

General introduction

Earth's continental crust is a unique feature of the inner Solar System (Figure 1.1). It witnesses the processes of differentiation that operated throughout Earth's history. Indeed, separation between mantle and core likely occurred within less than 30 Ma after Earth formation, whereas the genesis of continental crust is a continuous process through time. Because it is made up of buoyant quartzofeldspathic material that is hardly recyclable within the mantle, it recorded more than 4 billion years of geological history and thus represents a unique archive of the evolution of geodynamic processes that operated on Earth.

The chemical composition of the continental crust, especially regarding trace elements, is complementary to that of the depleted mantle, source of Mid-Ocean Ridge Basalts (MORBs), and both components are respectively enriched and depleted with respect to the primitive mantle (Figure 1.2). Such observations indicate that continental crust was extracted from the mantle throughout Earth's history, leaving behind a residual solid now represented by the depleted mantle. Apart from this statement, two important questions about the differentiation of continental crust are still unsolved so far:

(1) It is not yet clear whether crust extraction from the mantle was (i) a continuous process, which is supported by studies of the continental growth rate on the basis of isotopic constraints (colored curves in Figure 1.3) or (ii) an episodic process, as revealed by statistical distributions of U–Pb ages of juvenile granitoids (blue histogram of Figure 1.3). Whatever the mechanism considered, the rate of continental extraction clearly evolved through time, which calls for explanation.

(2) The mechanisms of continental crust differentiation likely evolved during Earth's history, as witnessed by a major change in the nature and composition of crustal granitoids at the end of Archaean times (2500 to 3000 Ma ago). Indeed, Archaean granitoids of the TTG series are clearly different from their post-archaeic counterparts (i.e. arc-related magmas). The transition between both rock types is witnessed by the emplacement within this short time range (2500–3000 Ma) of particular granitoids, such as sanukitoids, that likely record the evolution of crustal growth processes. Petrogenesis of sanukitoids is relatively well constrained, but some details are not yet clear, especially concerning their mantle origin and how they differentiate at shallow crustal levels. Both issues may have critical implications for our understanding of the growth and differentiation of continental crust, respectively. In

addition, sanukitoids are always associated with a wide variety of granitoids, and no comprehensive study has been carried so far to (i) clarify the typology of this late-Archaeon magmatism and (ii) understand its geodynamic implications.

This PhD work aims to figure out some of these questions, in particular concerning the classification and petrogenesis of late-Archaeon magmas, their spatial and temporal evolution within one craton and from a global point of view, and their significance for late-Archaeon geodynamic changes. For this purpose, I studied a wide range of granitoids emplaced at the northern edge of the Kaapvaal craton and in the Limpopo Belt in South Africa. Continental crust formed and evolved in this area between 3600 and 2000 Ma, such that it is particularly suitable to study late-Archaeon geodynamic changes.

Chapter 1

The Archaean-Proterozoic transition

As stated in the introduction, the nature and composition of crustal granitoids significantly changed at the Archaean-Proterozoic boundary, i.e. around 2500 Ma ago. As shown in Figure 1.4, this evolution is not only recorded by plutonic rocks, but also by a wide variety of geological parameters, including the nature of metamorphic and sedimentary rocks, as well as the presence of particular lithological associations, the structural features of the continental crust and even the composition of the atmosphere (the dashed lines represent uncertain occurrences or local anomalies for each parameter). These are summarized in this Chapter, which is only based on literature data (important references can be found in Figure captions).

Before detailing the nature of these changes for each one of these records, I wish to discuss terminology issues, i.e. the difference between Archaean-Proterozoic *boundary* and *transition*, to avoid any confusion. This is the purpose of Section 1.1.

1.1. Boundary or Transition?

The Archaean-Proterozoic *boundary* separates, in the geological timeline, Archaean times from the Proterozoic eon and its age has been historically defined at 2500 Ma. Such a definition is based on the chronostratigraphy of several cratonic areas worldwide, where the Archaean-Proterozoic *boundary* is often represented by an angular unconformity separating a plutonic or metamorphic, Archaean basement from wide sedimentary basins. An example of such an unconformity is provided in the picture of Figure 1.5 and its schematic interpretation : south of Polokwane in South Africa, conglomerates and sandstones of the Transvaal Supergroup, aged of ~2500 Ma, overlie the Archaean (~2780 Ma) Turfloop granite.

However, such a sharp definition is not representative of the geological reality. Indeed, in many Archaean provinces worldwide (notably the Pilbara and Kaapvaal cratons), the age of the first wide, stable sedimentary basins is much older than 2500 Ma. Furthermore, the late-Archaean evolution of each craton is characterized by a succession of geological events that contrast with typically “archaean” processes and give way to crust stabilization. Such an evolution is generally short-lived on any given craton but is found at different times in different places. Both observations define the Archaean-Proterozoic *transition* as a

diachronous evolution incorporating the major tectonic stabilization (“cratonization”) of all continental blocks at the end of the Archaean, i.e. between 3000 and 2500 Ma.

During the Archaean-Proterozoic *transition*, a number of geological parameters, which are listed and described in the following, significantly evolved.

1.2. Evolution of thermal regimes

1.2.1. Mantle temperature

Radioactive isotope decay (for the most part) as well as kinetic accretion energy and latent crystallization heat of the inner core contribute to Earth’s internal heat production. As shown in Figure 1.6a, the rate of internal heat production exponentially decreases since planetary accretion. A first-order consequence of this mechanism is the global cooling of Earth in general, and the mantle in particular.

Present-day temperature of the mantle is $\sim 1350^{\circ}\text{C}$ (Figure 1.6b) and Urey ratio (heat production over heat loss) is ~ 0.23 . Assuming that, in the past, heat loss was accommodated by similar processes as present-day plate tectonics with a similar Urey ratio leads to a “thermal catastrophe”, illustrated by unrealistically high mantle temperature predicted before 1 Ga (dashed line in Figure 1.6b). On the other hand, in order to fit the Archaean mantle temperatures ($1600\text{--}1800^{\circ}\text{C}$) deduced from chemical composition of komatiites, an Urey ratio of $0.7\text{--}0.8$ is required (Figure 1.6b), which is not consistent with present-day observations.

Such a paradox (“Urey ratio paradox”) was solved using new parametrizations of mantle thermal evolution. In particular, a model taking into account the growth of continents and assuming an Urey ratio roughly similar to the present-day value ($0.2\text{--}0.4$) adequately reproduces the evolution of mantle temperatures, deduced from the secular changes in the composition of oceanic basalts (Figure 1.6c). From this perspective, komatiites are unusual lavas, produced by melting of domains that are hotter than surrounding, normal mantle.

Interestingly, the thermal evolution shown in Figure 1.6c evidences a transition in thermal regime at ~ 2500 Ma. Indeed, during the Archaean, mantle temperature increases, while it decreases afterwards. This likely results from a global reorganization of heat loss mechanisms at the end of the Archaean, especially a change in plate tectonic processes. For example, one

model involves heat loss during the Archaean through a larger length of mid-ocean ridges resulting in smaller lithospheric plates (Figure 1.7).

1.2.2. Consequences on metamorphic gradients

Archaean terranes in one hand, and post-archaean ones on the other hand, are featured by sharply contrasted metamorphic record. As shown on Figure 1.8, Archaean greenstone belts are mainly high-temperature, low-pressure metamorphic domains (amphibolite to granulite facies), characterized by gradients exceeding $25^{\circ}\text{C}\cdot\text{km}^{-1}$. By contrast, P - T conditions during the Proterozoic and Phanerozoic exhibit a clear duality between high-temperature, low-pressure (granulite facies) and high-pressure, low-temperature (blueschist to eclogite facies) metamorphic gradients, mostly in the range 5 to $25^{\circ}\text{C}\cdot\text{km}^{-1}$. Such a duality in metamorphic regimes would be the hallmark of convergent tectonics where subduction (high-pressure metamorphism) coexists with crust thickening and intrusion of massive volumes of mantle-derived melt (high-temperature metamorphism).

These observations highlight that Archaean metamorphic belts experienced higher temperatures and lower pressures compared with their “modern” counterparts, which is a direct consequence of higher mantle temperature and thermal regimes. It must be noted that the occurrence of Archaean “eclogites” has been documented, especially from the Barberton area in South Africa, as well as the Kola Peninsula in northeastern Russia (red symbols, respectively labeled 1 and 2 in Figure 1.8). However, these rocks equilibrated at pressures (10–15 kbar) not as high as for present-day eclogites (>15 kbar, up to 50 kbar) and are rather high-pressure amphibolites. This observation clearly strengthens the conclusion that Archaean terranes are dominated by low- to moderate pressure, high-temperature metamorphism.

1.3. Continental record

1.3.1. Lithological changes

Archaean sedimentary sequences are dominated by immature clastic rocks such as greywackes and conglomerates, as well as purely chemical deposits (cherts, BIF). By contrast, mature siliciclastic rocks (pelites, quartzites...) and biogenic ones (carbonates) are much more common in post-archaean platform sequences. As shown in Figure 1.9, this evolution is not that sharp and rather continuous throughout Earth’s history, but lithological associations

typical of multicyclic sedimentary processes, as well as large stable basins (compared with small tectonized greenstone belts during the Archaean), only develop since the Paleoproterozoic.

On the other hand, the nature of magmatic rocks also changes across the Archaean-Proterozoic transition. Archaean greenstone belts are dominated by bimodal tholeiitic volcanism comprising a mafic to ultramafic component (basalts, komatiites) and a felsic (dacites and rhyolites), high-silica one. Calc-alkaline intermediate rocks such as andesites are relatively scarce, whereas they are conspicuous in post-Archaean volcanic associations, especially at convergent plate margins. In addition, typical alkaline magmas (intraplate basalts, carbonatites) are almost exclusively younger than 2500 Ma. On the other hand, Archaean plutonic rocks are also strikingly different compared with their post-Archaean counterparts. Indeed, Archaean terranes are mostly made up of plagioclase-rich granitoids of the Tonalite–Trondhjemite–Granodiorite (TTG) series, whereas post-Archaean felsic plutons mainly consist in “true” granites and granodiorites, richer in K-feldspar (Figure 1.10).

Finally, the Archaean record also lacks some rock associations typical of modern-style tectonics, such as ophiolites. Some Archaean lithologies have been interpreted as representing ophiolitic remnants, but in general, they do not exhibit the whole systematic lithostratigraphy of present-day ones. Indeed, the oldest, unequivocally complete ophiolitic complexes on Earth (Purtunig in Canada ; Jormua in Finland) are Paleoproterozoic in age (~2000 Ma).

1.3.2. Evolution of orogenic styles

The structural record of Earth’s continental also evolved across the Archaean-Proterozoic transition. Some Archaean structures, such as horizontal seismic reflectors in the deep crust of the Superior Province in Canada (Figure 1.11), could result from modern-style, horizontal plate tectonics, but this interpretation is not univocal. On the other hand, primary features of Archaean crust, such as typical dome-and-keel structures that are obvious in most cratons (such as the Pilbara; Figure 1.12a), are scarce in modern orogens and hardly accounted for by horizontal tectonics. They rather result from a global gravity-driven overturn of the crust, triggered by sinking of dense volcanic rocks within ductile TTG plutons affected by diapiric upwelling (Figure 1.12b). Such a “sagduction” process reflects mostly vertical tectonics.

On the other hand, the overall geometry of Archaean orogenic belts is very different compared with present-day ones (Figure 1.13). Modern-style plate tectonics generate continent-scale belts, featured by significant crustal thickening (>40 km, accommodated by a

deep crustal root and high topographic elevation) as well as crustal slices stacked through subhorizontal thrusts that focus the deformation. Such an orogenic style is referred to as a “Cold Orogen”. By contrast, Archaean “orogens” are characterized by homogeneously vertical, pervasive foliation over hundreds of kilometers, and parallel ductile shear zones that are associated to the transpressive accretion of small crustal blocks without significant thickening. These features relate them to “Ultra Hot Orogens”. All intermediates exist between these two situations, such as “Hot-” and “Mixed Hot Orogens” (Figure 1.13). A statistical study of the relative repartition of different orogenic types throughout Earth’s history reveals that transitional types are particularly common in the late-Archaean and early Proterozoic.

1.3.3. Secular evolution of crust composition

The secular evolution of the crust composition has firstly been addressed using detrital sediments. The comparison of Archaean and post-Archaean sediments revealed that the latter has less fractionated REE patterns (Figure 1.14a) than the former, as well as a pronounced Eu negative anomaly while Archean shales do not. Moreover, specific elemental ratios such as Th/Sc, K/Na (Figure 1.14b) increase across the Archaean-Proterozoic transition. All these changes indicate that the source of sediments turned from a relatively juvenile, K-poor and HREE-depleted source (i.e. the Archaean TTG) into a recycled component (K-rich granites) derived from intracrustal differentiation. In addition, the initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of marine carbonates rose up at the end of the Archaean (Figure 1.14b), reflecting a larger proportion of mature crust in the sedimentary material supplied to seawater.

Secondly, the compositional evolution of continental crust-forming rocks (including continental mafic rocks and granitoids) tracks the same global evolution. The Mg, Ni, Cr contents of mafic rocks continuously decrease throughout Earth’s history, whereas their Na and K concentrations as well as La/Yb ratios increase (Figure 1.15), reflecting smaller and smaller melting rates correlatively to decreasing mantle temperatures (see Section 1.2.1). The granitoids exhibit a sharper compositional evolution at ~2500 Ma. TTGs are sodic granitoids ($\text{K}_2\text{O}/\text{Na}_2\text{O} < 1$) whereas post-Archaean ones are potassic ($\text{K}_2\text{O}/\text{Na}_2\text{O} > 1$). In addition to this marked decrease in Na/K, La/Yb ratios and absolute Sr concentrations also dropped down, whereas Eu negative anomalies “appeared” (Figure 1.15). Such a sharp break reflects two different evolutions of petrogenetic processes giving rise to crustal granitoids: (1) during the Archaean, juvenile magmas (TTG) are produced by medium- to high-pressure melting of

mafic rocks, whereas post-Archaean juvenile granitoids derive from differentiation of metasomatized mantle-derived basalts, such that they are richer in incompatible and fluid-mobile elements (e.g. K); (2) since the late-Archaean, intracrustal differentiation emerged as a fundamental way to produce granitoid magmas: shallow melting in the plagioclase stability field is indeed depicted by increasing Eu negative anomaly and decreasing Sr contents through time.

1.4. Atmospheric changes

Several lines of evidence indicate that the Archaean atmosphere was significantly less rich in O₂ than the present-day one, especially the lack of oxidized soils older than 2500 Ma, reduced detrital minerals in Archaean sediments as well as mass-independent fractionation of S isotopes before ~2450 Ma (Figure 1.16). Specifically, the O₂ content of the Archaean atmosphere was likely below $2 \cdot 10^{-6}$ bars, i.e. more than 5 orders of magnitude lower than today. It rose up very quickly at ~2500 Ma, which is known as the Great Oxidation Event (GOE in Figure 1.16) probably owing to a bloom of biological, photosynthetic activity.

In turn, the Archaean atmosphere was also richer in CO₂ than is modern counterpart. Indeed, several observations (i.e. O isotopic composition of cherts; meteoric alteration of Archaean rocks) evidenced that the atmospheric temperature in the Archaean was similar as today. This is surprising given that the activity of the Sun was weaker in the Archaean. To account for this discrepancy, the only possibility is that greenhouse effect was stronger at that time, pointing to higher CO₂ concentrations in the Archaean atmosphere (Figure 1.16).

1.5. Evolution of other geological parameters

Several other parameters significantly evolved across the Archaean-Proterozoic transition, namely:

- The O isotopic composition of igneous zircons. $\delta^{18}\text{O}_{\text{V-SMOW}}$ is constantly close to mantle values (5–6‰) during the Archaean, whereas it continuously increases after 2500 Ma (up to 10–12‰), reflecting a larger and larger proportion of material that underwent interactions with hydrosphere (i.e. sediments) in the source of granitoid rocks.

- The composition of the lithospheric mantle. Indeed, xenoliths of mantle peridotite brought up by kimberlites in cratonic areas show much more melt-depleted composition (i.e. low CaO, Al₂O₃, FeO and high Mg#) compared with their modern counterparts. Moreover,

the Re depletion ages (T_{RD}) of cratonic mantle xenoliths spread between 2900 and 2500 Ma (Figure 1.18), i.e. during the Archaean-Proterozoic transition, suggesting a major melting event at that time.

- The geometry of the continental crust. Statistically, Archaean crust is ~20% thinner than Proterozoic crust and, in general, do not exhibit any basal layer with P-wave velocities higher than $7 \text{ km}\cdot\text{s}^{-1}$.

- The intensity of the magnetic field. Indeed, the latter suddenly increased within a short time span (less than 500 Ma) during the Archaean-Proterozoic transition before dropping down towards its present-day value (Figure 1.19). This shows that the dynamics of Earth's core were disrupted at that time.

1.6. Synthesis

At the light of data presented above, it appears clearly that the Archaean geological record is strikingly different compared with post-Archaean features, reflecting major geodynamic changes around the Archaean-Proterozoic transition. This PhD thesis aims to provide new insights into these geodynamic changes, in particular concerning the secular evolution of crustal granitoids. Indeed, as pointed out in Section 1.3.3, Archaean TTGs clearly differ in composition compared with post-Archaean granitoids, reflecting a global evolution of petrogenetic processes giving rise to the continental crust at the Archaean-Proterozoic transition. The geodynamic setting of crust formation during the Archaean is an ongoing matter of debate, but whatever the model considered, it has no consequences on the previous conclusion that crustal growth processes evolved at the Archaean-Proterozoic transition. Indeed, currently proposed models for genesis of TTG (i.e., schematically, extensive melting of hot subducted mafic crust or progressive recycling of a thick, magmatically or tectonically accreted mafic plateau) do not compare with post-Archaean sites of crustal growth.

1.7. Possible causes

Some authors already investigated the origin of the geodynamic changes at the Archaean-Proterozoic transition. Globally, the proposed causes for the changes can be split into two groups: “punctual” processes or “continuous” processes.

1.7.1. “Punctual” processes

This family of hypotheses involves “catastrophic” and short-lived events to account for the geodynamic changes at the Archaean-Proterozoic transition. The most popular one consists in considering global mantle overturns owing to the conjugate action of hot mantle plumes in one hand, and cold avalanches of subducted mafic crust on the other hand (Figure 1.20). Such a mechanism would account for the episodic growth of continental crust (as shown in Figure 1.3). From this perspective, the Archaean-Proterozoic transition thus represents the advent of such crustal growth processes, with the first mantle overturn occurring at ~2700 Ma.

Other, less common hypotheses consider a global reorganization of mantle convection processes (from two layers to one layer), or nucleation of inner core. The latter mechanism would have induced instability of the D” layer and, correlatively, an increase in the activity of mantle plumes, thus initiating the mantle overturn processes described above.

1.7.2. “Continuous” processes

These hypotheses are generally based on a unique “continuous” process, namely the progressive cooling of Earth. From this perspective, the abrupt character of the evolution for some geological parameters (e.g. composition of granitoids) would reflect a threshold effect within the global and continuous cooling of Earth, but the nature of this threshold is not well constrained so far. Based on the secular evolution of granitoid compositions, some authors proposed new interpretations that are further detailed in Section 2.4.3. Alternatively, some models consider that the Archaean-Proterozoic transition rather reflects the “local” stabilization of discrete pieces of continental crust owing to the cooling of Earth and decrease of crust productivity through time.

From a global perspective, the geodynamic changes at the Archaean-Proterozoic transition must result from very global processes, because of their occurrence within a relatively short time range (~500 Ma) compared with Earth’s history. On the other hand, some local parameters certainly exerted a significant influence, as the Archaean-Proterozoic transition is diachronous from a craton to another. This work aims to partly unravel this problem and propose new interpretations for the origin of late-Archaean geodynamic changes (see Chapter 7).

Chapter 2

Late-Archaean sanukitoids and associated granites

2.1. Sanukitoids

2.1.1. Historical definition

The first description (Shirey & Hanson, 1984) and subsequent definition (Stern *et al.*, 1989) of sanukitoids were based on studies of the late-Archaean (2680–2750 Ma) Rainy Lake and Roaring River suites from the Superior Province, Canada (Figure 2.1). Sanukitoids were named after their close similarity with high-Mg andesites of Miocene age from Japan, referred to as sanukites.

The first definition of sanukitoids, addressed by Stern *et al.* (1989), is based on a number of geochemical criteria:

- $\text{SiO}_2 = 55\text{--}60$ wt.%, $\text{K}_2\text{O} > 1$ wt.% and $\text{MgO} > 6$ wt.%;
- $\text{Mg\#} > 0.6$;
- $\text{Ni} > 100$ ppm; $\text{Cr} > 200$ ppm ; $\text{Sr} > 500$ ppm ; $\text{Ba} > 500$ ppm ;
- $\text{Rb/Sr} < 0.1$;
- Fractionated REE patterns and $\text{Eu}_N/\text{Eu}^* \sim 1$.

However, most of the late-Archaean rocks described as “sanukitoids” since this definition was initially published do not actually fulfill all these conditions. In particular, most of them are in fact felsic rocks, especially granodiorites and granites with $\text{SiO}_2 > 60$ wt.% (Figure 5.2). Surprisingly, less than 5% of all rocks (18 samples over ~600) described worldwide as “sanukitoids” fit the initial definition as expressed by Stern *et al.* (1989), only considering the SiO_2 and MgO contents (red dots in Figure 5.2). This shows that sanukitoids are in fact composite magmatic complexes, including a wide range of (ultra)mafic to felsic rocks, the latter being generally dominant in terms of volume.

Such a discrepancy between the historical definition and the field reality was solved recently by Heilimo *et al.* (2010) that introduced a new geochemical definition of the so-called “sanukitoid suite” (Table 2.1¹), i.e. sanukitoids *s.s.* and all their differentiation products

¹ In Table 2.1, « *critère qualitatif* » means “qualitative criterion”, and « *non spécifié* » means “not described”. In addition, “*pds.%*” states for “wt.%”.

(granodiorites and granites). Such a new definition includes a much larger proportion of rocks described as sanukitoids, and thus appears more reasonable.

2.1.2. Geographic repartition

The map of Figure 2.3 reports all occurrences of late-Archaean sanukitoids worldwide. Three levels of confidence for the assignment of each occurrence to sanukitoids are represented by the font style of the intrusion name:

- In black font: recognized, i.e. described as a sanukitoid intrusion in the literature.
- In grey font: not described as sanukitoid, but can be considered as so because of adequate petrographic features and chemical compositions consistent with the definition.
- In italic grey font: inferred on the basis of petrographic features, but not verified because geochemical data are either very scarce, unpublished or not available.

Here is an exhaustive list of sanukitoid occurrences worldwide and associated references:

→ **Superior Province** (Canada): many intrusions in the Berens River, Wabigoon, Quetico (Shirey & Hanson, 1984 ; Stern *et al.*, 1989 ; Sutcliffe *et al.*, 1990 ; Stern & Hanson, 1991 ; Stevenson *et al.*, 1999 ; Whalen *et al.*, 2004), Wawa (Sage *et al.*, 1996) and Abitibi (Sutcliffe *et al.*, 1990 ; Feng & Kerrich, 1992) subprovinces.

→ **Slave Craton and Rae Province** (Canada): Yellowknife granitoid suite (Davis & Hegner, 1992), Defeat suite (Davis & Bleeker, 1999) and Boothia suite (Hinchey *et al.*, 2011).

→ **Wyoming Province** (USA): Louis Lake and Bridger batholiths (Frost *et al.*, 1998).

→ **North Atlantic Craton** (Greenland): diorites in the Fiskefjord and Disko Bugt areas (Steenfelt *et al.*, 2005).

→ **Amazonian Shield** (Brazil): Rio Maria suite (Oliveira *et al.*, 2008).

→ **Baltic Shield** (Finland, Russia): by alphabetical order, the most important intrusions are : Arola, Elmus, Hautavaara, Ilomantsinjärvi, Kaapinsalmi, Kaartojärvet, Koitere, Kuittila, Kurgelampy, Kussamo, Lieksa, Loso, Nilisiä, Njuk, Panozero, Siikalahti, Sjargozero, Sysmäjärvi (Bibikova *et al.*, 2005 ; Halla, 2005 ; Kovalenko *et al.*, 2005 ; Lobach-Zhuchenko *et al.*, 2005, 2007, 2008 ; Samsonov *et al.*, 2005 ; Käpyaho *et al.*, 2006 ; Larionova *et al.*, 2007 ; Halla *et al.*, 2009 ; Heilimo *et al.*, 2010, 2011).

→ **Ukrainian Shield** (Ukraine): Osipenkovskii complex (Bibikova *et al.*, 2008).

→ **Tanzanian Shield** (Tanzania): lavas, Musoma-Mara greenstone belt (Manya *et al.*, 2007).

→ **Kalahari Craton** (Botswana, Zimbabwe, South Africa): Tati and Selkirk suites (Kampunzu *et al.*, 2003; Zhai *et al.*, 2006), Bulai pluton (this study).

→ **Antogil Craton** (Madagascar): Masoala suite (Schofield *et al.*, 2010).

→ **Dharwar Craton** (India): Dod gneisses, Closepet batholith (Jayananda *et al.*, 1995, 2000; Krogstad *et al.*, 1995 ; Moyen *et al.*, 1997, 2001 ; Sarvothaman, 2001).

→ **North China Craton** (China): Jiekouling, Anziling, Quinhuangdao plutons (Yang *et al.*, 2008) and Taishan diorites (Jahn *et al.*, 1988 ; Wang *et al.*, 2009).

→ **Pilbara Craton** (Australia): Sisters supersuite from the Mallina basin (Smithies, 2000; Smithies and Champion, 1999, 2000).

→ **Yilgarn Craton** (Australia): some plutons from the Norseman-Wiluna belt, especially the Liberty, Lawlers, Mt. Lucky and Porphyry granodiorites (Cassidy *et al.*, 1991; Champion & Sheraton, 1997).

→ **Terre Adélie Shield** (Antarctica): Port Martin granodiorite (Monnier, 1995).

2.1.3. Emplacement ages

Figure 2.4 presents the emplacement ages of all sanukitoid suites for which geochronological data are available. Two main observations arise from this compilation:

- Within a given craton or province, the emplacement ages of sanukitoids do not span over more than 100 Ma, and are rather in the range 25–50 Ma. As a result, the building of sanukitoid complexes is a relatively short-lived event, by contrast with, for instance, present-day calc-alkaline batholiths that emplace at convergent plate margins (sometimes more than 100 Ma of magmatic activity; *e.g.* Moyen and van Hunen, 2012).

- On the other hand, in all cratons worldwide, emplacement ages of sanukitoids span over ~500 Ma from 2500 to 3000 Ma.

This shows that the Archaean-Proterozoic transition seems to be a short-lived event locally, but rather progressive from a global point of view.

2.1.4. *Geology and petrography*

Sanukitoids are composite magmatic complexes, but the most common rock types are medium- to coarse-grained (0.1–5 cm) equigranular (Figure 2.5a–b) and porphyritic (Figure 2.5c–d) diorites and granodiorites. These rocks are systematically associated with very common microgranular mafic enclaves (MME; Figure 2.5) and felsic rocks (granites), and, less frequently, mafic to ultramafic bodies (lamprophyre dykes, gabbros, clinopyroxenites and, exceptionally, peridotites).

From a structural point of view, they are always intrusive within older rocks such as TTG and greenstone belts. Their intrusion is generally syn- to late-tectonic: some plutons show a pervasive deformation (Figure 2.5c) coeval with emplacement, and/or emplaced crust-scale structures such as major shear zones (*i.e.* Closepet batholith in India; Moyen *et al.*, 2001).

The mineralogy of sanukitoids is extremely homogeneous from a global perspective. These rocks are calc-alkaline: plagioclase and K-feldspar coexist in roughly equal proportions, even if the former dominates in mafic rocks, whereas the latter is more abundant in felsic terms. Interestingly, in many sanukitoid complexes (*e.g.* Stern, 1989 ; Stern & Hanson, 1991 ; Jayananda *et al.*, 2000 ; Smithies & Champion, 2000 ; Yang *et al.*, 2008 ; this study) plagioclase often has a very constant composition (An₁₅ to An₄₀) throughout the magmatic suite. Most common mafic minerals are both calcic (clinopyroxene and hornblende) and potassic (biotite). Orthopyroxene is occasional. Mafic minerals generally form aggregates, leading to a very typical speckled appearance. These aggregates also contain the (abundant) accessory phases that are systematically Fe–Ti oxides (both ilmenite and magnetite) apatite, allanite, zircon and sometimes titanite.

2.1.5. Geochemistry

Some aspects of the major-element geochemistry of late-Archaean sanukitoids are reported in Figure 2.6 (personal database of O. Laurent, available on request). These rocks are metaluminous (Figure 2.6a) and calc-alkaline (Figure 2.6b). They cover a wide range of SiO₂ contents, and are both magnesian (high Mg# compared with TTG; Figure 2.6c) and potassic (high-K calc-alkaline to shoshonitic affinities; Figure 2.6d). As shown in all panels of Figure 2.6, such compositions are clearly distinct from that of Archaean TTG, and is rather related to analogue late-orogenic, high-K and high-Mg calc-alkaline granitoid suites from the Proterozoic and Phanerozoic (for some examples, see for instance Debon & Lemmet, 1999; Bonin, 2004; Yang *et al.*, 2004 ; Fowler & Rollinson, 2012).

The trace-element signature of sanukitoid suites is somewhat ambivalent. They are both rich in transition elements (Ni, Cr, Co, V) and all incompatible elements (especially Ba and Sr, but also REE and HFSE). Their REE patterns are fractionated, and classical multi-element diagrams display typical troughs in Nb–Ta and Ti as well as a positive Pb anomaly. As a result, their patterns are globally parallel to that of Archaean TTG, but switched to more enriched values (Figure 2.7).

To summarize, the chemical composition of sanukitoids is intermediate between that of Archaean TTG (shape of trace element patterns) and post-Archaean calc-alkaline granitoids (major elements, richness in incompatible elements). Such a transitional affinity is well illustrated by the plot of Figure 2.8, where sanukitoids (dots) plot in-between the fields of TTG (fractionated REE patterns) and post-Archaean granitoids (potassic affinities).

2.2. Petrogenesis of the mafic sanukitoids

The petrogenesis of mafic end-members of the sanukitoid suite (*i.e.* [monzo]diorites) is well constrained, from both geochemical and experimental arguments.

2.2.1. Geochemical constraints

High Mg#, as well as transition element contents of the sanukitoids point to a mantle source (Shirey & Hanson, 1984). However, these rocks are also extremely enriched in incompatible elements (especially Sr, Ba, REE) that are depleted in the upper mantle and enriched in continental crust with respect to the primitive mantle. Such high concentrations can result neither from crustal contamination, neither extremely low-degree melting of a non-

enriched peridotite (Stern *et al.*, 1989). As a result, sanukitoid magmas necessary derive from interactions between the mantle and a component rich in incompatible elements.

Several authors argued that this component is represented by a TTG melt (Smithies & Champion, 2000 ; Moyen *et al.*, 2001 ; Martin *et al.*, 2005, 2009). This would account for the parallel trace-element patterns between sanukitoids and TTG (Figure 2.7). Furthermore, sanukitoids and TTG are respectively similar in composition to modern low- and high-silica adakites (Figure 2.8). Low-silica adakites derive from hybridation between high-silica adakites and mantle peridotite (*e.g.* Kelemen *et al.*, 1993 ; Yogodzinski *et al.*, 1995), which is consistent with the petrogenetic model inferred for sanukitoids.

2.2.2. Experimental constraints

Experimental interactions between TTG melts and peridotite have been carried out by Rapp *et al.* (1999, 2010). Those authors showed that melting of metabasalts without hybridation with peridotite yields liquids with major-element compositions similar to that of TTG (Figure 2.10). Subsequent interactions between such liquids and peridotitic material gives rise to melts that match the composition of natural sanukitoids (Figure 2.11), especially their higher Mg# and lower SiO₂ contents than TTG. Moreover, the composition of the experimental melts fit with that of natural samples not only for major elements, but also for trace elements (Rapp *et al.*, 2010; Figure 2.11).

Both lines of evidence strongly support the model inferred on the basis of geochemical constraints, implying that sanukitoids derive from interactions between TTG liquids and mantle peridotite.

2.3. Diversity of late-Archaean magmatism

Sanukitoids are obviously not the only kind of late-Archaean magmas: the plutonic record during the Archaean-Proterozoic transition is much more complex. In this Section, I would like to detail this diversity of late-Archaean magmatism. The latter could have critical implications for the geodynamic setting at that time and our understanding of the geodynamic changes at the Archaean-Proterozoic transition.

This diversity of late-Archaean magmas appears at two different levels:

- In the sanukitoid group itself (*i.e.* for rocks that match the definition proposed by Heilimo *et al.*, 2010), there are some subtle geochemical differences that need further investigation (see Section 2.3.1);
- Some late-Archaean granitoids do not match the definition of sanukitoids. Some of them are likely related to them (see Section 2.3.2) whereas others are clearly different in both composition and origin (see Section 2.3.3).

2.3.1. Low- and High-Ti sanukitoids

Within the Dharwar craton in Southern India, rocks from the Closepet batholith and Dod gneisses (Jayananda *et al.*, 1995, 2000 ; Krogstad *et al.*, 1995 ; Moyen *et al.*, 1997, 2001) both belong to the sanukitoid suite as defined by Heilimo *et al.* (2010). However, as shown in Figure 2.12, Closepet samples define a distinctly TiO₂-richer trend than the Dod samples, for a given level of silica saturation. Such discrimination between both groups also appears when considering petrography: the Closepet, high-Ti samples are dominantly porphyritic granitoids, whereas the low-Ti Dod gneisses are dark equigranular rocks.

A careful analysis of geochemical data from all sanukitoids worldwide reveal that such a dichotomy is not specific to the Dharwar craton and also concerns other late-Archaean terranes (especially the North China, Yilgarn and Kalahari craton, as well as the Superior and Wyoming Provinces). In particular, the whole sanukitoid database can be split into two equally represented subgroups, considering a value of 3.5 for the MgO/TiO₂ as a discriminating factor (Figure 2.13a). This led Martin *et al.* (2009) to introduce the terminology of low- and high-Ti sanukitoids to describe both groups.

Such a distinction is also relevant on the basis of trace elements: indeed, low-Ti sanukitoids display slightly more fractionated REE patterns and lower absolute REE concentrations than high-Ti ones (Figure 2.13b). In addition, high-Ti sanukitoids are richer in HFSE, but contain less transition elements than low-Ti sanukitoids (Figure 2.13c). By contrast, Sr and Ba contents are less discriminating: both groups are rich in these elements (Figure 2.13d). Importantly, this dichotomy is much more obvious for the mafic samples (SiO₂ <62 wt.%) than the felsic ones (Figure 2.13). This indicates that differences between low- and high-Ti sanukitoids result from mantle processes rather than shallow-level differentiation. Such issues will be investigated further in the scope of this PhD work (see Chapter 4, Section 4.2.2).

2.3.2. “Marginal” sanukitoids?

A number of late-Archaean magmatic complexes resemble sanukitoids: they are composite plutons, made up of a wide range of calc-alkaline plutonic rocks from diorites to granites, bearing clinopyroxene, amphibole and biotite as major mafic phases. However, these rocks show subtle geochemical differences with sanukitoids, such that they do not perfectly match the definition of the sanukitoid suite expressed by Heilimo *et al.* (2010), and present “marginal” composition compared with the latter (Figure 2.14). Such granitoids were reported from several cratons worldwide. They include both mafic and intermediate rocks from the Superior Province (Sage *et al.*, 1996 ; Whalen *et al.*, 2004), the North Atlantic Craton (Steenfelt *et al.*, 2005) and the Baltic Shield (Mikkola *et al.*, 2011a,b), as well as felsic granitoids (granodiorites and granites) from the Superior (Whalen *et al.*, 2004) and Wyoming (Frost *et al.*, 1998) Provinces, the Baltic Shield (Kovalenko *et al.*, 2005 ; Käpyaho *et al.*, 2006) and the Amazonian Shield (Almeida *et al.*, 2011).

In general, such rocks exhibit lower MgO and transition element contents than “true” sanukitoids, less fractionated REE patterns and slightly lower Sr and Ba contents (Figure 2.14). Nevertheless, their composition is quite diverse so that they cannot be grouped together into a unique, generic definition. Correlatively, it seems that there is no unique petrogenetic model to explain the origin of these granitoids. However, the authors that studied their origin so far systematically invoke the involvement of a sanukitoid component (*e.g.* Whalen *et al.*, 2004 ; Almeida *et al.*, 2011) or, such as sanukitoids, variously enriched mantle sources (*e.g.* Steenfelt *et al.*, 2005 ; Mikkola *et al.*, 2011b). In addition, there are often closely associated to “true” sanukitoids in space and time, such that a genetic link might exist between the latter and “marginal” sanukitoids. The origin of such a genetic link is further investigated in this study (Chapter 5).

2.3.3. Late-Archaean magmas unrelated to sanukitoids

Two kinds of late-Archaean granites are strictly unrelated to sanukitoids (either “true” or “marginal”). They include (1) biotite-bearing granites (extremely widespread in all cratons, even more than sanukitoids; Sylvester, 1994) and (2) two-mica leucogranites (relatively scarce) that generally lack any intermediate to mafic component (MME, diorites...). According to Moyen *et al.* (2003), these granite types derive from melting of pre-existing continental lithologies, respectively TTG and metasedimentary rocks, without involvement of the mantle in their origin. As shown in Figure 2.15, these granites are compositionally distinct

compared with both TTG and sanukitoids, as they display higher K/Na, A/CNK and Rb contents as well as lower Mg#, lower concentrations in Sr, HFSE (Zr), and transition elements (Ni).

Finally, some A-type (peralkaline) granites exceptionally occur in late-Archaean terranes. They outcrop, for example, in the Pilbara craton (Smithies & Champion, 2000), the Baltic Shield (Mikkola *et al.*, 2011a) and the Superior Province (Corfu *et al.*, 1989 ; Sutcliffe *et al.*, 1990). Their origin is poorly understood so far; they would derive from melting of a mafic, lower crustal source enriched in incompatible elements (Smithies & Champion, 2000) or a CO₂-rich metasomatized mantle (Mikkola *et al.*, 2011a).

2.3.4. Towards a new typology?

In the light of the considerations summarized in the previous sections, late-Archaean granitoids can be classified into six different groups:

- (1) Low-Ti sanukitoids (equigranular monzodiorites and granodiorites rich in MgO, transition elements as well as Sr and Ba);

- (2) High-Ti sanukitoids (porphyritic granodiorites and subordinate monzodiorites rich in TiO₂, HFSE, REE as well as Sr and Ba);

[Both groups (low- and high-Ti sanukitoids) match the definition of Heilimo *et al.* (2010) and thus, are referred to as “true” sanukitoids.]

- (3) “Marginal” sanukitoids, which resemble “true” sanukitoids in many ways and are generally associated to them but, because of subtle differences on the geochemical point of view, do not perfectly fit the definition proposed by Heilimo *et al.* (2010).

- (4) Slightly peraluminous biotite-bearing granites that derive from melting of older TTG gneisses.

- (5) Peraluminous two-mica leucogranites, which represent reworking of sediments.

- (6) Peralkaline granites that are exceptional and which origin is debated.

Whatever the group to which they belong, late-Archaean granites show strikingly different compositions compared with Archaean TTG. In particular, all these magmas are potassic in composition, with K₂O/Na₂O generally above unity, and in this way are rather related to post-Archaean granitoids. Therefore, it seems that the geodynamic evolution at the Archaean-Proterozoic transition was recorded by all these granitoids and not specifically sanukitoids.

2.4. Unsolved issues

2.4.1. Nature of interactions at mantle levels

As stated in Section 2.2, current models for the petrogenesis of sanukitoids imply interactions between mantle peridotite and a component rich in incompatible elements. However, some uncertainties still remain concerning:

- the nature of the metasomatic agent – although most of the authors argued that it is represented by a TTG melt, this cannot account for some particular chemical features of some sanukitoid samples. Indeed, some of them bear a moderate to strong Eu negative anomaly (Figure 2.16), even the most mafic ones, which cannot result from hybridation between mantle and TTG that both lack any anomaly. As a result, in the case of these samples, the metasomatic agent would have already displayed this negative anomaly. Some workers advanced that a number of other components, in particular fluid phases (*e.g.* Lobach-Zhuchenko *et al.*, 2005, 2008) or sediments (*e.g.* Halla, 2005) may have played a role in the origin of sanukitoids and would account, among other features, for this Eu anomaly.

- the physical process of hybridation, which could proceed either in one step (the melt percolating into the mantle assimilates peridotite and evolves in composition towards that of sanukitoids) or in two steps (the metasomatic fluid or liquid is entirely consumed by interactions with mantle peridotite, giving rise to a hybrid assemblage that subsequently melts to produce sanukitoid magmas). The distinction between both mechanism only rely on the relative mass fractions between the metasomatic melt (or fluid) and peridotite, *i.e.* the so-called melt:rock ratio (Rapp *et al.*, 1999). A melt:rock ratio above 1 results in one-step hybridation, whereas complete consumption of the metasomatic agent happens if the melt:rock ratio is in the range 0–1 (Figure 2.17). Those two distinct mechanisms would account for the origin of low- and high-Ti sanukitoids (Figure 2.17), as postulated by Martin *et al.* (2009). However, this assumption only relies on qualitative data and must be confirmed. Geochemical modeling carried in the scope of this study will provide new insights into this question (see Section 4.2.2).

2.4.2. Differentiation processes

The magmatic evolution of sanukitoid suites at shallow, crustal levels, are not well constrained, while it has critical implications for our understanding of how the continental

crust grows and differentiates. Indeed, Kelemen (1995) and Tatsumi (2008) highlighted that the average composition of the continental crust is close to that of sanukitoids.

At that time, two different models exist for the differentiation of sanukitoid suites: (1) fractional crystallization or partial melting (*e.g.* Stern & Hanson, 1991; Davis & Hegner, 1992; Sage *et al.*, 1996; Lobach-Zhuchenko *et al.*, 2005, 2008), which are almost always proposed on the basis of qualitative considerations or (2) crustal contamination of the mafic mantle-derived melts (Figure 2.18), which could have taken place by assimilation coupled with fractional crystallization (*e.g.* Stevenson *et al.*, 1999) or magma mixing between sanukitoids and crust-derived melts (*e.g.* Moyen *et al.*, 1997, 2001). However, only a few studies proposed quantitatively constrained models, such that a more comprehensive assessment of the differentiation mechanisms for sanukitoid suites is needed (see Chapter 4, Section 4.3).

2.4.3. Global significance of late-Archaean magmatism

Some authors already studied the significance of late-Archaean magmatism, but mainly on the basis of sanukitoids. For example, Halla *et al.* (2009) and Heilimo *et al.* (2010) proposed that the close spatial and temporal association of sanukitoids with low- and high-HREE TTG reflects a context of subduction beneath a thick oceanic plateau. Melting of the latter produced HREE-rich TTG, whereas magmas derived from melting of subducted oceanic crust, in the garnet stability field, are HREE-depleted. Subsequent slab breakoff would have triggered the melting of mantle hybridized by percolation of TTG melts throughout the subduction episode. Such a model provides strong constraints on the geodynamic setting for concomitant genesis of TTG and sanukitoid, and implies that the latter are subduction-related magmas. However, it does not account for their specific occurrence at the end of the Archaean.

To explain the peculiar emplacement of sanukitoids at the Archaean-Proterozoic transition, Moyen *et al.* (2003) and Martin *et al.* (2009) proposed a new model, in which TTG are produced by melting of subducted crust throughout the Archaean (Figure 2.19 – first cartoon). The secular evolution of TTG composition between 4000 and 2500 Ma reflects more and more interactions with the overlying mantle wedge (Martin & Moyen, 2002). From this perspective, at the end of the Archaean, because of a weaker and weaker thermal regime, only small volumes of TTG are generated by melting of subducted oceanic crust. Those small volumes will extensively interact with the mantle to give rise to sanukitoids (Figure 2.19 – second cartoon). Finally, after 2500 Ma, the mantle is not hot enough to induce melting of

subducted crust that rather dehydrates, generating fluids rich in mobile elements (Rb, Ba, K...). The latter will interact with the mantle and thus give rise to the source of present-day arc magmas (Figure 2.19 – third cartoon). Such a model explains the relative sharpness of the geodynamic changes at the Archaean-Proterozoic transition, and the presence of sanukitoids at that time. However, it relies on an extremely controversial paradigm, *i.e.* that Archaean TTG almost exclusively derive from melting of subducted oceanic crust. Such an assumption is not obvious, given that during the Archaean, subduction is unstable on the mechanical and thermal point of view because of higher thermal regimes (van Hunen & van den Berg, 2008 ; Moyen & van Hunen, 2012).

Consequently, the origin of geodynamic changes at the Archaean-Proterozoic transition needs to be better constrained. In addition, very few models were proposed to account for the late-Archaean occurrence of not only sanukitoids, but also the whole diversity of granitoids described in this Chapter. In the scope of this PhD thesis, I propose new insights into these issues, constraining precisely the origin of granitoid diversity at the Archaean-Proterozoic transition and studying the spatial and temporal evolution of this magmatism in one place (the Kaapvaal craton) and at the planetary scale. Such issues are discussed in Chapters 5 to 7.

Chapter 3

Geology of Archaean basement in South Africa

Subsequently to the problematic (Introduction and Chapter 1) and materials (Chapter 2) of this PhD work, this Chapter provides insights into its geological setting, *i.e.* the Kaapvaal craton and Limpopo belt in South Africa. Both crustal segments, together with the Zimbabwe craton further North, form the Kalahari craton (Figure 3.1). This block outcrops over the borders of South Africa, Botswana, Zimbabwe, Lesotho and Swaziland, but only the South African part will be described in this section.

3.1. Overview of the Limpopo-Kaapvaal system

The continental block represented by the Kaapvaal craton and the Limpopo belt is a unique record of Early Earth processes: this well-preserved, extensively exposed terrane indeed includes one of the oldest rocks on Earth (back to 3600–3700 Ma) and its evolution lasted until ~2000 Ma, such that it represents more than a billion years of Earth history.

3.1.1. Lateral and vertical boundaries

According to geophysical studies (seismic and aeromagnetic data), the Kaapvaal craton covers more than 10^6 km² (Figures 3.1 and 3.2). Exposures of Archaean rocks cover less than 20% of the overall craton surface, most of them being restricted within a 500-km long and 100 to 200 km-wide band parallel to the eastern edge of the craton (Figure 3.2). It is flanked by “mobile belts” (Figure 3.1) developed at its margin during the late-Archaean and Proterozoic. Among them, the Limpopo Mobile Belt is the most studied: it separates the Kaapvaal craton from the Zimbabwe craton, to the North (Figure 3.2) and likely results from collision between them either in the late-Archaean or Paleoproterozoic (see Section 3.4.4). The northern and southern limits of the belt with both cratons are well exposed, whereas its lateral extension is completely masked by proterozoic cover sequences.

The continental crust of the Kaapvaal craton is 30 to 40 km thick (Nguuri *et al.*, 2001; Figure 3.3a), except below the ultramafic Bushveld complex (up to 45 km). It is significantly thicker below the Limpopo Belt, with an average of 45 km and reaching up to 52 km (Figure 3.3a). On the other hand, the lithospheric mantle beneath the Kaapvaal craton and Limpopo belt is quite typical of cratonic roots (Fouch *et al.*, 2004): the depth of its lower limit is down

to 250–300 km (Figure 3.3b,c), and it is made up of cold, refractory rocks as revealed by elevated P- and S-wave velocities (Figure 3.3b,c).

3.1.2. Crustal architecture

The Archaean crust of the Kaapvaal craton is divided in four provinces, structurally and chronologically distinct (the so-called “blocks” in Figure 3.2 ; Eglington & Armstrong, 2004). They are separated from each other by major crust-scale lineaments:

- The Colesburg magnetic anomaly cuts the craton into two parts along a N–S direction, the westernmost one being referred to as the “Kimberley block”. Albeit the largest one, this block is not well known because of poor exposure.

The other lineaments separate the eastern part of the craton into three domains:

- The “Pietersburg block” is the northernmost part. It is bounded by the Limpopo belt to the North, and the Thabazimbi-Murchison lineament (TML) to the South, a major structure along which geological activity lasted more than 2500 Ma (Good & de Wit, 1997).

- The world-famous Barberton greenstone belt is cross-cut by the Inyoka-Inyoni structure and its western continuation (the Barberton lineament). Between the latter and the TML lies the “Witwatersrand block” that includes the homonym basin, host of the largest Au deposits on Earth, as well as granitoid outcrops, which are not well characterized.

- Finally, south of the Barberton lineament lies the Swaziland block. It hosts the oldest crustal rocks of the Kaapvaal craton, namely the Ancient Gneiss Complex of Swaziland which age ranges between 3700 and 3100 Ma.

The Limpopo belt distinguished from the Kaapvaal craton on the basis of its lithologies, structures and metamorphic conditions (see Section 3.4 for details). Schematically, it can be divided into three zones (Figure 3.2), separated by large shear zones (Mason, 1973):

- The Northern and Southern Marginal Zones (NMZ and SMZ) respectively bound the Zimbabwe and Kaapvaal cratons, and are separated from the latter by the Umlali and Hout River thrust zones. Even if they are part of the Limpopo Belt according to its original definition, these zones are now recognized as high-grade counterparts of the neighboring cratons.

- The Central Zone (CZ) lies in-between both Marginal Zones. It is structurally complex and underwent a polymetamorphic history that is not precisely unraveled so far. It represents most of the outcropping surface in the Limpopo Belt.

3.1.3. Geological evolution

The geological evolution of the Limpopo-Kaapvaal system can be synthesized in three successive episodes (*e.g.* de Wit *et al.*, 1992; Poujol *et al.*, 2003; Zeh *et al.*, 2009):

- From 3700 to 3100 Ma: formation and stabilization of a continental nucleus (exposed in the Swaziland and Witwatersrand blocks).
- From 3100 to 2650 Ma: accretion and collages of small crustal blocks to the northern and western margins of the craton (exposed in the Kimberley and Pietersburg blocks) together with intracratonic sedimentation (Witwatersrand basin).
- From 2650 to 2000 Ma: collision (possibly polycyclic) with the Zimbabwe craton to the North, giving rise to the Limpopo Mobile Belt.

The next three sections describe the Archaean terranes that are most representative of each one of these events, namely (1) the Barberton area (Section 3.2), (2) the Pietersburg block (Section 3.3) and (3) the Limpopo Belt (Section 3.4).

3.2. Paleoarchaean nucleus in the Barberton area

This section aims to describe the geology of exposed Archaean basement south of the TML (Figure 3.2), which is by far the best described segment of the Kaapvaal craton. In particular, the area of the Barberton greenstone belt has been extensively studied for decades.

3.2.1. Granitoids

Granitoid rocks exposed around the Barberton greenstone belt (Figure 3.4) can be classified into four groups, described in the chronological order in the following:

- The layered and migmatitic grey gneisses of the Ancient Gneiss Complex are polycyclic tonalites and thronthjemites of the TTG series, associated with minor amphibolites. They are as old as 3644 ± 4 Ma (Compston & Kröner, 1988), younger ages spreading between 3575 and 3250 Ma (Kröner *et al.*, 1989 ; Zeh *et al.*, 2009). Because of very complex deformation and migmatization, it is not yet clear if those ages represent successive emplacement events or metamorphic overprints.
- Massive TTG plutonism occurred around the Barberton greenstone belt between 3550 and 3200 Ma (*e.g.* Armstrong *et al.*, 1990 ; Kröner *et al.*, 1991, 1996 ; Kamo & Davis, 1994), in three successive steps: (1) 3540–3510 Ma (Steynsdorp pluton), (2) 3450–3440 Ma (Stolzburg, Theespruit, Doornhoek plutons) and (3) 3250–3210 Ma (Stentor, Kaap Valley,

Nelshoogte plutons). All these rocks are deformed, but the strain pattern is less and less pronounced towards younger ages. Just after the last third step, emplaced the monzonitic Dalmein pluton (~3215 Ma) that is not related to the TTG but which origin is poorly known.

- Very large batholiths emplaced between 3100 and 3050 Ma (Nelspruit, Salisbury Kop, Heerenveen, Mpuluzi and Pigg's Peak ; Kamo *et al.*, 1990 ; Kamo & Davis, 1994). They belong to the so-called GMS (Granite–Monzogranite–Syenite) suite, and are much more potassic than older TTG. Their origin is still unclear, but they would derive from melting of older TTG, possibly coupled to a juvenile mantle input (Robb *et al.*, 2006 ; Zeh *et al.*, 2009).

- After more than 200 Ma of magmatic shutdown, during which developed the volcano-sedimentary Pongola supergroup (grey dotted area in Figure 3.4), a number of small granitoid intrusions emplaced between 2870 and 2690 Ma (Maphalala & Kröner, 1993). They can be divided into two groups (Meyer *et al.*, 1994): (1) low-Ca granites (Sinceni, Godlwayo, Mooihoek, Mhlosheni, Spekboom) likely derived from metasedimentary sources and (2) high-Ca granites (Mpageni, Nzimane, Sicunusa, Kwetta, Ngwempisi, Hlatikulu, Mbabane) which source is rather a igneous, hornblende-bearing intermediate rock.

3.2.2. *The Barberton greenstone belt (BGB)*

The volcano-sedimentary sequence of the BGB emplaced between 3550 and 3200 Ma. It is a narrow, 120 km-long and 20 to 50-km wide basin (Figure 3.5), which depth does not exceed 8 km (de Beer *et al.*, 1988 ; Figure 3.6). The lithostratigraphic succession of the BGB (as described in Lowe, 1994; Lowe & Byerly, 1999 ; Hofmann *et al.*, 2004 ; Brandl *et al.*, 2006) consists in three groups:

- the Onverwacht group is a thick (initially up to 10 km) mafic volcanic package emplaced between 3550 and 3300 Ma. It is made up of komatiites in the lower part and tholeiitic basalts, interstratified with felsic lavas and pyroclastites, in the upper part. These lavas are associated with rare cherts (that contain possibly biogenic structures; Westall *et al.*, 2001) and clastic sediments. These associations, together with evidence for submarine outpouring (pillow-lavas), suggest that the Onverwacht group represent a well-preserved portion of Archaean oceanic crust.

- the overlying Fig Tree group is a siliciclastic unit (greywackes and pelites) associated to calc-alkaline felsic lavas, quite similar to present-day arc or back-arc associations. It emplaced between 3290 and 3220 Ma.

- at the top of the stratigraphy lies the Moodies group, almost exclusively made up of silicic detrital rocks. It consists in three well-sorted sequences, from conglomerates at the base to sandstones at the top. They would have emplaced between 3225 and 3500 Ma, in fluvial to deltaic environments fed by the erosion of elevated topography.

3.2.3. Structures and metamorphism

Three tectono-metamorphic episodes can be distinguished in the Barberton area:

- The oldest TTG (*i.e.* Ancient Gneiss Complex and Steynsdorp pluton) underwent at least one, possibly several episodes of deformation and migmatization between 3400 and 3550 Ma. Their detailed chronology and the conditions in which they took place are poorly known, because they were overprinted by successive events.

- Most of the structures and metamorphism are aged of 3300–3200 Ma. Associated deformation is represented by (1) subvertical faults and isoclinal folds in the volcano-sedimentary sequence of the BGB (Lowe, 1994 ; Figure 3.6), (2) subvertical lineaments in oldest TTGs (Kisters *et al.*, 2003), and (3) concentric foliation in the TTG that emplaced at 3250–3210 Ma (Kaat Valley, Nelshoogte, Stentor), acquired by syntectonic diapirism (Belcher *et al.*, 2005). Associated metamorphism did not exceed greenschist facies conditions in the core of the BGB (Cloete, 1999 ; yellow box in Figure 3.7). By contrast, its southwestern margin is featured by contrasted metamorphic gradients in both sides of the Inyoka fault (Figure 3.5): low-pressure ones in the northern part (brown boxes in Figure 3.7) and high-pressure ones in the southern part (blue boxes in Figure 3.7) (*e.g.* Kisters *et al.*, 2003 ; Dziggel *et al.*, 2005 ; Moyen *et al.*, 2006).

- Most recent structures are strike-slip faults and shear zones, coeval with intrusion of large batholiths of the GMS suite (~3100 Ma; see Section 3.2.1) (Brandl *et al.*, 2006).

3.2.4. Geological evolution

One model for the evolution of the BGB and associated plutonic rock is presented in Figure 3.8. This model is based on studies by de Wit *et al.* (1992), de Ronde & de Wit (1994), Lowe (1999), Brandl *et al.* (2006), Moyen *et al.* (2006), Zeh *et al.* (2009), Kisters *et al.* (2010). The arguments supporting this model will not be detailed here and the reader is referred to the abovementioned references for details. To summarize, this evolution firstly involves continental crust formation between 3700 and 3400 Ma by melting of mafic rocks similar to the Onverwacht group, giving rise to TTG of the Ancient Gneiss Complex as well

as Steynsdorp pluton and Stolzberg block (Figure 3.8a). The resulting protocrust was then involved into a north-verging subduction event at 3250–330 Ma, leading to TTG magmatism and deposition of the arc-like Fig Tree group (Figure 3.8b). “High-pressure” metamorphism occurred at ~3225 Ma, coeval with collision (Figure 3.8c), and subsequent exhumation allowed massive melting of the mafic package to produce 3200 Ma-old TTG and deposition of the Moodies group (Figure 3.8d). The large batholiths of the GMS group likely emplaced in the last exhumation stages, together with massive felsic rocks further north (between the BGB and the TML) as well as in the Johannesburg area: this magmatism contributed to crustal stabilization at ~3100 Ma.

3.3. The Pietersburg block

Published geochronological data suggest that the Pietersburg block, the northernmost terrane of the Kaapvaal craton (see Figure 3.2), evolved between ~3200 and ~2700 Ma (Poujol *et al.*, 2003), *i.e.* in continuity with the evolution of the Swaziland and Witwatersrand blocks outlined in Section 3.2. However, these data are scarce, and no systematic dating study of the whole domain was carried out so far. This is the objective of the article (now accepted in Precambrian Research with major revision) “Crustal growth and evolution in the northern Kaapvaal craton inferred by LA-ICP-MS dating of zircons from Meso- and Neoproterozoic granitoids”. This paper aims to synthesize the geology and available geochronological data of the Pietersburg block (referred to as the “MNK terrane” in the paper, after Zeh *et al.*, 2009), and precise those in order to better constrain its evolution.

The evolution of the Pietersburg block outlined in Section 3.3.5 will be further constrained by new data, especially geochemistry of granitoid rocks, in Chapter 6 of the present PhD thesis.

3.4. The Limpopo Belt

3.4.1. General features

The structural position of the Limpopo Belt and, furthermore, its architecture (see notably the cross-section of Figure 3.21) clearly indicates that it results from collision between the Zimbabwe and Kaapvaal cratons (*e.g.* Roering *et al.*, 1992). It is distinct from the latter from

three different points of view: (1) the structural framework of the Limpopo Belt is particular: it lacks any preferential large-scale orientation, and small slices (less than a few km) of greenstones are embedded within orthogneisses in a very complexly deformed fashion; (2) metamorphic conditions underwent by rocks of the Limpopo Belt reached granulite facies (up to 10 kbar and 1000°C) whereas they did not exceed lower amphibolite facies ($P < 6$ kbar, $T < 650^\circ\text{C}$) in adjacent cratons; and (3) while lithologies of the Marginal Zones can be considered as high-grade counterparts of the neighboring craton rocks, those of the Central Zone are clearly different in origin, indicating that the latter represents an exotic terrane with respect to the Zimbabwe and Kaapvaal blocks.

3.4.2. Marginal Zones

In this Section, I describe the lithologies, structures, metamorphism and that are typical of the Marginal Zones. The SMZ is described in more detail, because this study focuses on granitoid rocks that are intrusive at the suture between the SMZ and the Kaapvaal craton whereas the NMZ was not investigated at all.

Typical lithologies in the Marginal Zones are principally orthogneisses and metapelites:

- In the SMZ, both components are respectively represented by the Bavianskloof and Bandelierkop complexes (see Figure 3.23). Bavianskloof gneisses are volumetrically dominant; these are orthopyroxene-bearing tonalites, showing extensive deformation and migmatization. Bandelierkop gneisses are orthopyroxene-, garnet- and sillimanite-bearing metapelites (Kramers *et al.*, 2006). The protoliths of these rocks uncertainly emplaced between 3300 and 2800 Ma (Kröner *et al.*, 2000; Kreissig *et al.*, 2000).

- In the NMZ, orthogneisses are almost exclusive (~90% of the volume) and only associated with minor mafic granulites and remnants of BIF. Such lithologies are younger than in the SMZ, with ages in the range 2750–2625 Ma (Berger *et al.*, 1995).

In both cases, geochemical studies unequivocally demonstrated that the Marginal Zones are high-grade counterparts of the neighboring cratons (Taylor *et al.*, 1991 ; Berger *et al.*, 1995 ; Kreissig *et al.*, 2000). Indeed, Nd model ages for gneisses of the SMZ are identical to that of samples from the adjacent Kaapvaal craton, while rocks from the NMZ and the Zimbabwe craton show identical model ages (Figure 3.22).

Each Marginal Zone is separated from its neighboring craton by an internal verging, thrust or transpressive shear zone (see the cross-section of Figure 3.21): the Umlali-North Limpopo thrust zone in the case of the NMZ, and the Hout River shear zone in the case of the SMZ.

Structures in the NMZ are globally parallel to the Umlali-North Limpopo thrust zone. They are somewhat more complex in the SMZ, where large shear zones (*e.g.* Matok, Anaskraal, N'Tabalala shear zones), associated with the Hout River system, separate less intensely deformed crustal blocks (Figure 3.23) (Smit & van Reenen, 1997). Most of these structures developed during the D₂ event (see the Geological Setting of the article in Section 3.3), corresponding to exhumation of the SMZ rocks with respect to those of the Kaapvaal craton.

About metamorphism, rocks of the NMZ reached *P-T* conditions of 800–900°C and 8–9 kbar (Rollinson, 1989; Kamber & Biino, 1995). The *P-T* path in the SMZ is illustrated in Figure 3.24 (red arrow). Granulite-facies metamorphism took place at conditions of 850–1000°C and 8.5–9.5 kbar (Stevens & van Reenen, 1992 ; Perchuk *et al.*, 1996, 2000a ; Tsunogae *et al.*, 2004) (Metamorphic episode M₁ of Figure 3.24). After exhumation along the Hout River shear system (along different paths, illustrated by the thin dashed arrows in Figure 3.24), the granulites re-equilibrated at ~600°C and 4–6 kbar (Perchuk *et al.*, 2000a ; Smit *et al.*, 2001), while rocks of the footwall Kaapvaal craton underwent prograde metamorphism towards the same conditions (bold dashed arrows of Figure 3.24).

High-temperature, granulite-facies metamorphism is dated at 2691 ± 7 Ma in the SMZ (Kreissig *et al.*, 2001), which corresponds to the end of the northward thrusting event (D₁) bracketed between 2800 and 2700 Ma (see Geological setting of article in Section 3.3). Exhumation of the granulites occurred between 2700 and 2600 along the Hout River shear zone (Kreissig *et al.*, 2001). In the NMZ, the age of granulite-facies metamorphism is not well known, but is certainly in the range 2750–2625 Ma (Ridley, 1992). Exhumation of the granulites along the Umlali-North Limpopo thrust zone took place between 2650 and 2575 Ma (Mkweli *et al.*, 1995 ; Blekinsop *et al.*, 2004).

3.4.3. The Central Zone (CZ)

As stated earlier, the lithologies of the CZ can be related neither to those of the Zimbabwe craton, nor those of the Kaapvaal block. Typical lithologies of the CZ are:

- The Beit Bridge Complex (BBC), a very heterogeneous supracrustal sequence comprising granulite-facies metapelites (labeled “BBC” in Figure 3.25a), quartzites, BIF, calc-silicates and quartzofeldspathic gneisses. The latter (referred to as the Singelele gneisses ; Figure 3.25a) either represents metamorphosed terrigenous sediments, felsic volcanics or anatectic liquids. The BBC likely represents many successive sedimentary cycles, as illustrated by the huge range of absolute ages obtained from various of its

components (3300 to 2200 Ma; Barton & Sergeev, 1997 ; Jaeckel *et al.*, 1997 ; Kröner *et al.*, 1999 ; Buick *et al.*, 2003).

- The Messina Layered Intrusion is made up of meta-anorthosites and metagabbros aged of 3100 to 3200 Ma.

- The Sand River Gneisses (Figure 3.25b), a spectacular unit of banded and migmatized grey gneisses. These are typical TTG which emplaced between 3410 and 3170 Ma (Retief *et al.*, 1990 ; Tsunogae & Yurimoto, 1995 ; Jaeckel *et al.*, 1997 ; Kröner *et al.*, 1999 ; Zeh *et al.*, 2007, 2010). They represent either the basement of the BBC (*e.g.* Kramers *et al.*, 2006), or magmatic bodies intrusive within the latter (*e.g.* Hofmann *et al.*, 1998).

- Many granitoids are intrusive within the BBC, such as (1) the biotite-bearing, tonalitic Alldays gneisses (Figure 3.25c ; also known as Zanzibar or Verbaard gneisses), dated between 2670 and 2625 Ma (Jaeckel *et al.*, 1997 ; Kröner *et al.*, 1999); (2) the Bulai pluton, focus of this study (see Chapter 4 ; Figure 3.25d) at 2610–2580 Ma and (3) a heterogeneous set of undeformed leucogranite lenses and dykes, emplaced between 2050 and 2000 Ma (Jaeckel *et al.*, 1997 ; Kröner *et al.*, 1999 ; Zeh *et al.*, 2007, 2010).

Structurally, the CZ is bounded by two shear zones (the Triangle shear zone to the North, the Palala-Tshipise shear zone to the South ; see Figure 3.21) that are different compared with the bording thrusts of Marginal Zones in two ways: (1) they are vertical structures with a pure strike-slip movement and (2) their age is Paleoproterozoic (~2000 Ma ; Kamber *et al.*, 1995 ; Holzer *et al.*, 1998) rather than Archaean. On the other hand, rocks of the CZ display a very complex strain pattern at regional scale (see Figure 3.26) and at the outcrop scale. Closed structures (sheath folds ; Figure 3.26) and large shear zones (Baklykraal and Campbell structures ; Figure 3.26) are very common features. Little is known about these structures and their ages.

By contrast to rocks of the Marginal Zones that underwent a single P – T loop, those of the CZ are likely polymetamorphic. For instance, three successive migmatitic events can be distinguished:

- M_1 (~3140 Ma ; Zeh *et al.*, 2007): anatexis of the Sand River Gneisses.
- M_2 (~2650 Ma ; Jaeckel *et al.*, 1997 ; Kröner *et al.*, 1999) : anatectic Singelele leucogranite and Alldays gneisses.
- M_3 (~2000 ; various references) : undeformed melt patches.

The last two events are also represented by ages in metamorphic minerals from samples of the BBC (Figure 3.28). Those two groups of ages are problematic, because thermobarometric

studies evidence a unique P – T loop for the CZ (Figure 3.27a). Some of these studies were carried on samples yielding unequivocal Paleoproterozoic ages (*e.g.* Holzer *et al.*, 1998, 1999), whereas others result from the study of samples that were dated at ~2650 Ma, in the late-Archaean (*e.g.* Millonig *et al.*, 2008). These observations might have two different interpretations:

- Rocks of the CZ underwent two metamorphic events, one in the late-Archaean (~2650 Ma), one in the Paleoproterozoic (~2000 Ma), that are both characterized by identical (or at least similar) P – T loops (which is unlikely, but not impossible).

- As evidence in some (rare) samples (*e.g.* van Reenen *et al.*, 2008 ; Perchuk *et al.*, 2008), the P – T peak was reached in the late-Archaean (~2650 Ma), whereas the Paleoproterozoic event (~2000 Ma) is only related to a thermal event (isobaric heating) and subsequent, final exhumation (Figure 3.27b).

This issue is a considerable matter of debate (*e.g.* van Reenen *et al.*, 2008) and has important implications on the geological evolution of the Limpopo Belt.

3.4.4. Two different possible scenarios

No one doubts that the Limpopo Belt results from collision between the Zimbabwe and Kaapvaal cratons. Albeit very different from that of classical, present-day collisional orogens (*e.g.* the Himalayas), the geometry of the Limpopo belt is closely similar to that of “Hot-” and “Mixed-Hot Orogens” of Chardon *et al.* (2009 ; see Figure 1.13). The latter accommodate the collision between crustal blocks in the context of intermediate thermal regimes between Archaean and today (Cagnard *et al.*, 2011) such that it is very consistent with the late-Archaean to Paleoproterozoic age of the Limpopo belt.

However, because of geochronological problems about polymetamorphism in the CZ (see above), it is not yet clear if this collision occurred in the late-Archaean (~2650 Ma ; *e.g.* Roering *et al.*, 1992 ; Perchuk *et al.*, 2008) or the Paleoproterozoic (~2000 Ma ; Kamber *et al.*, 1995 ; Holzer *et al.*, 1998 ; Schaller *et al.*, 1999). Both models present they own advantages and discrepancies (see the details in van Reenen *et al.*, 2008) and this controversy is not yet solved, but such a discussion is beyond the scope of the present work.

Chapter 4

Petrogenesis of sanukitoids: the Bulai pluton

In this Chapter, I present new data on the late-Archaean Bulai pluton, from the Central Zone of the Limpopo Belt, which belongs to sanukitoid suites as expected from the definition of Heilimo *et al.* (2010). The goal of this Chapter is to provide new insights into:

- The petrogenetic processes at the origin of sanukitoid rocks at mantle levels, especially concerning the nature of the metasomatic agent and the physical conditions of hybridation between the latter and mantle peridotite (Section 3.2);
- How sanukitoid suites differentiate at crustal levels (Section 3.3).

4.1. Geological characterization of the Bulai pluton

Section 3.1 is simply a general presentation of the Bulai pluton (geology, structures, mineralogy, chemistry of minerals and conditions of emplacement). Such an introduction is grossly summarized in Section 3 of the Lithos paper. Below is a list of the Figures of Section 4.1, with important information associated with each one of those:

→ **Figure 4.1:** Geological sketch map of the Bulai pluton (can be found in the papers as well).

→ **Figure 4.2:** Specific map of the Three Sisters area, where sampling and fieldwork were extensively carried out. It particularly shows a large enclave of “enderbite” (pyroxene-bearing monzodiorite) on the southwestern flank of the inselberg, as well as emplacement of pegmatite and lamprophyre dykes and orientation of magmatic flow fabric.

→ **Figure 4.3:** Major rock types in the Bulai pluton: porphyritic granodiorite, undeformed (a,b), deformed in the margins of the pluton (c,d), associated with MME (e) and larger enclaves of enderbite (f,g), both showing diffuse contacts (h).

→ **Figure 4.4:** Minor rock types in the Bulai pluton: xenolith of Singelele gneisses (a) and BBC metapelites (b), comagmatic leucogranites (c), as well as lamprophyre (d,e) and pegmatite (e,f) dykes.

→ **Figure 4.5:** Mineralogy and textures in Bulai samples : MME (a), Cpx and Am in monzodiorite (b), complete pseudomorph of pyroxene in monzodiorite: oxide-, quartz- and amphibole-bearing euhedral aggregates (c), mafic clot in granodiorite (d), granoblastic texture of granodiorite (e) and partly recrystallized K-feldspar crystal in granodiorite (f). [for the origin of recrystallization textures, see Sections 3 and 4.1 of the Lithos paper].

→ **Figure 4.6:** Compositions of major minerals in samples of the Bulai pluton : (a) plagioclase, (b) amphibole, (c) biotite, (d) pyroxenes.

→ **Figure 4.7:** Illustration of the remarkably homogeneous composition of minerals in the Bulai pluton: composition of plagioclase, amphibole and biotite is plotted as a function of the SiO₂ content of the host rock.

These compositions are constant throughout the magmatic suite, which is similar to other sanukitoid suites worldwide (see Section 2.1.4). Symbols as in Figure 4.6.

→ **Figure 4.8:** P - T conditions of crystallization of 10 samples from the Bulai pluton, on the basis of different geothermobarometers. It is not yet clear whether these condition reflect magmatic emplacement or re-equilibration during the Paleoproterozoic metamorphic overprint.

4.2. Petrogenesis of mafic rocks: mantle processes

– *Section 4.2.1 : Lithos paper* –

(for Section 4.2.2., see below)

4.3. Petrogenesis of felsic rocks: crust processes

– *Precambrian Research paper* –

Explanation for models presented in Section 4.2.2

As discussed in Section 2.4.1, the origin of the mafic end-member of sanukitoid suites is not yet clear, in particular concerning the dichotomy between low- and high-Ti sanukitoids. Modeling presented in the Lithos paper (section 5.4) partly unraveled the origin of high-Ti sanukitoids (as the Bulai pluton belongs to this group), but a more systematic study is needed to account for the worldwide diversity of sanukitoid compositions. For this purpose, we built a geochemical model, detailed below, which results are compared with the worldwide database of sanukitoid compositions (data presented throughout Chapter 2).

• **The need for a new model**

Martin *et al.* (2009) already developed a geochemical model in order to understand how a unique mechanism (interaction between TTG and peridotite) could generate two groups of sanukitoids, clearly distinct on the basis of geochemistry. This model is based on that of Moyen (2009) for present-day adakites, and based on the batch melting equation 4.1, applied to a composition of metasomatized mantle (C_{mm}^i), itself calculated using the mass balance equation 4.2 (C_{TTG}^i and C_{π}^i represent the compositions of TTG and initial mantle peridotite, respectively, and a the mass fraction of TTG liquid). In this model, composition of different

components, as well as partition coefficients are fixed, so that the only variables are a and the mass fraction of liquid remaining after the interactions (F). Hence, Martin *et al.* (2009) demonstrated that the ratio F/a exerted an exclusive control on the geochemistry of the final liquid. From this point of view, the difference between low- and high-Ti sanukitoids is primarily the results of different F/a ratios during the interactions, respectively above and below 1 (Figure 4.9).

The problem of such a model is that it does not distinguish between one- or two-step hybridization between TTG and peridotite (see Section 2.4.1). Indeed, a F/a ratio of 1 (as an example) could either result from (1) a one-step process, where the mass of assimilated mantle is equal to that of new crystals formed from the hybrid liquid; or (2) a two-step process, if melting of the metasomatized mantle produces a mass fraction of liquid identical to that of the TTG liquids during the interactions. Moreover, if the ratio F/a was the only parameter controlling the composition of the final melt, all incompatible trace elements would be enriched in high-Ti sanukitoids compared with low-Ti sanukitoids (as illustrated in Figure 4.9). However, as pointed out in Section 2.3.1, low- and high-Ti sanukitoids present similar compositions in Sr, Ba, as well as Th and U that are all highly incompatible elements during mantle melting. As a result, the model developed by Martin *et al.* (2009) is not discriminating enough to account for the geochemical diversity of sanukitoids worldwide.

- **Experimental constraints on mantle hybridation processes**

We developed a more realistic model deduced from phase relationships in experimental studies of melt/rock interactions. Such studies demonstrated that hybridation mechanisms will be somewhat different depending on the relative proportion of TTG and peridotite, expressed as the “melt:rock ratio” ($R_{mr} = a / [1-a]$) of Rapp *et al.* (1999). This ratio equals the mass fraction of TTG divided by that of peridotite. In particular:

→ If $R_{mr} > 1$, the metasomatic liquid does not cross its solidus. In other words, it is not consumed by interactions and only assimilates peridotite. In turn, the hybrid liquid crystallized a pyroxenite assemblage containing garnet at high pressure ($P > 25$ kbar ; Rapp *et al.*, 1999, 2010) and hydrated minerals (amphibole, phlogopite) at lower pressure ($P < 25$ kbar ; Prouteau *et al.*, 2001). Such a process is adequately modeled by equations 4.1 and 4.2, if we consider that the melt equilibrates with residual pyroxenite. This assumption is confirmed by the good correlation between compositions of natural sanukitoids in one hand, and experimental melts on the other hand (see Section 2.2.1).

→ If $R_{mr} < 1$, and particularly if it is in the range 0.2 to 1 (Rapp *et al.*, 2006), the liquid is fully consumed by the interactions. It results in a hybrid peridotite or pyroxenite, containing metasomatic minerals such as secondary orthopyroxene, phlogopite, amphibole and sometimes, clinopyroxene (Sen & Dunn, 1994 ; Yaxley & Green, 1998 ; Prouteau, 1999 ; Rapp *et al.*, 1999). If such a hybrid assemblage melts, these phases will be the first ones to contribute to liquid production: specifically, amphibole melting starts between 980 and 1050°C, while phlogopite melts at 1050–1150°C (Rogers & Saunders, 1989 ; Schmidt & Poli, 1998 ; Conceição & Green, 2004). Clearly, such melting would be far from equilibrium. Moreover, amphibole melting produces melt together with peritectic garnet and clinopyroxene (Francis & Ludden, 1995 ; Dalpé & Baker, 2000) such that it must be modeled using an incongruent melting equation rather than the batch melting one (eq. 4.1).

In order to model both processes, we used the unique mass balance equation 4.2 to calculate the composition of the hybrid system. By contrast, the composition of sanukitoid melts were calculated (1) using the same batch melting equation 4.1 as Martin *et al.* (2009) for $R_{mr} > 1$ and (2) using the incongruent melting equation of Hertogen & Gijbels (1996) for $R_{mr} < 1$.

• Parameters of the models

Modeling is carried using a forward Monte Carlo calculation (similar to that of the Lithos paper; see Section 5.4 of the latter): compositions of melts and solids as well as partition coefficients are affected by an uncertainty range of $\pm 25\%$. The program runs a great number of calculations; for each one, it randomly selects a value within the assigned range for each variable. In general, the whole range of possibilities is statistically spanned after 10^4 to 10^5 runs.

Variables were selected as follows:

→ Initial composition of the mantle is that of depleted mantle (DMM) of Workman & Hart (2005).

→ Initial composition of TTG liquid is the average of Martin *et al.* (2005).

→ Partition coefficients in mantle minerals are those of Adam & Green (2006).

For the one-step model, we tested several mass fractions of TTG liquid in the range $a = 0.6$ – 0.9 ($R_{mr} = 1.5$ – 9). The mass fractions of resulting melts (F) were randomly chosen by the program at each step within the range 0.25–0.90, leading to global F/a ratios of 0.3 to 1.5. The modal composition of residual pyroxenite in equilibrium with the hybrid melt was estimated

using experiments of Rapp *et al.* (2010) at high pressure (45% Opx + 45% Cpx + 10% Grt) and Prouteau *et al.* (2001) at lower pressure (80% Opx + 10% Am + 10% Phl).

For the two-step model, we tested several mass fractions of TTG liquid in the range $a = 0.17\text{--}0.3$ ($R_{\text{mr}} = 0.2\text{--}0.4$). The incongruent melting equation need to assess the initial modal composition of metasomatized peridotite/pyroxenite. For this purpose, we used amphibole-forming reactions proposed by Sen & Dunn (1994) using a lherzolite as initial composition (75% Ol + 15% Opx + 10% Cpx). Final assemblages are harzburgites (45–60% Ol + 30–45% Opx + 0–5% Cpx, depending on the amount of metasomatic TTG) containing 5 to 10% modal amphibole. We tested the importance of phlogopite adding arbitrarily 1 to 3% modal of this mineral in metasomatized assemblages. We assumed that amphibole was the single melt-producing phase, and that it produced peritectic garnet and clinopyroxene in various relative proportions (Francis & Ludden, 1995). The value of the melting rate (F) was adjusted so that all amphibole is consumed (0–0.06). It leads to final F/a ratios in the range 0–0.35.

• Results

Results of the models are presented in Figure 4.10. The compositional range of calculated liquids represents all possible combinations tested according to the description above, and illustrated by colored fields. The symbols are natural mafic sanukitoids ($\text{SiO}_2 < 62$ wt.%).

It appears that the distinction between the two hybridation mechanisms is obvious from a geochemical point of view. Furthermore, it accounts for the dichotomy between low- and high-Ti sanukitoids. The high concentrations of the latter in HFSE and REE are well reproduced by two-step models. By contrast, the one-step hybridation produces liquids that are less rich in these elements, but contain more transition elements, such as low-Ti sanukitoids. Differences between both models are less contrasted considering Sr and Ba, as already observed on the basis of natural compositions.

In details (not shown), the composition of high-Ti sanukitoids is better reproduced if (1) garnet is absent from the peritectic assemblage and if (2) phlogopite is present as a residual phase. By contrast, the composition of low-Ti sanukitoids is better reproduced if (1) garnet is present in equilibrium with the hybrid melt and if (2) hydrated minerals are absent. These observations have critical implications about P – T genesis of both sanukitoid groups. Indeed, amphibole and phlogopite stability, but lack of garnet in equilibrium with high-Ti sanukitoids,

both imply pressures below 20 kbar and melting temperatures in the range 900–1100°C (Conceição & Green, 2004). On the other hand, equilibrium between low-Ti sanukitoids and garnet-bearing pyroxenites without hydrated phases imply higher pressures (> 25 kbar) and temperatures (1000–1200°C) (Rapp *et al.*, 1999, 2010) (Figure 4.11).

• **Nature of the metasomatic agent: does it have any influence?**

In order to test for the influence of different metasomatic agents than TTG, we carried out the same modeling but using either (1) a hydrous fluid (composition of Kessel *et al.*, 2005) or (2) a sediment-derived melt (composition similar as used in Section 5.4 of the Lithos paper). In the case of the hydrous fluid, only two-step hybridation has been modeled. Results are presented in Figure 4.12. They highlight two conclusions:

→ Melts derived from a fluid-metasomatized peridotite produces liquids that are close in composition to some low-Ti sanukitoids. Therefore, the latter could derive from two-step interactions, but only if the metasomatic agent is a fluid. However, the modeled liquids do not reproduce the overall geochemical variability of low-Ti sanukitoids, such that this process might not be very common and only concerns some samples such as the Bulai lamprophyres (see section 5.3 of the Lithos paper).

→ If the metasomatic agent is a felsic liquid, whatever its nature (TTG or sediment-derived), we obtain roughly similar results: in particular, two-step interactions always produce liquids richer in REE and HFSE and containing less transition elements than in the case of one-step hybridation. Only second-order variations are observed: modeled liquids are richer in Zr, Th and Nb and display higher Ba/Sr if the metasomatic melt derive from sediments. As a result, (1) the hybridation mechanism (one or two steps) exerts a first-order control on the geochemistry of sanukitoids (especially the distinction between low- and high-Ti) whereas (2) the nature of the metasomatic melt is only responsible for second-order variations.

Therefore, to connect with the modeling carried out by Martin *et al.* (2009), the discriminating parameter is not the F/a ratio, but only the value of a that controls the one of R_{mr} ($= a / [1-a]$).

• Implications for the physical nature of interactions

We propose that the difference between low- and high-Ti sanukitoids thus results from two distinct hybridation mechanisms, namely in one- or two steps, respectively. In natural systems, this distinction must rely on different propagation styles of the metasomatic melt (or fluid) (see Figure 4.13):

→ a high R_{mr} ratio (diagnostic of one-step processes) represents propagation in large conduits (0.1–10 m?), such that the ratio of volume to surface is very high for the felsic melt.

→ a low R_{mr} ratio (diagnostic of two-steps processes) represents percolation of the melt at grain boundaries or in very small veinlets (<0.1 m-wide), such that the ratio of volume to surface is extremely low.

Other parameters must obviously exert a control on the nature of interactions, such as temperature: if it is low, the melt would easily be consumed by reaction with peridotite, whereas high temperatures would sustain it above its solidus and thus favor one-step hybridation.

• Implications for geodynamic environment of sanukitoid genesis

As discussed in the Lithos paper (Section 6), interactions between felsic melts (or, occasionally, hydrous fluids) and mantle need that the source of these metasomatic agents must be located below a significant volume of peridotite. This geometry is easily achieved in subduction zones (Figure 4.14). Indeed, it provides a site for introduction of surface material within the mantle and, moreover, accounts for the presence of sedimentary material at depth. Indeed, genesis of some sanukitoids imply a sedimentary component (as proposed by Halla, 2005 ; Lobach-Zhuchenko *et al.*, 2008 ; the Lithos paper and the modeling presented above) that could hardly be introduced in the mantle owing to any other mechanism (such as delamination).

Nevertheless, it must be noted that this conclusion does not necessarily imply the existence of stable and long-lived subduction zones such as present-day ones. Indeed, as discussed in Chapter 2, sanukitoids represent relatively brief magmatic events, emplaced within shorter time ranges than modern calc-alkaline batholiths at convergent plate margins. These issues will be discussed further in Chapters 6 and 7.

Chapter 5

Petrogenesis of “marginal” sanukitoids: plutons of the Matok Group

In this Chapter, I present examples of the so-called “marginal sanukitoids” (introduced in Section 2.3.2) from the northernmost Kaapvaal craton, in South Africa, namely the Matok, Mashashane, Matlala and Moletsi plutons (referred together as the plutons of the Matok Group). Through the study of their petrogenesis, I will discuss:

- The genetic link between these rocks (especially the least differentiated end-member of the suite) and “true” sanukitoids, and their implication for the late-Archaean geodynamic changes (this is the aim of the paper presented in Section 5.2);
- How these “marginal” sanukitoids differentiate, in order to compare with “true” sanukitoid suites (such as the Bulai pluton) and provide insights into the origin of the “crustal” signature in granitoid rocks (this is the aim of the paper presented in Section 5.3).

5.1. Geological characterization of plutons of the Matok Group

As for the Bulai pluton in Chapter 4, this first section aims to present the geology, structures, mineralogy and chemistry of minerals that are typical of the Matok Group plutons. Once again, this Section is summarized in the following papers, especially in Section 5.2.4 and 5.3.3, such that I won't provide here any detailed synthesis of it. As for Section 4.1, I only list the Figures contained in Section 5.1 and the important information that they bring to the reader:

→ **Figure 5.1:** Geological sketch map of the Pietersburg block (can be found in papers of Section 3.3, 5.2 and 5.3).

→ **Figure 5.2:** Geological map of the Matok Group plutons, showing that the Matok pluton is made up of abundant intermediate rocks (diorites, granodiorites) whereas the Matlala, Mashashane and Moletsi plutons mostly consist in felsic lithologies (monzogranites). It also highlights that the Matok pluton is intrusive within the SMZ, whereas the other ones emplaced on the other side of the Hout River shear zone, in the Kaapvaal craton.

→ **Figure 5.3:** Major rock types in the Matok pluton: diorite (a), diorite enclaves within granodiorite (b), granodiorite (c,d), monzogranite (e), mingling relationships between diorite and granodiorite (f) and

deformation styles in granodiorite (g, h). Deformation only occurs locally, especially around the N'Tabalala and Matok shear zones (see Figure 5.9).

→ **Figure 5.4:** Major rock types in the Matlala pluton: pink monzogranite, typical of the southern part of the massif (a), equigranular grey monzogranites, typical of the northern part of the massif (b), mingling between two monzogranitic phases (c), pillowed enclave of monzogranite within granodiorite (d), dark MME and granodiorite enclaves in monzogranite (e, f), coarse-grained, angular enclave of amphibole-plagioclase-rich rock (g), xenolith of country rock (h).

→ **Figure 5.5:** Major rock types in the Mashashane and Moletsi plutons: mingling between two monzogranite phases at Moletsi (a), diorite enclaves within monzogranite at Moletsi (b), coarse-grained granodiorite with MME at Moletsi (c), porphyry-like monzogranite of Mashashane with small MME (d).

→ **Figure 5.6:** Petrography of rocks from the Matok Group plutons: pyroxenes, plagioclase and biotite in diorite at Matok (a), pyroxene-free, amphibole- and biotite-bearing granodiorite at Matok (b), zoned plagioclase in a granodiorite at Matok (c), amphibole + biotite + oxide + apatite mafic aggregate in a granodiorite at Moletsi (d), breakdown of amphibole into epidote + biotite assemblages at Moletsi (e), biotite + epidote + titanite mafic aggregate in a granodiorite at Matok (f), idem in a monzogranite at Matlala (g), fine-grained pseudomorph after amphibole : biotite + epidote + quartz assemblage in a monzogranite at Mashashane (h).

→ **Figure 5.7:** Composition of major minerals in rocks from the Matok Group plutons: (a) plagioclase, (b) pyroxenes, (c) alkali-calcic amphiboles, (d) calcic amphiboles, (e) biotite and (f) epidote. The composition of pyroxenes, amphiboles and biotites from the Bulai pluton are also shown: they systematically show higher Mg/Fe ratios than that of the Matok Group samples.

→ **Figure 5.8:** *P-T* conditions of crystallization of 20 samples (sorted by pluton) from the Matok Group plutons, on the basis of amphibole-plagioclase thermobarometry. Note that (1) samples of the Matok pluton crystallized at higher *P* and *T*, consistently with their structural position in granulites of the SMZ; (2) dashed lines depict the lowest pressure for the stability of epidote at given oxygen fugacity (Schmidt & Thompson, 1996); the presence of this mineral in the granodiorite and monzogranite samples imply very high fO_2 conditions at the time of emplacement, up to the HM buffer (further discussed in the papers).

5.2 and 5.3 : articles in preparation.

Chapter 6

Geodynamic evolution at the northern margin of the Kaapvaal craton

The aim of this chapter is to study the spatial and temporal evolution of granitoid magmatism in the Pietersburg block at the end of the Archaean, in order to build a consistent geodynamic model for its evolution. For this purpose, I'll present in more detail the nature and geochemistry of granitoids of the Pietersburg block that belong neither to "true", nor to "marginal" sanukitoid suites (Section 6.2). Secondly, I'll discuss the chronological evolution of petrogenetic processes and emplacement fashion in Section 6.3. Finally, in the light of this study and with the support of other geological evidence, I'll build a geodynamic model for the evolution of the Pietersburg block in the Meso- and Neoarchaean. In Section 6.5, this evolution is compared to a "modern" analogue, *i.e.* the Variscan crust of the French Massif Central, to discuss the differences and common features between both regimes.

6.1. Introduction

Not really interesting... It is just to provide a summary about all types of data used in this chapter: (1) field geology, (2) geochronology (already presented in Chapter 3, Section 3.3 – paper accepted to Prec. Res.), (3) whole-rock major, trace elements compositions as well as Sm–Nd isotopic data, from both our work and literature and (4) Lu–Hf isotopic data on separated zircons (work in progress in collaboration with Armin Zeh from the University of Frankfurt, Germany).

6.2. Granitoids of the Pietersburg block

Our field observations and sampling coupled to previous studies and geological maps reveal that granitoids outcropping in the Pietersburg block can be sorted into three groups:

- (1) TTGs;

- (2) Biotite-bearing granites, well represented in the Turfloop batholith such that I'll refer to them as "T-type" (for Turfloop-type) granites;
- (3) Ferroan, (pyroxene)-hornblende-biotite-epidote metaluminous granitoids that were already presented in the papers of Chapter 5. Because of the name of the plutons (Mashashane, Matlala, Matok, Moletsi), I'll refer to them as "M-type" granitoids.

The spatial distribution of the three different groups is shown in the map of Figure 6.1. All samples with geochemical information used in this Chapter are shown.

6.2.1. TTGs

TTGs cover the wider surface and belong to the Goudplaats–Hout River and Groot Letaba–Duiwelskloof gneiss units (see the geological setting and discussion of Section 3.3). They less frequently appear as xenoliths in younger plutons. Some of the TTG consist in well-defined intrusion, in particular around the Murchison greenstone belt (Maranda, Baderoukwe plutons). Finally, some are intimately associated with biotite granites in Turfloop batholith.

These rocks are always banded, grey orthogneisses, variously migmatized, displaying a systematic subvertical foliation (Figure 6.2a,b,c). They are sometimes rather homogeneous (Figure 6.2b), but in places, up to 6 different generations can be identified (Figure 6.2c), even if the post-emplacement deformation make the study of their relationships very difficult.

In terms of normative composition, TTGs of the Pietersburg block are trondhjemites (Figure 6.3) that are rich in quartz and plagioclase feldspar (An_{25-30} ; Bohlander, 1991). Both minerals are stretched along the foliation (Figure 6.2d) together with biotite that is the unique mafic mineral. In the SMZ, they also contain orthopyroxene, but it is of metamorphic origin and developed during granulite-facies high-temperature overprint (Bohlander *et al.*, 1992 ; Barton *et al.*, 1992 ; Stevens & van Reenen, 1992). Accessory minerals are magnetite, apatite, epidote (often as allanite) and zircon.

6.2.2. T-type granites

These granites principally outcrop in the Turfloop and Lekkersmaak batholiths (Figure 6.1), and as small intrusions, bodies, sheets and dykes within the TTGs (Robb *et al.*, 2006; see also Section 3.3 and Figure 6.4d) as well as xenoliths within younger plutons (Figure 6.4b).

These rocks sharply contrast with the TTG: they are much more homogeneous at the outcrop scale, and from an outcrop to another. They consist in equigranular, medium-grained (0.1–0.5 cm) granites (Figure 6.4a), locally displaying a layering of magmatic origin (Figure 6.4c) or rarely porphyritic textures. In general, they are not deformed, except at the edges of the large intrusions and in the vicinity of regional scale structures (*e.g.* large shear zones).

The modal composition of T-type granites is also different to that of TTG: K-feldspar is much more abundant and appears in equal or greater proportions than plagioclase, leading to clearly granitic compositions (Figure 6.5). The unique mafic mineral is biotite, and it is associated with the accessory phases: ilmenite, magnetite, epidote, zircon. Primary muscovite is a quite common feature in the Lekkersmaak pluton (Jaguin *et al.*, 2012) and occurs occasionally in Turfloop and other small intrusions.

6.2.3. *M-type granitoids*

M-type granitoids form the Mashashane, Matlala, Matok and Moletsi plutons that were extensively described and studied in Chapter 5, so that they will not be present in any further detail here.

Some important features must be noted:

- The ~2700 Ma-old (Poujol, 2001 ; Zeh *et al.*, 2009) Mashishimale pluton in the southern edge of the Murchison greenstone belt also belongs to this group (Figure 6.1).
- They are always intrusive in other granitoid types.
- It is the only granitoid type that is associated with significant volumes of intermediate to mafic rocks (granodiorites and diorites).
- They are undeformed, except along shear zones in the case of the Matok.
- They consist in calc-alkaline granitoids covering a wide range of modal compositions (Figure 6.6), bearing biotite, amphibole and pyroxene as mafic minerals, in different proportions depending of the level of silica saturation (see Chapter 5). Epidote and titanite are ubiquitous accessory minerals together with zircon, apatite and Fe–Ti oxides.

6.2.4. *Geochemical comparison between granitoid groups*

Data presented in this Section (detailed in Appendix 4) are from our work as well as already published studies (Kreissig *et al.*, 2000; Kröner *et al.*, 2000; Henderson *et al.*, 2000).

In terms of major-element geochemistry, the M-type granitoids cover a wider range of SiO₂ contents than TTGs and T-type granites that are restricted to SiO₂ contents above 65 and 69 wt.% (respectively) (Figure 6.7a,b). At given SiO₂, TTGs are richer in Na₂O, CaO and Al₂O₃ (Figure 6.7b,c) while their K₂O contents are twice to three times below that of T- and M-type granites (Figure 6.7a). In addition, TTG and T-type granites are slightly peraluminous, whereas M-type are clearly metaluminous (Figure 6.7d). As a result, Al saturation level and the alkali ratio are good discriminating factors between all granitoid groups (Figure 6.7d). Additionally, the M-type granitoids are much richer in FeO_t, MgO, TiO₂ and P₂O₅ than the others at a given SiO₂ (Figure 6.8a) and show mostly ferroan affinities in the classification of Frost *et al.* (2001) while TTGs and T-type granites are magnesian (Figure 6.8b).

The three granitoid groups are also distinct on the basis of trace element contents: TTG are richer in Sr and poorer in Ba than other types (Figure 6.9a). They also contain less Rb at given SiO₂ (Figure 6.9b). The M-type granitoids distinguish themselves by very high concentrations of REE and HFSE compared with the TTG and T-type granites (Figure 6.9c), and greater contents of transition elements (Figure 6.9d). Multi-element patterns are typical of continental material: they are fractionated with Nb–Ta and Ti troughs as well as positive a Pb anomaly. that of TTG display no negative Eu anomaly and a positive Sr one, whereas the T- and M-type granitoids are characterized by negative Eu and Sr anomalies (Figure 6.10). On the other hand, the TTG and T-type granites show moderately to highly fractionated REE patterns ($La_N/Yb_N = 10\text{--}50$), whereas M-type granitoids present homogeneous and moderate REE fractionation ($La_N/Yb_N \sim 10$; Figure 6.10). These differences between all granitoid types are highlighted in discriminating diagrams of Figure 6.11.

6.3. Evolution of the magmatic record

6.3.1. Geochronology

Geochronological data published so far in the Pietersburg block were already synthesized in the article accepted in prec. Res. (Section 3.3, see notable Figure 3.19). Five main magmatic pulses can be identified: (1) 3250–3350 Ma; (2) 2950–2970 Ma; (3) 2830–2850 Ma; (4) ~2780 Ma and (5) 2670–2700 Ma. The timeline of Figure 6.12 links this purely

geochronological synthesis with the granitoid typology adopted in this Chapter. The numbers in the boxes relate to the number of samples dated in the corresponding time range, and the hatched fields represent emplacement ages that are likely, on the basis of (1) inherited cores for TTG and (2) the need for a mixing end-member similar in composition to T-type granites in the genesis of M-type ones (see the article of Section 5.3).

To summarize the data presented in Figure 6.12, it appears that TTG emplace in successive, discrete magmatic events over more than 500 Ma between 3350 and 2780 Ma, whereas the T- and M-type granitoids successively emplaced within 150 Ma only, from ~2850 to ~2700 Ma.

Two important observations must be taken into consideration:

- (1) T-type granites of the Turfloop batholith, aged of 2780 Ma, are likely associated with coeval TTG. Indeed, as shown in Figure 6.13a, samples from this intrusion split into two groups, and samples of each group fall either in the TTG or the T-type granite fields. This unlikely result from sampling bias, as data from both this study and Henderson *et al.* (2000) show this dichotomy. Furthermore, the coeval association of these two granitoid types is revealed by the Rb–Sr whole-rock isochron drawn by Henderson *et al.* (2000) using samples from both groups (Figure 6.13b).

- (2) The magmatic event represented by M-type granitoids is likely much more widespread than previously thought. Indeed, as demonstrated in Sections 5.2 and 5.3, the mafic and intermediate rocks of the plutons are similar in composition to coeval lavas of the Platberg group, Ventersdorp supergroup, which cover more than $3 \cdot 10^5$ km² in central and northwestern South Africa.

6.3.2. Evolution of sources and petrogenetic processes

The origin of M-type granitoid has been extensively studied in Chapter 5: the felsic end-member of this magmatic suite derive from fractional crystallization, with minor assimilation of T-type granites, of intermediate to mafic melts, which themselves result from mixing of two magmas: dry, Fe-rich melts derived from mafic lower crust and sanukitoid magmas which source is the enriched mantle.

The petrogenesis of the other two granitoid groups can be assessed on the basis of their major- and trace element compositions:

- TTG of the Pietersburg block overlap in composition with medium- to high-pressure TTG of Moyen (2011) in Figure 6.14 (and are also relatively similar to low-HREE TTG of Halla *et al.*, 2009). Such magmas derive from partial melting of an amphibolitic source at pressures above 15 kbar (*e.g.* Martin, 1986, 1993; Rapp *et al.*, 1991 ; Smithies, 2000 ; Martin *et al.*, 2005 ; Halla *et al.*, 2009 ; Moyen, 2011). This is confirmed by major-element compositions of the Pietersburg TTG, which fit with that of experimental melts derived from low-K basaltic rocks (Figure 6.15), as well as trace-element modeling: the patterns of TTG are adequately reproduced by modeled melts derived either from high- (“model 1”) or medium-pressure (“model 2”) melting of amphibolites similar in composition to those of the Pietersburg block (Figure 6.16).

- On the other hand, the T-type granites are similar in composition with “potassic granites” in the classification of Moyen (2011), as shown in Figure 6.14. The origin of such magmas is classically related to melting of older TTG gneisses (*e.g.* Sylvester, 1994 ; Moyen *et al.*, 2003 ; Whalen *et al.*, 2004 ; Watkins *et al.*, 2007 ; Moyen, 2011). This is confirmed by their similarity, in terms of major-element composition, with experimental melts derived from tonalitic rocks analogue to Archaean TTG (Figure 6.15). Furthermore, trace-element modeling using phase relationships of the experimental study by Watkins *et al.* (2007) and considering the compositional range of Pietersburg TTG as the source, computes liquids which composition match that of the T-type granites (Figure 6.16).

These results are consistent with Sm–Nd and Lu–Hf data (Figure 6.17). Indeed, TTG are juvenile rocks (with positive ϵ_{Nd} and ϵ_{Hf}), indicating that they derive from melting of mafic rocks shortly after their extraction from the mantle. T-type granites display less radiogenic compositions in both Nd and Hf, but still positive (ϵ_{Hf}) or slightly negative (ϵ_{Nd}), showing that their source is represented by young crustal material such as the TTGs. Finally, M-type granitoids display homogeneously negative ϵ_{Nd} and ϵ_{Hf} , implying that their genesis involved significant amounts of ancient crust. The latter is represented by a sedimentary component responsible for mantle enrichment ultimately linked with the petrogenesis of all these granitoids (see articles of Section 5.2 and 5.3). Interestingly, all granitoids from the Pietersburg block, whatever the group into which they belong, fall within a unique crustal evolution trend for both Nd and Hf (grey arrows in Figure 6.17). This demonstrates that the Pietersburg block represents a portion of continental crust that was extracted from the mantle

at 3000–3200 Ma, and then successively reworked either by direct intracrustal melting (genesis of T-type granites) or coupled to introduction of sedimentary material within the mantle (genesis of M-type granitoids).

To summarize and conclude, the study of granitoids from the Pietersburg block shows the following evolution of sources and petrogenetic processes:

- (1) Moderate- to high-pressure melting of mafic rocks (amphibolites) homogeneously gave rise to TTG within a large time span between ~3350 and ~2780 Ma.
- (2) T-type granites result from intracrustal anatexis of the older TTGs;
- (3) M-type granites result from the interactions between crust-derived melts (T-type granites as well as mafic lower crust-derived liquids) with ones derived from enriched mantle.

From a global point of view, this indicates that generation of juvenile continental crust (TTG) dominates the magmatic record during >500 Ma, whereas after ~2800 Ma, recycling of this crust prevails, either through direct intracrustal differentiation (T-type) or contribution of detrital material hybridized with the mantle (M-type).

6.3.3. *Towards a dominant structural control?*

The map of Figure 6.18 presents the different granitoids types, distinguished from each other on the basis of both typology and geochronology.

This map reveals that:

- TTG and T-type granites emplaced before 2800 Ma emplaced throughout the Pietersburg block, from the SMZ to the North to the Murchison belt to the South. As a result, their emplacement pattern is geographically random and was not likely controlled by any structural discontinuity.

- T-type granites at 2780 Ma emplaced exclusively in the vicinity of the Thabazimbi–Murchison Lineament (Lekkersmaak pluton) and its satellite, the Kudu’s River Lineament (Turfloop batholith and, further east, associated small intrusions within the TTG). Importantly, the TML also accommodated the emplacement of the Gaborone Granite Suite, that emplaced at the same time (*e.g.* Grobler & Walraven, 1993 ; Zeh *et al.*, 2009) but ~350 km to the West of our study area (Figure 6.19). The TML was likely active at ~2780 Ma (Good & de Wit, 1997), such that the associated magmatic event was strongly controlled by tectonic activity along the TML and its satellites. These lineaments are parallel to, and

associated with greenstone belts, suggesting that they represent sutures zones corresponding to the closure of former volcano-sedimentary basins.

- M-type granitoids emplaced at ~2700 Ma also intruded around a major crust-scale structure, namely the Hout River Shear Zone (HRSZ). Such as T-type granites for the TML, their emplacement is coeval to tectonic activity along the HRSZ that started at ~2700 Ma (Kreissig *et al.*, 2001) to accommodate exhumation of the SMZ granulites.

To summarize, the structural context of granitoid emplacement significantly evolved in the Pietersburg block at ~2800 Ma. No structural control was exerted before that time, whereas subsequently, granitoids emplaced along major, active tectonic structures likely representing terrane boundaries or major crustal discontinuities.

6.4. A global geodynamic model

The model developed in this section integrates all data presented in the previous ones, and also relies on further geological evidence from the literature. It is basically the same as synthesized in Section 5.2.7.1 at the end of the first paper of Chapter 5; the French description of this model is more detailed, as it is supported by exhaustive literature review and better constrained by the previous study of magmatic evolution in the Pietersburg block, presented in Sections 6.2 and 6.3. Consequently, I do not provide another English summary here, as it is possible to follow the different steps of the geodynamic model simply considering Figures 6.21 to 6.25 that are presented in the chronological order.

It must be noted that this model has to be better constrained by further studies, and the interpretations presented in this section are opened to discussion.

An important feature of this model has to be taken into consideration: at ~2800 Ma, there is a dramatic change of geodynamic regimes. Before that time, emplacement of juvenile TTG prevails over melting of older crust; tectonic and deformation styles are also typical of Archaean geodynamics. By contrast, the granitoid record after 2800 Ma is dominated by recycling products (T- and M-type granitoids) and tectonic processes that are closer to present-day ones (focusing of deformation along large-scale structures such as thrusts and shear zones; subduction-like evolution; collision; sediment deposition in large basins...).

6.5. Comparison with a “modern” analogue: the French Massif Central (FMC)

In this Section, I compared the geodynamic evolution of the Pietersburg block in the late-Archaean with that of the FMC in the Paleozoic. The goal of this comparison is to highlight the commons and differences between both regimes and discuss their origin. The FMC was chosen for his good geological characterization and deep level of erosion that allow good exposure and thus, good knowledge of granitoid rocks.

6.5.1. Evolution of the FMC: a synopsis

A general and simplified geological map of the FMC is provided in Figure 6.26.

The goal of this section is not to extensively describe the geology of the FMC, but rather synthesize the succession of geological events that led to its present-day structure (Figure 6.27). It is based on published reviews (Matte, 1986 ; Lardeaux *et al.*, 2001 ; Ledru *et al.*, 2001 ; Faure *et al.*, 2009 ; Berger *et al.*, 2010 ; Melleton *et al.*, 2010).

- **Ordovician** (450–500 Ma): opening of oceanic basin, associated mafic magmatism and deposition of detrital sequences.
- **Silurian** (400–450 Ma): convergence and subduction (D_0), as recorded by remnants of high-pressure metamorphic rocks of that age.
- **Devonian to lower Carboniferous** (340–390 Ma): continent-continent collision characterized by nappe stacking (D_1 and D_2 thrusts) and associated migmatization.
- **Middle Carboniferous** (320–340 Ma): transition from convergent to divergent tectonics (D_3): thrusting occurs in the external parts, whereas extension occurs in the core.
- **Upper Carboniferous** (320–290 Ma): general extension, massive anatexis of middle to lower crustal segments (development of the Velay anatectic complex) and emplacement of huge volume of granitoids as well as small intra-continental, coal-bearing basins.
- **Permian** (290–250 Ma): emplacement of post-orogenic granites and large sedimentary basins.

In order to compare the granitoids of the Pietersburg block with those of the FMC, I describe the latter in more detail. On the basis of unpublished geochemical data (compiled by J.F. Moyen and collaborators), synthesized in Figure 6.28, those granitoids can be classified into three groups:

- Peraluminous, either biotite-cordierite-bearing or two mica-bearing S-type granitoids, derived from dehydration or water-present melting of metasediments, respectively.
- Subalkaline (*i.e.* calc-alkaline) monzogranites and granodiorites, generally amphibole- and biotite-bearing, close to I-type granites in terms of petrography but also bearing a peraluminous signature.
- Intermediate to mafic calc-alkaline rocks that are both magnesian (high mg#) and rich in K₂O (2–6 wt.%), locally referred to as “vaugnerites”. They are notably rich in incompatible elements, even more than the two previous granitoid types.

Most of these magmas emplaced during extension and late-orogenic collapse in the Middle to Upper Carboniferous (340–290 Ma), but some of them locally intruded at ~400 and ~350 Ma.

6.5.2. *Common features between Pietersburg block and FMC*

The sequence of tectonic and magmatic events is very similar in both areas as shown in Figure 6.29. In particular, the successive order and the respective absolute duration of each phase are surprisingly similar between the Pietersburg block and the FMC. Main magmatic events occurred at the time within this sequence, *i.e.* at the transition between subduction in collision in one hand, and collision and exhumation on the other hand. Moreover, the same sequence of tectonic regimes is observed in both cases, starting with convergent tectonics, then giving way to extension and eventually to strike-slip faulting. Finally, each orogenic event ends by a period of crust stabilization, associated to intra-continental sedimentation dominated by detrital sequences.

Secondly, the magmatic record is also similar in terms of petrogenetic processes, which is fundamental for our understanding of the evolution of crustal growth processes. Indeed, both in the Pietersburg block and the FMC, the granitoids emplaced during the subduction-collision cycle are characterized by two distinct petrogenetic processes (Figure 6.30):

- (1) Intracrustal differentiation, *i.e.* melting of pre-existing crustal lithologies: peraluminous granites in the FMC, T-type granites in the Pietersburg block. The low Fe and Mg contents of these rocks attest of their crustal origin (Figure 6.30).
- (2) Melting of hybrid mantle sources, previously enriched in incompatible elements during subduction event prior to collision. Those magmas are represented by vaugnerites in

the FMC that are both rich in Fe+Mg and incompatible elements (hatched fields in Figure 6.30). They are not directly represented in the Pietersburg block, but the M-type granitoids attest of their presence, as they represent mixtures between such magmas and a crustal end-member. Their analogues in the FMC might be represented by the subalkaline granites (red fields in Figure 6.30).

Moreover, in both Archaean and modern orogenic settings, the granites derived from intracrustal differentiation formed throughout the collision process, whereas those implying the involvement of enriched mantle emplaced specifically at the end of it.

To summarize, the late-Archaean (<2800 Ma) evolution Pietersburg block and that of the FMC are both characterized by a typical sequence of geological events, and similar petrogenetic processes involved in granitoid petrogenesis. This shows that the tectonic regime responsible for the late-Archaean evolution of the Pietersburg block is closer to “modern-style” geodynamic processes than typical “Archaean” ones.

6.5.3. Differences between Pietersburg block and FMC

Although the geological sequences and magmatic record are similar between the Pietersburg block and the FMC, some major differences exist:

- High-pressure metamorphic relicts lack in the Pietersburg block (Figure 6.29). This could result for a preservation bias, or inefficient exhumation processes for such rocks during Archaean times. In addition, granulite-facies metamorphism took place in both settings, but at different times: it occurred at early stages of the collision in the FMC, whereas it seems to be linked with exhumation in the Pietersburg block.

- The magmatic episode that features the onset of collision is represented by much more granitoids, in terms of volume, in the Pietersburg block than in the FMC. This possibly results from higher geothermal gradients during the Archaean: indeed, the lower crust likely reached higher temperatures in the Pietersburg block (>800°C) than in the FMC (no more than 800°C) such that the melting rates, and correlatively the amount of granitoid melts, was necessarily greater.

- Finally, the crust-derived granitoids show different compositions in both areas. Specifically, they are slightly peraluminous biotite granites (T-type) in the Pietersburg block, whereas they consist in strongly peraluminous, biotite-cordierite or two-mica granites in the

FMC (Figure 6.31²). This is easily accounted for by the difference in the composition of the upper crust between the late-Archaeon (mostly TTG) and the Paleozoic (mostly metasedimentary rocks). Even if Paleozoic crust certainly contains high volumes of metaigneous rocks, the lower thermal regimes at that time (compared with Archaean settings) did not allow their partial melting and significant contribution to the genesis of granitoid rocks.

6.5.4. Summary

The late-Archaeon (<2800 Ma) evolution of the Pietersburg block is much closer to modern-style, subduction-collision orogenic events than typically Archaean tectonics, concerning (1) the sequence of events; (2) the relative and absolute duration of these events and (3) the magmatic record, especially the petrogenetic processes involved in the origin of granitoid rocks. Observed differences would largely result from greater thermal regimes in the Archaean.

² In the left bottom apex of the triangle in Figure 6.31, FM = FeO + MgO (wt.%) and SB = Sr + Ba (ppm).

Chapter 7

Magmatism and geodynamic changes at the Archaean-Proterozoic Transition

This Chapter aims to (1) summarize the results and interpretations presented in this PhD thesis, (2) understand the evolution and significance of granitoid magmatism, at the planetary scale, for the late-Archaean geodynamic changes and (3) assess why these changes occurred at that time of Earth's history. Finally, I will propose a global perspective of the secular evolution of crustal growth mechanisms.

7.1. Nature of geodynamic changes at the Archaean-Proterozoic Transition

7.1.1. Late-Archaean magmatism: a synthesis

A synthetic typology of late-Archaean magmatism (Figure 7.1) can be addressed as follows:

- “True” sanukitoids (or sanukitoids *s.s.*) include (1) low-Ti sanukitoids; (2) high-Ti sanukitoids and (3) the sanukitoid suite. They respectively derive from (1) one-step hybridation of a felsic melt with mantle peridotite; (2) melting of a metasomatic amphibole-, phlogopite-bearing and orthopyroxene-rich peridotite or pyroxenite, produced by interactions between felsic melts (possibly aqueous fluids, in some cases) and pristine peridotite; (3) differentiation of the latter two end-member by fractional crystallization (less likely partial melting) at shallow, crustal levels.

As a result, “true” sanukitoids primarily derive from interactions between mantle peridotite and a component rich in incompatible elements. Such petrogenetic model is very close to that of modern arc magmas. However, in the case of sanukitoids, the metasomatic agent is more often a felsic melt (TTG, or derived from terrigenous sediments) whereas, in the case of arc magmas, it is dominated by fluid phases.

- Crust-derived granites, *i.e.* derived from melting of any older crustal lithology such as TTG (more often), metasediments or metaigneous mafic rocks.
- “Marginal” sanukitoids, which result from interactions between “true” sanukitoids and the latter crust-derived liquids at shallow, crustal levels.

7.1.2. A diachronous, but systematic evolution

A careful examination of the late-Archaean magmatic record in every craton worldwide (initiated by Heilimo *et al.*, 2011 and further explored here) shows that the evolution of this magmatism follows a very systematic sequence. As a general observation, the evolution of late-Archaean magmatism is organized in two distinct phases: (1) the **first phase** lasts over more than 200 Ma and up to 500 Ma. It homogeneously consists in emplacement of TTG and, locally, of their melting products; (2) the **second phase** is much more short-lived (20–120 Ma) and is marked by considerable diversification of petrogenetic processes at the origin of granitoid magmas. Indeed, this event is characterized by the emplacement of all granitoid types mentioned in Section 7.1.1 (“true” and “marginal” sanukitoids, as well as “crustal” granites and not only derived from TTG). In some cratons (Superior Province, Baltic and Amazonian Shields, Dharwar craton), the second event follows itself a typical evolution: (1) low-Ti sanukitoids firstly emplace, followed within a short time by (2) high-Ti and/or “marginal” sanukitoids as well as anatectic granites.

The most representative examples of this sequence are summarized in Figure 7.2. The detailed magmatic evolution of each late-Archaean province is not reported in this summary but is part of the French text; to synthesize, such an evolution can be observed in the following cratons (presented in the same order as in the French text, such that references can be found in the latter):

- Superior Province, Canada (see also Figure 7.2);
- Baltic Shield, Finland–Russia (see also Figure 7.2);
- Dharwar craton, India (see also Figure 7.2);
- Pilbara craton, Australia (see also Figure 7.2);
- North China craton (see also Figure 7.2);
- Amazonian Shield, Brazil (see also Figure 7.2);

- Zimbabwe and Kaapvaal cratons, South Africa, Zimbabwe, Botswana (see also Figure 7.2);

- North Atlantic craton;
- Yilgarn craton, Australia;
- Antogil craton, Madagascar.

In the details, the magma types emplaced during the second phase, and their respective chronology, can be slightly different from a craton to another. On the other hand, the respective duration of phases 1 and 2 are similar in different places. Furthermore, such evolution took place within the same time range at the planetary scale (3000–2500 Ma). As a result, it appears that the geodynamic changes at the Archaean-Proterozoic transition were controlled by both local and global parameters.

The origin of such evolution is discussed in the following section.

7.1.3. Initiation of modern-style subduction-collision cycles

As suggested by previous studies (*e.g.* Smithies & Champion, 2000 ; Moyen *et al.*, 2001 ; Whalen *et al.*, 2004 ; Käpyaho *et al.*, 2006 ; Percival *et al.*, 2006 ; Halla *et al.*, 2009), we propose that the evolution of granitoid magmatism at the end of the Archaean reflects the initiation of subduction and collision cycles similar in many aspects to present-day ones. This assumption is supported by several lines of evidence:

- (1) Whether Archaean TTG are generated or not in subduction regimes is still a matter of debate. However, the origin of “true” and “marginal” sanukitoids unequivocally points to such a geodynamic setting. Indeed, their petrogenesis involves the interaction between mantle peridotite and felsic melts (or aqueous fluids) either derived from metabasalts (*i.e.* TTG) or metasedimentary material. The easiest way to introduce the latter into the mantle is clearly subduction, rather than delamination of lower crust that would only affect highly refractory, dry and unlikely supracrustal material.

- (2) In some cratons, such as the Baltic Shield, sanukitoids outcrop over large surfaces and are organized along linear structures up to 500 km-long (Figure 7.3), which recalls the present-day magmatic “arcs” that develop at convergent plate margins.

- (3) “True” sanukitoids are mostly juvenile rocks with ϵ_{Nd} in the range -1 to $+4$ (Figure 7.4). This implies that mantle enrichment and mantle melting occurred within a short

time span, such that the hybrid component did not evolve towards less radiogenic values. Such an observation is compatible with mantle enrichment during subduction, subsequently and shortly followed by magma genesis and emplacement during collision, within an unique geodynamic cycle.

- (4) In almost all cratons, the emplacement of a variety of granitoids in the late-Archaean is coeval with a major tectono-metamorphic event, likely related to collision (high-temperature and moderate- to low-pressure conditions).

- (5) The duration of each magmatic phase (as defined in Section 7.1.2, *i.e.* emplacement of TTG and subsequent genesis of “true”, “marginal” sanukitoids and anatectic granites) is consistent with those of present-day, subduction and collision events, respectively.

- (6) From this perspective, TTG are generated by melting of subducted slab during the first phase, while low-Ti sanukitoids emplaced at the transition between subduction and collision and finally, high-Ti, “marginal” sanukitoids as well as crust-derived granites mark the end of collision and onset of post-orogenic exhumation (Figure 7.5). Such a sequential emplacement of different granitoid types is recorded in many cratons (*i.e.* Superior Province, Baltic Shield, Dharwar craton). It is not reproduced in some other terranes, such as the Kaapvaal craton (that lacks “true” sanukitoids – see Chapter 6), but this might only result from the local geometry of subduction-collision as well as local characteristics of the continental crust.

- (7) Late-Archaean granitoids are basically derived from the interplay of two different petrogenetic processes: (1) contribution of hybrid mantle sources and (2) melting of pre-existing continental crust. Such an association is also very typical of present-day, late- to post-collisional settings (*e.g.* Bonin, 2004), as pointed out in Section 6.5 (comparison between Pietersburg block and the Paleozoic, French Massif Central). Hence, each type of late-Archaean granitoids has its own counterparts in post-Archaean magmatic record in late-collisional contexts: (1) for instance, “true” sanukitoids compare with Mg-K magmas of the Western Alps, Corsica (Debon & Lemmet, 1999; Bonin, 2004) and Central Europe (Janousek *et al.*, 2004; Slaby and Martin, 2008; Parat *et al.*, 2010) as well as Ba-Sr-rich granitoids of the Caledonian Belt (Fowler & Rollinson, 2012), etc.; (2) differentiation products of “true” sanukitoids are similar to very common high-K calc-alkaline granodiorites and granites (see Section 4.3) while (3) “marginal” sanukitoids resemble a wide variety of post-Archaean

granitoids, including the “oxidized ferroan granites” (Eby, 1992; Dall’Agnol & Oliveira, 2007); (4) widespread peraluminous S-types granites are equivalents of late-Archaean anatectic granites. They are different in terms of chemical compositions because the dominant crustal lithologies are not the same in late-Archaean (TTG) and post-Archaean (metasedimentary rocks) settings.

To conclude, all these arguments support that the late-Archaean geodynamic changes reflect the initiation of subduction-collision cycles and, by extension, the onset of modern-style plate tectonic processes.

7.2. Consequences for crustal evolution

7.2.1. Formation of the first supercontinents?

Plate tectonics processes and the Wilson cycle imply that continental blocks constantly derive, and their periodical welding gives rise to large supercontinents. Evidence for past supercontinents are unequivocal since ~1800 Ma (with the earliest well-defined one, referred to as “Nuna” or “Columbia”), but the age of the older ones are poorly constrained because of the lack of data before ~2000 Ma. The first supercontinents could be as old as 3600 Ma (“Vaalbaara”; Zegers *et al.*, 2005), or possibly 3000 Ma (“Ur”; Rogers & Santhosh, 2003) and 2700 – 2500 Ma (“Kenorland” of Williams *et al.*, 1991; “Superia” and “Sclavia” of Bleeker, 2003).

The formation of supercontinents relies on that of modern-style tectonic processes. Because we proposed that the latter started between 3000 and 2500 Ma, as recorded by the granitoid evolution, it seems that the first supercontinents welded during this range of time. From that perspective, the late-Archaean emplacement of a diversity of granitoids (“true” and “marginal” sanukitoids + anatectic granites) within each craton marks the assembly of a supercontinent. Applying this theory, we observe that the ages of these late-Archaean magmatic associations group into three clusters (Figure 7.6):

- 2950–2850 Ma (Amazonian and Ukrainian Shields, Pilbara craton);
- 2750–2625 Ma (Baltic Shield, Superior, Wyoming provinces, Zimbabwe, Slave and Kalahari cratons);
- 2600–2500 Ma (Dharwar, Antogil and North China craton).

This favors the assembly of at least three supercontinents between 3000 and 2500 Ma, as previously suggested by Bleeker (2003). Such an assumption must be tested by detailed chronostratigraphic studies to confirm the correlations between the concerned cratons.

7.2.2. *Evolution of the crustal growth rate*

Archaean TTG are principally juvenile rocks, as they directly derive from young metabasaltic material either by partial melting (*e.g.* Martin, 1986, 1994; Atherton & Petford, 1993; Smithies, 2000; Rapp *et al.*, 2003; Smithies *et al.*, 2005; Bédard, 2006) or fractional crystallization (*e.g.* Kamber *et al.*, 2002; Kleinhanns *et al.*, 2003). As they form up to 65% of Archaean continental crust (*e.g.* Moyen, 2011), it is classically considered that the Archaean represents a fundamental period of crustal growth and extraction from the mantle. Recycling processes certainly occurred at that time (*e.g.* Friend & Nutman, 2005; Sanchez-Garrido *et al.*, 2011) but were clearly subordinate compared with juvenile, TTG magmatism.

By contrast, the late-Archaean granitoids studied in this work principally evidence crustal recycling, at two different levels: (1) by classical intracrustal differentiation, as recorded by purely crust-derived anatectic granites; and (2) by introduction of continental material (sediments) within the mantle, which partly contributes to mantle enrichment and subsequent genesis of hybrid, mafic to intermediate melts (*i.e.* “true” and “marginal” sanukitoids). The concept of this duality in recycling processes is summarized in Figure 7.7. This late-Archaean magmatism is clearly a widespread manifestation (see Section 7.1.2) and in each craton, its products are as important as late-Archaean (3000–2500 Ma) TTG in terms of volume. As a result, the Archaean-Proterozoic transition represents the time when recycling processes became as important as juvenile processes during the secular evolution of the continental crust.

This conclusion is in good agreement with studies of the secular evolution of the crustal growth rate (Figure 7.8; references in the caption). Indeed, most curves show an important break in the range 3000 to 2500 Ma, generally interpreted as reflecting the transition from Archaean to modern tectonic processes and the onset of subduction regimes on Earth (Dhuime *et al.*, 2011). This is very consistent with our interpretation of the late-Archaean magmatic record at the global scale, as it would result from initiation of subduction-collision cycles.

7.2.3. Role in “cratonization” processes

Late-Archaean magmatism (and other geological events) is followed by a period of crust stabilization, as witnessed by a major magmatic shutdown that lasted more than 200 Ma (*e.g.* Condie *et al.*, 2009). This is consistent with our interpretation of the late-Archaean evolution of granitoids, as subduction and collision cycles are always followed by a period of crust stabilization. On the other hand, the addition to large volumes of juvenile material at the end of the Archaean (*i.e.* TTG during stage 1, then sanukitoids during stage 2) likely contributed to a major episode of crustal growth at that time. As a result, the volume of continental crust significantly increased during this period, which favored its differentiation in one hand, and stabilization on the other hand. Crustal differentiation also occurred through the emplacement and evolution of sanukitoid suites (see Section 4.3). All these parameters led to the stabilization, by the end of the Archaean, of large, relatively thick (~30 km) and buoyant continental blocks, now referred to as “cratons”.

On the other hand, these cratons also bear a thick (up to 300 km) lithospheric keel characterized by cold, dense and refractory peridotite. This keel formed throughout the Archaean by melting and removal of the liquids (ultimately leading to continental crust formation) as revealed by the highly refractory modal and chemical composition of cratonic peridotites, brought up as xenoliths by kimberlites (*e.g.* Ionov *et al.*, 2010). On the other hand, these peridotites display anomalously high orthopyroxene contents (Figure 7.9a), and trace-element contents in their minerals are typical of metasomatic processes (*e.g.* Grégoire *et al.*, 2003 ; Ionov *et al.*, 2010). These observations, coupled to Re–Os clustering around ~2500 Ma (*e.g.* Carlson *et al.*, 2005), argues in favor of a large-scale metasomatic event in the late-Archaean which could be related to the initiation of subduction processes and genesis of sanukitoids. Consistently, experimental studies highlighted that the hybrid mantle left after sanukitoid genesis contains ortho-, clinopyroxene and garnet which major- and trace element compositions are similar to that in natural cratonic mantle xenoliths (Figure 7.9b ; Rapp *et al.*, 2010).

7.3. The origin of geodynamic changes

In this Section, I propose some reasons for the occurrence of geodynamic changes specifically at the Archaean-Proterozoic transition.

7.3.1. *Thinning of oceanic plates*

Because the Archaean mantle was hotter than today (see Section 1.2), and because melt productivity of the mantle is a linear function of its temperature (*e.g.* Langmuir *et al.*, 1992), it is classically considered that Archaean oceanic crust was thicker than today to accommodate for this higher productivity (*e.g.* Bickle, 1986 ; Foley *et al.*, 2003). From a mechanical point of view, it is much more difficult to introduce into the mantle a thick oceanic lithosphere than another one of similar composition, but thinner (Abbott *et al.*, 1994). Through the study of present-day cases, Abbott *et al.* (1994) demonstrated that if the oceanic crust is, in average, thicker than 10 km, it will not be able to “subduct” and will be tectonically “subcreted” (“flat” subduction ; Figure 7.10). Based on the thermal evolution of the mantle, Abbott *et al.* (1994) predicted that before 3000 Ma, >99% of oceanic crust on Earth was thicker than 10 km such that classical, present-day subduction was impossible (Figure 7.10). By contrast, between 3000 and 2000 Ma, the oceanic crust progressively thinned because of the global cooling of the mantle, such that >50% of it was thin enough to undergo “classical” subduction (Figure 7.10). This shows that this period of time represents a threshold in the cooling and thinning history of oceanic crust, and specifically, the time when it became able to undergo subduction in a modern fashion.

Interestingly, this parameter well explains the “local” character of the Archaean-Proterozoic transition, because the thickness of the oceanic crust was likely very different from one place to another at that time.

7.3.2. *Evolution of the rheology and volume of continental crust*

As stated in Section 7.2.2, the Archaean represents a time of major crustal growth. Therefore, a large volume of continental crust was already formed by the end of the Archaean. At that time, there were likely a number of large and stable crustal blocks that could undergo subduction at the margins, as suggested by the magmatic record. Furthermore, this increase of the continental volume, coupled to the cooling and strengthening of the crust, allowed the

stabilization of large surfaces able to sustain elevated topography at the end of the Archaean (Rey & Coltice, Figure 7.11a). This was not the case before, as Archaean crust was hotter and more ductile such that it accommodated deformation by “flowing” rather than thickening (Thébaud & Rey, 2012). This effect also has implications for the hypsometry of the continental crust: the Archaean-Proterozoic transition also represents the time when emerged crustal blocks started to represent >5% of Earth’s surface (Flament *et al.*, 2008 ; Figure 7.11b).

All these observations indicate that, at the end of the Archaean, Earth’s surface was likely characterized by a number of stiff, voluminous and emerged continental blocks. This favored continental collision in a similar way as today; correlatively, this allowed thickening and differentiation of the continental crust and, ultimately, its stabilization ; finally, the emersion of continental crust initiated major mass transfers through the sedimentary cycle and introduction of the detrital products within the mantle by the starting subduction, as reflected by the petrogenesis of some sanukitoids.

7.3.3. *Diminution of juvenile productivity*

It is likely that Archaean TTG did not exclusively generated through subduction-like processes (such as “flat” or anomalously “hot” subduction zones as suggested by Martin, 1986). However, a given proportion of them certainly did, because their composition requires high-pressure melting (in the garnet and rutile stability fields, and far away from that of plagioclase) of mafic rocks and subsequent interactions with the mantle (*e.g.* Martin & Moyen, 2002). Because of a large proportion of sanukitoids derive from hybridization between mantle and TTG, they likely mark the time when juvenile productivity through melting of metabasaltic rocks has become so weak that the low volumes of produced melts extensively interacted with peridotite, and, in some cases, were completely consumed by the interactions (*e.g.* Martin *et al.*, 2009 ; see also Section 4.2.1). So, the Archaean-Proterozoic transition represents the time when, as stated in Section 7.2.2, recycling processes increased in importance, but also featured by the progressive waning of juvenile magmatism (Figure 7.12).

This is why sanukitoids specifically occurred at the Archaean-Proterozoic transition and are scarce afterwards. Indeed, they emplaced at the time when the subducted oceanic crust was still hot enough to melt, but not sufficiently to avoid extensive interactions with

peridotite. After the end of the Archaean, because of even lower thermal regimes, the subducted crust generally reaches its dehydration curve before its solidus. Consequently, it only produces aqueous fluids that contribute to the metasomatism of the mantle source of arc magmas and syn- to post-collisional granitoids.

7.3.4. An unifying theory: the global cooling of Earth

All reasons invoked to account for the geodynamic changes at the Archaean-Proterozoic transition fit within the scope of an unique process: the global and progressive cooling of Earth. Indeed, it accounts for:

- (1) the decrease of melt productivity from the mantle and, correlatively, of the thickness of oceanic plates;
- (2) the strengthening and emersion of continental blocks;
- (3) the waning of juvenile magmatism.

These observations explain the ambivalent nature of geodynamic changes at the Archaean-Proterozoic transition: the control exerted by the global cooling of Earth accounts for the homogenous sequence of events worldwide (especially concerning the magmatic record) and its occurrence within a relatively short time range (~500 Ma) compared with Earth's history. Nevertheless, this global parameter has a local influence on the three parameters listed above, which explains that the Archaean-Proterozoic transition is specifically different from a craton to another in terms of chronology and nature of its expressions.

7.3.5. Link with other witnesses of the geodynamic changes

The theory presented above accounts for a number of other changes listed in Chapter 1:

- The cooling of Earth also explains the evolution of orogenic styles (Section 1.3.2) that depend of the volume, temperature and rheology of continental crust.
- It also explains the evolution of the metamorphic record (Section 1.2.2), as metamorphic gradients continuously decrease since the Archaean. Furthermore, the duality of metamorphic regimes (HT-LP coupled to HP-LT), which is the hallmark of modern-style plate tectonics (*e.g.* Brown, 2006), only appears after the end of the Archaean, which is consistent with our conclusion that these modern-style processes started at that time.

- Because of (1) the emersion of continents and (2) the initiation of subduction, the importance of erosion, transport and deposition cycles in the global geochemical balance increased at the Archaean-Proterozoic transition. This is consistent with (1) the increase of the $\delta^{18}\text{O}$ signature of igneous zircons, owing to increasing sedimentary provenance (Section 1.5); (2) the evolution of sedimentary lithologies, with appearance of shelf lithologies at the end of the Archaean; and (3) the isotopic signature of some oceanic island basalts (OIB) that require the storage of “enriched” component within the mantle during more than 2000 Ma.

- The geodynamic evolution at the end of the Archaean also explains the evolution of the atmosphere (Section 1.4). Indeed, initiation of modern-style plate tectonics also contributed to the progressive storage of atmospheric CO_2 into the lithosphere, and to its recycling within the mantle. This had influence on the advent of a habitable climate on Earth and, ultimately, to the development of life.

7.4. A model for the secular continental crust evolution

The model presented here is not an exhaustive review of the available data on this topic, but rather my synthetic interpretation of them. It is based on a number of literature reviews and published data, the references of which can be found at the end of the first paragraph of this section in the French version of the manuscript.

7.4.1. Archaean processes

As discussed earlier, the Archaean represents a time of important crustal growth. The geodynamic setting in which the continental crust formed at that time is still extensively debated. On the basis of geochemical and experimental data, it is now well established that the TTG series (which represent >50% of the volume of Archaean continental crust) derive from differentiation of a young mafic source, within a wide range of pressure (5–40 kbar). This differentiation could either have concerned:

- fragments of oceanic crust foundering within the mantle, which recalls modern subduction but which is definitely not given its instability during Archaean times;
- stacked slices of oceanic crust, favored by the thickness and buoyancy of the latter, which base reaches the condition of anatexis and/or delaminates within the mantle;

- several episodes of melting of a thick, magmatically accreted oceanic plateau on top of a mantle “plume”.

Those three processes are not mutually exclusive and likely operated simultaneously (Figure 7.13) or successively during the Archaean, to generate the considerable volumes of TTG still preserved in Archaean terranes. Such geodynamic mechanisms led to the extraction of more than 50% of the present-day volume of continental crust, as well as the development of thick, depleted and buoyant lithospheric mantle roots beneath these proto-continents. In the context of a hotter Archaean Earth, these crustal blocks progressively enlarged by lateral accretion, because the crust was too hot and too ductile to undergo significant thickening. This effect accounts for the typical structures of Archaean crust, such as vertical faults, shear zones and regional foliations, as well as dome-and-keel structures that represent tectonic reworking of this hot and ductile material.

7.4.2. Evolution at the Archaean-Proterozoic transition

By the end of the Archaean (from 3000 to 2500 Ma), the global and progressive cooling of Earth induced (1) a thinning of oceanic lithosphere and (2) a strengthening of continental crust, leading to the thermal and mechanical stability of subduction and collision processes. Consequently, during the late-Archaean, crustal blocks accommodated the deformation by the development of collisional belts, subsequent to subduction, and leading to significant thickening and differentiation of the crust (Figure 7.14). As a result, the geodynamic changes at that time likely reflect the initiation of modern-style subduction-collision cycles on Earth. This explains the evolution of the magmatic record: the granitoids emplaced at the Archaean-Proterozoic transition sharply contrast with Archaean TTG and derive from hybrid mantle sources (metasomatized by subduction-related components) as well as pre-existing crustal lithologies. This duality of petrogenetic process is very similar to that of present-day collisional settings.

Both petrogenetic mechanisms also show that the Archaean-Proterozoic transition represents the advent of crustal recycling in the source of granitoid magmas. It progressively replaces juvenile magmatism, which waning is marked by the decreasing melt productivity in subduction zones, leading to more and more interactions with the mantle and genesis of sanukitoid magmas at that time.

7.4.3. Evolution from 2500 Ma to today

Present-day plate tectonics are featured by the genesis and recycling of oceanic crust at mid-ocean ridges and subduction zones, respectively (Figure 7.15). Such evolution controls the periodicity of the Wilson cycle and, therefore, the formation of continental crust. In this context, production of juvenile continental crust is restricted to subduction and collision contexts (Figure 7.15), by melting of subduction-metasomatized mantle sources either (1) during subduction itself (flux melting of the mantle) or (2) shortly after, during the post-orogenic collapse following collision (decompression melting and/or slab breakoff) (Figure 7.15).

In oceanic or continental arcs, the volume of juvenile crust is globally balanced by the volume of recycled crust owing to (1) erosion, deposition and subduction and (2) intense thermal erosion of the lower crust. As a result, the only juvenile addition to the volume of continental crust is represented by the late-orogenic magmatism. In such settings, the petrogenetic mechanisms at the origin of granitoid rocks are similar to late-Archaean ones, showing that the crustal growth and differentiation processes did not evolve significantly since the end of the Archaean. These processes are dominated by recycling of older crustal lithologies and minor contribution of juvenile material, which explains that crustal growth rates are much less elevated than during the Archaean.

These observations explain the peculiar composition of the continental crust. Indeed, it has always been a problem, because it is intermediate (~60 wt.% SiO₂) and thus, not in equilibrium with “normal” peridotitic mantle. Such composition is very close to that of sanukitoids, which solves the paradox: indeed, such as sanukitoids, the continental crust is not in equilibrium with “normal” mantle, but rather with “hybridized” mantle, enriched in water and other incompatible elements during subduction processes that started at the end of the Archaean.

General conclusions

A summary of the general conclusions of this work would be fairly similar to the extended abstract presented in the paper version of the manuscript. As a result, I will not present it here and rather refer the reader to this abstract.

Appendixes

• **Appendix 1** reports the analytical methods used in this work. Reasonable summaries can be found in the articles joined to the manuscript (especially the Lithos paper in Chapter 4, as well as the two drafts in preparation in Chapter 5). For more information, please have a look at the Figures of the appendix dedicated to analytical methods, a list of which is provided below with a short explanation:

→ **Figure A.1:** MFW triangle of Ohta & Arai (2007) showing that all samples considered in this study plot within the “igneous trend” and do not show any evidence for significant weathering.

→ **Figure A.2:** Sketch of the Jobin-Yvon Ultima-C ICP-AES of the Laboratoire Magmas et Volcans that has been used for major-element analyses.

→ **Table A.1:** External reproducibility of (1) standard BHVO-1 and (2) samples of this study for major elements measured at Laboratoire Magmas et Volcans.

→ **Figure A.3:** Protocol of acid digestion realized to dissolve sample powders for purposes of trace element and isotopic analyses. The lower part of the figure represents the attack under high-pressure bombs to dissolve refractory minerals such as zircon.

→ **Figure A.4:** Ratio of concentrations for a sample analyzed after classical digestion without using high-pressure bomb, and after digestion using high-pressure bomb.

→ **Figure A.5:** Sketch of the Agilent 7500cs ICP-MS of the Laboratoire Magmas et Volcans that has been used for trace element (as well as U–Pb isotopic) analyses.

→ **Figure A.6:** Plots of reference concentrations of natural standards G-2, RGM-1 and DR-N as a function of the average of measured values. For all elements, concentrations plot along the 1:1 line.

→ **Table A.2:** External reproducibility of standards G-2, RGM-1 and DR-N for trace elements measured at Laboratoire Magmas et Volcans.

→ **Figure A.7:** Comparison between concentrations measured at Laboratoire Magmas et Volcans and at AcmeLabs (Vancouver, Canada – used for Bulai and Matok plutons) for 11 samples of the Matok pluton and 34 minor and trace elements. Differences observed are (1) for Pb, which is very low in AcmeLabs analyses because of an incomplete digestion in Aqua Regia for this element); (2) for Zr, Hf, and Y, which are interpreted as power heterogeneities with respect to zircon; and (3) for Cr, Co and Ni, which concentrations are much higher in AcmeLabs analyses because in this case, powders were obtained after crushing into a tungsten (rather than agate at LMV) swing mill.

→ **Figure A.8:** Purification of Sm and Nd for purposes of isotopic analyses. This protocol of chromatographic extraction is run after digestion of the sample as described in Figure A.3.

→ **Figure A.9:** Sketch of the Thermo Finnigan Triton TIMS of the Laboratoire Magmas et Volcans that has been used for Sm–Nd isotopic analyses.

→ **Table A.3:** Cup configuration for the analyses of Sm and Nd isotopes using the Thermo Finnigan Triton of the Laboratoire Magmas et Volcans. ^aMass measured for interference corrections; ^bPossible interference.

→ **Figure A.10:** Summary of standard measurements for Nd and Sm in the course of the study.

→ **Table A.4:** Mass of Nd and Sm measured in total procedure blanks. The number in italics represents the ratio between blank and sample, considering a mass of Nd and Sm in the sample of 2 µg and 300 ng, respectively, which are the expected masses after realization of aliquots before chemical purification.

→ **Figure A.11:** Sketch of the Resonetics M-50E laser ablation system of the Laboratoire Magmas et Volcans that has been used for U–Pb *in situ* dating of separated zircons.

→ **Figure A.12:** Results of measurements of natural zircon standards GJ-1 (calibration standard) and 91500 (external standard) in the course of the study. Reference ages are 599 Ma for GJ-1 and 1065 Ma for 91500.

• **Appendix 2** (pages 479 to 483) presents the method applied to select partition coefficients for purposes of modeling (the resulting database is presented in the article of

Section 4.3). For each element and for three different melt compositions (mafic, intermediate and felsic with arbitrary cuts at 60 and 70 wt.% SiO₂ to separate the groups from each other), I selected a range of partition coefficients based on the published variability as reported in the GERM database (<http://earthref.com/GERM/>).

For a given element, a given mineral and a given compositional range, four different cases were experienced:

- A lot of data ($n > 5$) were available and cover a relatively restricted range (Figure A.13). In this case, the extreme values are defined by the average value, affected of a 1σ uncertainty.

- A lot of data ($n > 5$) were available but their variability is very high, either because of (1) a few numbers of outliers (Figure A.14a) or (2) a homogeneous spread of the data (Figure A.14b). In the first case, we simply excluded the outliers from the calculation of the average. In the second one, we used the median rather than the average, still affected of a 1σ uncertainty, to define the final range.

- A few data ($n = 2-5$) are available. In this case, the retained range roughly corresponds to the most extreme published values (Figure A.15).

- Sometimes, only 1, or even no value was available. In this case, we used the same range as for the adjacent melt composition.

- **Appendix 3** gives the GPS positioning of all sampling sites.

- **Appendix 4** presents all data obtained in this work (except major element analyses of minerals, reported in Master thesis of Laurent, 2009; Rapopo, 2010 and Pauzat, 2011, as well as Lu–Hf analyses of zircon, as it is work in progress in collaboration with A. Zeh from the University of Frankfurt).