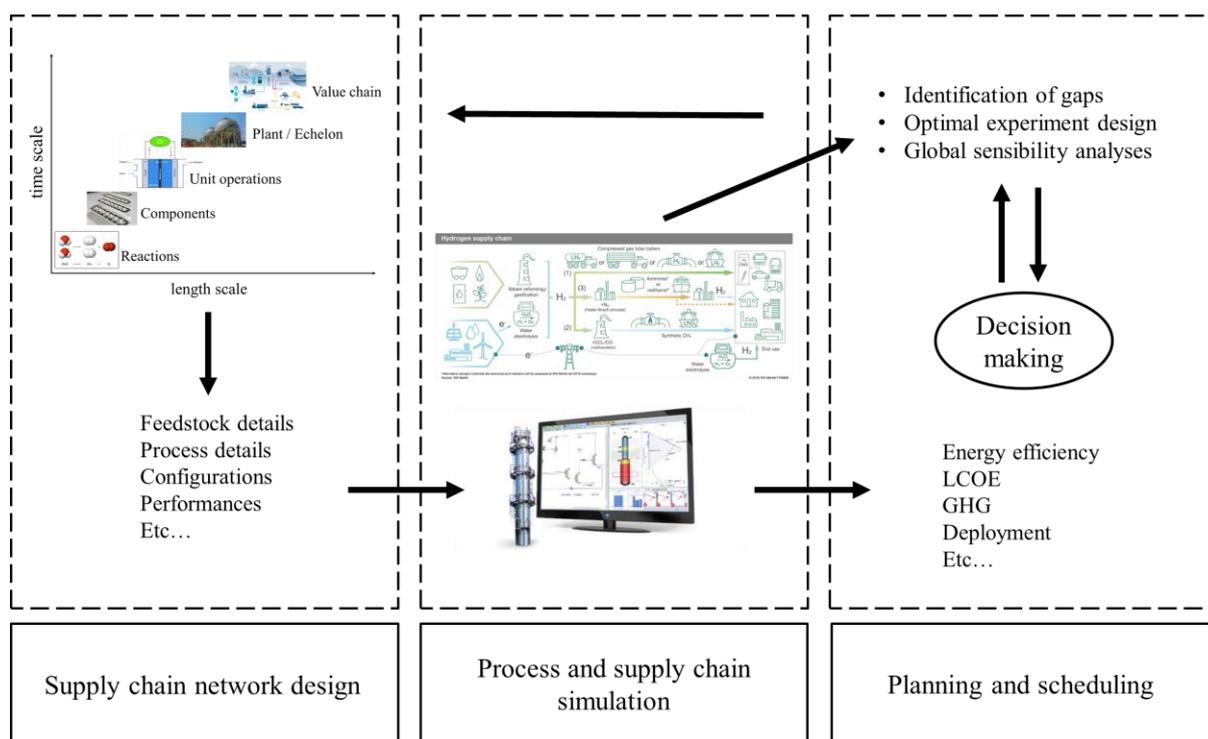


Figure 9.2. Multi-scale modeling for holistic energy supply chains (adapted from (Hosseini and Shah, 2011))

Although multi-scale modeling is a promising technique for integrating all aspects of processing at different scales in the PtG supply chain, the simultaneous resolution of process models inserted in the supply chain still faces prohibitive computational time problems. This could be overcome by using "off-line" process models to predict the possible range of production and efficiency of each operating unit and interconnect them before inserting them into the supply chain model.

Finally, the next challenge will also be to operate the PtG supply chain (the so-called supply chain planning and scheduling problem) and develop an integrated approach that can reconcile both levels, the GEOM formulation used in this work and the operational aspects that are usually addressed with LoPOM models. In this sense, more specificities on the distribution and local transport of hydrogen, the specific location of refueling stations, among others have to be addressed.

We hope to have contributed to a better understanding of the modeling of PtG energy systems, but there is still a lot of work to be explored. Making a successful energy transition suggests that there will be a range of processes and vectors to be hybridized, that the solution will not be unique, which reinforces the interest in an optimization approach.

CONCLUSIONS ET PERSPECTIVES GÉNÉRALES

Conclusions

Le méthane renouvelable a un rôle majeur à jouer dans le système énergétique futur et le Power-to-Gas (PtG) est une technologie clé pour répondre à cette demande car il fournit une solution pour produire à grande échelle des molécules vertes d'hydrogène (H₂) ou de méthane (CH₄). La stratégie de plusieurs pays et régions pour leur transition énergétique a intégré ces systèmes pour atteindre l'objectif d'une décarbonisation profonde du secteur de l'énergie. En ce sens, la stratégie à adopter pour la mise en œuvre des concepts PtG peut varier considérablement selon le territoire à considérer, et doit être adaptée à différents scénarios et à l'interaction avec diverses technologies. L'élaboration d'une méthodologie pour la conception de tels systèmes et le déploiement des infrastructures correspondantes a été au cœur de ce travail de doctorat.

Étant donné que les systèmes énergétiques sont de plus en plus diversifiés sur le plan technologique, répartis dans l'espace et variables dans le temps, ces travaux ont proposé un cadre méthodologique pour l'optimisation spatiale, temporelle et multi-objectifs des chaînes d'approvisionnement en énergie sur la base des concepts PtG.

La région de l'Occitanie a été considérée comme un cas d'étude mais l'approche développée est suffisamment générique pour être reproduite sur d'autres territoires et avec d'autres technologies. Un horizon temporel compris entre 2035 et 2050 a été considéré comme période de déploiement avec une discrétisation de 5 ans. Plusieurs sources d'énergie pour la production d'hydrogène ont été identifiées (solaire, éolienne, hydraulique, gaz naturel et électricité importée par le biais du réseau électrique), ainsi que des sources de CO₂ (biogénique et industriel) pour la production de méthane par méthanation. Une attention particulière a été accordée à la satisfaction des demandes d'hydrogène et de méthane, en tenant compte de sources d'énergie fossiles et renouvelables. Des optimisations mono et multi-objectifs ont été réalisées avec plusieurs fonctions objectifs considérées soit isolément soit en combinaison selon le scénario étudié : coût, impact environnemental, production totale de méthane renouvelable, et exergie perdue et détruite.

La revue de la littérature a souligné que seules quelques études ont abordé les aspects logistiques des chaînes de PtG, en particulier la double production d'hydrogène et de méthane avec les problèmes qui y sont associés, c'est-à-dire la détermination de l'emplacement, de la taille et du nombre d'unités des stations de production, de stockage, de transport et de ravitaillement. Le cadre proposé a élargi le champ des travaux antérieurs réalisés au sein de notre groupe de recherche. La gestion de la dualité hydrogène/méthane complexifie considérablement la modélisation de la chaîne logistique en augmentant le nombre de contraintes et de critères d'un point de vue mathématique.

Le problème MILP a été résolu en utilisant le solveur CPLEX pour l'optimisation mono-objectif dans l'environnement GAMS. Dans le cas de l'optimisation multi-objectifs, la méthode dite « augmented ϵ -constraint » a été appliquée pour obtenir l'ensemble des solutions optimales (front de Pareto). La méthode m-TOPSIS a ensuite été utilisée pour choisir la solution qui représente le meilleur compromis. Les informations quantitatives fournies par le modèle d'optimisation permettent ensuite d'étudier le déploiement de l'an HMSC pour un scénario de transition planifié. Les principales contributions du point de vue de la modélisation peuvent être résumées comme suit :

- L'extension de la HSC à une HMSC, intégrant la demande de méthane renouvelable par méthanation.
- L'incorporation d'autres fonctions objectifs telles que la production totale de méthane et la destruction et la perte d'énergie.
- La reformulation de la fonction objectif de coût annuel total pour tenir compte du taux d'actualisation, qui a ensuite permis de calculer le LCOE.
- La différenciation des sources d'énergie entraîne une série de contraintes supplémentaires pour répondre aux exigences de la demande d'hydrogène et de méthane.

Les principaux échelons du HMSC ont été décrits dans un chapitre consacré à ce sujet, du point de vue des procédés et de la technologie. L'accent a été mis sur les concepts de PtG (c'est-à-dire la production d'hydrogène vert et de méthane renouvelable) et mis en perspective en intégrant des alternatives fossiles avec le procédé de reformage à la vapeur (Steam Methane Reforming, SMR) pour le gaz naturel, en tenant compte de la réglementation environnementale (c'est-à-dire le prix du carbone). L'utilisation des technologies sélectionnées a été également justifiée, compte tenu de leur maturité technologique ou de leur adéquation à l'étude de cas.

Les valeurs du LCOE et des GES pour l'hydrogène et le méthane des systèmes PtG, selon l'étude de cas, sont indiquées dans le Table 0.1. L'un des principaux défis que doivent relever les systèmes PtG est leur viabilité économique par rapport aux autres options, en raison des investissements et des coûts de matières premières (en particulier l'électricité) importants qu'ils impliquent. Pourtant, ces technologies sont également des éléments clés de la décarbonisation.

Table 0.1. LCOE et GES pour l'hydrogène et le méthane issus de systèmes PtG

Vecteur énergétique	LCOE [€/MWh]	GES [gCO ₂ -eq/kWh]
Hydrogène	107-177	32-41
Méthane	133-181	96-139

Dans la méthodologie proposée, une analyse exergetique a permis d'identifier la destruction (irréversibilités) et la perte (sous-produits) d'exergie. Dans la version actuelle du modèle, la prise en compte de la perte d'exergie tout au long de l'HMSC comme critère d'optimisation favorise l'utilisation des combustibles fossiles. Elle suggère que le système devrait être élargi en interagissant avec d'autres systèmes (c'est-à-dire la méthanisation, l'industrie de l'acier et du ciment) ou en valorisant certains sous-produits (c'est-à-dire l'oxygène), afin de promouvoir l'utilisation de sources renouvelables.

La configuration de l'an HMSC change considérablement en fonction de la fonction objectif. D'une part, la minimisation des TAC implique généralement une HMSC centralisé, en raison de l'économie d'échelle. D'autre part, la minimisation des GES favorise la décentralisation avec une utilisation maximale des matières premières à faible émission de carbone.

En ce qui concerne l'interaction entre les sources d'énergie fossiles et renouvelables, il faut souligner que l'hydrogène vert nécessite une réglementation environnementale stricte (prix du carbone élevé) pour remplacer l'hydrogène gris. Cependant, pour un prix du carbone plus bas, les rôles des deux options sont complémentaires et permettent de satisfaire la demande estimée. Pour le méthane renouvelable, des objectifs ambitieux de réduction des GES sont nécessaires pour lui conférer un rôle significatif. Sa contribution peut être mesurée non seulement en termes de viabilité économique, mais aussi en termes de services supplémentaires de flexibilité du réseau et de contribution à la réalisation d'une économie circulaire en récupérant le CO₂ émis par d'autres procédés. La contribution de la PtG à la flexibilité doit être évaluée au moyen d'une nouvelle mesure de la flexibilité.

Leçons tirées

En général, on peut dire que les problèmes multi-objectifs sont plus complexes que les problèmes mono-objectifs dont le coût de calcul est plus élevé. Dans cette étude, les aspects spatiaux et multi-périodes rendent le problème plus complexe. Un problème à quatre périodes utilisant 13 grilles comme divisions géographiques a permis d'obtenir une première solution intéressante pour la planification stratégique dans une phase d'investissement. En ce sens, une augmentation de la complexité du problème serait plus réaliste mais nécessiterait également des efforts de calcul importants.

La méthodologie proposée apporte une solution opérationnelle et efficace aux orientations mises en évidence dans les feuilles de route proposées par l'ADEME et les institutions locales, qui montrent les

objectifs finaux à atteindre sans indiquer les aspects logistiques liés au déploiement du HMSC sur le territoire. Le cadre a été conçu dans un but de généralité pour être adapté à d'autres régions et peut être modifié pour intégrer d'autres technologies, indicateurs et critères. Cependant, il est important de noter que les paramètres impliqués nécessitent des hypothèses qui dépassent le cadre de la méthodologie. Une validation par un ensemble d'experts est intéressante pour améliorer les solutions qui seront obtenues avec le modèle.

Perspectives

Enfin, plusieurs perspectives peuvent être suggérées pour améliorer le cadre méthodologique proposé :

- Les formes physiques gazeuses et liquides de l'hydrogène pour le transport, les autres options de production, les modes de transport et de stockage peuvent être envisagées pour étendre le modèle. L'équipe de recherche du LGC se tourne en particulier vers cette perspective pour obtenir un meilleur niveau de granularité pour les options de stockage. Le cas d'un territoire insulaire (la Corse) avec ses contraintes spécifiques est actuellement à l'étude (Cristofari et al., 2014).
- Dans ce travail, les concepts "hydrogène vert" et "hydrogène gris" ont été intégrés à l'an HMSC. Cependant, une option intéressante serait l'incorporation de l'"hydrogène bleu". Cela implique l'utilisation du captage et du stockage du carbone (CSC) et donc la réduction des émissions de GES associées au processus de SMR. Le développement d'installations de production avec le CSC est considéré comme une exigence essentielle pour produire de l'hydrogène à faible teneur en carbone et à faible coût. Toutefois, l'élaboration d'une norme crédible est nécessaire pour démontrer que l'hydrogène produit est suffisamment durable. Ce modèle pourrait alors servir de base à l'attribution d'un système de certification du méthane et de l'hydrogène verts.
- Une demande en hydrogène pour de l'électromobilité (FCEV) et une demande de méthane qui est injecté directement dans le réseau de gaz naturel ont été utilisées. Pourtant, l'hydrogène a le potentiel d'être utilisé dans diverses applications de conversion et de consommation finale dans les secteurs de l'électricité, du gaz, des transports (c'est-à-dire les navires et les avions), du résidentiel et de l'industrie, qui pourraient être étudiées.
- Dans cette étude, seule une variation globale du prix du carbone a été prise en compte. Une analyse plus détaillée des différents mécanismes de régulation et de leur influence sur la configuration et les indicateurs du HMSC pourrait donc être une perspective intéressante à explorer.
- Une autre perspective intéressante pourrait être d'étendre le modèle HMSC à une chaîne logistique énergétique basée sur les concepts de Power-to-X. L'hydrogène est au centre de plusieurs options de couplage sectoriel, notamment le PtG (pour le réseau de gaz), Power-to-Heat, Power-to-Liquids, et Power-to-Ammonia, ce qui accroît les possibilités de transfert

d'énergie entre les secteurs, et donc des économies de coûts et d'amélioration de l'efficacité des ressources (c'est-à-dire de réduire au minimum la perte et la destruction d'énergie). Un exemple des options possibles pour l'extension du modèle est présenté dans la Figure 0.1.

- Un autre aspect à aborder est la variabilité de la demande et des coûts de l'énergie, du CO₂, etc. Un travail plus approfondi sur ce sujet pourrait enrichir considérablement les résultats obtenus. Nous avons montré que le coût de l'électricité influence de manière significative le coût final de l'hydrogène et la configuration de l'HMSC. En ce sens, la variation saisonnière des sources d'énergie renouvelables d'un point de vue multi-période (discrétisation par échelon mensuel) peut être intéressante à aborder.

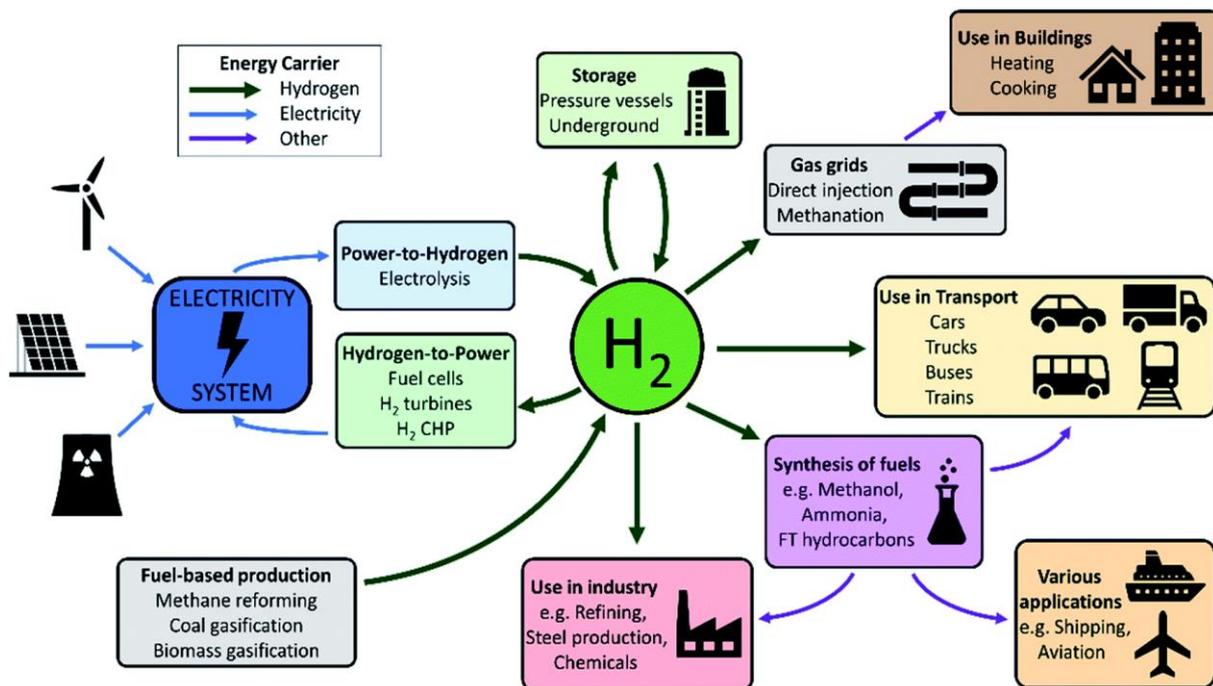


Figure 0.1. Chaîne logistique de l'énergie impliquant les concepts de Power-to-X (Quarton et al., 2020)

- Intégration des modèles de procédés et des modèles de chaîne logistique. Il convient de mentionner que dans le domaine de la recherche opérationnelle et de l'approche systémique du génie des procédés, un très grand nombre de travaux ont été entrepris pour résoudre à la fois le problème de la conception des réseaux d'infrastructure et l'optimisation des réseaux existants. Toutefois, dans la plupart des travaux réalisés à ce jour, les performances de chaque brique de procédés sont figées. Comme il existe au stade actuel de développement, une grande incertitude quant à l'efficacité de chaque brique de procédé du PtG lorsqu'elle sera à un stade de maturité avancé, ces incertitudes devraient être prises en compte dans le modèle de chaîne logistique. Un

exemple schématique de modélisation multi-échelle pour une chaîne logistique énergétique globale est présenté à la Figure 0.2.

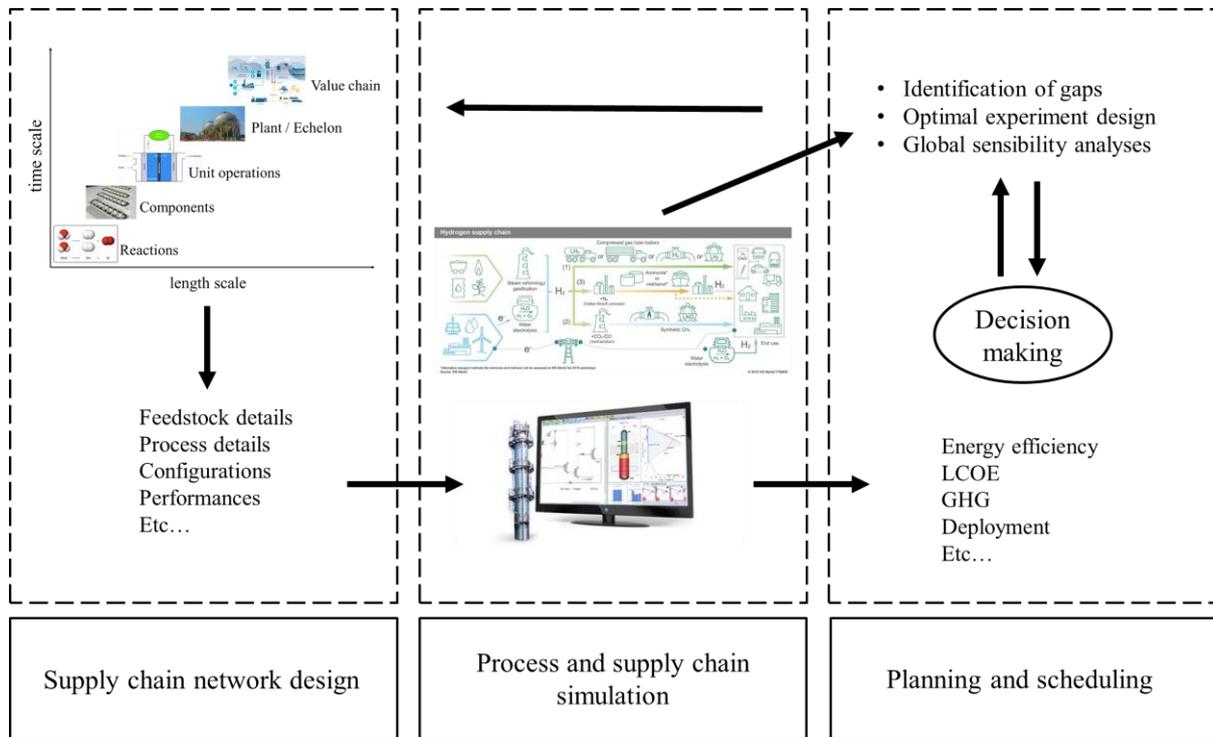


Figure 0.2. Modélisation multi-échelle pour une chaîne d'approvisionnement énergétique globale (adapté de (Hosseini and Shah, 2011))

Bien que la modélisation multi-échelle soit une technique prometteuse pour intégrer tous les aspects du traitement à différentes échelles dans la chaîne logistique PtG, la résolution simultanée de modèles de procédés insérés dans la chaîne logistique se heurte encore à des problèmes de temps de calcul prohibitifs. Cela pourrait être levé par l'utilisation de modèles de procédés « off-line » afin de prévoir la gamme possible de production et d'efficacité de chaque unité d'exploitation et de les interconnecter avant de les insérer dans le modèle de chaîne logistique.

Enfin, le prochain défi consistera également à exploiter la chaîne logistique des PtG (problème de planification et d'ordonnancement de la chaîne logistique) et à élaborer une approche intégrée permettant de concilier les deux niveaux, la formulation GEOM utilisée dans ce travail et les aspects opérationnels qui sont généralement traités avec les modèles LoPOM. Cela nécessite de prendre en compte les spécificités sur la distribution et le transport local de l'hydrogène ainsi que sur l'emplacement des stations-services, entre autres.

Nous espérons avoir contribué à une meilleure compréhension de la modélisation des systèmes énergétiques PtG, même si beaucoup de travail reste à explorer. Réussir la transition énergétique suggère

qu'il y aura un panel de procédés et de vecteurs à hybrider, que la solution ne sera pas unique, ce qui renforce l'intérêt d'une approche d'optimisation.

DATABASE

Table A.1. – Average distances between grids [km]

Grid	1	2	3	4	5	6	7	8	9	10	11	12	13
1	0	81.7	232	285	87.4	166	237	198	339	155	199	160	139
2	81.7	0	238	200	93.1	171	153	204	292	241	115	166	144
3	232	238	0	228	151	230	172	112	107	310	273	77	130
4	285	200	228	0	291	369	58	402	145	439	205	256	342
5	87.4	93.1	151	291	0	78.9	254	114	255	155	207	76.3	53.9
6	166	171	230	369	78.9	0	321	149	335	72.8	284	157	87.5
7	237	153	172	58	254	321	0	351	196	388	154	198	291
8	198	204	112	402	114	149	351	0	217	264	317	112	59.8
9	339	292	107	145	255	335	196	217	0	417	297	181	234
10	155	241	310	439	155	72.8	388	264	417	0	352	235	203
11	199	115	273	205	207	284	154	317	297	352	0	280	258
12	160	166	77	256	76.3	157	198	112	181	235	280	0	77.3
13	139	144	130	342	53.9	87.5	291	59.8	234	203	258	77.3	0

Table A.2. – Total demand for hydrogen (transport sector) [kg/year]

Period	Grid												
	1	2	3	4	5	6	7	8	9	10	11	12	13
2035	2,239,710	3,072,822	4,464,587	6,165,209	17,089,172	3,110,589	9,376,225	2,676,210	1,087,665	3,424,191	3,927,228	5,242,233	3,754,744
2040	3,144,492	5,124,474	6,250,844	10,281,573	23,931,196	4,348,044	15,636,510	3,750,506	1,523,126	4,793,020	6,549,345	7,344,211	5,254,946
2045	3,814,074	7,176,123	7,596,365	14,397,936	29,048,413	5,269,780	21,896,793	4,547,224	1,848,954	5,822,820	9,171,461	8,920,695	6,380,099
2050	4,483,656	8,711,241	8,941,885	17,477,947	34,165,628	6,191,514	26,580,961	5,343,942	2,174,779	6,852,620	11,133,423	10,497,179	7,505,250

Table A.3 – Total demand for methane

Period	[Kg/year]
2035	67,200,793
2040	91,998,170
2045	99,664,649
2050	107,991,361

Table A.4. – General data: a) for the supply chain, b) Low heating value (LHV), c) molar mass

a) For the supply chain

Network operating period	8000 h per year
Discount rate	5.25%

b) Low heating value (LHV)

Energy vector	LHV [MJ/kg]
Hydrogen	119.96
Methane	50.00

c) Molar mass

Energy vector	Molar Mass [g/mol]
Carbon dioxide	44.01
Hydrogen (diatomic)	2.00
Methane	16.04
Oxygen (diatomic)	16.00

Table A.5. – General for hydrogen production: a) capacities, b) Rate of utilization of primary energy source, c) investment cost, and d) Fixed cost

a) capacities

Liquid H ₂ (<i>i</i>)	$PCap_{p,ij}^{min}$ Minimum production capacity of plant type <i>p</i> size <i>j</i> for product form <i>i</i>				$PCap_{p,ij}^{max}$ Maximum production capacity of plant type <i>p</i> size <i>j</i> for product form <i>i</i>			
	small	medium	large	unit	small	medium	large	unit
Technology (<i>p</i>)				(ton per				(ton per year)
Steam methane reforming	650	1150	1640	year)	730	1275	1825	
Electrolysis	200	0	360		1005	0	1805	

b) Rate of utilization of primary energy source (MWh/kgH₂)

Process	γ [MWh/kgH ₂]
SMR	55.8
Electrolysis	53.6

c) Investment cost (M€)

Process	Period	Size		
		small	med	large
SMR	2035-2050	1,050,000	1,700,000	2,280,000
	2035	2,137,500		5,054,460
Electrolysis	2040	2,747,100		6,496,000
	2045	2,485,500		5,877,300
	2050	1,962,200		4,640,000

d) Fixed cost (M€ per year)

Process	Period	Size		
		small	med	large
SMR	2035-2050	52,500	85,000	114,000
	2035	106,875		25,000
Electrolysis	2040	137,355		21,000
	2045	124,275		19,000
	2050	98,110		15,000

Table A.6 – General information for transportation (De-León Almaraz, 2014)

Liquid H ₂	mini	small	medium	large
$SCap_{sji}^{min}$ - Minimum storage capacity of storage type s size j for product form i (kg)	50*	500*	10000	200000
$SCap_{sji}^{max}$ - Maximum storage capacity of storage type s size j for product form i (kg)	450*	9500	150000	540000
SCC_{sij} - Capital cost of establishing storage type s size j storing product form i (\$)	802165*	5000000	33000000	122000000
USC_{sij} - Unit storage cost for product form i at storage type s size j (\$ per kg per day)	0.064	0.032	0.01	0.005
GW^{stock}_i - Storage global warming potential form i			(704 g CO ₂ -eq per kg of H ₂) [▲]	
β - Storage holding period-average number of days worth of stock			(1 day)	

Table A.7. – General information for storage (De-León Almaraz, 2014)

Parameter		units	Reference
W_l	Weight of transportation mode l	40	ton
DW_l	Driver wage of transportation mode l	14.57	US\$ per hour
FE_l	Fuel economy of transportation mode l	2.3	km per liter
FP_l	Fuel price of transportation mode l	1.5	US\$ per liter
GE_l	General expenses of transportation mode l	8.22	US\$ per day
LUT_l	Load and unload time of product for transportation mode l	2	hours per trip
ME_l	Maintenance expenses of transportation mode l	0.126*	US\$ per km
SP_l	Average speed of transportation mode l	66.8	km per hour
TMA_l	Availability of transportation mode l	18	hours per day
GW^{Trans}_l	Global warming potential of transportation mode l	62	g CO ₂ per tonne-km
RT_l	Risk level of transportation mode l	1.33	Units
$TCap_{il}$	Capacity of transportation mode l transporting product form i	3500	kg per trip
Q^{min}_{il}	Minimum flow rate of product form i by transportation mode l	3500	kg per day
Q^{max}_{il}	Maximum flow rate of product form i by transportation mode l	960000	kg per day
TMC_{il}	Cost of establishing transportation mode l transporting product form i	500000	US\$

Table A.8. – General information for refueling station: a) Capacity and b) capital cost

a) Capacity

Size	Capacity [Kg/year]
small	599330
med	999005
large	1198660

b) Capital cost

Size	Period			
	2035	2040	2045	2050
small	2,941,839	2,714,671	2,420,690	2,205,538
medium	4,451,859	4,072,635	3,693,410	3,314,186
large	5,182,752	4,738,989	4,295,225	3,851,462

Table A.9. – General information for methanation: a) capacity and b) capital cost.

	small	large
Capacity [kg/year]	2,522,878	55,503,316
Capital cost [€]	2,000,000	14,300,000

Injection of Methane into the grid: 0.09 €/kg

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LISTE OF FIGURES

Figure 1.1. Main energy conversion pathways (power-to-gas, power-to-power, and gas-to-gas) in a renewable energy integrated energy system (Brandon and Kurban, 2017)	18
Figure 1.2. Number of papers based on an initial search in Web of Science™ (WoS) using the keywords “renewable hydrogen and methane supply chains” (December, the 5 th 2020)	19
Figure 0.1 Principales voies de conversion de l'énergie (power-to-gas, power-to-power and gas-to-gas) dans un système intégré d'énergie renouvelable (Brandon and Kurban, 2017)	28
Figure 0.2 Nombre d'articles basés sur une première recherche sur le site web de Science™ (WoS) à l'aide des mots clés “renewable hydrogen and methane supply chains” (5 Décembre 2020)	30
Figure 2.1. General HMSC including feedstock, product, physical form, transport, storage, and end-uses	39
Figure 2.2. The step conversion efficiencies for the hydrogen and methane supply pathways being considered (SMR+CCS efficiency of 75%, transport and distribution (T&D) loss of 5% (distribution only), residential boiler efficiency 84%, and residential micro-CHP fuel cell efficiency 88%) (Brandon and Kurban, 2017).....	40
Figure 2.3 Simplified process flow diagram for hydrogen production by steam methane reforming (Basile et al., 2015a).....	42
Figure 2.4 Gasification block diagram and governing reaction (de Leon Almaraz, 2014).	43
Figure 2.5. Schematic illustration of alkaline water electrolysis (Shiva Kumar and Himabindu, 2019)	45
Figure 2.6. Schematic illustration of PEM water electrolysis (Shiva Kumar and Himabindu, 2019)	47
Figure 2.7. Schematic illustration of Solid Oxide electrolysis (Shiva Kumar and Himabindu, 2019).	48
Figure 2.8. Principle of the methanation reaction (Energo, n.d.)	50
Figure 2.9. Typical methanation process chain (Energo, n.d.)	50
Figure 2.10. Process flow diagram for biological methanation with a separate reactor (Götz et al., 2014)	51

Figure 2.11. Example of liquid hydrogen tank for space applications (NASA, 2015).....	52
Figure 2.12. Limits on hydrogen blending into national gas grids around the world, and the relationship between energy content, carbon savings, and hydrogen injection mixtures (right) (Staffell et al., 2019).	53
Figure 2.13. Hydrogen transportation options for refueling stations: (a) gaseous hydrogen delivery and (b) liquid hydrogen delivery (Mayyas and Mann, 2019).	54
Figure 2.14. Layout of the components of a hydrogen refueling station supplied by an on-site production unit (Reddi et al., 2016).	55
Figure 2.15. Main shades of hydrogen (IRENA, 2020)	55
Figure 2.16. Biomethane production with CH ₄ synthesis via methanation (Collet et al., 2017).....	57
Figure 2.17. Selected technologies and echelons for HMSC modeling	59
Figure 3.1. Generic HMSC modeling with a multi-period approach	68
Figure 3.2. Solution methods for mono-objective models (modified from (Li et al., 2019)).....	71
Figure 3.3 Solution methods for multi-objective models (modified from (Li et al., 2019))	72
Figure 3.4. Illustration of Pareto optimality in two-objective optimization (modified from (Xin, 2013)).....	73
Figure 3.5. Generic decision matrix for application of the TOPSIS method (from (González- Rodríguez, 2020))	76
Figure 4.1. a) Administrative regions in France; b) Departments in the Occitania region.....	85
Figure 4.2. Hydrogen and methane demand in the period 2035-2050	86
Figure 4.3. Regional electricity availability in 2035-2050.....	87
Figure 4.4. Available energy in each grid by source.....	88
Figure 4.5. Regional CO ₂ availability for methane production in 2035-2050.....	88
Figure 5.1. HMSC supply chain based for the case study	97
Figure 5.2. Methodology framework for the HMSC (Chapter 4)	98
Figure 5.3. Possible grids configurations: A) without transportation and B) with transportation.....	104
Figure 5.4. Superstructure of the multi-period model for the Methane Supply Chain (MSC).....	108
Figure 5.5. Methane Supply Chain Structure for low demand.....	111
Figure 5.6. Methane Supply Chain Structure for high demand.....	112
Figure 5.7. Production of methane in each grid (2050).....	112
Figure 5.8. Available energy and energy consumption (by sources) in each grid (2050).....	114
Figure 5.9. Available CO ₂ and CO ₂ consumption (from the grid and imported) in each grid (2050).....	114
<i>Figure 5.10. LCOE of Methane for: 1) Low demand, 2) High demand, 3) Low cost of electricity, 4) High cost of CO₂ and 5) High capacity of hydrogen storage. The LCOE is constituted by Facility Capital (FCCh) and Operating (FOCh) Cost of Hydrogen, Cost of Electricity (ESCh), Transportation Capital (TCCh) and operating cost</i>	

<i>(TOCh), Storage Capital (FCCs) and Operating (FOCs) Cost of Hydrogen, Facility Capital (FCCg) and operating (FOCg) of Methane and Cost of CO₂ source (CO₂SC).</i>	115
Figure 6.1. HMSC supply chain based on PtG concepts.....	121
Figure 6.2. Methodology framework for the HMSC.....	123
Figure 6.3. Superstructure of the multi-period model for the HMSC	129
Figure 6.4. Supply chain configuration for min TAC.....	133
Figure 6.5. Supply chain configuration for min GWP	133
Figure 6.6. Pareto front obtained through the AUGMECON method. « TOPSIS solution » and « TOPSIS solution' »	134
Figure 6.7. Supply chain configuration for TOPSIS solution	135
Figure 6.8. Production of methane (PtM), hydrogen for PtM (PtH _{CH₄}), and hydrogen for transportation (PtH _{trans}) in each grid (2050)	136
Figure 6.9. Electricity consumption for min TAC, min GWP, and TOPSIS solution.....	137
Figure 6.10. CO ₂ consumption for min TAC, min GWP, and TOPSIS solution	137
Figure 6.11. GWP of hydrogen associated with each solution per year, compared to PtH (Wind): Hydrogen produced from wind energy source (Collet et al., 2017); PtH (PV): Hydrogen produced from solar source (Collet et al., 2017); and PtH (Network): Hydrogen produced from French electricity network (Reiter and Lindorfer, 2015)	138
Figure 6.12. GWP of methane associated with each solution per year, compared to PtM (Wind): Methane produced from wind energy source (Collet et al., 2017); PtM (PV): Methane produced from solar source (Collet et al., 2017); and PtM (Network): Methane produced from French electricity network (Reiter and Lindorfer, 2015)	139
Figure 6.13. LCOE for methane according to discount rate (r), electricity cost (ESC), and CO ₂ cost (CSC). The LCOE is constituted by Facility Capital (FCCh) and Operating (FOCh) Cost of Hydrogen, Cost of Electricity (ESC), Transportation Capital (TCCh) and operating cost (TOCh), Storage Capital (FCCs), and Operating (FOCs) Cost of Hydrogen, Facility Capital (FCCf) and operating (FOCf) of Refueling station.	140
Figure 6.14. LCOE for methane according to discount rate (r), electricity cost (ESC), and CO ₂ cost (CSC). The LCOE is constituted by Facility Capital (FCCh) and Operating (FOCh) Cost of Hydrogen, Cost of Electricity (ESC), Transportation Capital (TCCh) and operating cost (TOCh), Storage Capital (FCCs), and Operating (FOCs) Cost of Hydrogen, Facility Capital (FCCg) and operating (FOCg) of Methane, Cost of CO ₂ source (CSC) and the cost of injection into the natural gas network (FOCj).....	140
Figure 7.1. HMSC studied in this chapter	145
Figure 7.2. Methodology framework for the HMSC.....	146
Figure 7.3. Superstructure of the multi-period model for the HMSC	150
Figure 7.4. Supply chain configuration for low CP	152

Figure 7.5. Supply chain configuration for high CP	152
Figure 7.6. Pareto front obtained through the AUGMECON method and TOPSIS solution.....	155
Figure 7.7. Supply chain configuration for TOPSIS solution	155
Figure 7.8. Production of hydrogen in each grid (2050)	156
Figure 7.9. Production of methane in each grid for TOPSIS Solution (2050)	156
Figure 7.10. Electricity consumption for low and high CP, and m-TOPSIS solutions	157
Figure 7.11. CO ₂ consumption for TOPSIS solution	157
Figure 7.12. LCOE for methane according to discount rate (r), energy source cost (ESC), and Carbon Price (CP). The LCOE is constituted by Capital Costs (CC), Operating Costs (OC) ESC, and CP	158
Figure 8.1. Methodology framework for HMSC.....	164
Figure 8.2. General case for equipment or process involved in the HMSC, involving exergy as resource (R), product (P), and exergy lost as waste (I) and due to irreversibilities (D).....	166
Figure 8.3. Network structure of the HMSC for TOPSIS solution	169
Figure 9.1. Energy supply chain involving Power-to-X concepts (Quarton et al., 2020)	174
Figure 9.2. Multi-scale modeling for holistic energy supply chains (adapted from (Hosseini and Shah, 2011)).....	175
Figure 0.1. Chaîne logistique de l'énergie impliquant les concepts de Power-to-X (Quarton et al., 2020).....	180
Figure 0.2. Modélisation multi-échelle pour une chaîne d'approvisionnement énergétique globale (adapté de (Hosseini and Shah, 2011)).....	181

LISTE OF TABLES

Table 2.1. Summary of the key operational parameters of Alkaline, PEM, and Solid Oxide Electrolysis.....	46
Table 2.2 Summary of the key operational parameters of chemical and biological methanation process	49
Table 2.3. Selected technologies for HMSC modeling	60
Table 2.4. Three main types of HMSC models	63
Table 3.1. Strategic decisions in HMSC models	70
Table 4.1. Main techno-economic assumptions of the case study.....	83
Table 4.2. Primary energy, CO ₂ cost, and carbon price	84
Table 4.3. GWP for electricity and CO ₂ sources	84
Table 5.1. Mono-objective optimization results of methane supply chain.....	110
Table 5.2. Parameters for sensitivity analysis	115
Table 6.1. Main results for the solutions obtained through the proposed methodology	132
Table 6.2. LCOE and GWP for hydrogen and methane.....	134
Table 6.3. Values used to apply the m-TOPSIS method.....	134
Table 6.4. Computational results for the optimization	135
Table 6.5. Parameters for sensitivity analysis	139
Table 7.1. Main results for the solutions obtained through the proposed methodology	154
Table 7.2. Computational results for the optimization	155
Table 7.3. Parameters for sensitivity analysis	157
Table 7.4. LCOE and GWP for hydrogen and methane.....	159
Table 8.1. Typical exergy yield for the main processes involved in the HMSC.....	167
Table 8.2. Optimization results of the HMSC.....	168
Table 8.3. Computational results for the optimization	169
Table 9.1. LCOE and GHG for hydrogen and methane from PtG systems	172
Table 0.1. LCOE et GES pour l'hydrogène et le méthane issus de systèmes PtG.....	178