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Technological and Archaeological studies of the Middle-Elamite and Neo-Elamite colored glazed ceramics (1500-540 BCE)

Ali Aarab

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présentée par

Ali Aarab

pour obtenir le grade de :

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Technological and Archaeological studies of the Middle-Elamite and Neo-Elamite colored glazed ceramics (1500-540 BCE)

Soutenue le 19 décembre 2022

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Chapter One: Introduction

1.1-What is Elam?

Elam is an artificial construct, a name coined by Mesopotamian scribes, gazing across the alluvium towards the Iranian plateau, who imposed it from outside on the disparate regions of highland southwest Iran and its peoples; Potts suggested that the sumerogram NIM meaning simply ‘high’, often accompanied by the determinative KI denoting ‘land, country’ (D. T. Potts, 1999). However, Desset suggested that it should be better for epigraphists to stop translating systematically the cuneiform sign NIM by Elam (which gives the impression that we know what the Mesopotamian scribes were meaning, i.e. the modern idea of Elam) and keep in the translation NIM (Desset, 2017). Probably Elam was the word that Mesopotamian scribes used to call their foreign eastern neighbors, similar to Shimashki, and Marhashi people. However, ‘Elam’ was not a place (or a unique place), and Elamites (as the word that is accepted in modern times) called themselves ‘Hatamti’ (König, 1965).

It seems that during the 19th century, scholars used different words to express “Elam”. Perhaps one of the first uses of the word “Elam” can be attributed to F.H. Weissbach and H. Winkler (Basello, 2001). But what if researchers in the 19th century suggested another word instead of ‘Elam’? Maybe the more correct word is ‘Hatamti’.

Nevertheless, I will use ‘Elam’ in the present study since ‘Elam’ is an accepted word among scholars, and the reader does not get confused.

The Elam could be divided into four different periods: Proto Elam, Old Elam, Middle Elam, and Neo Elam, which were present in the time range between 3200 to 540 BC (Table 1.1), especially in the southwest of Iran. It seems that Elam consists of two main parts: the “Highlands”, and the “Lowlands” (Alizadeh, 2010), and the two main centers of “Anshan” and “Susa” can be considered related to them.

Table 1.1- The timeline of Elam

Period	Date (B.C)	Description
Proto-Elamite	3200-2700	-
Old Elamite	2700	First war in recorded history between Elam and Sumer; Sumer wins.
Old Elamite	2400-2100	The Awan Dynasty
	2100-1970	Shimashki Dynasty
	1970-1600	Sukkalmah Dynasty
Middle Elamite	1500-1400	Kidinuid Dynasty
	1400-1210	Igihalkid Dynasty
	1210-1100	Shutrukid Dynasty
Neo Elamite	1100-770	Neo Elamite I
	770-646	Neo Elamite II
	646-539	Neo Elamite III

1.2-Elamite glaze and its origin

When there is a discussion about the first glazes, undoubtedly vitreous materials will be associated in the mind. Stones such as turquoise and lapis lazuli were precious objects in ancient times but their access was not possible for all civilizations. Therefore, the first vitreous materials should be considered for the production of materials that will mimic precious stones. The first

vitreous materials were glazed stones, mainly quartz and steatite, and faience which consists of a ground quartz body coated with a glaze. In all cases, the glazes were of the alkali-lime type, initially with copper added to produce a blue color. Although the precise chronology and geographical origin are not known, it is clear that these vitreous materials were being used in the Near East and Egypt from the 4th millennium BC onward to produce small objects such as beads, scarabs, seals, and amulets (Tite et al., 2002). However, from another point of view, it is possible to discuss the origin of glaze, which is a coating deposited on ceramic bodies. Regarding the Elamite period, it seems that the advancements of vitreous materials and ceramics together led to the formation of glaze. It should not be overlooked that this art itself was not invented all at once, but it also had its roots, so it is necessary to look at the activities related to this art in Elam and its neighboring lands that were carried out before. The first use of colored clay cones in the decoration of buildings in Mesopotamia in the middle of the 4th millennium BC, the Sumerians erected a series of huge religious and memorial buildings in the holy city of Inanna, the Lady of Heaven (Hameeuw, 2015). The surface of the columns and walls in this columned hall are decorated with clay cones in a way that reminds the old and traditional buildings made of reeds. In this way, the surface of the walls and columns is covered by thousands of small colored clay cones that were covered by geometric motifs. The surface of these cones is painted with black, white, and red colors, sometimes a slurry of these colors is applied to them and fired in the kiln. Such a technique can be considered one of the basic forms of decoration with glazed bricks, which was seen in abundance later in the Elamite period, especially in the Chogha Zanbil ziggurat. It is also possible to refer to the painted pottery in the pre-Elamite period as another type of pre-glazed ceramics, which can be seen in Iran since the Neolithic period.

The relationship between the first glaze makers and metalworkers can be seen in Abu Matar site in the Middle East (Peltenburg, 1971). Glazed surfaces visible in the metallurgical kilns support this hypothesis that the glazes were first created unintentionally in copper casting furnaces. It is likely that the ancient metallurgists noticed the turquoise glow caused by the pouring of its melt on the quartz stones during the process of melting copper. Although the above-mentioned event probably did not happen frequently, it is important because it was the first time that mankind gathered all the necessary factors to make a glaze in one place. The story of the art of glazing begins with the formation of these accidentally glazed stones. In addition, during the Elamite

period, Untash Napirisha, the king of Elam in Chogha Zanbil, mentioned one of the first references to “glaze” among the ancient texts:

“I, Untash-Napirisha, son of Humban-Numena, king of Anshan and Susa, desirous that my life be continually one of prosperity, that the extinction of my lineage not be granted when it shall be judged (?), with this intention I built a temple of baked bricks, a high temple of glazed bricks; I gave it to the god Inshushinak of the Sacred Precinct. I raised a ziggurat. May the work which I created, as an offering, be agreeable to Inshushinak”(P. Jones, 2014).

So, like many other studies related to ancient materials, it is difficult to determine the first place where glazes were produced. However, the advances in the elaboration of vitreous materials and ceramics have led to the production of glaze. During the current research, we will try to deal with the evidence of the first glazes in Iran. Also, what will be considered in the present research is the possibility of the influence of trade networks on Elamite glaze making that itself can be a part of the origin, and one should not ignore the influence of trade on glaze making.

1.3-Studied areas, archaeological sites, and samples

The province of Khuzestan, which occupies an area of 63,213 km² in southwestern Iran, has ca. 4 million inhabitants. It is located between 48°E and 49.5°E longitudes and between 31°N and 32°N latitudes. Topographic elevations in the province vary between 0 and 3740 m. The climate of the study area varies from arid to humid. The northern parts of the province experience cold weather, whereas the southern parts experience tropical weather. Summer is from April to September, whereas winter is from October to March. The annual mean of maximum summer temperatures in the province is about 50 °C (in July) and the minimum winter temperature is 9 °C (in March). The annual amounts of rainfall are 150–256 mm in the south and 995–1100 mm in the north, and about 70% of annual rainfall events occur from February to April (Zarasvandi et al., 2011).

Since many researchers consider the first evidence of glazes in Iran to be in the Elam civilization and one of the main centers of the Elam civilization was in Khuzestan province, therefore, three sites at Haft Tepe, Chogha Zanbil, and Jubaji were selected from the Southwest of Iran. According to figure 1.1 in terms of soil taxonomy, all three studied areas are located in the

area of inceptisols. This is very important for the ceramic bodies because inceptisols often occur on more resistant quartz-rich deposits where the rate and degree of weathering are somewhat retarded (Palmer, 2005) and, according to figure 1.2, the studied areas are located in the range or border of feature forming conglomerate.

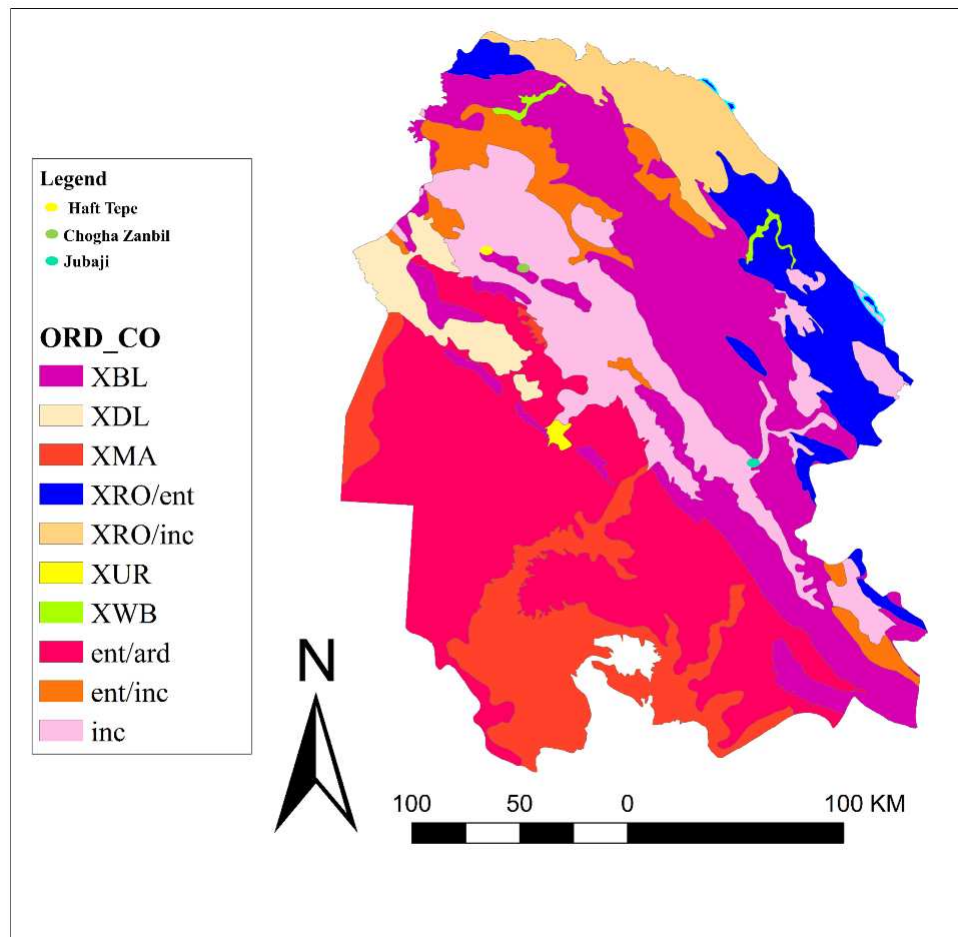


Figure 1.1- Soil Taxonomy of the Khuzestan and the location of: Haft Tepe, Chogha Zanbil, and Jubaji* (inc = Inceptisols, ent= Entisols, ard= Aridisols)

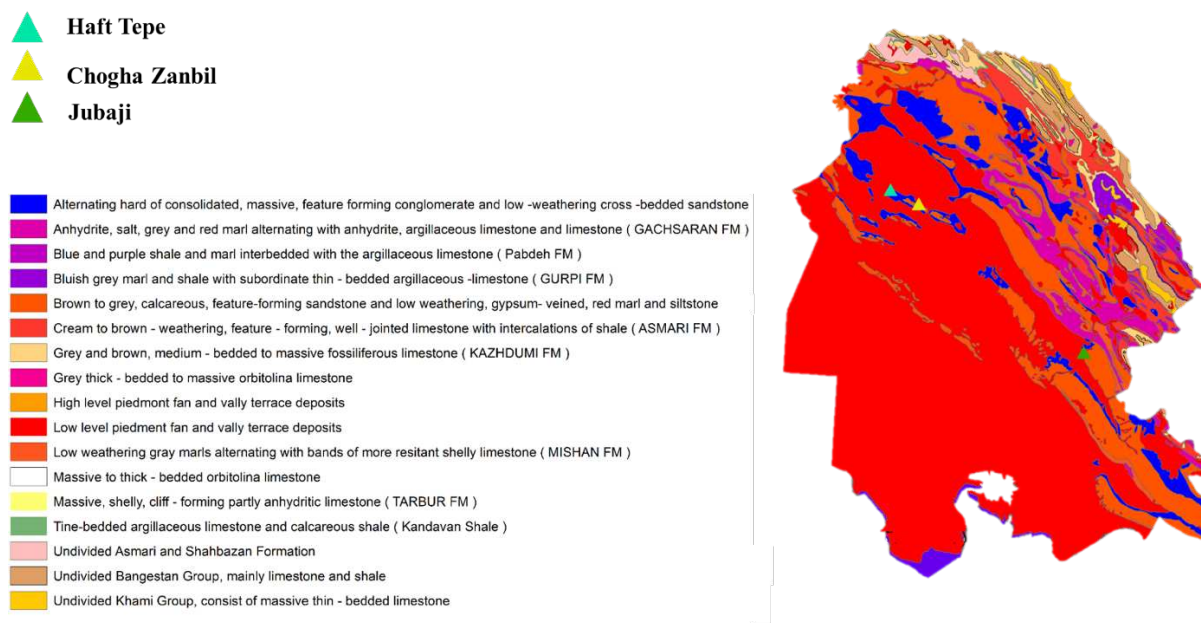


Figure 1.2- Geological map of Khuzestan

***Table 1.2-** Soil Taxonomy subgroups (According to the figure 1)

Soil Taxonomy subgroups (abbreviations)	Soil management group
XBL; XDL; XWB	Undifferentiated non-soil areas
XRO	Rock outcrops and very shallow soils
XMA	Poorly drained soils of wetlands and floodplains

All glazes studied in this research were collected from archaeological excavations. The Haft Tepe samples were collected from Ezzat Negahban's archaeological excavations (Negahban, 1991), which are now kept in the Iran National Museum. The Chogha Zanbil's glazes were collected from Ghirshman's archaeological excavation (Ghirshman, 1961), and Chogha Zanbil samples were collected from the Haft Tepe museum. And the most recent area is Jubaji with a date of 585 to 539 BC, and the samples of this area are the result of Shishegar excavations (Shishegar, 2015). But there were two major problems in collecting the samples: first, the Neo Elamite period, for which no detailed information is available from this period between 1000 to 585 BC, and Jubaji is a site that is dated after this period. To compensate for this gap, two samples from the Qalaichi area from the 8th century BC were selected. The second problem was related to the Haft Tepe, which has been inhabited from the Elamite period to the Islamic period and it seems that it has

always been of special importance. Therefore, the overlapping of the ancient layers caused problems in the dating of several of glazed samples between the Elam and Parthian-Sasanian periods, except for the three samples that Ezzat Negahban directly refers to their discovery from the layers of Elam. But solving these problems is one of the goals that have been tried to be faced in the present research.

1.4-Technological studies on Elamite glazes

Perhaps one of the first studies on Elamite glazes is related to Chogha Zanbil. The glazes that many researchers believed should be considered the first glazes of the Middle East region (Moorey, 1994; Peltenburg, 1971). But what about the Elamite glazes found in Haft Tepe, which are older than the Chogha Zanbil glazes? This is one of the questions that, if answered, can make the date of the first glazes in the Middle East a little older. In this way, studies were conducted on the glazes of Chogha Zanbil from the Louvre Museum (Caubet & Pierrat-Bonnefois, 2005), which of course were influenced by the view that the first glazes of the Middle East were sought in Chogha Zanbil. This issue is important because the site of Haft Tepe is in the same period as the Nuzi site in Mesopotamia (mid-14th millennium BC). Nuzi is known as one of the first glass production centers in the world (Shortland et al., 2017). In this way, Haft Tepe will be important as an area that has glazed objects in the same period as Nuzi. Of course, in another more recent study that analyzes Elamite glazes, the presence of glazes in Haft Tepe during the Elamite period is correctly mentioned, but in this study, Elamite glazes from Haft Tepe are not analyzed (Raith et al., 2022), and in this article, the only glazes that can be categorized as the Elamite glazes with certainty are Chogha Zanbil samples. Susa has always been problematic because of the disturbance of the stratigraphy of the Elamite layers (Caubet & Bouquillon, 2007a; Caubet & Pierrat-Bonnefois, 2005). This is exactly the problem that is pointed out in Holakooei's article about Neo Elamite glazes (Holakooei, 2014). How can it be said with certainty that the Susa glazes belong to the Elamite period? This is the same problem that exists in the present study on Haft Tepe glazes. But to better understand the Elamite glazing of Haft Tepe, in addition to the glazes that were found from the Elamite layers, we tried to select samples from the newer periods of Haft Tepe (Parthian-Sasanian) for comparison. To be able to discuss with more certainty about the Elamite glazes of

Haft Tepe as much as possible. It seems that without knowing the Achaemenid glazes of Susa or the Parthian-Sasanian glazes of Haft Tepe, it will be difficult to discuss with confidence about the Elamite glazes of these areas. Also during the Elamite period, there were glasses, which are known as glass cylinders, and analyzes have been done on these glasses from Chogha Zanbil, which indicated the presence of silver in these glasses is one of the interesting cases of this study (Emami et al., 2020). It should also be mentioned the iron age glazes of Iran and Mesopotamia, which can be very important because they belong to the same period as the Neo Elamite period. In this regard, it should be mentioned the studies carried out by Holakooei, led to the identification of opacifiers in Iron Age glazes (Holakooei, 2013; Holakooei, Soldi, et al., 2017). But how can the use of glaze refer to the trade relations between Elam and their northern neighbors in the Iron Age?

It seems that Elamite glazes were mainly limited to Khuzestan province and it was only in Fars province (Tall-i Malyan) (Carter & Deaver, 1996) that Elamite glazes were mentioned in a place outside of Khuzestan province, of course, not many studies have been done on Tall-i Malyan Elamite glazes so far.

At the beginning of the study in chapter 2, an attempt was made to study ancient texts that deal with how to make the glaze. Such texts can remind the background of glaze-making in Iran. In chapter 3, the terminology of color and colored materials during the Elamite period is discussed; It has also been tried to discuss the names of the toponyms that supplied colored minerals to Elam. In the next chapter (chapter 4) analytical methods are discussed to study ceramic bodies. In the last chapter (chapter 5), the glazes have been studied, and one of the most important analyzes performed on them is LA-ICP-MS analysis.

Chapter Two:

Glaze making in the texts of the Islamic period of Iran; with emphasis on plant ash and coloring agents of the glazes

Abstract of the chapter

Glaze-making is one of the artistic manifestations that can be considered a part of Islamic sciences and technologies due to its connection with light or paint. The alkaline glaze is one of the types of glazes that has a long history in Iran and, according to the texts of the Islamic period, it was made using *Shakhar*, which is the ash of plants. Due to their high alkali content (sodium and potassium), plant ashes have acted as a flux in making the glaze. In this research, an attempt was made to remake alkaline glazes based on ancient texts explaining glaze making in the Islamic period. The results of recreation following the instructions in the texts of the Islamic period indicated that those instructions for making glaze are quite practical. However, the two types of plants that are mentioned in the texts for making glaze are *Shura* and *Ushnan*. The properties of the above plants in the glaze can be examined in more detail with XRD, EPMA, and ICP-MS analyses. On the other hand, such analyzes can help the history of science or technology studies not only recreate past manufacturing processes but also scientifically verify the validity of old texts. According to this study, the two treatises of Kashani and Nishabouri are more specialized than other treatises regarding glaze making. Also, by comparing the coloring agents in Mina'i glazes and Islamic texts, it seems that there is a complete connection between texts and glazes.

2.1-Introduction

Glaze in ancient Iran had high diversity since Iran, and its neighbors can be considered as one of the first regions to make a glaze on ceramics (Holakooei, 2016: 49). The Islamic period can be considered as the flourishing period of some glaze-making techniques in Iran look like lustre glazes. Unfortunately, there are no written documents describing the method of glaze production during the pre-Islam period. Therefore, the manuscripts written during the Islamic period can be considered the only documents that refer to the method of making glaze in Iran. These texts not only help us to have a comprehensive view of glazing during the Islamic period in Iran but also, as the most ancient texts documenting glazing processes, they can be considered an important source of information to address crucial questions on pre-Islamic technologies (type of raw materials and their provenance, commercial routes, elaboration processes).

Table 2.1- The list of treatises studied in this research

Number	The name of the treatise	Author	Date
1	Al - jamāhir Fī-ma'rifat Al-jawāhir	al-Birūnī	973-1048 A.D
2	Bayān al-Şanā'āt	Tiflīsī	12 th century
3	<i>Javāhir-Nāma Niẓāmī</i>	Neyshābūrī	12 th century
4	<i>Arā'yis al-Javāhir va Nafā'yis al- Aṭā'yib</i>	Kashani	1301 A.D
5	Majmū'at al-Şanāya	Unknown	1582 A.D
6	On the manufacture of modern Kashi earthenware tiles and vases	Isfahani	1888 A.D
7	<i>Kashf al-Şanāya</i>	Ḥusaynī	19 th century

Instructions for making glaze have been explained many times in old Iranian texts under the title of making the glaze. In Table 2.1, the treatises that were tried to be studied in this research are introduced. In some old Iranian texts, making glaze is mentioned as a part of the Alchemy, which means, the transmutation of other metals into gold; although even Arabian scientists were aware that “not all that glitters is gold” (Pradell, 2016: 34). This industry was developed by alchemists

who used the two basic concepts of dye (color) and elixir (transformation) that have an essential place in their studies. Therefore, various texts are available that have been dealing with making glazes.

In the present paper, an attempt is made to investigate the instructions for making glaze in these texts and to compare the ancient writings to modern glaze-making knowledge. To know the traditional method of glaze making, researchers have three sources at their disposal:

ancient texts that are all related to the Islamic period and are the objects of the present study; archeological glazes remaining in Iran in the form of bricks and glazed vessels from the Middle Elam to the Islamic periods and laboratory investigations on these ancient materials that can provide useful information on the elaboration techniques;

the limited number of traditional glaze production workshops, of which some are still active.

In the present study, we have considered all Persian texts related to glaze making, especially alkaline glazes. In addition to this literature survey, it should be noted that the ancient materials left from the past can also be considered as a kind of text because they look like the texts, and the material culture can be considered as data in the study of the history of science or technology, and analyzing these materials will also help researchers in the history of science and technology, just as much as recreating and reproducing a science or technology according to the instructions of various ancient treatises.

2.2-Glaze and its constituents

2.2.1- General glaze information

The glaze is a thin layer of glass or glass mixed with crystals that are continuously applied to the surface of a ceramic body (Pradell & Molera, 2020). The term enamel designs more specifically a glaze layer on a glass or a metal. In the old Islamic texts, al-Biruni also mentions very precisely that glaze is a kind of glass (al- Biruni, 1965).

In addition to enhancing the esthetic appearance of ceramics, glazes make the decorative motifs created on pottery last longer. The glaze must fulfill important criteria such as increasing waterproof, stability against cracking, resistance to acids and alkalis, removing roughness, and

porosity surface resistance required to prevent the penetration of moisture and the alteration of the material by chemical reactions (Pradell & Molera, 2020).

The presence of two major ingredients is essential in the synthesis of the glaze mixture. First, a glass network-forming agent is the main component. In ancient and modern glasses, silica SiO_2 is the major network former, occurring in nature as the mineral quartz. Old Persian texts indicate that silica is mostly found in the form of “blour” (crystal) or “sang-e blour”(rock crystal). Neyshabouri also mentioned the existence of different names for this stone and believes that different names have been proposed for it in different regions, for example, “shekar- sang” in Khorasan, or “sang-e atash-e sepid” in Iraq, and it is mentioned in mineralogy books under the name of “hajar-e maha” (Porter, 2004: 348). In ancient Persian texts, network formers also in the glaze are named the "asl" in the meaning of “principle” (Afshar & Daryāgasht, 2003: 345; al-Biruni, 1965: 98). In addition, Porter suggested that “asl” should be more or less pure silica (Porter, 2004: 348).

Due to the high melting point of silica ($>1700\text{ }^\circ\text{C}$), a melting aid (flux) is required and is provided by alkalis, lead, or borax (Chavarría, 1998: 21). These fluxes or melting agents are added to silica to decrease the melting point of the mixture, towards temperature that can be reached in traditional furnaces with an estimate around $1000\text{-}1100^\circ\text{C}$ (Klimscha et al., 2021). The glaze that is obtained using only silica and an alkali flux is transparent and colorless. Additional raw materials (limestone/shell, feldspars) are used to better control the viscosity or the glaze properties. Finally, to achieve an opaque or colored glaze, minerals containing chromophores are added.

Materials that make the opaque glaze without being dissolved in the glaze mixture or its melt at high temperature would be dispersed in the glaze and be separated into crystals and fine grains when cooled and causing the glaze not to let light pass through it and making it appear opaque (Cai et al., 2018).

To color, the glaze, dye oxides are used in the glaze mixture. Each oxide produces a specific color that can be modified by other components constituting the glaze (Chavarría, 1998).

There are several ways to classify the parameters that can alter the glazes, so these categories may have something in common. So glazes can be classified according to the method of preparation (raw or fritt), cooking temperature, type of fluxes, firing condition, raw materials used, gloss, transparency, and texture.

In this study, traditional glazes are divided into two categories based on the type of fluxing agent: alkaline or lead glazes.

2.2.2- Alkaline glaze

The use of alkaline oxides together with MgO (magnesium oxide), ZnO (zinc oxide), and B₂O₃ (boron oxide) leads to the formation of an alkaline glaze. The properties of these alkaline glazes include being glassy, soft, scratchy, prone to cracking, and creating bright colors by adding coloring oxides.

Alkali glazes are those obtained using alkali oxides (Na₂O, K₂O) as fluxing agents, adding small amounts of CaO as a stabilizer and firing at a low temperature, typically ~ 900-1050°C. The fluxes used were obtained from natural alkali salts (hydroxides, carbonates, sulfates) among nine which natron¹ is historically the most important, also obtained from ashes of salt-tolerant plants (Pradell & Molera, 2020).

2.2.3- Lead glaze

In lead glazes, lead oxide PbO plays a major role in flux so that lead and silica can form the desired glaze on their own. PbO also has disadvantages as a glaze constituent. So it must be fired in a strongly oxidizing atmosphere because lead is readily reduced. It volatilizes above 1200°C (Eppler & Eppler, 2000). Lead glazes are more transparent and materials such as barium oxide are used to opacify them (Shelby, 2020). Due to the presence of other fluxes in lead glazes, in addition to the main flux (lead oxide), lead glazes can also be divided into pure lead glazes, alkaline lead, boron lead, zinc-lead, and lead- tin.

2.3-Alkaline glaze in Islamic texts

As mentioned above, one of the main types of glaze is alkaline. Alkaline glazes have a significant occurrence among historical glazes. The source of alkali in such glazes can be minerals,

plant ashes, or even part of plants (Gradmann, 2016). The main source of alkali for common glazes in Iran is plant ash. An extended vocabulary has been encountered in texts of the Islamic period to describe alkaline substances. They are designated as Shakhar, Shakhireh, Alkali, Kaliab, Qalyab, Qali, Kahar, Kala, and Qaliyeh (Wulff, 1966). Among all these names, Shakhar is mentioned more than others.

Abu Rihan al-Biruni describes Shakhar or Qali (both of them in the meaning of plant ash) as follows:

” After burning the plant, it is necessary to pour water into the ashes. The way to make it is to make the Haloxylon tree and Shura plant wet and set it on fire. When it is burnt and comes together, water is poured on it, and when the water reaches [the ashes], its mass becomes hard, and it would become shakhar. In the Roman Latin language, it is called Estoriquin; it is called alkali in Syriac. It is called Shakhar in Persian and Saji in Hindi “ (al-Bīrūnī et al., 1973).

al-Mukaddasi also refers to Ushnan (Qali or Shakhar) and mentions it as being exported from Aleppo province in Syria (Barkoudah & Henderson, 2006: 300). Neyshabouri and Kashani also point out the cleansing properties of shakhar (like soap) for rubies and emeralds (Afshar & Daryāgasht, 2003; Kashani, 1966: 42-51). Kashani, regarding the use of shakhar in glaze, mentioned that shakhar is obtained by burning Ushnan plant and introduces the best type of Shura from Ushnani, which if it is broken, the middle of it is red and has a pungent odor. He also indicates that Shakhar of each region of Iran has its characteristics, including the Shakhar of Tabriz (northwest of Iran). It seems that he is referring to Shakhar of Tabriz because he wrote the main part of his treatise when he was in Tabriz, in this case, he mentioned for one MAN stone, one MAN and a half Shakhar are needed in glaze-making (each MAN is equal to about 3 kg) (Kashani, 1966: 339-342).

Also, in a version called *Majmū‘at al-Ṣanāya*, the author of which is not known, but according to some scholars, it is probably related to the Safavid period (Holakoei, 2016). Shakhar is called Shaji or Saji salt (Majmū‘at al-Ṣanāya, 1582: 9). Wulff also refers to the method of making alkali or shakhar indicating that the preparation of alkali is done by alkali burners (qala), most of whom live in the city of Qom. He believes that alkali from Qom was famous for its quality during the Qajar period (1798-1925). He also refers to the Ushnan plant or Oshnon as the main substance of alkali, although he also mentioned that sometimes other salt tolerant plants were used for this

purpose (Wulff, 1966). For making plant ash glasses, Henderson believed in that the range of plant species suitable for making glasses is quite wide. Three species of halophytic plants belonging to the Chenopodiaceae family could have been used for glass production: *Salsola kali*, *Salsola soda*, and *Hammada scoparia* (Henderson, 2003). Allan suggested, Ushnan in the Islamic texts is *Salsola Soda*, and Shura is *Salsola Tragus* (Allan, 1973). But, Ashtor and Cevdalli suggested Ushnan as the *Hammada Scoparia* plant (Ashtor & Cevdalli, 1983). According to Allan (Allan, 1973), Ushnan corresponds to the plant *Salsola Soda*, and Shura corresponds to *Salsola Tragus*. Wulff then describes how alkali is made as follows:

"The person who prepares the plant ash throws all these plants that are not still dried in a pit with an opening diameter of one meter and a depression of two meters and burns it from under and throws the plant on the fire until it burns slowly and with a low flame and a lot of heat."

The ash obtained from this method is then baked in a special furnace called calcine (color furnace) with a tube that heats from below to be calcined. Each time, about 5 to 7 kg of this ash is calcined in a kiln by stirring it with a skewer, and 5 kg of Shura ingots are then stored (Wulff, 1966: 145).

In the glaze production workshops in Maybod in northeastern Iran, they also use a method almost similar to the one mentioned by Wulff to produce shakhar:

"in the vicinity of Aghda (a small locality located 40 km northwest of Meybod), the inhabitants collect the green plants of Ushnan (Salsola soda), and the collect in a hole 1 m in diameter and 0.6 to 0.7 m deep; we kick it fire; the plant's char and the sap "caramelizes"; cover the hole with soil put out the fire and this agglomerate of soapwort and earth is brought to Meybod by truck, once every two years. "

Ali Mohammad In the treatise *On the manufacture of Modern Kashi Earthenware Tiles and Vases in Imitation of the Ancient* (Isfahani, 1888), Ali Mohammad Isfahani also believes that alkali is obtained from the ashes of the desert plant (Shura) and mentions that, in Naein and Ardestan (in central of Iran), very good samples are obtained from this plant. Isfahani explains the process of preparing alkali from Shura plant:

"Take some of the alkalies we mentioned earlier {what we mentioned earlier is the ashes of the desert plant}. Small pot, put it on the fire, and bring it to a boil (add water as needed).

After boiling, pour it into an earthenware bowl and leave it overnight. The next morning, on the alkaline ink, you see crystals and twigs that look like candy or snow. Take this ink that our master called ink of alkali, take it ... » (Isfahani, 1888: 333).

There are two interesting points about shakhar. One of which is about mixing Shura and Ushnan plants. Allan believed that, following information in Kashani's treatise, we must not mix Ushnan and Shura (Allan, 1973: 116). Also, Olmer believed that plant ash should not be soaked or washed before use (Olmer, 1908: 44-45).

However, regarding these two points, it should be said that in none of the Kashani's texts regarding the production of glaze or enamel, which was studied in this research, the issue of mixing or not mixing Shura and Ushnan plants has been mentioned. It can only be said that in texts such as al-Sidna Abu Rihan al-Biruni and the treatise of Ali Mohammad Isfahani, the plant Shura is mentioned and the rest of the texts mention Ushnan as the main plant for making alkaline glazes. Also, regarding washing the ashes of plants to make the glaze, again, in the text of al-Sidna and Ali Mohammad Isfahani's treatise, the presence of water in making Shakhar is mentioned, and in the rest of the instructions, there is no reference to water.

2.4-Recreation of alkali flux and alkaline glaze

Therefore, after reviewing the texts of the Islamic period regarding the production of alkaline glazes, new questions were raised in the present study: Is the difference between using water or not when preparing plant ashes for a glaze? Is the use of water mentioned in some texts and not in others can be useful in producing better glaze or not?

Do all the items mentioned in the texts of the Islamic period lead to the production of glass or glaze? If the answer to this question is yes, then what is the difference between glass or glaze made through the Ushnan plant and the Shura plant? In another sense how is it possible to examine this sentence: "mixing these two plants (the mixing of Shura and Ushnan) will destroy the glaze" according to Allan?

2.4.1- Preparation of plant ashes

To answer the above questions, we followed the instructions mentioned in the texts of the Islamic period, as described above, and we prepared alkali using both Shura and Ushnan plants, which are the best plants that can be used for glaze production according to the texts. Finally, we will use the prepared alkali flux to produce alkaline glazes based on Islamic texts. Therefore, the ashes of both plants were prepared according to two different instructions. First, according to the instructions given in most texts and especially those used during the Qajar period (Wulff, 1966), and, secondly, based on the instructions according to al-Biruni (al-Biruni, 1965: 826). The main difference between the two instructions was the use or not of water when preparing the ash.

A question that was asked at the beginning of this section is about how to prepare plant ash as Shakhar or alkali agent in the glaze. In some texts, the use of water is mentioned, and in others not. At the same time, the percentage of elements in each of the ashes should be examined to obtain a more accurate knowledge of the texts and treatises on alkaline glazes. Therefore, some Shura and Ushnan plants were collected from the edge of the central desert of Iran and between the provinces of Isfahan and Qom, which were also considered by many texts. Both plants are saline and drought-tolerant plants that often grow in saline and alkaline areas.

According to the instructions mentioned in the book of Al-Sidana by Abu Rayhan al-Biruni and what Wulf brought about the Qalya (plant ash) makers from Qom, both Ushnan and Shura were prepared and reduced to ashes. The obtained ashes were then sent to the laboratory for ICP-MS analysis. The results of ICP-MS analysis show that in two plants, Ushnan and Shura, which have been prepared by the way of Qajar artisans, the percentages of sodium (Na) and potassium (K), as the two alkaline agents, are very high. The proportion of potassium in the ashes of Shura and Ushnan, which were prepared by the method of Qajar craftsmen, according to Wulf's book, is much higher than the Ushnan ashes, which were prepared by the method mentioned in the book of Al-Sadina Abu Rihan al-Biruni using water washing of the ashes. So here it seems that Olmer's remark is perfectly valid and the ashes of these plants should not come into contact with water or be washed in any way (Olmer, 1908). According to Tables 2.2, and 2.3, we can also pay attention to the differences between Ushnan and Shura.

Table 2.2- Results of ICP-MS analysis on the ash of Ushnan and Shura plants (ppm)

Compound	Ushnan (ICP-MS)- ppm	Shura (ICP-MS)- ppm	Washed Ushnan (ICP-MS) ppm
Ag	1.99	1.72	<0.5
Al	576.54	973.37	485.52
As	22.33	2.96	14.58
B	2319.09	4250.86	2355.20
Ba	7.95	48.97	8.52
Ca	>10000	>10000	5579.22
Cr	1.91	<0.5	<0.5
Cu	5.97	3.33	2.29
Fe	698.81	232.82	40.03
Ga	<0.5	2.26	<0.5
K	>10000	>10000	3678.02
Li	2.10	3.06	1.75
Mg	3449.30	3474.23	1508.52
Mn	66.60	262.89	68.14
Mo	<0.5	0.86	<0.5
Na	>10000	>10000	>10000
Nb	<0.5	<0.5	<0.5
Nd	<0.5	<0.5	<0.5
Ni	<0.5	1.58	<0.5
P	664.02	665.81	252.98
Pb	<0.5	<0.5	1.69
Rb	3.92	15.37	4.81
S	1601.39	2237.97	4373.08
Sr	389.66	678.69	288.76
Ti	9.73	56.34	14.41
V	4.63	5.27	3.05
Zn	151.09	35.22	46.85
Zr	6.96	36.94	6.81
Cl	-	-	-
Si	-	-	-

Table 2.3- Results of EPMA on the ash of Shura and Ushnan plants (wt%)

Compound	Ushnan (EPMA)%	Shura (EPMA)%
Ag	-	-
Al	0.12	1.43
As	-	-
B	-	-
Ba	-	-
Ca	20.57	57.55
Cr	-	-
Cu	-	-
Fe	-	-
Ga	-	-
K	35.38	8.40
Li	-	-
Mg	6.92	10.81
Mn	-	-
Mo	-	-
Na	13.55	10.31
Nb	-	-
Nd	-	-
Ni	-	-
P	1.22	0.69
Pb	-	-
Rb	-	-
S	4.47	3.66
Sr	-	-
Ti	-	-
V	-	-
Zn	-	-
Zr	-	-
Cl	17.45	2.25
Si	0.26	4.75

2.4.2- Preparation of glazes

Also, For the glaze/glass elaboration, according to the texts of the Islamic period, in most cases, the amount of plant ash (shakhar) equals the amount of silica (flint), and shakhar has been considered a bit more (Kashani, 1966: 340). Therefore, in the present study, the ratio of 1 to 1.2 was considered for base glass¹ and shakhar, respectively; this ratio was chosen according to Brill's article, which is related to glassmaking during the Assyrian period (R. H. Brill, 1972). One was a mixture of Ushnan ashes and silica and the other was a mixture of Shura ashes and silica.

After placing the mixture in the oven at 90 ° C for 5 hours and drying the mixture completely, both compounds were first heated to 800 ° C in the air in an electric furnace, and the synthesis were obtained in a Pt crucible. The results can be seen in Figure 2.1, both of which led to the production of glass or glaze, which was also mentioned in the texts. But the most interesting thing about glass was the color difference between them, which has been less discussed so far. Thus, differences in the plant ashes used to make alkaline glazes can lead to differences in the color of the glass or the obtained glaze. This was an interesting finding in studies on alkaline glaze that has rarely been mentioned so far. In the texts of the Islamic period, when talking about color in glaze, only mineral pigments are mentioned to make different colors, and contemporary research has not mentioned the color differences of different plants in alkaline glazes, which of course, is one of the main reasons is that the most contemporary research focuses on making lead-based glazes. However, this is a point that has been mentioned in pre-Islamic texts. Indeed, during the Assyrian empire, *Zuku* the word meant gold-colored glass (Thavapalan, 2019a). In the texts related to the Assyrian period, the production of alkaline glaze with plants is mentioned (Oppenheim, 1970; Brill, 1972), they mentioned a base glass for making other glasses, and the color of this base glass must be amber yellow (Thavapalan, 2020: 369). The amber yellow color was finally obtained in this research by using the Ushnan plant, and this color was completely different from the color obtained from a glass made with the Shura plant. Brill (1972) suggested that the amber color was undoubtedly caused by the high sulfate content and the high proportion of unburned carbonaceous material in the particular plant ash used. The glass experienced a strongly reducing internal atmosphere during the heating, and the sulfate was reduced to sulfide which formed a "carbon

¹ The composition of base glass (weight): 0.5% MgO, 0.5% CaO, 0.5% Na₂O, 0.5% K₂O, 0.5% Fe₂O₃, 95% SiO₂, 2.5% Al₂O₃.

amber" color. This carbon amber is due to the presence of a complex formed between traces of iron and sulfur in the glass (R. H. Brill, 1972). In addition, according to table 2.2 the content of iron in the Ushnan is higher than Shura. Therefore, it is possible to raise the possibility that the plant ash that was used to produce glass in the Assyrian text² could have been the Ushnan plant, a plant that has been mentioned many times in the texts of the Islamic period for the production of glaze.

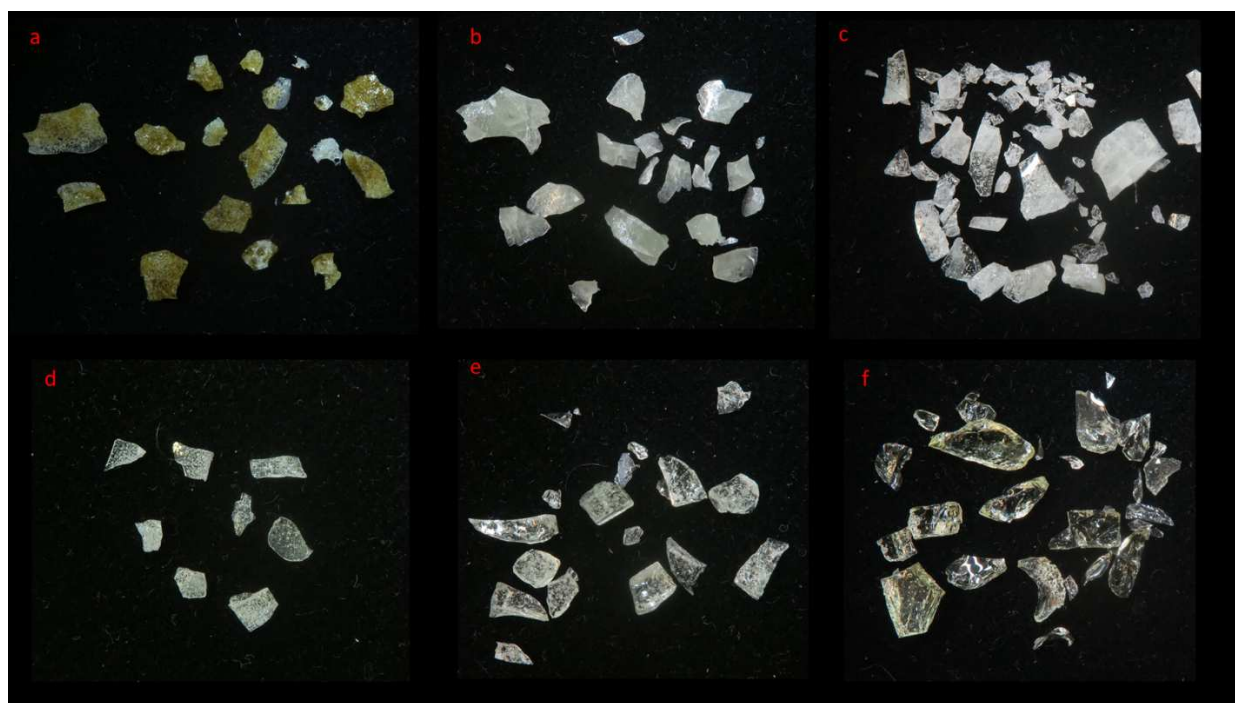


Figure 2.1- Alkaline glass made according to the instructions of Islamic texts; a, glass made of Ushnan plant ash (800 degrees centigrade) [which is amber yellow]; b, glass made of Ushnan plant ash (900 degrees centigrade); c, glass made by Shura plant ash (900 degrees centigrade); d, glass made by Ushnan plant ash (1100 degrees centigrade); e, glass made by Shura plant ash (1100 degrees centigrade); d, glass made by mixing Ushnan and Shura plant ashes (900 degrees centigrade).

² During the Neo-Assyrian period, "ahussu/uhullu" was used together with Zuku (base glass) in the meaning of soda ash (Thavapalan, 2020).

XRD analysis was performed on the fabricated and heated glass up to 800, 900, and 1100°C for sixteen hours to measure and compare the influence of using different plant ashes (Shura or Ushnan) in making glazes or glasses. The reason for choosing the two temperatures at 900 and 1100 °C is based on Brill's article on Assyrian glass-making texts (R. H. Brill, 1972). The diffraction patterns indicate that the samples are not completely amorphous since Bragg peaks are visible. This indicates that incomplete melting of the raw materials was obtained, preventing getting a clear, transparent glass. However, the melting was sufficient to get a glaze. As shown in Figure 2.2, the glass obtained from the Ushnan plant ash is more amorphous than the glass made from the Shura plant ash.

But another important question was the possibility combination of these two plants (Ushnan and Shura), which is mentioned in Allan's article that he tried to translate the Kashani's treatise (Allan, 1973: 116). As shown in Figure 2.2, the amorphous percent in the sample in which Shura and Ushnan are mixed is less than the amount of Ushnan alone but greater than the Shura alone. This means that the sample resulting from the combination of the two plants is less amorphous than the one resulting from Ushnan, but more than the one resulting from the Shura. So why did Kashani mentioned that two plants should not be mixed and Allan also points out in response to the fact that adding Shura to Ushnan and mixing them will destroy the glaze. However, this point is mentioned only in Allan's paper and in none of Kashani's treatises not mentioned while the mixing of the two plants still leads to the formation of glass and the amorphous phase. Nevertheless, why Allan mentioned that mixing these plant ashes makes the glazes rot? Which edition and text of Kashani's treatise did he refers to? Anyway, what was found in this study is that the mixing of Shura and Ushnan plants does not prevent the formation of a glaze or a glassy phase and will incidentally lead to the formation of a material with a significant amount of amorphous phase.

Now it is necessary to once again examine the text of Kashani in more detail regarding the fifth substance used in the making of glaze, namely shakhar. As mentioned earlier, Kashani says the following about this substance in the composition of the glaze:

” The fifth is Shakhar, which is called Qaliyah. For having a shakhar, you must burn the ripe form of Ushnan without adding anything to that, only you can add Shura, because it's very similar to Ushnan, and the best Ushnan has a strong smell, and the middle of its stem is red.”(Kashani, 1966: 339).

Allan's interpretation of this part of Kashani's text is as follows:

“The fifth is shakhar, which they call qali, which is made by burning pure, fully-grown ushnan plants, not mixed with shura, which is like ushnan. [If it is (mixed with shura), it makes it rotten.] The best shakhar is that which has a red-colored center when broken, with a strong smell.” (Allan, 1973: 112).

But by paying more attention to this part of Kashani's text, it can be seen that Kashani does not mention the deterioration of the glaze if Ushnan and Shura are mixed. Incidentally, what can be deduced from this text is that Shura, like Ushnan, leads to glaze because it is very similar to Ushnan. Thus, we can speak of a mistake in understanding Kashani's treatise, which unfortunately has been repeated many times in various articles and research since 1973. But here, the remaking of alkaline glaze and the mixing of two plant ashes and its positive result in XRD analysis caused the authors of the present study to re-examine this part of Kashani's text more carefully.

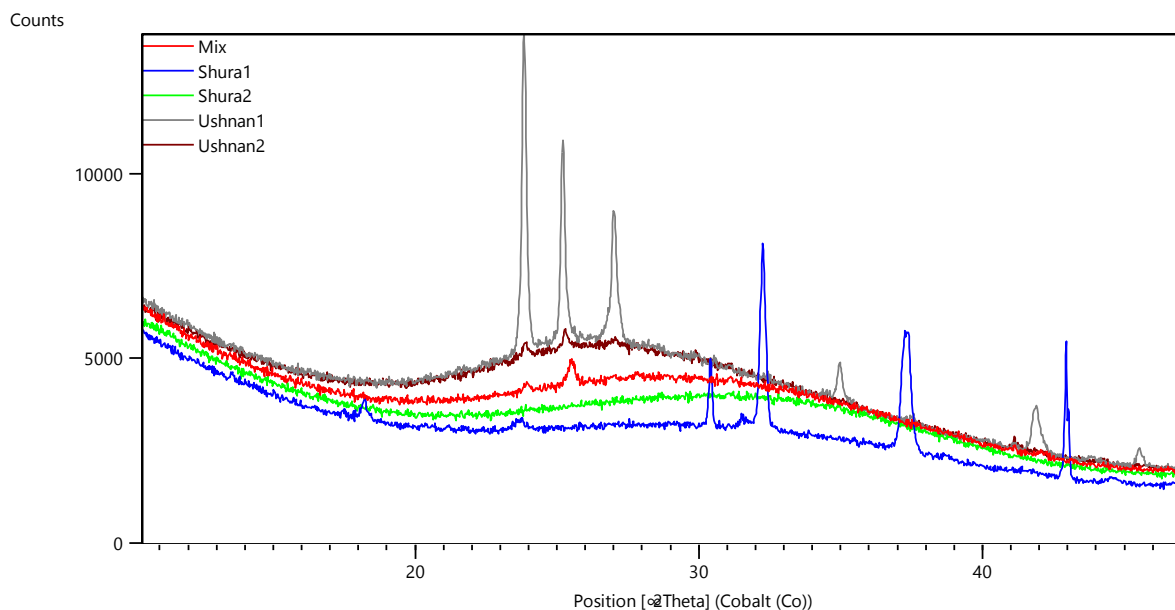


Figure 2.2 XRD result on glass samples produced based on ancient texts with Ushnan plant, Shura, and their mix; Shura1, firing temperature 900 degrees centigrade; Shura2, firing temperature 1100 degrees centigrade; Ushnan1, firing temperature 900 degrees centigrade, Ushnan2, firing temperature 1100 degrees centigrade, and Mix, Shura and Ushnan mixing, firing temperature 900 degrees centigrade.

2.5-Coloration in glazes according to ancient texts

2.5.1- Opacifier in Islamic texts (white glazes)

Neyshabouri well refers to two different categories of glaze, i.e., opaque glaze and transparent glaze (Afshar & Daryāgasht, 2003). Al-Biruni identified red lead as the color agent in the white glaze (al-Biruni, 1965), and Tiflīsī mentioned that by adding lead-tin oxide to transparent glaze, you could have opaque white glaze (Tiflīsī, 1956).

Most Islamic texts refer to the use of lead to produce white glaze. However, Kashani also mentioned the use of Shakhar in the production of white glaze in this way (Kashani, 1966):

“lead is prepared in three parts and tin is prepared in one-third. Then, lead and tin are molten together to produce lead and tin oxides, and in the second phase, the mixture of qamṣarī stone and the Shakhar (plant ash) is fired with the product of the first step, and after cooling down, three MAN³ of this mixture should be mixed with one MAN of lead-tin oxides”.

For melting lead and tin together, the lead should be molten in the kiln for a while, and the tin is then added on top of it. The mixture should then be placed in the kiln for half a day and cooled in water (Kashani, 1966: 342-343). Ali Mohammad Isfahani suggested the same method for making a white glaze, only the amount of lead and tin is different. In this way, one MAN of lead and a quarter of a MAN of tin are needed (Isfahani, 1888). It was interesting for us that Neyshabouri refers to the Talc as one of the main materials in the white opaque glazes (Afshar & Daryāgasht, 2003: 350).

2.5.2- Turquoise glaze in Islamic texts

Ali Mohammad Isfahani described how to produce turquoise glaze (Isfahani, 1888). He referred to the burnt copper pulp, one part of which should be well beaten and mixed with half of the broken stone to obtain finally a turquoise color.

³ Each MAN is about 3 Kg.

The instruction of Majmū‘at al-Şanāya treatise (1582) is very similar to Isfahani’s recipe for making turquoise glaze: ten components of transparent glaze with one component of burnt copper and naturally placed in the furnace will lead to turquoise color (Majmū‘at al-Şanāya, 1582: 22).

Neyshabouri describes a different recipe for making the turquoise glaze, requiring 100 parts of quartz stone, two parts of Soleimani stone, one part of copper rust, half part of burnt zinc, and one part of plant ash (Afshar & Daryāgasht, 2003). In this description, in addition to copper rust, Neyshabouri refers to two parts of Soleimani stones. Though it must be noted that there is a lot of discussion about Soleimani stone, according to Kashani’s manuscript (Kashani, 1966), we can likely conclude that Soleimani stone refers to cobalt ores. Indeed, he mentioned that you could find Soleimani stone in Qamsar, which is one of the famous cobalt mines in Iran. Therefore, it can be guessed that Soleimani stone in the Islamic texts is the meaning of cobalt stone.

2.5.3- Yellow glaze in Islamic texts

The Kashf al-Sanaye treatise mentioned the use of white lead, quartz, and Shakhar to produce yellow glaze (Ḥusaynī, 19th Century: 34). Alternatively, Ali Mohammad Isfahani refers to other compounds as the cause of yellow color (Isfahani, 1888). Ocher is a rock composed of clays and iron oxides (Fe_2O_3) that gives its specific color from yellow to brown and red. This rock is also mentioned in the Neyshabouri recipe as one of the compounds giving yellow color to the glaze. For the yellow color in the glazes, Neyshabouri, in chapter four of his treatise suggested at least three different recipes:

" Three parts of Copper sulfide and one part of Tin oxide must be mixed with vinegar,"
and the second one

"Two parts of manganese oxide, eight parts of sulfur, five parts of hematite, one part of lead oxide (massicot) and seven parts of silver sulfide must be mixed with vinegar" (Afshar & Daryāgasht, 2003).

"One part of silica and one part of red lead and a little white alkali should be melt together after that you can get a brilliant yellow like a yellow sapphire" (Porter, 2004: 348).

Also, Ali Mohammad Isfahani, in the continuation of his treatise, mentions 16 parts of lead and 1 part of tin for making the yellow glaze, in the polychrome technique (Isfahani, 1888). In the

book Majmū‘at al-Şanāya, the use of two components of lead and one component of Shura is mentioned regarding yellow glaze. Abu Rayhan al-Biruni considers lead as the cause of the yellow color in the glaze (al-Biruni, 1965). Also, in the two treatises of Kashani and Nishabouri, the use of gold to create a yellow color in the glaze is mentioned. For this purpose, Kashani refers to the

“gold sheets that are first kept in papers impregnated in plaster powder, then they are cut and glued on the glaze with a natural glue called “Sirish,” and they have painted around with hematite. This material should be fired in the kiln for half a day”(Kashani, 1966).

But Nishabouri only mentions that:

“the gold used must be powdered and gum arabic must be used to stick it” (Afshar & Daryāgasht, 2003).

In none of the Islamic texts related to glaze making, the use of antimony as a yellow color agent in the glaze is mentioned. It seems that some writers of ancient texts knew about antimony. For example, Kashani only refers to the treatment of *athmad* (antimony) for eye health and deals with its mines in Iran. No analytical data is supporting the application of antimony in making Iranian yellow glazes in Islamic Persia (Allan, 1973; Holakoei, 2016).

2.5.4- Red Glaze in Islamic texts

Ḥusaynī said the following to produce red glaze in his book *Kashfat al-Sana'i*:

“Quartz 10 parts, shakhar, tin oxide, mercury, and sulfur one part from each other” (Ḥusaynī, 19th Century: 34).

Neyshabouri for red color mentioned:

“ They take five derams of maghnistyö (manganese oxide), two and a half derams of green zāj (Iron sulfate). Two and a half derams⁴ of white gugird (sulfur), two and a half derams of silver that had been burnt with yellow gūgird (yellow sulfur). They grind and sieve all these, mix with old vinegar, and then apply on glass and ceramics and others. They paint with it whatever they wish and leave it to dry. Then they put (the vessel in the kiln) as described before and increase the fire by not more than the level which mentioned before.

⁴ A unit of measurement

After firing, they leave it to cool. Then they bring (the vessel out of the kiln) and wash it clean so that a deep red color comes out.” (Afshar & Daryāgasht, 2003; Watson, 2020).

Tiflīsī considers ferric sulfate (or copper sulfate?) to be the cause of the red color in the glaze (Tiflīsī, 1956: 331).

2.5.5- Black and dark colors in glaze based on texts

Tiflīsī considers the addition of iron oxide to transparent glaze as the cause of the black color in the glaze (Tiflīsī, 1956: 331). Abu Rayhan al-Biruni also introduces iron oxide as the cause of the black color in glaze (al-Biruni, 1965: 99). Ali Mohammad Isfahani mentioned the use of manganese in making black glaze (Isfahani, 1888). He also refers to the juniper color obtained with al-Hadid saffron (iron hydroxide). Ali Mohammad introduces the method of preparing al-Hadid saffron by placing a piece of iron in acid for a few days (Isfahani, 1888). Kashani, like Ali Mohammad Isfahani, refers to manganese as the cause of the black color in the glaze. In addition to manganese, Kashani suggested: “*Mozarrad*”(chromite) as the agent of black color, too (Kashani, 1966).

2.5.6- Green and blue color in glaze based on texts

Abu Rayhan al-Biruni considers the cause of the green color in the glaze to be burnt copper or copper oxide (al-Biruni, 1965: 100). Tiflīsī also refers to white lead and copper oxide as the green color agent in glaze (Tiflīsī, 1956: 331). Neyshabouri also refers to the combination of copper oxide, copper acetate, and lead oxide in making green glaze (Afshar & Daryāgasht, 2003: 346). Neyshabouri, also believed that white tin and antimony can be used to opaque these colors (Porter, 2004: 349).

For making the blue glaze, Ḥusaynī mentioned:

"Red lead and crushed quartz ten parts, and 1 part of cobalt oxide must be mixed"
(Ḥusaynī, 19th Century).

For blue glazes, Tiflīsī suggested:

, "*One liter of white lead must be mixed with one Deram (about 2.5 grams) of crushed Chinese Iron to have a lapis lazuli color.*" (Tiflīsī, 1956).

2.5.7-Comparing color agents in the Islamic glazes and texts

Fifteen samples of Islamic period glazes were selected (figure 2.3) to compare coloring agents in the Islamic glazes and texts. All the samples are Mina'i glazes that are collected from Iran National Museum. These kinds of glazes are chosen because most of them have more than three colors, so these glazes are suitable for comparing color agents in the texts and glazes. For this purpose, PIXE analysis was first performed on the samples at the Atomic Energy Organization of Iran. After that, p-XRF analysis at the Institute of Archeology of the University of Tehran was used to better distinguish the colors on the glaze. Because the beam diameter of the p-XRF was smaller than that of PIXE, for this purpose, it might be better to use the micro-PIXE method, that beam diameter of that is less than 10 microns. Finally, the results of these analyzes and the comparison of the coloring agent in the texts and samples of Mina'i glazes belonging to the 12th and 13th centuries are presented in table 2.4. According to this table, for the red color, it seems that Neyshabouri's recipe is very close to the glaze samples. In addition, Kashani's suggestion is similar to yellow glaze samples, then his recipe for black color has the best similarity with black lines in one of the glazes. For the black color, Isfahani's suggestion was found in the glazes, and Hussain's suggestion is very close to the chemical composition of blue glaze samples, and for green glaze samples, we can find the same suggestion in Tiflīsī's treatise.

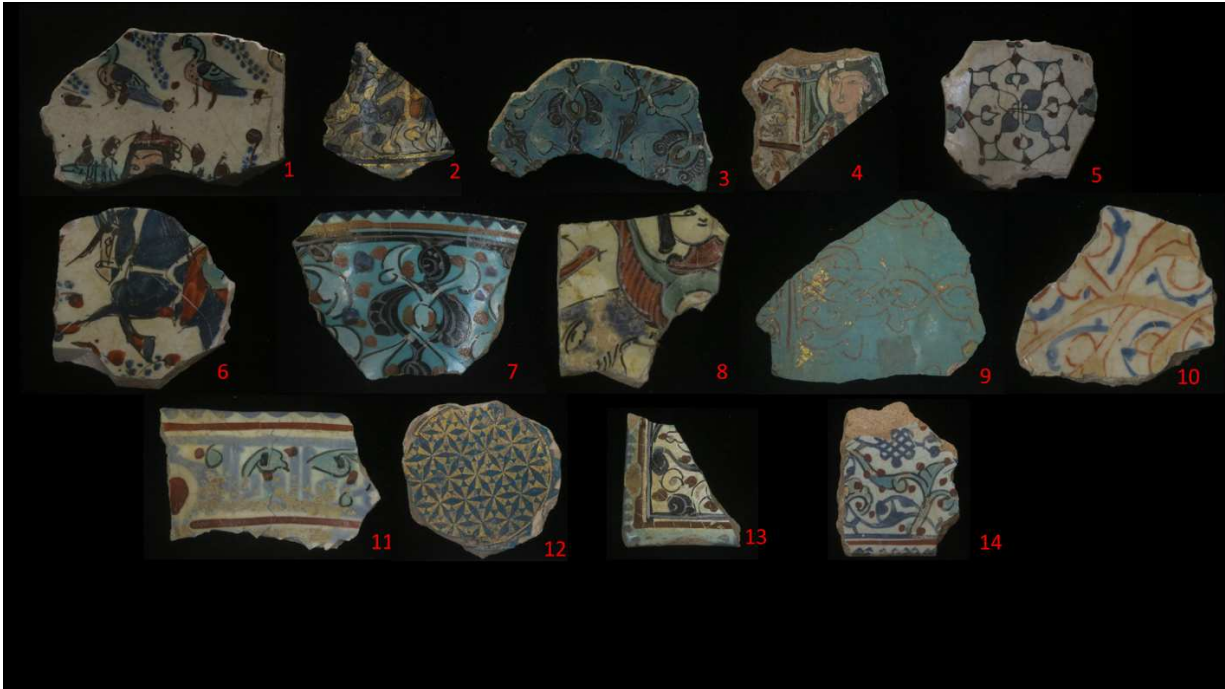


Figure 2.3- Mina'i glazes, and number of each sample

Table2.4- PIXE and p-XRF analysis of Mina'i glazes (12th and 13th century)

Sample/Analyze	Al2O3	SiO2	K2O	CaO	Cr2O3	MnO	Fe2O3	CoO	CuO	SnO	Au	PbO	Na2O	MgO
5/PIXE	3.4	57.55	1.56	3.7	0.37	0.08	6.81	0.28	0.12	5.78	-	17.25	-	-
1/PIXE	5.51	58.2	0.83	1.1	3.9	-	1.5	-	0.3	1.9	-	11.57	12.9	2.28
2/p-XRF	4.15	49.2	-	5.02	-	0.05	5.94	0.19	0.27	4.45	-	18.49	5.03	4.41
8-p-XRF	5.04	57.1	-	6.2	3.05	0.09	3.22	0.06	0.22	8.1	-	15.69	-	4.14
9/PIXE	6.21	44.94	1.32	3.1	-	0.01	0.37	0.96	-	5.08	-	17.16	4.72	1.9
4/p-XRF	8.36	60.8	-	3.1	0.05	0.26	0.58	0.003	0.09	0.79	-	2.96	13.3	9.40
7/PIXE	-	57.72	0.91	4.74	0.05	0.02	0.43	-	1.18	2.98	-	17.2	5.51	6
14/p-XRF	1.81	50.06	-	4.87	0.06	0.1	1.4	1.36	0.71	5.7	-	30.52	-	3.76
10/PIXE	5.02	54.28	0.69	1.76	-	-	0.02	0.5	0.05	2.78	4.74	16.42	8.6	4.57
12/p-XRF	3.3	50.63	-	4.8	-	0.09	0.94	0.04	4.59	6.2	4.37	20.14	-	3.6
2/p-XRF	7.09	54.04	-	4.3	-	0.05	1.2	0.01	0.22	4.35	3.92	15.33	6.01	4.38
8/PIXE	5.98	57.12	0.48	1.83	0.45	0.03	0.53	-	0.64	1.75	-	15.26	12.29	2.73
8/p-XRF	3.27	50.8	-	3.3	0.40	0.07	1.58	0.03	1.72	4.03	-	21.3	7.23	5.9
9/p-XRF	6.7	47.62	-	5.3	-	0.03	0.81	-	2.15	4.9	-	18.37	8.11	4.2
10/p-XRF	2.89	47.08	-	4.06	0.03	0.03	0.55	-	0.13	9.72	-	18.62	12.54	8.31

2.6-Conclusion

Islamic texts are one of the primary sources for reviving traditional Iranian glaze-making. In this study, an attempt was made to remake the Iranian alkaline glaze based on the texts, which showed the accuracy of the texts in dealing with the instructions for making a glaze. In the end, what was achieved in this research was alkaline glass, which was fully adapted to the instructions of texts and treatises of the Islamic period, and was a successful result of the production of glaze or glass based on past texts. We also verify a statement in Abulghasim Kashani's treatise. Indeed, for many years, scholars, according to Allan's translation of Kashani's treatise, have believed that the mixing of Shura and Ushnan ashes leads to the destruction of alkaline glazes. However, in this study, it was found, based on our recreation of glazes, that such mixing can give an alkaline glaze. It seems that this discrepancy is due to a mistake in translating a part of Kashani's treatise, which over the years, it had misled various researchers.

In addition, according to the comparison of texts and glazes, it seems that among all the treatises which were introduced during this research, Kashani's treatise is more specialized than others ones. Indeed, in this book, there are some recipes for glazes for which the color is very special, and only in this treatise, we can find the detail of yellow glaze with gold sheets and black color with chromium. Also, in the end, by studying Mina'i glazes as one of the glazes that have a high color variety in the Islamic era, it was found that many texts are in harmony with ancient glazes, and the coloring agents left in the glaze are similar to what is given in Islamic texts.

Chapter Three: Elamite colors and colored materials, meaning and origin

Abstract of the chapter

Color agent in the glaze is one of the main factors. Therefore, the study of the terminology of Elamite colors can provide a clearer perspective for the study of glazes. For this purpose, the terminology of Elamite glazes was studied and it was determined that some of the modern Persian words used in the meaning of different colors have Elamite origins. Also, one of the colored materials that are frequently repeated in Elamite texts can be considered the Marhashi stone. Since Marhashi along with Shimashki were toponyms that provided colored materials and colored stones for Elam, therefore their location has been very important for scholars. In the continuation of the research, an attempt was made to determine the location of ancient Shimashki by using linear Elamite writing. In this way, the eastern regions of Iran are a more suitable area for the Shimashki toponym.

3.1-Introduction

One of the most important goals of archaeology is studying the types of living in ancient times. These types can be categorized into many different subsets in the topic of ancient technology. For example, pottery, metals, glasses, obsidians, and buildings can be categorized into these subsets. These kinds of technologies are important for archaeologists. Moreover, in the meantime, studying colors and pigments have a vital role in the such investigation because colors can be very useful in archaeologists' analysis of ancient people. This is accepted, studying colors are part of cultural artifacts research. With the progress of this subject, the Archaeological journal of Cambridge University assigned several issues to ancient colors and pigments (Gage, 1999). In addition to that, in 2002 a congress was held about the coloring in ancient times (A. Jones & MacGregor, 2002). These kinds of works can be named the first studies on colors in ancient

societies. But this was just the beginning of a such investigation. Recently, a new term boomed in the world of archaeology; this term was “The Archeology of color” (Miller, 2022). But it seems that a close term to Archaeology of color is Anthropology of color, which was used some years before and there is some similarity between them (MacLaury et al., 2007: 8).

Regarding the background of the studies conducted on color in Iran and its neighboring lands, Nagel’s dissertation could be mentioned as it was related to Achaemenid color and pigments (Nagel, 2013). Meanwhile, the study conducted by Warburton, regarding ancient Egypt and Akkadian periods, is also one of the studies related to the present research (Warburton, 2012). Finally, the study related to the terminology of colors in Mesopotamia written by Thavapalan should be mentioned as a land that had a lot of connection with Elam (Thavapalan, 2020).

This research tries to discuss polychrome as a very important part of the archaeology of Elam. Imagining the world of Elam without color can be seen as a betrayal of the artists who made many efforts to produce a colorful world during that period.

3.2-The Terminology of colors during the Elamite period

The Elamite words for colors were often used for expressing the color of clothes. It should also be noted that there is only one dictionary for the Elamite language (Hinz & Koch, 1987), which makes studying Elamite words difficult. Therefore, in the current research, it has tried to use the words related to color in Mesopotamia to study the terminology of colors in the Elam period more accurately.

3.2.1-White color

The earliest word, which can be related to white color in ancient times is “light”. This word signified a term denoting something closer to “white color” (Warburton, 2014: 1). The term which is used in the Elamite era for white is “BABBAR.BABBAR”. This term was also used in Sumerian color terms for white color during the 3rd-millennium BC (Warburton, 2012: 3). Basello believed that this is a Sumerian logogram, which was also used in the Akkadian period (Basello, 2011: 1). But other researches agreed that the white color term which was used during

the Akkadian period was “Pešum” (Warburton, 2014: 3). It was mentioned that the color terms in Elamite era often were used for expressing the color of clothes. For the white color term, this is true, and “BABBAR.BABBAR” was used after “Kuktu” which means dress.

In addition to that, Pesu/ Pesum was used as a term for the white color in the Akkadian period. In the Elamite language, there is a term, which has been suggested by Hinz and Koch, as the translation of roasted flour or brown flour. The mentioned term is “Pistiya” and they mentioned another term “Pisti” without any meaning (Hinz & Koch, 1987: 198). Now in the Persian language, there is a similar term that is related to white color. The term is “Piseh/Pistak” with this meaning in the Dehkhoda Persian dictionary: All the colors are mixed by white color (Dehkhoda, 1998), and in another famous Persian Dictionary, this term is translated as the skin disease with white spots (Moein, 2007). So it seems that “Pisit” without any meaning in the Elamite dictionary can be used for white color during the Elamite era.

3.2.2-Light red color

In all ancient languages, there is a very clear tendency for a division of the reddish continuum of color experiences into several hues (Warburton, 2014: 2). Also, in the Elamite era, it seems there are different terms for different spectra of red color. The term which was used for the light red color in the Elamite era is “Burnu”; Basello believed in that it is possible to compare this term to “Babrunnu” which was used as a term for brown horses in Nuzi, and he suggested that “Bur” was an Elamite term which was used for brown color (Basello, 2011: 1). In addition to that, there is a similar term in Akkadian color terms, “Burumu”, which Warburton suggested could mean blue color (Warburton, 2007: 233). But “Bur” now is common in the modern Persian language as light red color. So it is possible that “Bur” was used in the Elamite era as light red color too. In addition, this term was used for “Tukli” and “Kuktu” with the meaning of clothes and dress. In addition, it seems that there are some special terms for colored clothes during the Elamite period. For example, “dabarrium” is an Elamite form of a Hurrian word “tawarriwa” (Thavapalan, 2020). The New Assyrian form of this word is “tabrimu” which appears in Akkadian texts in “tabarru” form (Thavapaln, 2019: 294). Basello suggested that the Elamite form of this word is “tabarrum”

(Basello, 2011: 2); but in Elamite texts, it seems that “da-bar-ri-um” was used as the meaning of “light red wool” which is similar to Akkadian “tabarru”.

3.2.3-Dark blue color

Some of the color words preserved in the earliest Semitic languages (e.g., uqnu, “lapis lazuli” or “dark blue”) are loanwords for materials from other unknown older languages. In Akkadian (uqnu) and Egyptian (xsbj), terms for lapis lazuli designated “dark blue (Warburton, 2014: 2). In addition to that, in the Elamite language, some researchers believed that “Kapautaka”, designating lapis lazuli, was used for dark blue color too (Tavernier, 2007: 36). Kapautaka: not kaputka, as Scheil normalizes it, which qa-ba-u is related to Skt. kapota- "pigeon, gray (color of a pigeon)", and giving Modern Persian “Kaboud” “gray-blue (Table 3.1) “. Though Scheil, p. 29, calls attention to the fact that this term is now found to include several related varieties of stones (Kent, 1931: 207). On the other hand, there are some Elamite texts with the term “Kaput” without any meaning (Hinz and Koch, 1987: 835). The text is: qa-pu-ut li-ut-tur- $\{e(?)\}$ har-ip-na; this text probably can be useful for dark blue during the Elamite era.

3.2.4-Light Blue/Green color

As mentioned in dark blue color, some colors originated from associations with precious materials. It seems that during the Elamite era the term of turquoise was used for light blue/ green color too.

3.2.5-Black color

In Mesopotamian inscriptions “agurru” is a term for brick which is followed by a metal or color word, the expression signifies “glazed (-colored) brick”. For example, “agurru kaspi ebbi” means, shining (colored) glazed brick (Thavapalan, 2019: 43). In addition to that, in Elamite language “erien” is a term for brick, and in some texts followed by this term” Siyah”. Hinz and

Koch believed that the meaning of this phrase is fired brick (Hinz & Koch, 1987: 181). But, “Siyah” in Modern Persian is used for black color. So, it may be possible that like what happened in Mesopotamian inscriptions for brick and colored brick, happened in Elam too. In other words, probably the phrase “erien siyah” means a black brick that is fired.

3.2.6-Magenta color

In Akkadian texts, “argamanu” is a term that was suggested for purple color. But, this term is very similar to “arghavani” as a modern Persian term for magenta color. In addition to that, Hinz and Koch suggested: “har-qa-ma-an-{na}” as a similar term to “argamannu” which was used for purple color too, in the Elamite language (Hinz & Koch, 1987: 633). So it seems that magenta is a better suggestion for the meaning of this term. It is clear that magenta and purple are very similar, but there are also minor differences between these colors. Also, this term was followed by “kuktu” (which was discussed earlier) in some Elamite inscriptions.

3.2.7-Versicolored

In Akkadian texts, the term which was used for versicolored is “Barmu”, and Thavapalan believed that “Barmu” is not etymologically related to any one of the ancient near east or Mediterranean words to express versicolored or mixed concerning color (Thavapalan, 2019: 79), but in Elamite language “Birmu” is a term which is related to Akkadian “Barmu”. Some Elamite language researchers suggested versicolored as the meaning of “Birmu” (Hinz & Koch, 1987: 211). In addition to that, the “Birmu” is followed by “Kuktu” like some other Elamite words which were related to color.

3.3-Colored materials as the color terms

During the Elamite period, there were colored materials that evoked a certain color in the mind. In other words, it seems that most of the time the Elamites used colored materials instead of words to express color. In this regard, should refer to a text dated to the Middle Elamite period from Chogha Zanbil:

“*a-na-ku mun-taš-DINGIR.GAL i-na li-bi-it-ti ša KÙ.GI ša KÙ.BABBAR (2) ša ^{na}KA ù ^{na}UD.UD.AŠ ku-ku-un-na-a e-pu-uš-ma (3) a-na DINGIR.GAL ù dMÜŠ.EREN ša si-ia-an-ku-uk ad-di-in (...)*”(Malbran-Labat, 1995: 77; Potts, 2010: 50).

“*I, Untaš-Napiriša, built the kukunâ (= el. Kukunum) in (glazed) bricks of **gold, silver, obsidian and alabaster**, and I gave them to the Napiriša and Inšušinak gods of siyan-kuk*“(Basello, 2011: 3).

If pay attention to the translation of the words marked in bold, it is clear that the presence of such words in the description of bricks puts them in the meaning of color. In other words, gold means golden color, silver means silver color, obsidian means black color, and alabaster means white color. However, such uses of colored materials in the meaning of color make research difficult for researchers. Duise (Marhashi) stone should be mentioned as an example. During the Elamite era, some stones were traded between Elam and Mesopotamia, because Mesopotamia was a region that did not have many colored and various stones. “Duhsi/Duhsu” or “Marhasi” is an important example of these colored stones. The ancient Babylonians believed that “Dusu” and “Marhasi” stones came from the land of Marhasi, said to be situated east of Anshan (Talle Malyan in Fars province of Iran) and from the mountain of ZAR.DU.A (Thavapalan, 2019a: 254). In the Elamite language there not has been mentioned to this term yet. But there are two texts, which can be compared with Akkadian texts about “Duhsi/Duhsu”. The mentioned Akkadian text is:

“5/6 MA.NA 7 2/3 SU.KU.BABBAR

1/3 MA.NA 7 1/2 SU KU.SI

1 2/3 SU KU.SI ru-su-um

16 ZA.GIN1 DUH.SU.A pa-ra-ah-si

Si-ip-rum ga-an-rum sa su-ri-in UTM.

The translation of this text is: 5/6 minas and 7 2/3 shekels of silver, 1/3 minas and 7 1/2 shekels of gold, 1 2/3 shekels of red gold, sixteen (a piece of) lapis lazuli, one (a piece of) duhsu –stone from Marhasi, the complete work of an emblem of Samas... (Thavapaln, 2019: 257).

And the mentioned Elamite text is:

“KU.BABBAR.lg.na du-is-se 4 an-qa-ap KU. GI.lg. na-ha-rak.... “.

The translation has been suggested for this text is: Silver, four gold eyelets (?) Pressed onto its border (?) (Koch and Hinz, 1987: 62).

So it seems that these texts have some similarities and it is possible to compare Elamite “Duise” with Akkadin “Duhsi/Duhsu. In addition, there are three forms for this term, the Sumerian form is “DUH.SI.A”, the Hurrian form is “tuhsiwa” and the Hebrew form is “tahas” and the late form of the word in the Neo-Assyrian and Neo- Babylonian period appear without the medical “h” and with the long vowel at the end” dusu” (Thavapalan, 2019a: 244-245).

There is some hypothesis about the color and type of this stone. Steinkeller believed in that “Duhsu” is mineral chlorite, which must be denoted pale green or blue-green (Steinkeller, 2006: 1-5), and it is generally accepted that the color of this stone is pale green (Abrahimi 2014). Oppenheim thought the color of this stone must be greenish-yellow or yellow (Oppenheim, 1948). In addition to that, Thavapalan has suggested that the color of this stone is probably yellow and this stone refers to calcite (Thavapalan, 2019a: 260). It means that the Elamite “Duise” is Alabaster stone that was used during the third Millennium BC. in Mesopotamia, Elam, BMAC⁵, and east of the Iranian plateau. As mentioned before “Duise” is related to Marhashi toponym, and Alabaster stones were found a lot in the east of Iran (from northeast to southeast).

⁵ Bactria-Margiana Archaeological Complex

Most of the researchers who have studied the location of Marhashi have located the area of this name in the east of Iran. But another toponym, which according to many researchers can be located in the east of Iran, is Shimashki. It should be noted that both of these names are part of the regions where the colored materials of Elam were supplied them. These two toponyms have been the main suppliers of gold, silver, tin, and precious stones in Elam and Mesopotamia. Therefore, finding their location is one of the most important things in the present research. In the following, it tried to deal with the new evidence that can help to find these two toponyms and finally the location of the color stone of “Duise”.

For this purpose, linear Elamite writing has been used.

Table 3.1- Elamite words to express colors and compare it with modern Persian words

Elamite White	Elamite Red	Elamite Blue	Elamite Magenta	Versicolored
BABBAR.BABBAR	Burnu	Kaputka	Argamanu	Birmu
Pesu/ Pesum	Bur	-	-	-
Modern Persian White	Modern Persian Red	Modern Persian Blue	Modern Persian Magenta	Modern Persian Versicolored
Piss/Pisit	Bur	Kaboud	Argavani	Rangarang

3.4-Linear Elamite

Linear Elamite is a partially deciphered syllabic writing system invented in <Elam> from 22nd century BC. onwards. It was used to write down royal inscriptions in Elamite language, in contrast to cuneiform writing which prior to <Middle Elamite> period displayed if culturally Elamite, though linguistically Akkadian texts (the only exception being the <treaty of Naram-Sin>

using cuneiform writing for an Elamite content). This dichotomy has led to the hypothesis that LE⁶ was developed to emphasize Elam's cultural unity and independence from Mesopotamia after the inauguration of <Puzur-Inshushinak> at Susa, who had run successful raids to conquer highland Elamite provinces such as Kimash and Shimashki (Kienast & Gelb, 1990: 321-324), and thus seems to have united lowland and highland Elam. An alternative point of view (Tavernier, 2010: 205) is that by creating a genuinely Elamite script besides the cuneiform writing borrowed from Mesopotamia, Puzur-Inshushinak aimed to emphasize the two axes, i.e. lowlands and highlands, of his new kingdom.

3.5-Text Corpus

A total of 51 (41 if collated) LE inscriptions or fragments of inscriptions are known today (OCLEI 2020). They comprise a total of 1731 sign tokens dispensed to 99 sign types. Each sign type has 1 to 9 graphic variants, the champion being sign 81𐎠 with its variant forms 𐎠;𐎠;𐎠;𐎠;𐎠;𐎠 and 𐎠 (Plachtzik et al., 2017: Tab. 7). However, this proposed syllabary is merely a working hypothesis, as with every progressive step in decipherment, not to speak publications of new inscriptions, its scores as well as its token/type relation will slightly change. Of the 51 texts and fragments, 44 are written in sinistroverse and 7 in dextroverse direction (in this article, sequences are depicted dextroversely for reasons of comparability). The LE text corpus can be classified into three sub-corpora, each being coherent in itself but differing considerably from the others. While on a graphemic level all three use the same syllabary, distinct patterns are applied on a syntactic level, i.e. the three sub-corpora choose different sign sequences (parts of speech) to combine them in a different order. Thus, only a few short sequences such as 𐎠𐎠𐎠 (with its supposed graphical variants 𐎠𐎠𐎠; 𐎠𐎠𐎠 in Susian texts and 𐎠𐎠𐎠; 𐎠𐎠𐎠; 𐎠𐎠𐎠; 𐎠𐎠𐎠; 𐎠𐎠𐎠 in Central Elamite texts, see Desset, 2018: Fig. 20) with a total of twelve occurrences of this example, appear across the sub-corpora, while within one sub-corpus sequences as long as 18 signs are repeated in different inscriptions (Mäder et al., 2017: 70). On a grammatical level though, one feature is shared by all three sub-corpora: the use of common suffixes which appear to be linguistically Elamite (see below "Commonly accepted sound values"). This leads to the assumption that all three sub-corpora

⁶ Linear Elamite

represent the Elamite language or an idiom closely related to it, but refer to different topics, mention different kings or gods and follow different formulae and scribal traditions, not to mention the geographical and presumed diachronical gap between them.

3.5.1- The Western Elamite (Lowlands) Sub-Corpus

This sub-corpus, coined Susa in OCLEI, consists of 18 stone or clay inscriptions (with a total of 553 signs) excavated at Susa. The length of the texts reaches from 3 (^{Susa}T) to 66 (^{Susa}H) signs. Most are royal inscriptions often mentioning Susa's protection god Inshushinak together with king Puzur-Inshushinak as a donator or founder. As they persistently allude to this Susa-based Elamite ruler who came to power 2150 BC., this sub-corpus is the only one to be dated with certainty. It is probably older than the other two and marks the beginning of LE writing tradition.

3.5.2- The Central Elamite (Highlands) Sub-Corpus

The second sub-corpus, coined Mah, Phoe, Schø, Time, and Pers in OCLEI, consists of 24 inscriptions or fragments (with 1133 signs totally) offered by the Mahboubian, Phoenix Ancient Arts, Schøyen, and Timeline Auctions collections, or, in the case of ^{Pers}Q, found in a grave near Persepolis. While for the Phoe, Schø, and Time group no original provenance is indicated, the Mah inscriptions are claimed to originate from Kam-Firouz in the north of Shiraz, Iran (Desset, 2018: 109). The highland inscriptions, all accurately imbossed on hammered silver vessels (so-called <Gunagi> jars), are likely to be royal inscriptions as well, as they mention the kings <Ebarat> and <Shilhaha> according to an unconfirmed reading proposed by Desset (2018: 132–133). If the readings of these royal names turn out to be true, the Central Elamite sub-corpus must be dated to the early Sukkalmahat, i.e. up to two centuries later than the Susian texts.

3.5.3- The Eastern Elamite (Elamo-Bactrian / Elamo-Jiroftian) Sub-Corpus





The third sub-corpus consists of only eight short inscriptions, the length of which ranges between two and eleven signs: ^{Sha}S is incised on a pot's rim excavated at <Shahdad>. ^{GonurP'} is a controlled surface find from <Gonur Depe> and was incised before firing on a locally manufactured <Namazga> V–IV sherd (Kločkov, 1995: 54), which makes it autochthonous. ChrisG' is a gold seal from a private collection with a typical Bactrian bird-man and the four LE letters  on it. ^{Liga}V is a steatite seal of the Dilmun / Persian Gulf / Elamo-harappan (Ascalone, 2011: 420) type with the letters  incised, linking it linguistically, though not iconographically, to the before mentioned Bactrian seal ChrisG'. Compared to the latter, it shows only minor graphic variation and the lack of the suffix sign.  (For an attempted, but unconfirmed interpretation of these two inscriptions see Tab. 2.). The Eastern Elamite sub-corpus further includes the tablets found at Jiroft (^{Jir}). These are unusual insofar as a LE word (name?) of four to eleven signs seems to conclude or subscribe, be it at the last line or the reverse of each tablet, a text written in a mysterious and undeciphered notation system. The latter is sometimes called a "writing system" (Desset, 2014), though, regarding the fact that only 12 different sign types (added by 6 diacriticized, i.e. dotted, variants) are present, it remains doubtful whether it depicts the sounds of human language or it is to be considered as another notation system, be it numerical or other. At any rate, the present LE sub-corpus builds a bridge between Elam and its Eastern neighboring cultures. It is also the only sub-corpus with a distinctive feature on the palaeographical level: Instead of the angular shape  known from the other areas, Elamo-Bactrian texts consistently have the rounded form for this suffix sign (Mäder, 2020: Tab. 1). Although several inscriptions came to us by regular archaeological activity (i.e. ^{Sha}S, ^{GonurP'}, and, though with doubts, the Jiroft / Konar Sandal tablets), no closer dating frame than 2150–1900 BC. can be given.

Table 3.2- Statistical properties of the three geographical/chronological sub-corpora.

	Western Elamite (Lowlands)	Central Elamite (Highlands)	Eastern Elamite (Bactria / Konar Sandal)	Overall
Date	2150 BC	1950-1900 BC (?)	2200-1900 BC	2150-1900 BC
Provenance sigla of archaeologically reported texts/fragments (number)	Susa (18)	Pers (1)	Sha / Jir / Gonur (6)	25
Provenance sigla for texts/fragments stemming from art collections (number)	---	Mah / Phoe / Schø / Time (24)	Liga / Chris (2)	26
Number of texts counting each fragment	18	25	8	51
Number of texts collating fragments (thereof dextroverse texts)	18 2 ^(SusaB; SusaP)	25 2 ^(MahYb; MahN')	8 3 ^(JirB'r; JirC'r; ChrisG')	41 (7)
Readable signs	553 (32%)	1133 (65.5%)	45 (2.5%)	1731 (100%)
Average text length	31	45	6	28
Longest text (readable signs)	SusaH(66)	MahZ(160)	JirD(11)	
Material	Stone / Clay	'Gunagi' Silver Vessels	Clay / Steatite / Gold	
Persons mentioned	Puzur- Inshushinak,	Ebarat (?), Shilhaha (?)	Title of an unnamed high official (?)	

	Shinpi-Ishhuk			
Gods mentioned	Inshushinak	Napirisha	---	
Places mentioned	Susa	---	Shimashki (?)	
Purpose	Royal Inscriptions	Royal Inscriptions	Signatures/Labels/Seals	

3.6-Shimashki toponym, and Eastern Elamite sub-Corpus

Shimashki is one of the place-names related to Elam, and since 1960, many researchers have been looking for its location. Therefore, various opinions have been expressed about the location of Shimashki. Some researchers believe that Shimashki must be located somewhere in the eastern neighborhood of the third series of Ur (Mesopotamia) and accessible to them (Aarab, 2017). Walter Hintz considers the location of Shimashki to be the mountains of Lorestan in the north of Khuzestan, and more precisely, today's Khorramabad city (Hinz, 1962: 3-12). Kupper considers Shimashki to be the northern neighbor of Khuzestan (Kupper, 1969: 27). De Miroschedji also mentions the mountains in the east and northeast of Susa as the place of Shimashki (De Miroschedji, 1980: 137-138). Herzfeld considers Shimashki as a region between Golpayegan and Isfahan city (central Iran) (Herzfeld & Walser, 1968: 179-180). Also, Zadok mentions the area between Persia and the Caspian Sea as Shimashki (cited by Potts, 1999: 103) and Vallat considers the Kerman province region as the place of Shimashki (Vallat, 1985: 50-52). These cases are only part of the hypothesis about the name of Shimashki and its location.

Shimashki is a region that consists of six important cities. These six cities are Zabšali, Šigriš, Yabulmat, Alumiddatum, Karta, and Šatilu (Potts, 1999). Also, Shimashki and Marhashi are the toponyms that have a high potential for Elam and Mesopotamia in preparing metal ores (Steinkeller, 1988: 199). What should be mentioned about metal mines is that Mesopotamia and Elam are not very rich in terms of the capacity of metal mines. Therefore, for the supply of metal materials, trade connections with other regions have been the only solution (Kohl et al., 1978). But the two documents that have been examined in the present study (Figures 1 and 2) may include the name of Shimashki. Of course, this issue is raised only as a hypothesis here. In this way, a sign for the phonetic value of "Shi" has already been discovered "𐎶" (Table 3) and it has been used in

these two seals. Also, the sign that should be considered as "Ki" in these two stamps to obtain the phrase Shimashki or a phrase close to it, is "𐎧". While this sign is very similar to the sign "𐎧" mentioned for the phonetic value of "ir/ri" in Table 3, their differences are clear. However, they may have the same phonetic value. But regarding the "𐎧" sign, it should be said that it was mostly in the suffix position (mostly in the suffix position at the end of inscriptions and sometimes at the end of a word (Mäder et al., 2017). Now Elamite cuneiform inscriptions have been found where "Ki" is used as a suffix in Elamite writing. So it is possible that the sign "𐎧" can be considered as a suffix. So far several suffixes of linear Elamite writing have been identified (Mäder et al., 2017: 84), but the suffix "Ki" is still not known. Therefore, there is a possibility that "𐎧" is "Ki". If this assumption is considered correct, then the phrase engraved on the golden seal (Figure 1) is the same as Shimashki, and the other phrase engraved on the Persian-Gulf seal (Figure 2) is Shimash. Shimashki was discussed before. But regarding Shimash, it should be said that this toponym appears only once in the written sources of the Neo-Assyrian period, and that was in the time of Sargon II. In that inscription, Sargon II mentions the range of his conquests from Kashmar to Shimash and mentions the location of Shimash near the far eastern Medes (Luckenbill, 1927: 61).

But are Shimash and Shimashki the same? This claim was first stated by Herzfeld and Walser (1968: 180). Of course, in his stele of 716 BC, Sargon II made another reference to the distant lands of Eastern Media, and, in that stele, he mentions the place name "Sigris" as one of those places (Levine, 1973). The similarity of the name "Sigris" in the new Assyrian period with "Šigriš" of Elam as one of the six main regions of Shimashki, which was explained above, is a topic that has led some researchers to consider the two toponyms as one (Hinz, 1972). Since both documents are among the prominent seals, the first one is a golden seal with the image of the bird-man of the 3rd and early 2nd millennium BC engraved on it along with the possible name of Shimashki (Figure 3.1), and the second seal is one of the seals known as Persian Gulf along with the possible name of Shimah (Figure 3.2).

Regarding the first seal with the image of a bird-man holding two goats in his hands, it should be said that this role has a significant frequency in the BMAC culture and Eastern Iran in the 3rd and 2nd millennium BC, and there is also a similar role on many seals (Figure 3.3) (Aruz, 1999: 18). In general, the image can be introduced with three elements in its details: a) The muscular body of the bird-man and emphasizing it, b) The presence of a shawl on the waist of this bird-man, which was one of the common cases in this era. c) There is a circular necklace on the neck of this

bird-man. Both items (a) and (b) are seen in abundance in BMAC culture and chlorite stone works of Jiroft (Figure 3.4), but item (c), i.e. a necklace with a large circular ring, is found only in Jiroft chlorite stone motifs (Figure 3.5). This type of necklace is emphasized in Jiroft chlorite stone motifs, and sometimes this emphasis is added with a decorative stone. By doing p-XRF analysis on these necklaces, it was found that this emphasis was not only in terms of appearance but also in terms of the type and material used; this stone is a type of turquoise stone, which was considered a valuable stone in that period. In addition, such a necklace has not been reported in this category of motifs in BMAC culture. However, regarding seal number 2, it should be said that such seals are known as Dilmun, Indus, and Persian Gulf seals. The seal discussed in this research is of the type of seals of the Persian Gulf, the geographical distribution of this type of seal is high and has been obtained from Kerman, Susa, Western Iran, Indus Valley, Bahrain, Kuwait (Failaka), Emirates (Tal Abraq) and Mesopotamia. This seal is a unique example of a seal in the collection of Dilmun, Indus and, Persian Gulf seals, and it has not been reported so far. The only seals that have a significant similarity with the seal in question are two other seals that were also obtained from Iran and are of the type of Persian Gulf seals that have written inscriptions on them (Figure 3.6). Of course, one of the other essential things in the studied seal is the object that is at the bottom of the image and in front of the cow's mouth. The presence of such an object among all the seals of Dilmun, Indus and, Persian Gulf types was observed only in two examples of Indus seals (Figure 6). Although according to a not very accurate and reliable report, the location of the studied seal was reported to be in the western region of the Iranian Plateau (Winkelmann, 1999), the similarities of this seal with the seals of the eastern region, including the Indus Valley as stated above, is undeniable. Also, the study of the stone sources of Dilmun, Indus and, Persian Gulf seals shows that the stone source of the studied seal, which is a type of chlorite stone, does not exist in Central Asia and the BMAC cultural area, nor in the west of Iran (Olijdam & David-Cuny, 2018: Fig 5). All the mentioned cases, in the end, show the greater similarity of this seal to the seals of the eastern regions. In addition, the extensive trade connections between Elam and BMAC culture is a topic that Potts also mentioned in detail (Potts, 2008). Also, according to the above, a sample of Elamite writing has been obtained from the site of Gunur Tepe in Central Asia, which itself can be another sign of this strong connection between Elam and Central Asia in the studied period. In addition, the existence of rich connections between the eastern regions (Kerman) and northeastern Iran (BMAC culture) is a topic that many researchers have discussed (Kuz'mina, 2007). It should

be noted that the seal is a commercial item and there is a possibility of finding it in any region that was part of the trade network in the late third and early second millennium BC. However, by looking carefully at the seals, what seems clear is the similarity of their artistic style with eastern regions such as BMAC culture, Harappa-Mohenjo Daro, and most importantly Kerman civilizations, which makes it possible that they were made in these regions.



Figure 3.1- A golden seal with the name Shimashki, and the image of a bird-man and its representation in OCLEI (elamicon.org)



Figure 3.2- Persian Gulf seal with the name Shimashki and its presentation method in OCLEI (elamicon.org)



Figure 3.3- The image of man-bird in Kerman (left) and BMAC culture (right) (Winkelmann, 1999: 21)



Figure 3.4- Right side, BMAC ax with emphasis on muscles and shawl on waist (Metropolitan Museum); The left side of the Jiroft civilization soapstone (chlorite) vessel with emphasis on the muscles and shawl on the waist (Perrot, 2008: Plate1)

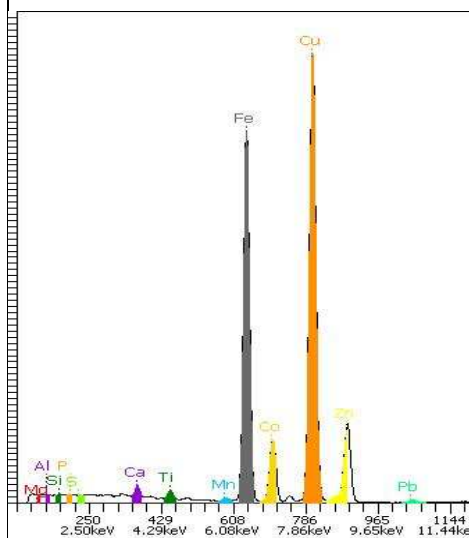
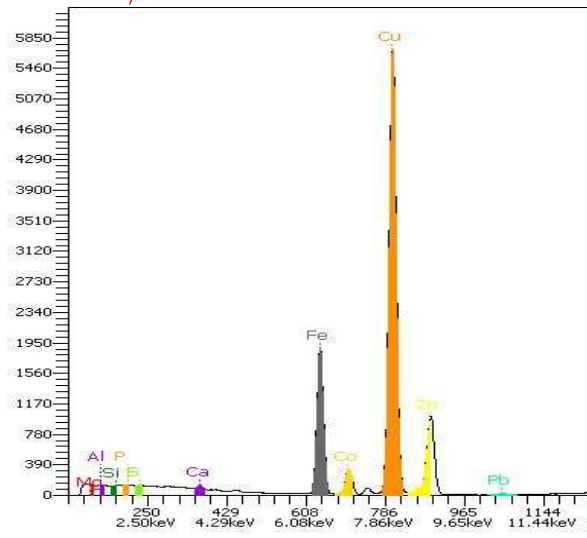




Figure 3.5- The recurring image of the necklace in the motifs of the so-called wrestlers who lift two animals with both hands and its emphasis in the motifs of Jiraf vessels and the use of turquoise stone in the necklace.

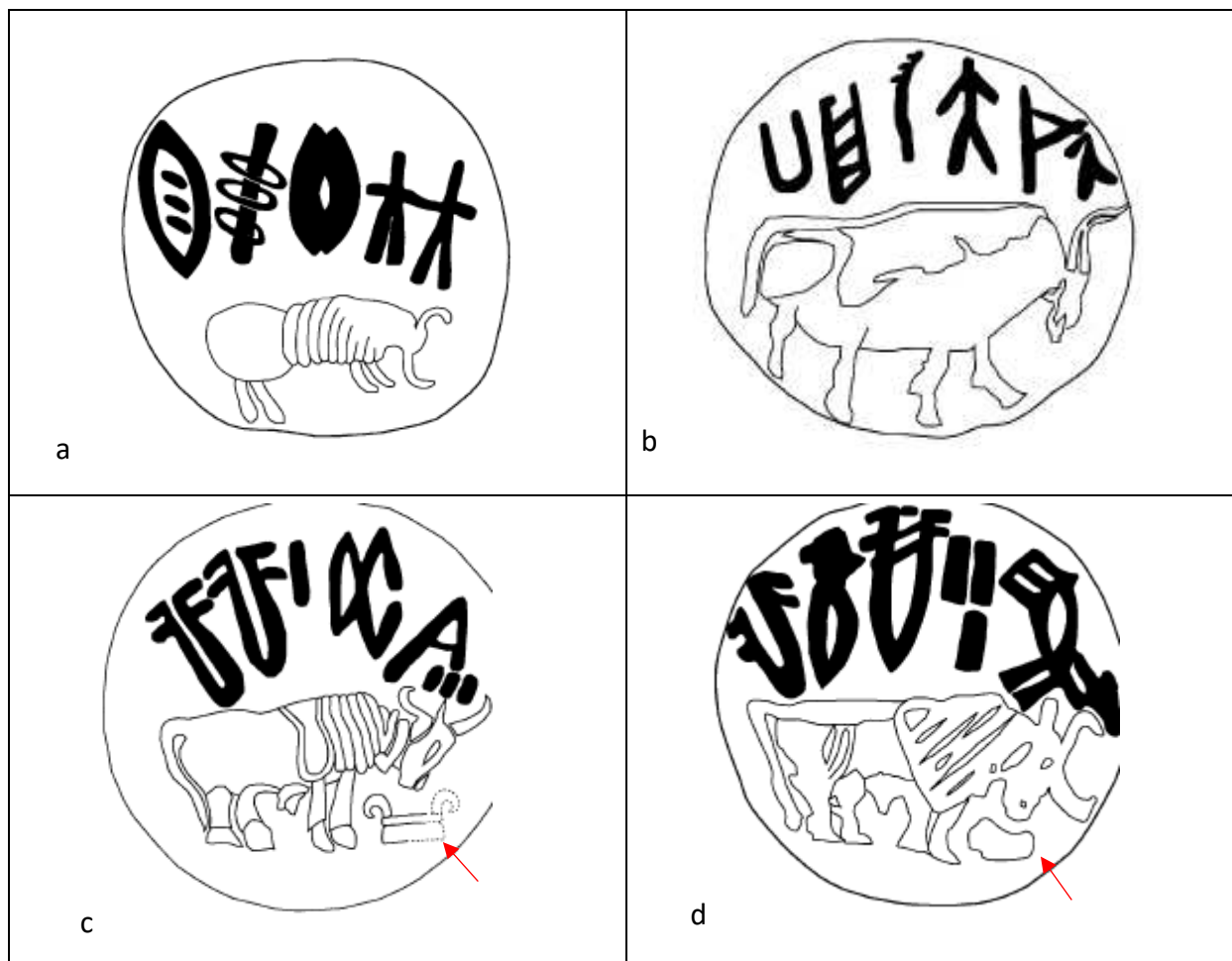


Figure 3.6- (a) and (b) seals of the Persian Gulf type with Indus inscriptions obtained from western Iran; (c) and (d) Indus Valley seals and an object in front of the cow's face (Laursen, 2010: 111).

3.7-Authenticity of the Artefacts

As Tab. 2 shows, 26 out of 51 texts or fragments came to us from private collections and thus raise doubts being modern forgeries. The most common way to provide (circumstantial) evidence is to check if the syntactical pattern (rank-frequency distribution, suffix position probabilities, etc.) of a text in question matches the statistics established from archaeologically secured inscriptions. Such examination turned out negative for ^{Time}M' (even distribution instead of a near-hyperbolic rank-frequency distribution according to Zipf's law), and ^{Phoe}W (unexpected behaviour of suffix signs, and, as an additional problem, the use of pseudo-variants, e.g. 𑀵𑀺𑀓𑀺 for the otherwise well-

known 𐎶𐎠𐎫 hal-me-ni-k sequence, despite the fact that 𐎶 and 𐎠 are two different types with different syntactical functions; a typical forger's error) (Mäder et al., 2017: 94). The most common, and maybe only way to provide hard evidence in favour of the authenticity of an artefact, is to identify fairly specific sign sequences that otherwise occur exclusively on inscriptions published a good time after the artefact in question. This has been done for ^{Phoe}A', which furthermore displays the paradigmatic behaviour of typical suffix signs including the facultative application of the suffix me besides the consequent application of all other suffix signs (ibid.: 91ff.). For the 24 ^{Mah} inscriptions, no positive authenticity proof can naturally be established as they have been the latest to come to light. Desset (2018) reaches a positive conclusion after metallurgical examination and coherent syntactical behaviour of sequences across the Central Elamite sub-corpus. Finally, there remains doubt concerning the excavation reports of the Jiroft tablets (Muscarella, 2018).

3.8-Historical Descendance

A genealogical derivation from <Proto-Elamite> is to be considered from a theoretical as well as from an archaeological standpoint: In theory of writing systems, only four to seven scripts are endogenic, i.e. have been invented independently from any existing ancestor (Woods, 2010: 15), all of which are logo-syllabic. A stimulus-free development of a syllabic script like LE is thus unseen in history. Archaeologically, the only known predecessor with Elamite background are the Proto-Elamite administrative texts. A palaeographical comparison between LE and <Proto-Elamite> reveals a formal identity of a number of xx sign types, i.e. x% of the 99 LE sign types and x% of the 393 Proto-Elamite sign types. Furthermore, the feature of inscribed signs known from Proto-Elamite is also applied in LE (see below Table 3.5).

Sound value attributions for 15 signs have been proposed based on the detection of proper names (PN). All commonly accepted sound attributions rely on the sole bilingua ^{Susa}A, the LE version of which happens to translate the first column of the akk. version. As the relevant proper names ^{NAP}Inshushinak; Puzur-I(n)shushinak; halmenik-Shushenri a) occur in the expected order judging from the composition of the akk. version, b) are intentionally not interrupted by line breaks, and c) use syllables which do partially overlap, these sound values (Table 3.3) were accepted early in LE research. Three PN identifications, ^{NAP}Inshushinak, Puzur-I(n)shushinak, and

Shinpi-hi-Ishhuk, had been proposed by Bork (1905: 327-328), and got slightly modified by Frank (1912), and Meriggi (1971: 206). The halmenik-Shushenri sequence was offered by Hinz (1962: 6) as soon as the translation of akk. ensi as el. halmenik 'governor' was certain enough.

Table 3.3- Commonly accepted sound values

Proper name induced from Akkadian version of Susa A	Sign	Accepted sound value
Based on the ^{NAP} Inshushinak Sequence (𐎠𐎢𐎣𐎤𐎥𐎦) Susa A I 5-10	𐎠	NAP (divine determinative)
	𐎢	in
	𐎣	shu
	𐎤	shi
	𐎥	(u)sh
	𐎦	na
	𐎧𐎣	k
Based on the Puzur-I(n)shushinak sequence (𐎠𐎢-𐎠𐎢𐎣𐎤) Susa A II 1-7	𐎠	pu
	𐎢	uz/zu
Based on the halmenik-Shushenri sequence (𐎠𐎢𐎣𐎤- 𐎠𐎢𐎣𐎤) Susa A III 1-8	𐎠	ha(l)
	𐎢	me
	𐎣	ni
	𐎤	she
	𐎥	en
	𐎦	ir/ri

In recent times, and largely due to the publication of the Mahboubian texts, some of these sound values have been corroborated by syntactic evidence: A) The orthographic ambiguity of the shu-shi/(u)sh-na-k sequence (𐎠𐎢𐎣𐎤 *shu-shi-na-k* occurring in 9 different inscriptions vs. 𐎠𐎢𐎣𐎤 *shu-(u)sh-na-k* found in 4 different inscriptions) perfectly replicates the orthographic variation known from cuneiform Elamite. In consequence, 𐎥(u)sh must be categorized as commonly accepted as well. B) Quantitative syntactic analysis (see below) identified 6 signs with a highly elevated probability of being suffixes as they are over-represented at word end positions: 𐎢;𐎣; 𐎤; 𐎥 𐎣 and 𐎦. Four of them have accepted sound values which had been established independently based on PN: 𐎥(u)sh 𐎦 ir/ri 𐎣 k and 𐎢 me. At the same time, -sh, -r, -k, and -me are indeed suffix syllables in cuneiform Elamite (see Table 3.4). Such a triangular relationship of evidence is

hard to explain by coincidence. C) The value *me* for 𐎠 is confirmed by the fact that both the cuneiform Elamite syllable *me* (the suffix for abstract nouns) and the LE sign 𐎠 are optional in their actual use: Cuneiform Elamite shows free variation between *kušik-me u-me* vs. *kušik u-me* 'my construction' or *sunki-me u-me* vs. *sunki u-me* 'my kingdom'. This pattern is reflected in LE sequences like 𐎠 𐎠 𐎠 vs. 𐎠 𐎠 𐎠 (Mäder et al., 2017: tab. 12-14).

Table 3.4- Additional sound value proposals

Sequence	Tentative reading	Tentative translation	Attestations	Contributor	Sign	Proposed sound value
𐎠𐎠-𐎠𐎠𐎠𐎠	Puzur I(n)shushi-Nak TEM _{TI} -x-x	PN followed by male determinative	8	(Desset, 2012: 19)	𐎠	TEM _{TI}
𐎠𐎠𐎠	tu-ni-sh	'he dedicated'	3	(Meriggi, 1971: 207)	𐎠	tu
𐎠𐎠 vs. 𐎠𐎠 𐎠𐎠𐎠 vs. 𐎠𐎠𐎠 𐎠𐎠𐎠-𐎠-𐎠𐎠𐎠	x-(u)sh 𐎠 x-ish xx-(u)sh vs. xx-ish ... Shi-in-pi-hi-ish-hu-k	Verbal suffix <i>ish</i> in paradigmatic distribution with <i>ush</i> (3. sg. perf.). (PN; Father of Puzur-I.)	21+8	(Mäder et al., 2017: 88; 62)	𐎠	ish
𐎠𐎠𐎠-𐎠-𐎠𐎠𐎠 𐎠𐎠𐎠-𐎠-𐎠𐎠𐎠	Shi-in-pi-hi-ish-hu-k or Shi-in-pish-huk-sha-a-k	(PN; Father of Puzur-I.) or 'PN's son'	8	(Bork, 1905; Vallat, 2011: 188)	𐎠	hi / huk
					𐎠	hu / (h)a
					𐎠	pi / pish
𐎠𐎠𐎠	ta-k-ki-me	'life'	3	(Mäder et al., 2017: 70)	𐎠	t(a)
					𐎠	ki
𐎠𐎠(𐎠)𐎠𐎠	Na-pi-(ir-)ri-sha	DN	9(3)	(Desset, 2018)	𐎠	pi
					𐎠	ri
					𐎠	sha
𐎠𐎠𐎠	Shi-(i)l-ha-ha	PN	3	(Desset, 2018: 133)	𐎠	(i)l

𐎧...	ú (text initial pronoun)	'I'	5	(Desset, 2018: 138)	𐎧	u
𐎧...	e (text initial exclam.)	'oh!'	6	(Desset, 2018: 132)	𐎧	e/ia
𐎧𐎧𐎧𐎧	E-ba-r-at	PN	3	(Desset, 2018: 132)	𐎧	ba
					𐎧	at
𐎧𐎧𐎧𐎧	^{HAL} Shi-mash-ki	GN	2	(Mäder, 2020: 6)	𐎧	Hal
					𐎧	mash
					𐎧	ki

Table 3.5- The six signs with the highest suffix probability, i.e. the clearest statistical overrepresentation at word ends. Adducing the accepted (= unmarked) or proposed (= marked with "?") sound values from Table 3.1 or Table 3.2 respectively, they happen to match the Elamite suffix syllables known from cuneiform texts.

Sign	Accepted or proposed values	Linguistic suffix known from Cuneiform Elamite
𐎧	k	-k (Class suffix 1. sg. / suffix for perf. participle)
𐎧	ta?	-t (Class suffix 2. sg.)
𐎧	ri/ir	-r (Class suffix 3. sg.)
𐎧	me	-me (Class suffix for inanimate nouns)
𐎧	(u)š	-š (Verbal suffix 3. sg. perf.)
𐎧	ih/iš ?	-h (Verbal suffix 1. sg. perf.)

3.9-Possible Interference between Shape and Sound

Desset (2018: 140; Fig. 45) adduces shu; (u)sh; sha; shi, pointing to the fact that the graphical shape (lines and dots) may relate to their phonetic values (palatal sibilant). Similar claims have been made by Hinz (1969) for inserted signs (Table 3-6). Though Hinz' sound value proposals are outdated, it is still conceivable that such signs might be somehow phonetically or functionally related:

period both from a linguistic and archaeological point of view. Shimashki is a place that has been introduced as a very influential area in the studied periods and countless scholars have tried to locate it, and different areas have been suggested for this important place. However, one of the most important pieces of evidence that can always be a guide in the location of ancient toponyms is the inscriptions that were obtained from a specific geographical area and contain the name of that place. Of course, in this research, only one hypothesis was proposed that it is possible to consider two seals related to Shimashki toponym. In this way, the eastern regions of Elam were probably more connected with Shimashki than other places. Now it is a little early to say that Shimashki is located in the east of Elam. However, such cases can indicate the importance of studying the linear Elamite writing for a more accurate reconstruction of the Elamite periods.

Chapter Four: Combined studies on glazed ceramic bodies of Middle and Neo Elamite periods (1500-539 BC)

Abstract of the chapter

XRD analysis was done on ceramic bodies and based on the PCA plot on the XRD results, ceramic bodies can be divided into four groups. A group of ceramics has a high amount of quartz, and these samples are placed in the category of siliceous pottery based on petrographic studies. Mordenite was also identified in one of the samples of this group, which was previously reported in Elamite ceramics. Another category is calcareous ceramic bodies, in which pieces of primary calcite have also been observed. Thus, it can be said that their firing temperature was probably less than 800 degrees centigrade. Also, according to the EPMA results, it was found that the amount of calcium in these samples is more than in other samples. Also, based on petrography, another group of ceramic bodies can be placed in the voids-plant matter group. Most of the ceramic bodies of Middle Elam were placed in this group. Also, it is possible to raise the possibility of firing twice or different firing of some ceramic bodies due to the presence of cristobalite in the samples of the fourth category.

4.1-Introduction

Elam is an artificial construct, a name coined by Mesopotamian scribes, gazing across the alluvium towards the Iranian plateau, that designates the disparate regions of highland southwest Iran and its peoples (Figure 4.1). In Sumerian sources dating to the middle of the third millennium BC, the name Elam was written with the sumerogram NIM meaning simply 'high', often accompanied by the determinative KI denoting land and country. Elam appears in the Elamite language as ^{hal}*Hatamti*, *hal Hatamti*, or *Hatamti* (D. T. Potts, 1999). Walther Hinz suggested that this term was composed of *hal*, meaning 'land', *tamt*, meaning 'gracious lord' (Hinz, 1963). The Elam period is of great importance considering that it was the beginning of the entry of some parts of the Iranian plateau into the historical period. It seems that the Middle Elamite period (1400-

1200 BC) is the first time that artisans used glazes on ceramics in Iran, an industry that conveys an important aspect of artistic and architectural developments in Mesopotamia and Iran. Therefore, studying the glazed ceramics of this period is of utmost importance. So far, some analysis has been done on Elamite unglazed ceramics (Emami & Trettin, 2012). However, recently, a study has been conducted on the glazed ceramic bodies of Chogha Zanbil from the Middle Elamite period. The authors concluded that the ceramic bodies were produced from calcareous clays rich in fragments of limestone, flint, grog, and organic temper, and they mentioned that the glaze was added to the biscuit-fired bodies, not the dried green bodies (Raith et al., 2022).

One of the available documents to obtain information on the raw materials and the elaboration processes of the ceramics of the Elamite period is the written sources that have survived from that period. In the Elamite language, there are different words to describe baked and mud ceramic bodies, which are mainly used to introduce mudbricks, baked bricks, and glazed bricks. Information about these words is provided in Table 4.1 (D. T. Potts, 2010). The most astonishing point about the frequency of different words for the ceramic bodies referring to Elamite bricks is the variety of terms that were used. This linguistic diversity in a period underlines the diversity in the ceramic production techniques in Elam. For instance, in the twelfth century BC, King Shutruk Nahunte I (~1184-1150 BC) considered himself the creator of a new brick processing, which is called *akti*, also known as siliceous bodies (G. P. Basello & Rossi, 2012). Therefore, the difference in ceramic glazed can be one of the main questions of this research. Also, since in this research only the Elam period is considered, it is possible to pay attention to the elaboration techniques and firing conditions of glazed ceramic bodies from the Middle Elamite period to the Neo Elamite period.

The purpose of this study is to compare ceramic bodies of glazed samples from the Middle Elamite period to the Neo Elamite period (about 1000 years) in Iran. In this way, we can also talk about the process of making ceramic bodies of glazed samples in Iran. Samples that can be considered the first glazes on ceramic bodies in Iran, and therefore it is very important to examine the techniques of making, chemical composition, and firing temperature of their ceramic bodies.

Table 4.1- Terms used in the Elamite language to designate different bricks (D. T. Potts, 2010)

Period	Mudbrick terms	Baked brick terms	Glazed brick terms	Siliceous bricks
Old Elam	-	erientumimma	-	-
Middle Elam-I (1400-1200 BC)	-	upat hussipme	upqumimma, ubqumiya	akti
Middle Elam-II (1200-1100 BC)	Halatimma, halatni	upatimma, erientum-ia, erientumma	upat aktiya kuramma karrah	upat aktini

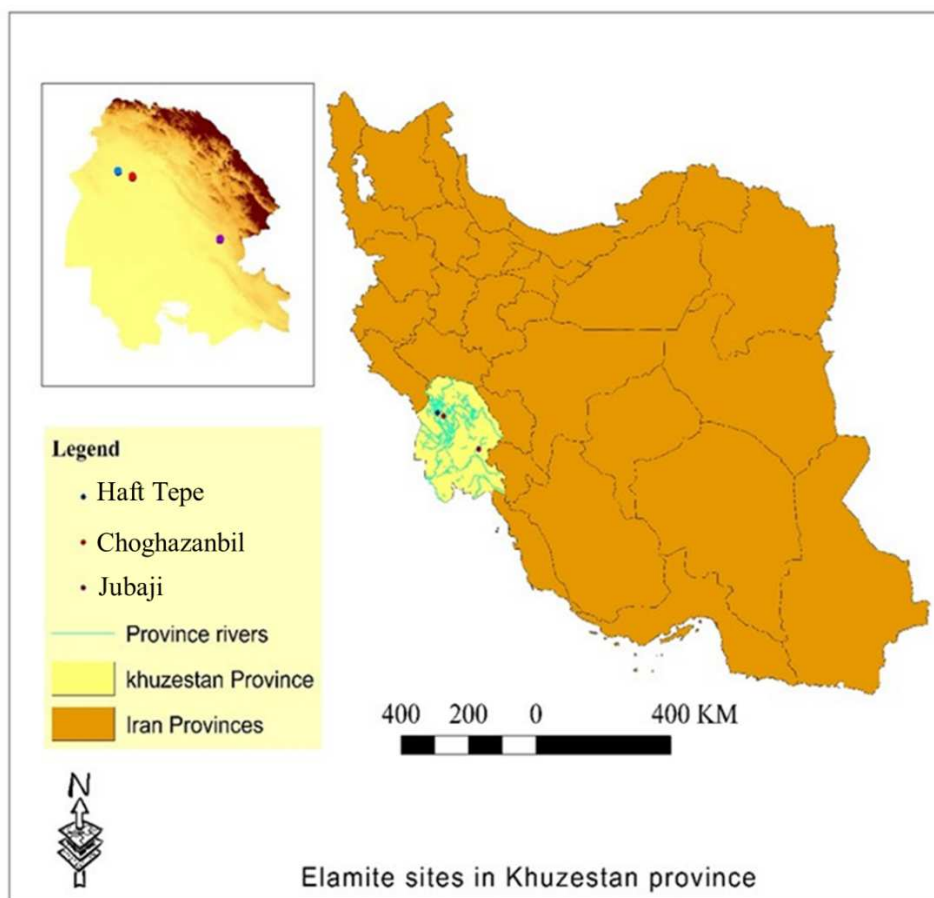


Figure 4.1. Location of studied Elamite sites








4.2- Materials

To study glazed ceramic bodies related to the Elamite period, 29 samples were collected from three index sites of the Elamite period (Haft Tepe, Chogha Zanbil, and Jubaji) in Khuzestan province in southwestern Iran (Figure 4.1). Regarding the studied samples, they can be considered related to the two periods of Middle Elam and Neo Elam. Information about the approximate time of the studied ceramics and the number of samples is given in Table 4.2. All glazed ceramic bodies are from archeological excavations in the Khuzestan province of Iran. Samples of the Chogha Zanbil ziggurat have been collected during archeological excavations conducted by Ghirshman (Ghirshman, 1961), and Neo Elamite glazes are obtained from Jubaji site excavations (Ahmadinia & Shishegar, 2019). The glazed ceramic bodies of Haft Tepe site were obtained from the

archeological excavations under the directorship of Negahban (Negahban, 1991). Regarding Haft Tepe glazed ceramics, it should be said that these samples can be considered one of the first ceramic glazes in Iran. Apart from these three sites, Susa is another Elamite site with glazed ceramics. However, one should consider the dating of glazed Elamite artifacts at Susa has always been problematic because of the disturbance of the stratigraphy of the Elamite layers (Holakooei, 2014).

Regarding the first glazes in the ancient world, it seems that there was a close connection between the first glazes and metal smelting furnaces. For example, in the site of Tell Abu Matar in the Middle East (valley of Beersheba), copper smelting leads to the formation of a glaze at the surface of the oven, which is reminiscent of the earliest glazes that have a blue color due to copper (Peltenburg, 1971). It was then suggested a relationship between the metalworkers and the first glaze makers. Also, archeological excavations in Haft Tepe have reported evidence of the use of glaze on ceramic bodies (Negahban, 1991). Although their use seems very limited, according to the evidence obtained so far, the spools from the Haft Tepe related to Tepti Ahar (king of Elam from the 15th century BC) (G. Basello, 2012) can be considered the first use of glazed objects in the Elamite period. Two samples of Haft Tepe, investigated in the present study, are spools found in the Elamite layer of Haft Tepe. But even in Haft Tepe, samples of glazed bricks have not been obtained, only the small glazed objects have been obtained, and thus the beginning of using glazed bricks can be considered related to Chogha Zanbil. In this study, five glazed bricks of Chogha Zanbil have also been studied. The specimens of Jubaji are all obtained from Elamite tombs, and all belong to the vessels buried with the deceased buried in the grave. To study the samples more efficiently, the abbreviation of ancient sites was used in this research. Thus, “CZT” has been used as an abbreviation for Chogha Zanbil, “HT” as an abbreviation for Haft Tepe, and “J” as an abbreviation for Jubaji.

Table 4.2- Sample of glazed bodies studied in the present study. CZT, J and HT indicates samples fro Chogha Zanbil, Jubaji and Haft Tepe, respectively.


Number	Sample	Possible use	Glaze color	Period	Photo
1	CZT-4007	Building decoration	White	Middle Elam (1300 B.C)	
2	J- 2505	Pottery	Yellow/White	Neo Elam (540 B.C)	
3	CZT-4012	Building decoration	Blue/White	Middle Elam (1300 B.C)	
4	J- 7009	Pottery	Green	Neo Elam (540 B.C)	
5	CZT-4011	Building decoration	White	Middle Elam (1300 B.C)	
6	J- 2009	Pottery	Green	Neo Elam (540 B.C)	
7	J -2001	Pottery	White	Neo Elam (540 B.C)	
8	J- 2005	Pottery	Blue/White	Neo Elam (540 B.C)	

9	J- 7008	Pottery	Blue	Neo Elam (540 B.C)	
10	J- 2607	Pottery	Yellow/White	Neo Elam (540 B.C)	
11	CZT-4004	Building decoration	Blue	Middle Elam (1300 B.C)	
12	CZT-4002	Building decoration	White	Middle Elam (1300 B.C)	
13	CZT-4010	Building decoration	Blue	Middle Elam (1300 B.C)	
14	J- 2003-a	Pottery	White	Neo Elam (540 B.C)	
15	J- 2501	Pottery	Green	Neo Elam (540 B.C)	
16	CZT-4006	Building decoration	White/Blue	Middle Elam (1300 B.C)	

17	HT-1016	Spool (knob)	White/Blue	Middle Elam (1500 B.C)	
18	HT-1001	Spool (knob)	White	Middle Elam (1500 B.C)	
19	CZT-4005	Pottery	Blue	Middle Elam (1300 B.C)	
20	CZT-4013	Building decoration	White/Black	Middle Elam (1300 B.C)	
21	CZT-4001	Pottery	White/Blue	Middle Elam (1300 B.C)	
22	J2003-b	Pottery	Yellow/White	Neo Elam (540 B.C)	
23	J- 2004	Pottery	Yellow	Neo Elam (540 B.C)	
24	CZT-4009	Building decoration	Blue/Green	Middle Elam (1300 B.C)	

25	CZT-4014	Building decoration	Blue	Middle Elam (1300 B.C)	
26	J- 2503	Pottery	White/Blue	Neo Elam (540 B.C)	
27	J- 2007	Pottery	Green	Neo Elam (540 B.C)	
28	J- 2502	Pottery	White/Yellow	Neo Elam (540 B.C)	
29	J- 2006	Pottery	Blue	Neo Elam (540 B.C)	
30	J-2002	Pottery	Blue/White	Neo Elam (540 B.C)	
31	QA-3001 ⁷	Building decoration	Blue	Iron Age (850 B.C)	
33	CZT-4008	Building decoration	Blue/White	Middle Elam (1300 B.C)	

⁷ This sample were collected from Qalaichi site in the northwest of Iran.

34	J-2008	Pottery	Green/Yellow	Neo Elam (540 B.C)	
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4.3- Methods

4.3.1- X-ray diffraction

To perform X-ray diffraction (HT-XRD) measurements, we have used a PANalytical X'Pert PRO (Amelo, Netherlands) diffractometer. A Co K α radiation operates at 40kV and 40mA. Diffraction patterns were collected using a X-Celerator detector over an angular range $10^\circ \leq 2\theta \leq 90^\circ$, using an acquisition time of 45 min. For all samples, the zones for XRD analysis were chosen from the middle part of the ceramics and finely crushed. XRD is an essential tool in mineralogy for identifying, quantifying, and characterizing minerals in complex mineral assemblages. Its application to ancient ceramics, which are a mixture of clay minerals, additive minerals, and their transformation products, yields information on the mineral composition of objects (Stanjek & Häusler, 2004). Diffractogram analysis was performed using X-Pert High Score Plus (Panalytical) software to semi-quantitatively identify the crystalline phases from their peak locations and intensities using ICDD patterns. The data obtained from the X-ray diffraction analyses of the samples were submitted to the multivariate statistical procedure of principal components analysis (PCA). This multivariate analysis technique was used due to the fact that it provides an exceptionally useful tool for grouping samples together and illustrating links between variables (Holakoei, 2016). PCA was prepared based on HighScore Plus software.

4.3.2- Electron Probe Micro Analysis

EPMA (Electron Probe Micro Analysis) was used to investigate the chemical composition of the 20 selected ceramic bodies. A CAMECA SX50 electron microprobe, equipped with wavelength-dispersive spectrometers (WDS), at the Camparis Center (Sorbonne Université, Paris, France), was used. Two operating conditions were determined: 15 keV accelerating voltage and 10 nA beam current for main elements (Na, Mg, Al, P, K, Ca, Ti, Fe: $K\alpha$) and 25 keV – 299 nA for minor elements. The beam was focused to a spot of 40 μm for point analysis and limited counting time was used to limit the alkali migration. Standards used for calibration were albite, diopside, orthoclase, apatite, MnTi, Fe_2O_3 , PbS, BaSO_4 , FeS_2 , Cr_2O_3 , NiO, ZnS, SrSiO_3 , ZrSiO_4 and metals (V, Co, Cu). Samples were mounted in epoxy resin, polished and then carbon-coated. Ten points of measurements were taken at random in the ceramic bodies to give an average value of the chemical composition.

4.3.3-Thin-section petrography

Another analysis performed on the Middle and Neo Elam samples was a petrographic study for which 12 thin sections were prepared for the study, and the samples were studied through a polarizing microscope. The standard thickness for a thin section is 30 microns, which has been used in this study. Quartz is often used as a reference material to ensure the sample material is at the appropriate thickness. Quartz is both abundant and its optical properties are well-known, making it an excellent reference source for the pottery.

Ceramic petrography incorporates methodology from sedimentology and sedimentary petrography, such as the description of particle shape and texture. Ceramic petrography draws upon principles from the microscopic study of soils or ‘soil micromorphology’ to describe the nature of the matrix as well as the pores or ‘voids’ that occur in ceramic artefacts (Quinn, 2013).

4.3.4- Scanning Electron Microscope

Scanning Electron Microscope (SEM) analysis was used to observe the microstructure of bodies and glazes. The instrument was a Zeiss Ultra55 equipped with a field emission gun. The analysis was performed only on samples with a different structure from the rest of the samples based on the petrographic study. SEM micrographs have been principally used to examine firing temperature and structure of potteries (Maniatis & Tite, 1981; Freestone & Middleton, 1987). Energy Dispersive Spectroscopy (EDS) microanalysis has also been widely applied in identifying ancient bodies and glazes constituents and in determining their chemical compositions qualitatively and quantitatively (Holakooei, 2016). EDXS point analysis was performed using an EDXS QUANTAX system equipped with a silicon drift detector XFlash 4010 (Bruker).

4.4- Results

4.4.1- X-ray diffraction analysis

X-ray diffraction analysis (XRD) was conducted on 29 glazed ceramics to determine the mineralogy of the bodies. The crystalline phases of Middle Elamite and Neo Elamite samples were searched and selected via their best-preferred orientation by means of the 2θ position on diffractograms (Figures 4.2, 4.3). The major crystalline phases in the matrix are quartz, diopside, enstatite, calcite, akermanite, albite, and gehlenite. In the Elamite glazed ceramics, quartz is the basic component of all samples. Diopside and enstatite, which are members of the pyroxene group, develop in increasing amounts above a temperature of 840 °C (Dirksen & Bally, 1997). Diopside drastically appears at 950°C (Moroni & Conti, 2006) and their main diffraction peaks increase significantly at higher temperatures (Cultrone et al., 2001). It was proposed that the presence of diopside in the ceramic bodies could be associated with carbonates (maybe dolomite) in the clayey material. The reaction of carbonates with quartz can allow the formation of diopside at high temperatures (Holakooei, 2016). In addition, crystal structures resembling enstatite are formed from Mg-silicates, like talc, or clay minerals, such as Mg-rich smectite, via a topotactic phase transformation at temperatures above 700°C to 1000°C (Rösch et al., 2009). From 850°C gehlenite

starts to crystallize as revealed by XRD patterns (Rathossi & Pontikes, 2010). In lamps fired at a temperature higher than 1000°C or for prolonged retention at the maximum temperature, the gehlenite content is expected to have diminished as it reacts with residual quartz to give anorthite and wollastonite (Rathossi et al., 2017). Akermanite is stable between 744°C and 875°C only (Dirksen & Bally, 1997). Javanshah believed that the presence of albite in ancient ceramics indicates that the samples were fired around 900°C (Javanshah, 2017). However, albite, like quartz and feldspars (orthoclase and microcline), can be present at all temperatures because they are resistant even at high temperatures (Portilla-Mendoza et al., 2020). In addition, albite undergoes a triclinic-to-monoclinic phase transition near 980°C leading to the formation of “high albite” which has a disordered Al-Si arrangement (in contrast to the “low albite”) (E. Grammatikakis et al., 2019).

According to the minerals observed in XRD analysis, it seems that there are different firing temperatures for the ceramics and, in a general view, the presence of these minerals is a signature of a firing temperature between 750°C to 1000°C. According to the above, it can be said that the firing temperature in samples J-2502, J-2002, J-2009, and J-2007 is higher than the rest of the samples. Indeed, high-temperature minerals such as diopside have been identified in these samples, and some minerals that are stable in lower temperatures, such as akermanite and calcite, have not been identified. In addition, the XRD pattern in samples CZT-4006, CZT-4013, and J-2003 is completely different from the others. However, to clarify the issue of the firing temperature and different structures of the samples, it is necessary to perform other analyses on the samples, which will be discussed below.

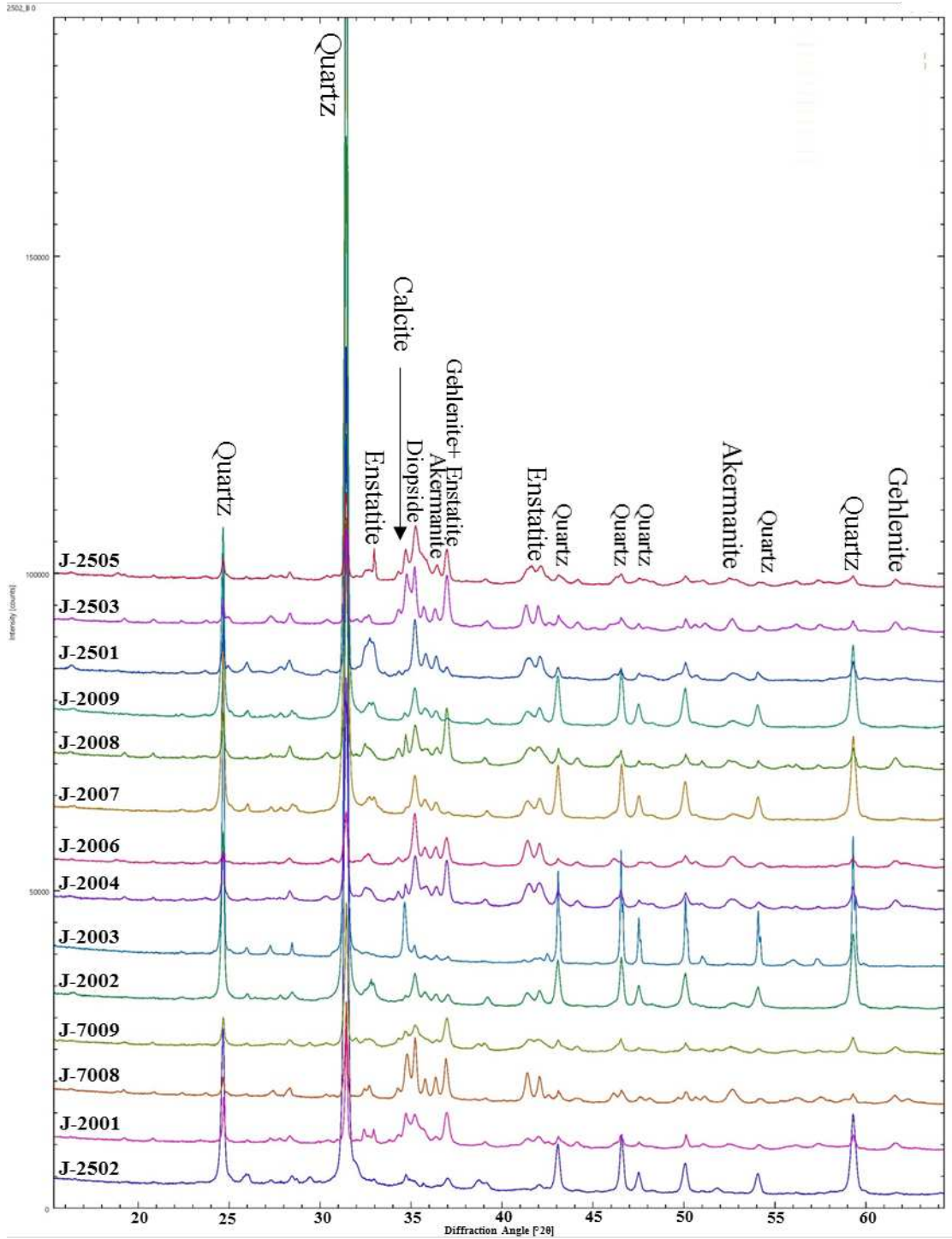


Figure 4.2- XRD diffractograms obtained on ceramic bodies of the Neo Elamite samples from Jubaji with the crystalline phases.

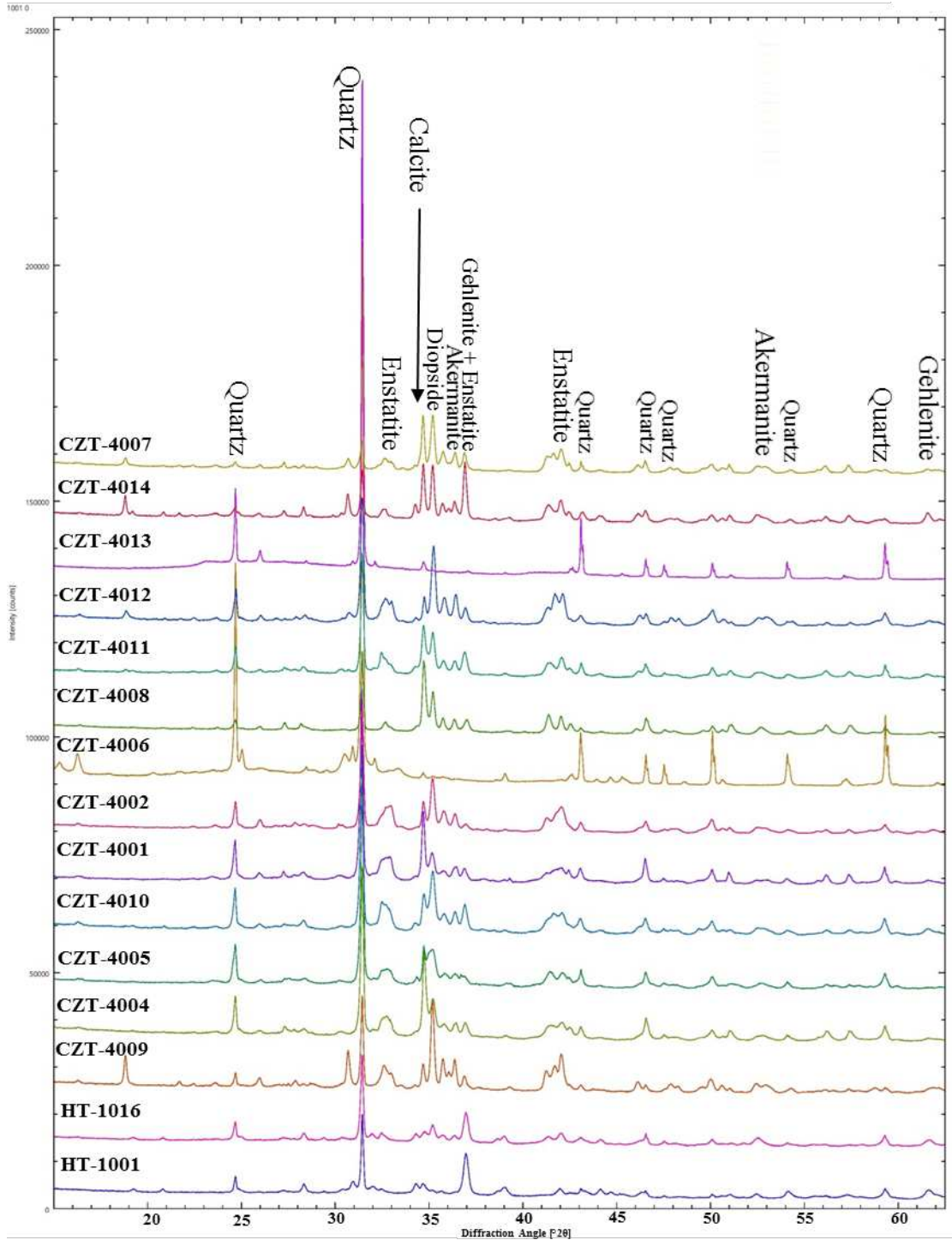


Figure 4.3- XRD diffractograms obtained on ceramic bodies of the Neo Elamite samples from Chogha Zanbil and Haft Tepe with the crystalline phases.

4.4.2-Thin section petrographic investigation

For optical mineralogy, thin sections of ceramic fragments were prepared and their structure and composition were observed with a polarizing microscope (Olympus), which uses transmitted plane-polarized (PPL) or cross-polarized (XP) light.

According to the petrographic analysis, all the samples were divided into two main categories based on their composition and texture. The first category is the moderately sorted texture which indicates that the mineral sizes are similar, and the second one is poorly sorted angular texture which indicates that the mineral sizes are mixed. In other words, the ceramic samples can be categorized into four different groups: siliceous type, quartz-mudstone tempered type, voids-plant matter type, and calcareous type (Figure 4.4).

4.4.2.1-Siliceous type

This group includes three samples (CZT-4006, CZT-4013, and J-2003 in figure 4.4), and its mineralogy is characterized mainly by the presence of quartz fragments. The size, angular shape, and distribution of quartz crystals are different in these ceramics. In other words, the size of quartz in CZT-4006 is bigger than in other samples, and quartz in two samples J-2003 and CZT-4006 has sharper angles than sample CZT-4013. In addition to quartz fragments, there are some needle-shaped crystals in these samples, and the nature of these needles was one of our questions about this group of fabrics. Due to this feature, this group of ceramics was classified in poorly sorted angular texture. Siliceous bodies were used in Elam in a local manifestation as architectural decor. This type of building decoration remained characteristic of Elamite architecture until the end of the Neo-Elamite Period (c. 1000–540 BC) (Fügert & Gries, 2018). It seems that this word (siliceous) or similar words such as quartz-based include a wide range of ceramic bodies of the Elamite period (Roach, 2009; Basello & Rossi, 2012).

4.4.2.2-Quartz-mudstone tempered type

This group consists of two samples (J-2009, and J-2502 in figure 4.4) and both of them are dated to the Neo Elamite period. They are composed of angular mudstone fragments and quartz as

temper. In this fabric group, the proportion of quartz crystals is higher than in other moderate texture groups.

4.4.2.3-Void-plant matter type

Most of the Chogha Zanbil samples can be categorized in this group (for example, samples CZT-4004, and CZT-4011 in figure 4). Petrographic images identified the frequent use of tempering agents, namely organic matter, which, however, decayed or burnt out. In this group, some voids are characteristic of tempering with plant matter. Organic inclusions sometimes show preserved vegetal microstructures (plant remains), and we observed burnt traces of these organic matter in the ceramic. But numerous grains are only made of opaque black matter featuring micro-cracks and are hollow in the middle (carbonaceous residues) (Fronteau & van den Bel, 2021). The high quantities of organic matter in the Chogha Zanbil samples may suggest it was added to the clay by potters at this site.

4.4.2.4-Calcareous type

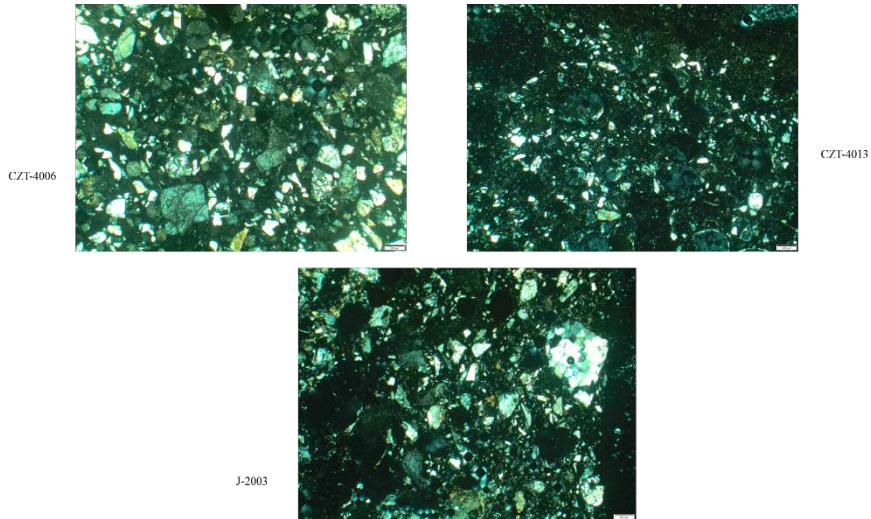
This group can be divided into two subgroups. Two samples of the first subgroup are shown in figure 4 (HT-1001, and J-7009) in which limestone fragments are used as a temper. The second subgroup is composed of samples without limestone fragments but with calcareous fabric, as an example sample J-7008 in figure 4. In this fabric group and the previous one (void-plant matter type), some primary and secondary calcite crystals were determined (according to the characteristics of primary and secondary minerals in petrography). Generally, primary calcite is associated with a rather low firing temperature (less than 750°C) which does not allow the calcite to be de-carbonated. But the dimensions of the grains and the firing atmosphere have a strong influence on the calcite stability, so it can also be found when firing temperatures reached up to 800°C. Secondary calcite, formed after the ceramic firing, may be reformed (re-carbonated) calcite, precipitated calcite, or calcite from alteration (Fabbri et al., 2014). The origin of these secondary calcite crystals is diverse and several pathways for their crystallization are proposed in the literature (Rathossi et al., 2017):

(i) filtration of calcareous aqueous solutions during burial conditions, followed by the recrystallization of secondary calcite,

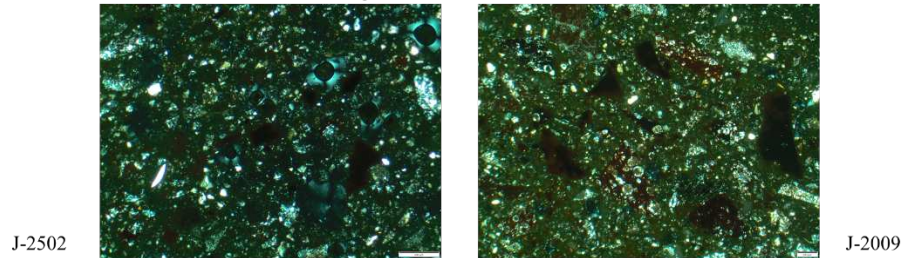
(ii) secondary calcite formed by the decomposition of gehlenite under post-burial chemical leaching,

(iii) re-carbonation of portlandite $[\text{Ca}(\text{OH})_2]$ generated by the hydration of unreacted lime (CaO).

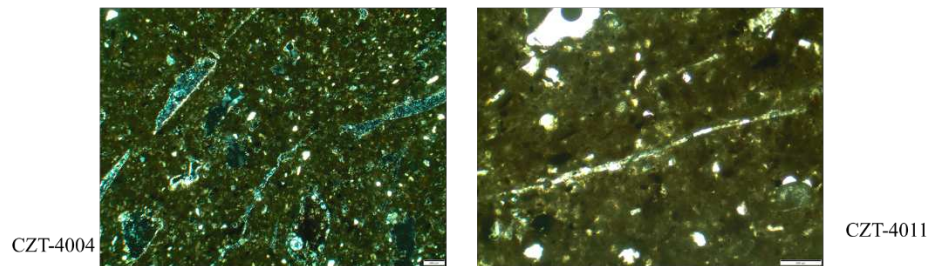
Siliceous fabric



Quartz-Mudstone fabric



Voids-Plant matter fabric



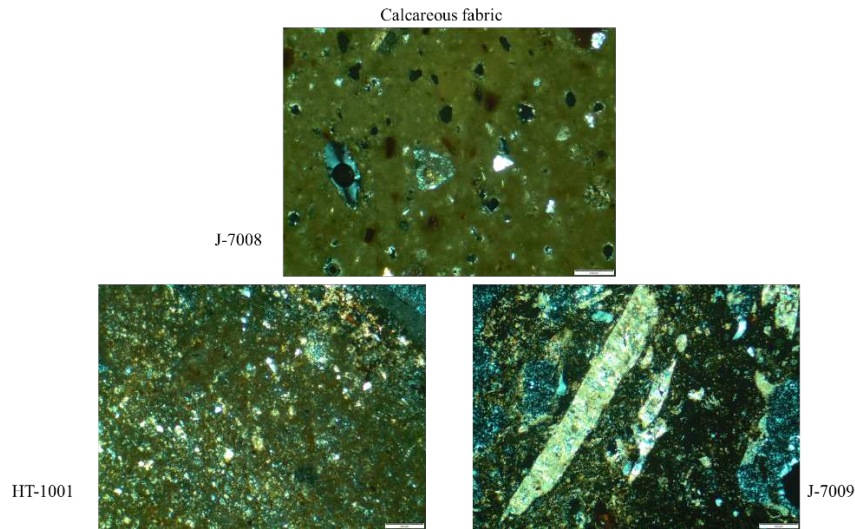


Figure 4.4- Petrographic images showing different types of ceramic bodies.

4.4.2- Electron Probe Micro Analyzer

EPMA analysis was performed on the samples to investigate the chemical composition of the samples. According to Table 4.3, related to EPMA measurements on ceramic bodies, the first important result is the high content of SiO₂ in seven samples (J-2002, J-2003, J-2502, J-2009, J2007, CZT-4006, and CZT-4013). This is interesting because these seven samples correspond to the quartz-mudstone tempered fabric and the siliceous fabric according to the petrographic analysis above. To better understand the difference between the ceramic bodies of the Elamite period, the plot of Al₂O₃, SiO₂, and MgO + CaO for the samples were drawn according to the EPMA results (Figure 4.5). In this ternary diagram, the four samples (CZT-4006, CZT-4013, J-2502, and J-2003) identified are separated from the other ones at the top of the plot, in the SiO₂-rich domain. In other words, these seven samples are located beyond the conode quartz/anorthite or close to it which is related to the non-calcareous field (Noll & Heimann, 2016).

Also, in the SiO₂/Al₂O₃/(CaO+MgO) ternary diagram described by Noll (p224 in (Noll & Heimann, 2016)), the conode triangle diopside/wollastonite-gehlenite-anorthite is related to the very lime-rich samples. Three samples (HT-1001, HT-1016, and J-2001) are located in this triangle (Figure 4.5). Sample HT-1001, according to the petrographic analysis, contain a significant number of limestone fragments (the first group of calcareous type). Then, it is clear that the EPMA

results and $\text{SiO}_2/\text{Al}_2\text{O}_3/(\text{CaO}+\text{MgO})$ ternary diagram confirmed the petrographic results. A salient feature that emerges from the ternary diagram is an important scattering of samples along the SiO_2 - $(\text{CaO}+\text{MgO})$ line. Such an observation indicates that the raw clayey matrices have rather constant Al_2O_3 contents and the differences in chemical composition are mainly due to variations in the $\text{SiO}_2/(\text{CaO}+\text{MgO})$ ratios within the matrices (Heydarian et al., 2020).

According to the proportion of calcium in the samples, it is clear that the percentage of CaO in samples CZT-4006, J-2003, and CZT-4013 (Siliceous fabric group based on petrography) is less than 6wt%. In addition, these three samples cannot be categorized as calcareous potteries (Maniatis, 2009; Maniatis & Tite, 1981; Madrid i Fernández & Sinner, 2019). However, according to the XRD diffractograms, the amount of amorphous phase in these samples is higher compared to other samples. Indeed, in the case of pottery made from calcareous clays, the development of vitrification is inhibited by the formation of crystalline phases with high melting temperatures, that is calcium aluminosilicates (anorthite, gehlenite) and calcium/magnesium silicates (wollastonite, diopside) (Maniatis & Tite, 1981).

Table 4.3- Chemical composition of the samples (wt%)

sample number	sample name	MgO	Al ₂ O ₃	Si ₂ O	CaO	P ₂ O ₅	K ₂ O	TiO ₂	FeO	Petrographic fabric	MgO + CaO
2	J- 2505	4.54	11.20	55.9 2	14.6 8	0.44	1.90	0.66	6.21	NS ⁸	19.2 2
3	CZT- 4012	6.12	12.06	53.2 7	17.8 7	0.47	0.99	0.69	6.44	NS	23.9 9
4	J- 7009	4.37	12.99	48.1 5	22.1 8	0.55	2.43	0.67	5.83	Calcareous fabric-1	26.5 5
5	CZT- 4011	5.97	10.39	47.9 5	25.9 8	0.74	1.21	0.56	5.66	Voids-Plant matter fabric	31.9 4
6	J- 2009	3.59	8.15	68.3 7	9.92	0.28	0.98	0.31	6.10	Quartz-Mudstone fabric	13.5 1
7	J- 2001	5.13	13.77	43.0 0	25.6 2	0.78	2.89	0.56	6.11	NS	30.7 5
8	J- 2005	12.63	13.04	45.4 4	14.8 0	0.61	1.63	0.55	6.21	NS	27.4 3
9	J- 7008	3.15	13.74	50.9 0	19.4 9	0.45	5.35	0.46	3.91	Calcareous fabric-2	22.6 4
10	J- 2607	5.59	12.75	46.4 7	21.8 3	0.63	2.17	0.64	6.85	NS	27.4 2

⁸ Not Studied

11	CZT- 4004	6.24	12.36	50.2 2	19.1 6	0.58	1.88	0.64	6.68	Voids-Plant matter fabric	25.4 0
12	CZT- 4002	6.10	11.30	49.5 7	21.8 5	0.59	1.65	0.76	6.40	Voids-Plant matter fabric	27.9 5
13	CZT- 4010	6.64	12.73	46.8 3	23.1 1	0.67	1.74	0.63	6.29	Voids-Plant matter fabric	29.7 5
14	J- 2003 (a)	2.56	6.39	77.1 1	4.59	0.54	2.72	0.22	2.59	Siliceous fabric	7.15
15	J- 2501	4.16	12.05	57.4 9	12.3 5	0.53	1.70	0.72	5.67	NS	16.5 2
16	CZT- 4006	3.04	1.30	69.7 0	5.55	0.29	3.25	0.06	1.45	Siliceous fabric	8.59
17	HT- 1016	4.04	9.05	43.9 8	34.4 6	0.85	0.98	0.35	4.44	Calcareous fabric-1	38.5 1
18	HT- 1001	3.52	11.42	44.0 0	27.2 4	0.82	2.48	0.76	5.48	Calcareous fabric-1	30.7 7
19	CZT-4005	4.32	11.09	60.6 2	11.0 3	0.39	2.08	0.75	6.28	NS	15.3 5
20	CZT-4013	2.11	2.44	85.5 0	4.74	0.12	0.68	0.17	2.03	Siliceous fabric	6.85
21	CZT-4001	5.69	10.57	62.5 6	10.4 2	0.41	1.86	0.67	6.27	NS	16.1 1
22	J-2003 (b)	1.51	4.38	83.1 6	5.46	0.16	1.44	0.18	1.72	Siliceous fabric	6.97
23	J-2004	4.40	12.86	56.9 3	13.6 9	0.44	3.39	0.94	6.06	NS	18.0 9
24	CZT-4009	6.67	11.72	46.2 1	23.6 0	1.00	1.03	0.91	6.11	NS	30.2 6
25	CZT-4014	7.21	11.53	48.4 0	20.8 4	1.02	1.37	0.96	5.90	NS	28.0 5
26	J-2503	3.98	12.02	48.4 8	22.1 0	1.26	2.56	0.98	6.25	NS	26.0 8
27	J-2007	5.63	7.28	65.2 5	11.3 7	1.50	1.23	0.47	4.97	NS	17.0 1
28	J-2502	1.77	5.16	75.8 2	9.22	0.90	1.29	0.24	3.63	Quartz- Mudstone fabric	11.0 0
29	J-2006	2.56	12.37	51.9 4	21.1 4	1.20	3.10	0.54	4.20	NS	23.7 0

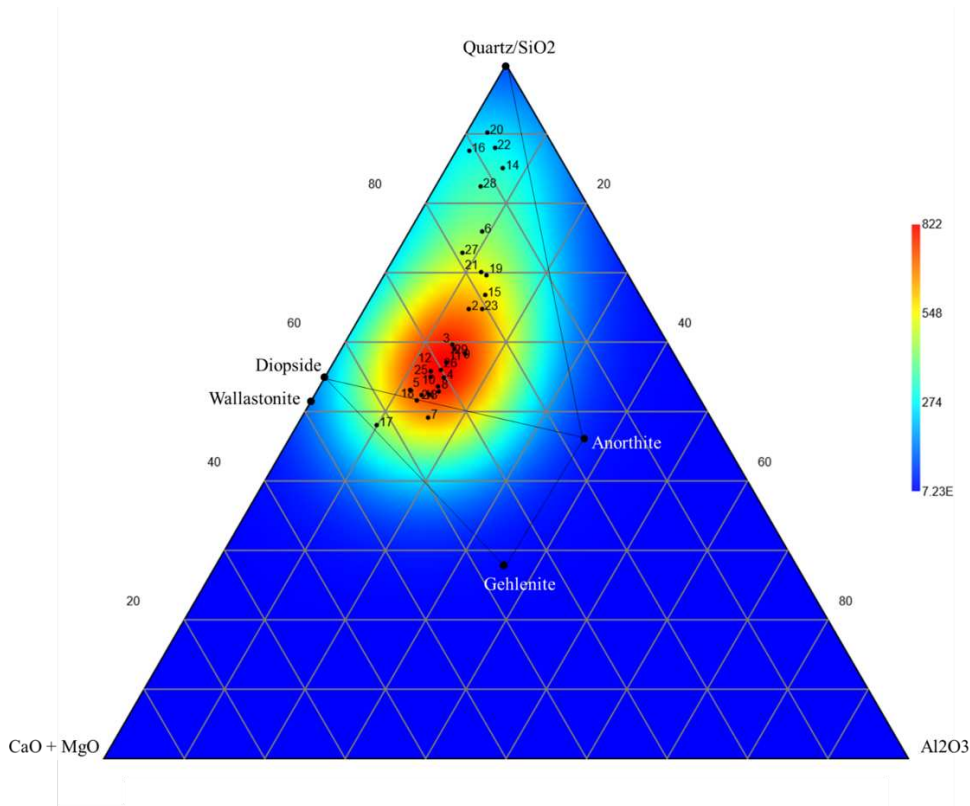


Figure 4.5- Bulk chemical composition of the different analyzed ceramic bodies plotted on SiO₂-Al₂O₃-(CaO + MgO). Source: After Noll & Heimann, 2016.

4.5- Discussion

A Principal components analysis (PCA) study was performed on the results of XRD patterns of the studied samples (Figure 4.6). Highscore plus software was used to prepare PCA diagrams on the samples. The PCA method in studies of ceramic bodies on XRD results can lead to a better understanding of the classification of samples. This is a method aiming to compress the original, *n*-dimensional hyperspace into a new principal component (PC) space of reduced dimensionality. All the original data points are projected in the new PC space and their new co-ordinate values are called scores or principal components (PCs) (Papachristodoulou et al., 2019). According to the PCA diagram on the ceramic bodies of Middle Elam and Neo Elam, the samples can be divided into four groups. The largest group, marked in green in Figure 4.6, includes 11 samples from Middle Elam and 7 samples from Neo Elam. However, the other group, marked in red in Figure 4.6, has 4 samples and the samples are only from the Neo Elam, and the third group, marked in

blue in Figure 6, which is very close to the previous group, consists of 3 samples. This latter group includes samples with the numbers CZT-4013 and CZT-4006 and J-2003. The last group, which is marked in gray color in Figure 4.6, consists of 4 samples, and this group is closer to the green group in comparison to other ones. The PCA results are also shown in the form of a dendrogram with the names of the samples in Figure 4.7, through which the relationship between the samples can be more clearly understood.

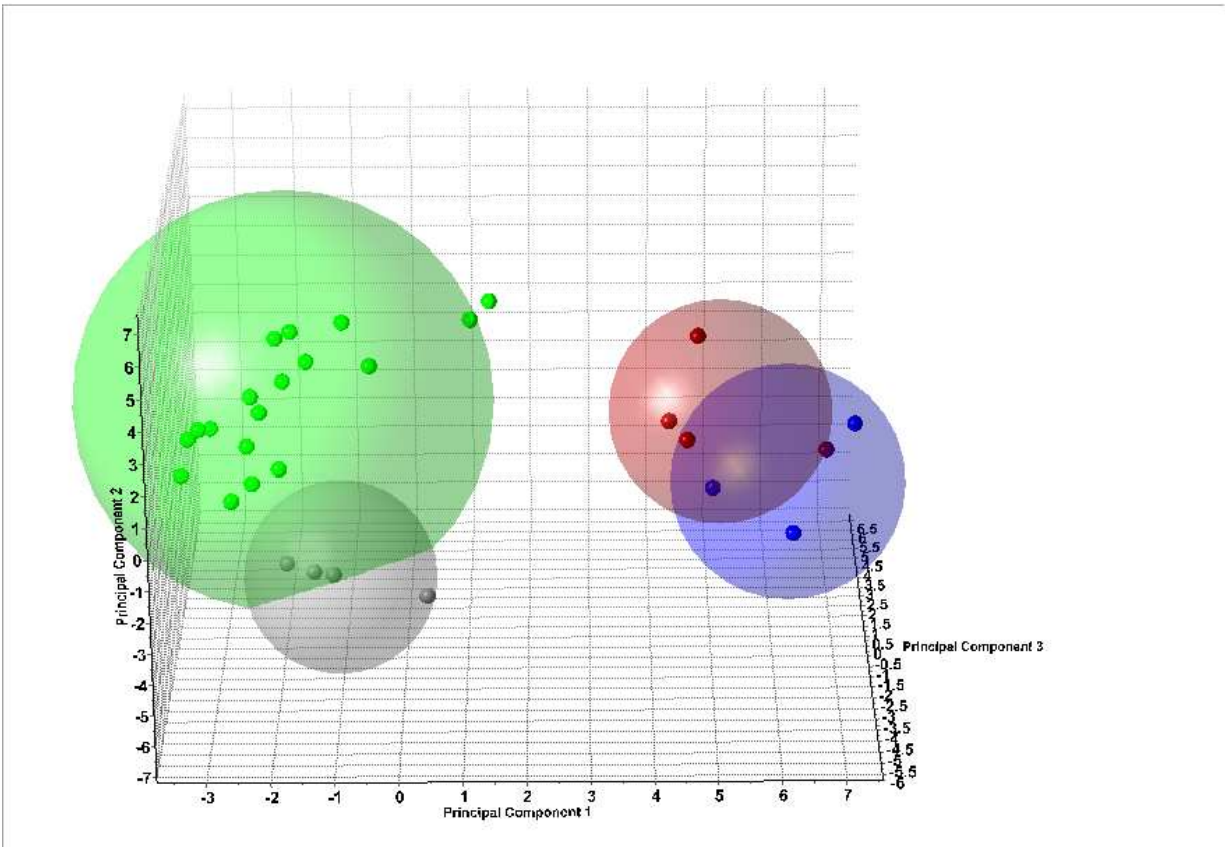


Figure 4.6- PCA plot of ceramic bodies (73.87% var) based on XRD diffratograms.

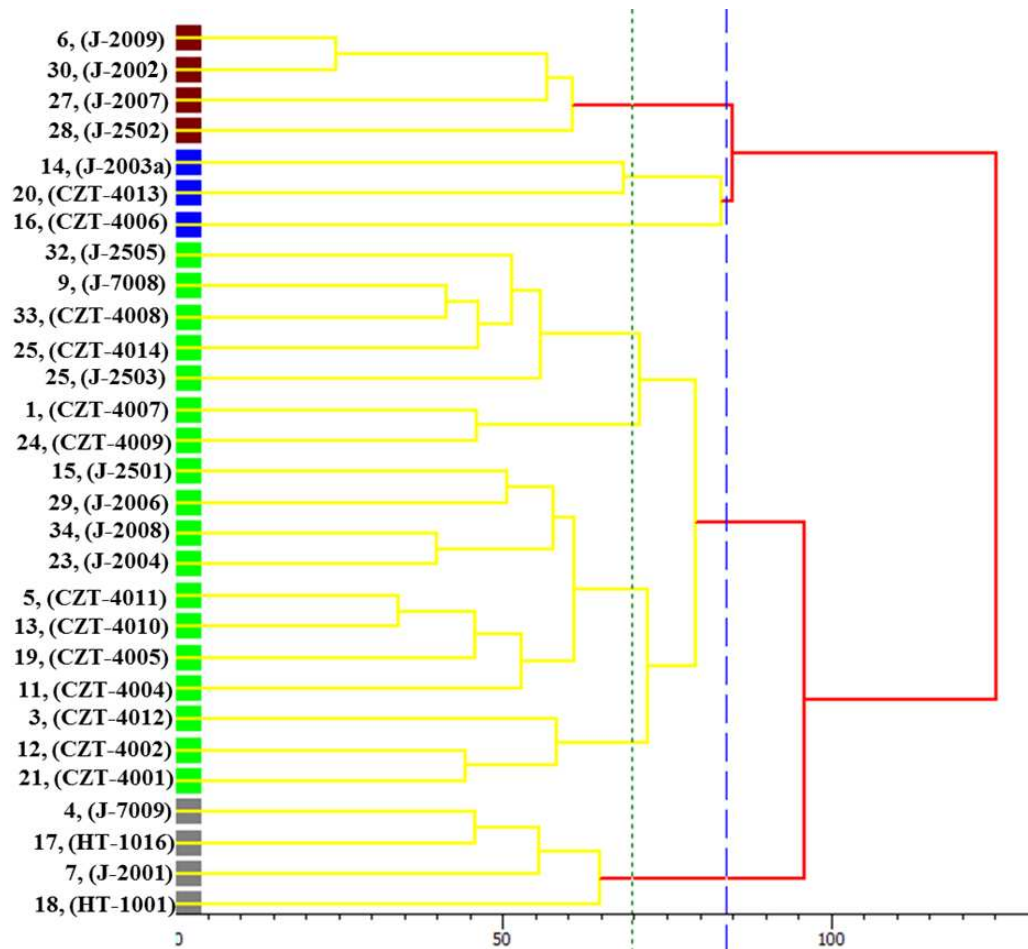


Figure 4.7- Dendrogram of PCA results derived from XRD diffractograms.

According to, the XRD results of the CZT-4006 sample, it seems that this sample is very atypical compared to other samples (this sample is located in the blue group in the PCA plot in Figure 4.6). In other words, in this sample, there are peaks related to mordenite (Figure 4.8). This point is exciting because, during the Elamite era, the presence of mordenite in manufacturing ceramics that are related to Elamite seals was also mentioned (Sax & Middleton, 1989; Lahanier, 1976). Mordenite is commonly associated with heulandite and both typically occur as the products of the alteration of fragments of volcanic glass (Sax & Middleton, 1989). Maybe that is why we observe a high amorphous content in this ceramic body. The closest samples to the CZT-4006 according to figures 4.6 and 4.7 are the CZT-4013 and J-2003 samples, but in these samples, it is very difficult to identify the mordenite peaks. Also according to the SEM images from the J-2003 sample, there is some altered glass structure in the ceramic body together with calcium carbonate

(Figure 4.9), such structures may be related to the firing temperature of the ceramic body. In other words, when the pottery was fired to higher temperatures (850-1050 degree centigrade), the amount of glass increased, this issue is dominant in non-calcareous ceramic bodies (Maniatis & Tite, 1981). In addition, the presence of calcium carbonate in the archaeological ceramics could be related to the post-depositional alteration, and the presence of zeolite is linked to the crystallization of free minerals left by the alteration of the glassy phase in overfired pottery (Gilstrap et al., 2021). Therefore, the presence of calcium carbonate and altered glasses in the ceramic body of “J-2003” may be related to the two factors of high firing temperature and ceramic post-depositional alteration.

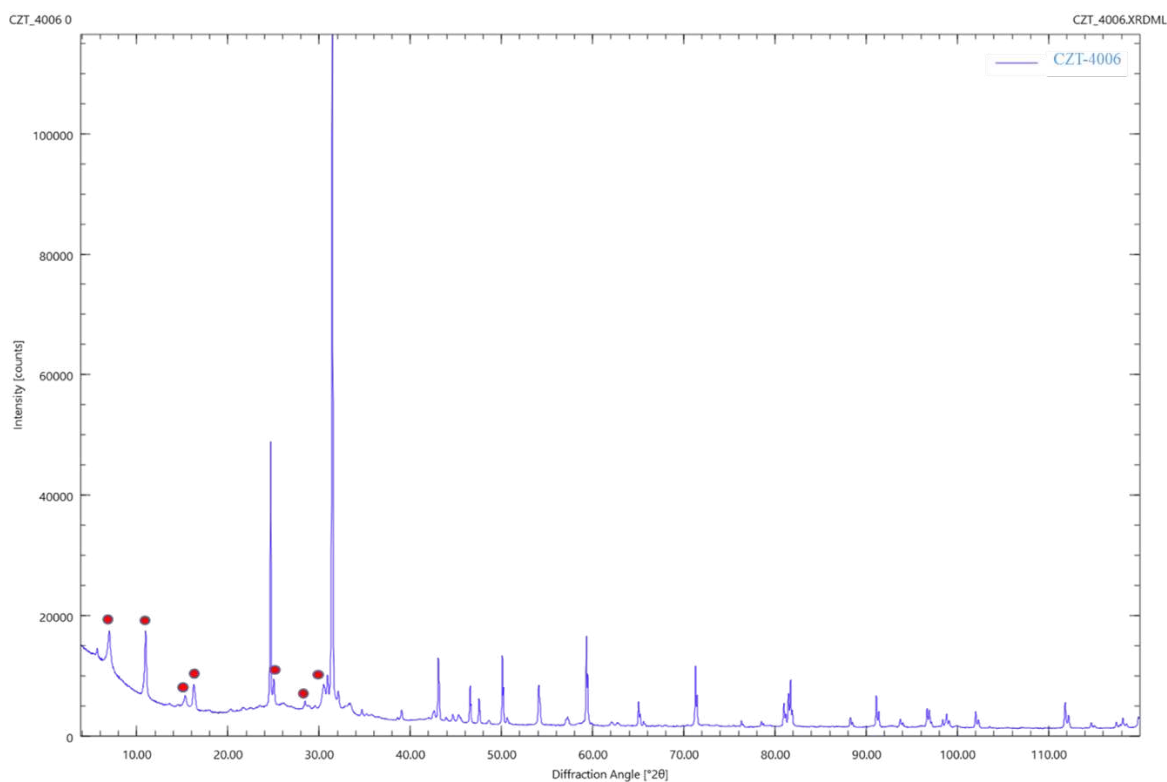


Figure 4.8- X-ray diffraction pattern of mordenite in sample CZT-4006

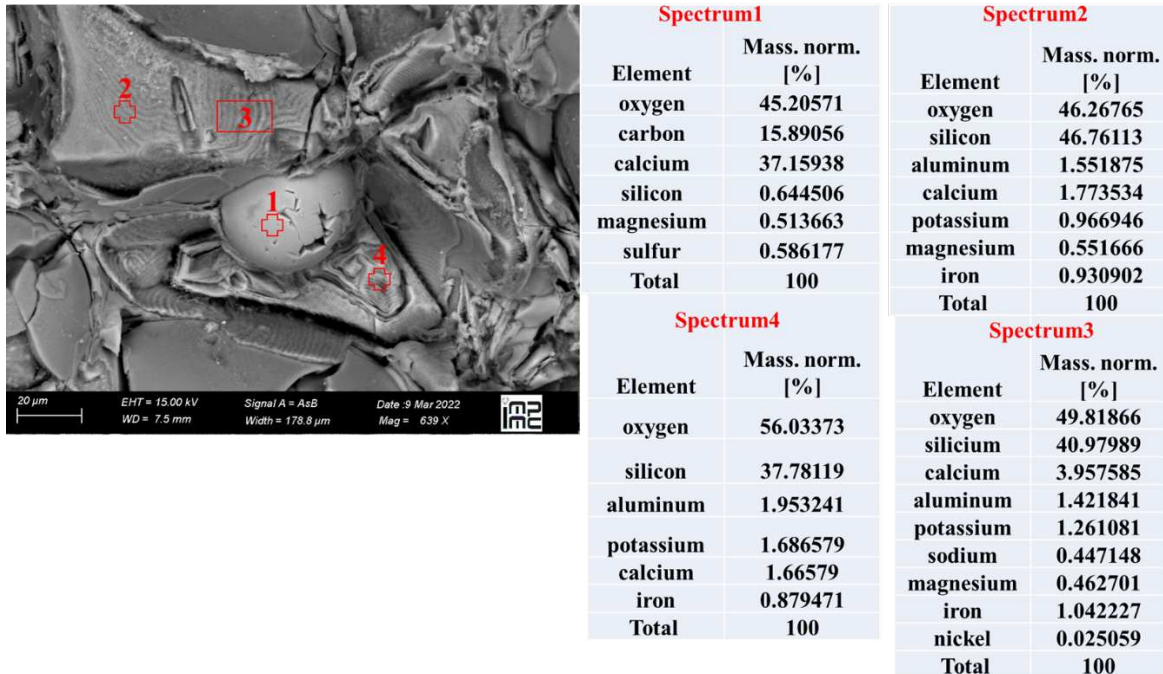


Figure 4.9- SEM image of altered glass and calcium carbonate zones in sample J-2003, and the EDS result of each spectrum.

Concerning the samples which are located in the red group of the PCA plot in Figure 6, all these samples are categorized in the quartz-mudstone fabric according to the petrographic analysis. In addition to that, these samples in the $\text{SiO}_2/\text{Al}_2\text{O}_3/(\text{CaO}+\text{MgO})$ ternary plot are located at the top of the diagram, close to the SiO_2 vertex. Petrographic images indicate that there are some incompletely melted quartz grains in these ceramics, all edges except the center of the quartz grain has been converted to cristobalite (Figure 4.10). This cristobalite is evidence of the long firing time and slow cooling time (Reedy, 2016). In Song Jun Chinese ceramics, this kind of cristobalite is determined as the horse-tooth morphological cristobalite which is related to the heat treatment process (Yang et al., 2015). This kind of cristobalite is only shaped after a long firing time and a long soaking time of the ceramics. However, in the Elam period, it did not seem to be expected to have a kiln to control the temperature for a long time completely. An alternative to long firing or soaking time could be related to the firing conditions. In other words, samples that are fired twice (the first firing of the ceramic alone and the second firing when the glaze is added) could show the same behavior of a long firing and soaking time (Holakooei et al., 2014). Therefore, we can propose that, only during the Neo Elamite period, there are some new techniques that artisans try

to fire the glazed ceramics twice. It is very interesting because as mentioned in the first part of this paper, there are some different words for mudbrick and baked brick during the Elamite period (Table 4.1). According to the terminology of bricks and glazes during the Elamite period, Potts believed that some middle Elamite bricks were inscribed after glazing but before firing (D. T. Potts, 2010). In other words, in some Elamite inscribed bricks, they used the Akkadian term *Libittu*, which is related to mudbrick. However, after this word, they used some colors (silver, gold, blue-green, and white) related to glazes, so it means that Elamite people added the glaze on mudbrick, not baked- brick, so they fired brick and glaze once together. Also, according to the red group of the PCA plot, it seems that, during the Neo Elamite period, some techniques will be added. In these 4 samples, the content of SiO₂ is high; however, we must pay attention that all the percentage of SiO₂ is not related to quartz, and in these samples, some cristobalite is identified too.

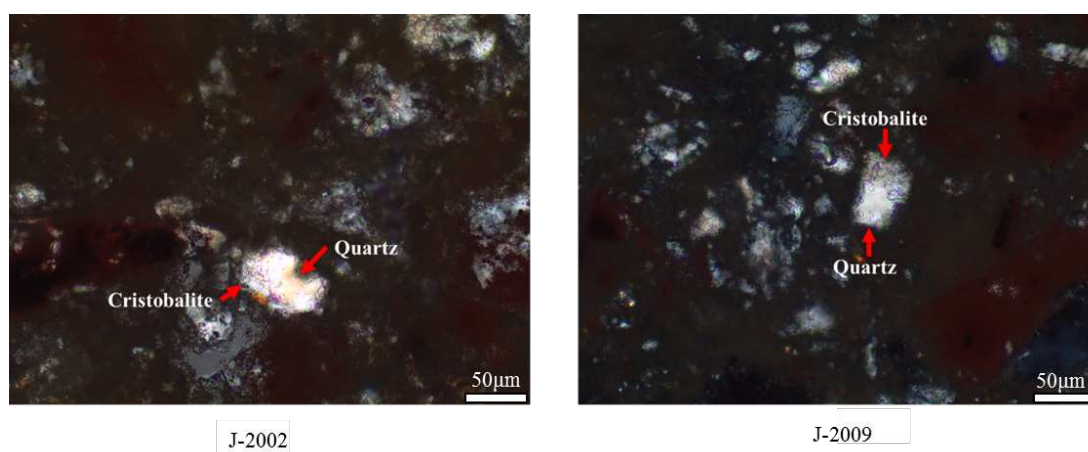


Figure 4.10- Quartz cores with cristobalite edges in Neo Elamite samples.

Based on the wares of Song dynasty (Reedy, 2016: Fig.4)

For the 4 samples which are located in the gray group of the PCA plot in Figure 4.6, it must be said that these samples correspond to the first group of calcareous fabrics (the calcareous fabric with lime fragments). Also, three samples (HT-1001, HT-1016, and J-2001) of this group are located in an area that is related to the very reach lime samples. So as it turns out, the XRD, petrographic, and EPMA data all confirm each other.

The remaining samples are located in the green group of the PCA plot. According to the petrographic study, these samples are voids-plant matter fabric and calcareous fabric. As

mentioned before, these fabrics can be categorized into the main calcareous fabric. However, because the plant matter is used in some samples, they are separated from others. As shown in the $\text{SiO}_2/\text{Al}_2\text{O}_3/(\text{CaO}+\text{MgO})$ ternary diagram, most of the samples are located in the quartz/ anorthite/ diopside triangle, away from the quartz/anorthite line. This means that these samples can be categorized as calcareous ceramics (Noll & Heimann, 2016).

4.6-Conclusion

The study of ceramic bodies that have glaze makes it possible to examine the samples with a more comprehensive view. For this purpose, XRD, EPMA, SEM-EDS, and petrography analyzes were performed on the studied samples. Based on the studies, the firing temperature of pottery can be considered in the range of 750 to 1000 degrees centigrade. Also, the samples were divided into 4 main groups based on their thin-section study. These 4 groups are siliceous samples, calcareous samples, voids-plant matter samples, and quartz-mudstone samples. The results of EPMA and the noll plot also confirmed this division. As it is clear from the presented division, in two of the four studied groups, the amount of quartz is very high. Of course, this issue is related to the discussion in the first chapter about the geology of the studied areas. It seems that the ceramic bodies of Neo Elam samples have more differences than the ceramic bodies of Middle Elam. In other words, the ceramic bodies of middle Elam are almost all very similar to each other, we can see the diversity of the ceramic bodies of Neo Elam. Of course, it should be noted that some of the ceramic bodies have been altered. However, according to the PCA study of ceramic bodies, the two Jubaji samples (2009 and 2002 samples) can be considered very different from other samples. These two samples are placed on the top of the dendrogram chart. In these two samples, cristobalite was also observed, and the presence of cristobalite can indicate the different firing conditions of these two samples compared to other samples. The variety of ceramic bodies of the Neo-Elam period suggests two hypotheses, firstly, in this period, the techniques of making and firing ceramic bodies were more than those of the Middle-Elam period. Secondly, Trade can also be one of the factors.

Chapter Five: Scientific research on the Elamite glazes from Khuzestan province (Southwestern Iran)

Abstract

Finally, in the last chapter, Elamite glazes were studied. Regarding Elamite glazes, it can be said that all of them are alkaline glazes and the percentage of lead is more than 3% only in Parthian and Sasanian samples, and in the periods of Middle Elam and Neo Elam, the amount of lead was always less than three percent. Haft Tepe can be considered the beginning of enamel production in Iran, which can be one of the first places in the world where enamel production was carried out. Turquoise color is the most abundant in Elamite glazes, and copper 2+ is used for this purpose. There are differences between the glazes of Middle Elam and Neo Elam, and the solution to finding the root of these differences is somewhere beyond Elam and to the north of it. For this purpose, the iron age glazes of Qalaichi area were also analyzed. The two major differences between Qalaichi glazes and middle Elamite glazes were the presence of sodium antimonate as an opacifier in Qalaichi glazes, which was not observed in middle Elam glazes, and also the formation of nanoparticles in Qalaichi glazes, which were not present in middle Elam glazes.

5.1-Introduction

The beginning of using glazes on ceramics in Iran has always been associated with the naming of two important Elamite sites: Haft Tepe and Chogha Zanbil sites, both of which are located in Khuzestan province in the southwest of Iran. The use of glazed objects has been reported in the excavations of Haft Tepe (Negahban, 1991) and Chogha Zanbil. According to the dating in the Middle Elam period (Haft Tepe 15th and 14th centuries BC and Chogha Zanbil 13th – 10th centuries BC), it can be said that the production of glazes in Iran probably started from the Haft Tepe and has flourished at Chogha Zanbil. The first glazed ceramic bodies in Elam correspond to spools found in Haft Tepe which are very similar to Elamite knobs and nails (Basello, 2012). However,

after Haft Tepe and Chogha Zanbil, during the Neo Elamite period in Khuzestan province was one of the centers of Elam civilization, glazed ceramics were also reported at Susa and Jubaji sites (Ahmadinia & Shishegar, 2019; Holakooei, 2014). The only Elam site where glazed ceramics have been obtained beyond Khuzestan province is Tall-i Malyan in Fars province (Sumner & Reiner, 1974). However, at the same time as the Neo Elamite period in the northwest of the Iranian plateau, glazed ceramics were also found at Qalaichi, Tepe Rabat, Ziwiye, and Hasanlu sites (Holakooei, Soldi, et al., 2017; Raith et al., 2022). Also, among the neighbors of Elam, the use of glaze was also common during the period of Neo-Assyria and Neo-Babylon (Alloteau et al., 2022). Thus, in the middle of the second millennium to the middle of the first millennium BC, which is the period studied in the present research, there were four branches of glaze production on ceramic bodies: a) Elam, b) Iron Age in Northwestern Iran, c) Neo Assyrian and d) Neo Babylon (Figure 5.1).

In the current research, the focus of the study is on the period of Elam. This period covers from the middle of the second millennium to the middle of the first millennium BC. There has been less effort to comprehensively pay attention to the glazes of this period. Therefore, paying special attention to the glazes of Elamite sites in Khuzestan province can give a better point of view about the beginning and flourishing of glazes on ceramic bodies in this region. For this purpose, three Elamite sites at Haft Tepe, Chogha Zanbil, and Jubaji were selected to collect glazed materials. The reason for not choosing Susa is that the dating of glazed Elamite artifacts at Susa has always been problematic because of the disturbance of the stratigraphy of the Elamite layers (Caubet, 2005).

During the Elamite period, different words were used to express different types of glazes and ceramic bodies related to glaze (Malbran-Labat, 1995; D. T. Potts, 2010). It should be noted that during the Elamite period, there were colored materials that evoked a certain color in the mind. In other words, it seems that, most of the time, Elamites used colored materials instead of words to express a specific color. In this regard, we could refer to a text dated to the Middle Elamite period from Chogha Zanbil:

“a-na-ku mun-taš-DINGIR.GAL i-na li-bi-it-ti ša KÙ.GI ša KÙ.BABBAR (2) ša ^{na}KA ù ^{na}UD.UD.AŠ ku-ku-un-na-a e-pu-uš-ma (3) a-na DINGIR.GAL ù dMÙŠ.EREN ša si-ia-an-ku-uk ad-di-in (...)” (Malbran-Labat, 1995: 77; Potts, 2010: 50).

*“I, Untaš-Napiriša, built the kukunâ (= el. Kukunum) in (glazed) bricks of **gold, silver, obsidian and alabaster**, and I gave them to the Napiriša and Inšušinak gods of siyan-kuk*
“(Basello, 2011).

If you pay attention to the translation of the words marked in bold, it is clear that the presence of such words in the description of bricks puts them in the meaning of colored glazes. In other words, gold means golden color, silver means silver color, obsidian means black color, and alabaster means white color. Such uses of colored materials in the designation of colored glazes were common during the Elamite period. However, this point is also important, when talking about metals in Elam and Mesopotamia. The lack of metal resources in these lands (Nezafati et al., 2006) should also be mentioned. In other words, since in glazes, metal oxides play a decisive role as coloring agents, attention should be paid to this lack of metal sources. Therefore, an important question arises, where did the Elamites get the metal oxides they used in the glazes? For this purpose, it is necessary to refer to some very important toponyms, for instance, “Marhashi” and “Shimashki” that are mentioned in Elamite and Mesopotamian texts as the lands supplying metals ores (Nezafati & Pernicka, 2012; Potts, 1993), whose their exact locations are not yet known.

This research aims to gain a better understanding of the technology of glaze-making from the Middle Elamite to the Neo Elamite period. Investigating and studying the production process of Elamite glazes is important because, in the time between these two periods (Middle Elam and Neo Elam), the making of glazes went beyond the civilization of Elam, and in the northwest of Iran, the empires of Assyria and Babylon also expanded. In other words, in the first millennium BC, the production and use of glaze became very popular in Iran and Iraq. This issue itself is one of the reasons for the importance of the current research. So, by studying the glazes of Middle Elam and Neo Elam, there is an opportunity to investigate how the use of glaze spread from the Elamite period to the Achaemenid period.

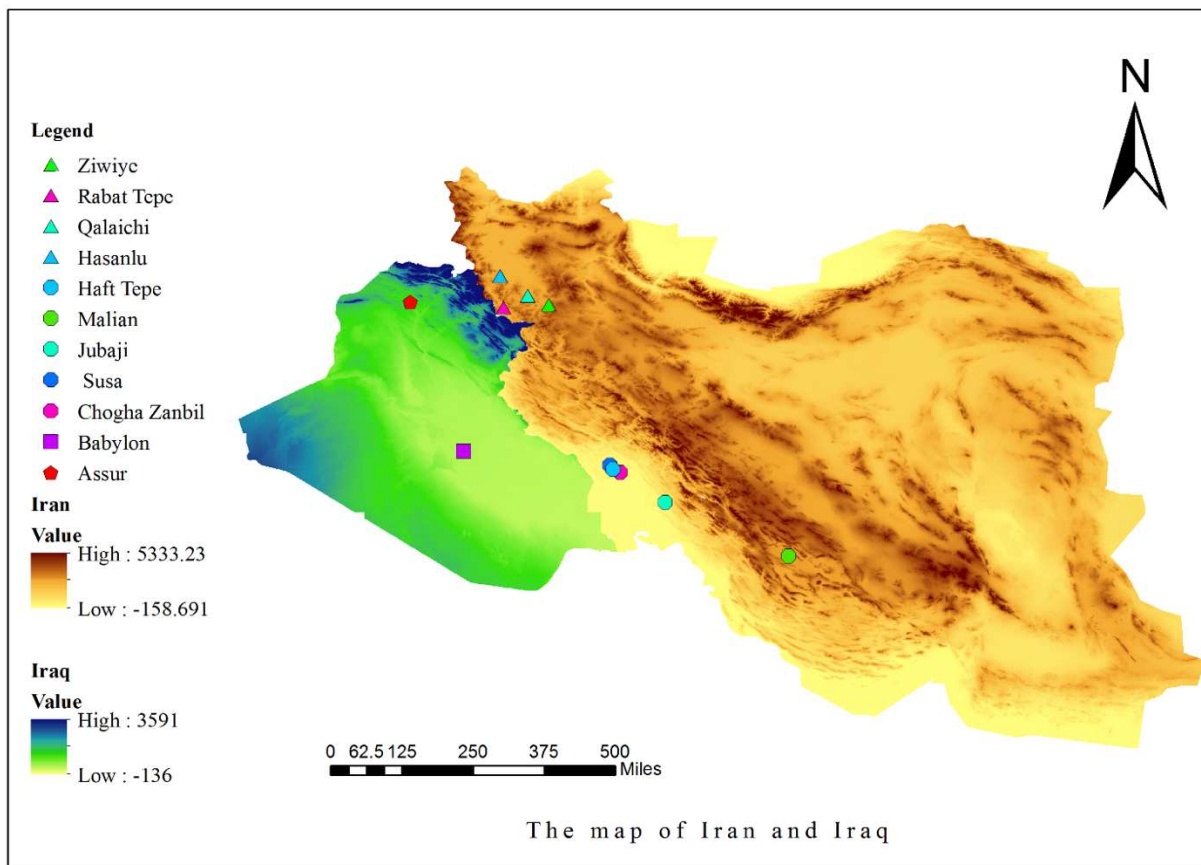


Figure 5.1- The map of Iran and Iraq, with the ancient sites which were mentioned in the text

5.2-Samples and analytical methods

Fifty-four glazed ceramics were selected from four areas at Haft Tepe, Chogha Zanbil, Qalaichi, and Jubaji to study their glazes (selected samples are shown in figure 5.2). The selected glazes were all the result of scientific archaeological excavations. The samples which are selected from the Haft Tepe are from Ezzat Negahban’s archaeological excavations (Negahban, 1991), the Chogha Zanbil’s samples are from Ghirshman’s archaeological excavations (Ghirshman, 1961), and the glazed pottery of Jubaji was also the result of the excavations of Arman Shishegar (Ahmadinia & Shishegar, 2019). The Qalaichi glazes are selected from the archaeological excavation of Bahman Kargar in the northwest of Iran (Kargar, 2004). The glazed ceramics collected from Chogha Zanbil are all related to the architectural decorations of the Chogha Zanbil ziggurat, which can be confidently considered to be dated from the Middle Elamite period. The

glazed ceramics investigated in the present study from the Jubaji site are also related to the vessels found in the graves and were selected from the Elamite graves. However, in the case of Haft Tepe, which was discussed earlier, it should be said that the dating of the found objects will not be as easy as the other two sites. Among the samples collected from Haft Tepe, three of them according to Ezzat Negahban's excavations, were obtained from the Elamite layer, which now are stored in the Iran National Museum. However, the number of glazed ceramics obtained from Haft Tepe is very significant, and many of them are reported from newer layers related to the Parthian-Sasanian and early Islamic periods (Table 5.1). However, due to the oldness of the excavations of Haft Tepe by Ezzat Negahban, and the overlapping of ancient layers, some information about the samples have been lost or not mentioned. Therefore, two samples of Haft Tepe glazes do not have a specific period, so it is not possible to determine whether they are Elamite glazes or more recent Parthian-Sasanian or early Islamic glazes. In this way, considering the importance of the Haft Tepe, the first glazed ceramics of Iran were obtained from that site, as well as the existence of samples that can be confidently said to be Elamite or not. The samples without information were also added to the studied collection so that perhaps their periods can be discussed and some of the information of these samples can be recovered during this study. Therefore, in this research, there is a chance to be able to examine the glaze-making in the southwest of Iran from the period of Elam to early Islam.

5.2.1- LA-ICP-MS

Laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) was undertaken at the Institut de Recherche sur les Archéomatériaux (IRAMAT-CEB) using a Resonetics M50E excimer laser and a Thermo Fischer Scientific ELEMENT XR mass spectrometer. The laser operates at 193 nm, a 5 mJ energy and 10 Hz pulse frequency. The standard analytical protocol was used for all analyses that captures the signals in counts-per-second of 58 isotopes including major, minor, trace and rare earth elements. Each tessera was placed in a standard Resonetic S155 cell and ablated for 20 s pre-ablation and the acquisition of 9 mass scans over 27 s with a beam diameter between 50 μm and 100 μm and an argon/helium gas flow of 1 l/min Ar + 0.65 l/min He.

5.2.2- Raman

Raman measurements were performed on the prepared samples using a LabRam Infinity (Jobin-Yvon-Horiba) Raman spectrometer. For our analyses, a 532 nm Nd:YAG laser source was used with a maximum power of 10 mW and a spectral resolution of 2.5 cm^{-1} . The instrument is combined with an Olympus BX-40 confocal microscope. At a magnification of $500\times$ and area of $5 \times 5 \mu\text{m}^2$ is analysed at a depth of about $10 \mu\text{m}$. A calibration on the silicon peak at 520 cm^{-1} was carried out before each set of data acquisitions (on average 10 per sample) with a laser aperture of $300 \mu\text{m}$, an average acquisition time of 10 s, and repetitions of 10–30 times depending on the intensity of the spectrum.

5.2.3- Uv-visible

Ultraviolet-visible-Near Infrared spectroscopy was carried out on glazed samples. Samples were illuminated using two lamps as light source, a 30 W deuterium lamp for the UV spectral region and 20 W halogen lamp emitting in the visible and infrared regions. Optical fibres guide the light from the light sources to a plano-convex lens, which focused the light to the samples to a spot size of around 3 mm of diameter. The light was collected using an integration sphere with an aperture of 6 mm, which was connected via optical fibres to two spectrometers: the AvaSpec-3648 and the AvaSpec-256-NIR1.7 from Avantes. Spectra were thus recorded between 200 and 1650 nm with a spectral resolution of 1.4 nm.



Figure 5.2- Representative glazed ceramics of Middle and Neo Elamite periods

Table 5.1- The studies glaze information

Number of the sample	Name of sample	Color of the glaze	Possible use	Dating
1	CZT- 4011	White	Building decoration	Middle Elam
2	CZT- 4002	White	Building decoration	Middle Elam
3	CZT- 4013	White	Building decoration	Middle Elam
4	HT- 1015	White	Pottery	Parthian-Sasanian

5	J- 2009	Light green	Pottery	Neo Elam
6	J- 2502	White	Pottery	Neo Elam
7	J- 2001	White	Pottery	Neo Elam
8	J- 2003	Yellowish white	Pottery	Neo Elam
9	J- 2002	Green	Pottery	Neo Elam
10	J -2009	Green	Pottery	Neo Elam
11	J- 7009	White	Pottery	Neo Elam
12	HT- 1629	White	Pottery	Islamic period
13	QA- 3001	Blue	Glazed tile	Iron Age
14	HT- 1016	White	Spool	Middle Elam
15	QA- 3002	Blue	Glazed tile	Iron Age
16	HT- 1005	White (Yellowish white)	Pottery	Parthian- Sasanian
17	HT- 1001	White	Spool	Middle Elam
18	HT- 1005	White	Pottery	Parthian- Sasanian
19	HT- 1017	Green	Pottery	Parthian- Sasanian
20	HT- 1004	Green	Pottery	Parthian- Sasanian
21	HT- 1628	White	Pottery	Parthian- Sasanian
22	J- 2008	Green	Pottery	Neo Elam
23	HT- 1011	Blue	Pottery	Parthian- Sasanian
24	J- 2004	Yellow	Pottery	Neo Elam
25	HT- 2006	White	Pottery	Parthian- Sasanian
26	J- 2501	Green	Pottery	Neo Elam
27	CZT- 4014	Blue	Building decoration	Middle Elam
28	CZT- 4004	Blue	Building decoration	Middle Elam
29	CZT- 4003	Blue	Building decoration	Middle Elam
30	CZT- 4005	Blue	Building decoration	Middle Elam
31	CZT- 4002	Blue	Building decoration	Middle Elam
32	CZT- 4006	Blue	Building decoration	Middle Elam
33	CZT- 4007	White	Building decoration	Middle Elam

34	CZT- 4009	Greenish blue	Building decoration	Middle Elam
35	CZT- 4012	Blue	Building decoration	Middle Elam
36	CZT- 4013	Black	Building decoration	Middle Elam
37	CZT- 4010	Blue	Building decoration	Middle Elam
38	CZT- 4001	Blue	Building decoration	Middle Elam
39	HT- 1005	Blue	Pottery	Parthian-Sasanian
40	HT- 1006	Greenish blue	Pottery	Parthian-Sasanian
41	HT- 1006	Yellow	Pottery	Unknown
42	HT- 1008	Green	Pottery	Parthian-Sasanian
43	HT- 1009	Yellow	Pottery	Parthian-Sasanian
44	HT- 1015	Black	Pottery	Parthian-Sasanian
45	HT- 1017	Light green	Pottery	Parthian-Sasanian
46	HT- 1018	Blue	Pottery	Unknown
47	HT- 1860	Blue	Pottery	Middle Elam
48	HT- 1901	Black	Pottery	Parthian-Sasanian
49	HT- 1992	Blue	Pottery	Parthian-Sasanian
50	J- 2005	Blue	Pottery	Neo Elam
51	J- 2008	Green	Pottery	Neo Elam
52	J- 2501	Light green	Pottery	Neo Elam
53	J- 2585	Blue (Greenish blue)	Pottery	Neo Elam
54	J- 7008	Blue	Pottery	Neo Elam

5.3-Results

5.3.1-Main composition

The elemental composition of the glaze matrix is analyzed by LA-ICP-MS (Table 5.2). First, it should be mentioned that the glaze is all highly altered which is evidenced by the low alkali (Na_2O) and high silica (SiO_2) contents that are indicative of lixiviation processes as Na^+ ion is the most mobile element in the glaze and can be easily leached out. According to the main fluxes, i.e. oxide agents used to decrease the melting temperature (mainly alkali oxides and PbO), all the studied glazes can be categorized into two main groups. The alkaline glazes (less than 3 wt% PbO) and the alkaline-lead glazes (between 3 wt% to 11 wt% PbO) (Figure 3). As shown in Figure 3, most of the studied samples are classified in the category of alkaline glazes. Also, all the samples that are classified in the category of lead-alkali glazes are glaze samples found at Haft Tepe.

An interesting thing to note is sample number 41 (HT-1006), as this sample is located in a range that can represent natron glazes according to Figure 5.3, and the lead content is less than 3 wt% in this sample, which can be categorized as the low lead-alkali glazes too. Also, silver is used in this yellow glaze sample (Ag: 7.80 wt%), and Colombari suggested silver yellow in glazes started in the 12th century A.D (Colombari, 2013). This means that this glaze belongs to the Haft Tepe Islamic period layers.

Moreover, according to Ezzat Negahban's report, sample HT-1017 is related to Parthian-Sassanian layers. Since this sample is classified in the lead-alkaline glaze category, this suggests the possibility that all lead-alkaline glazes in Haft Tepe can be related to the periods after Elam.

All three known Elamite samples of Haft Tepe (HT-1001, HT-1016, and HT-1860) are also placed in the category of alkaline glazes. In this way, it is possible to understand that studying the chemical composition of glazes can help archaeologists in some cases as a dating method.

According to Figure 3, in several samples classified in the category of alkaline glazes, the amount of alkaline elements is very low. In such a way, they cannot even be considered as a glaze. The reason for this seems to be the alteration of Elamite glazes. Most glazes with very low alkaline elements are either completely white in color or have a whitish layer in some parts of them. Also,

the presence of altered whitish parts in Assyrian and Babylonian glazes was mentioned before (Alloteau et al., 2022).

Table 5.2- LA-ICP-MS analysis of the studied glazes (wt%)

Number of the sample	Name of sample	Na ₂ O	MgO	Al ₂ O ₃	SiO ₂	K ₂ O	CaO	Fe ₂ O ₃	CuO	Sb ₂ O ₃	PbO
		wt									ppm
1	CZT- 4011	0.10	6.87	1.14	87.93	0.20	1.56	0.75	0.74	3.0	13.2
2	CZT- 4002	0.14	8.84	0.66	74.55	0.20	13.40	0.75	0.63	3.0	150.7
3	CZT- 4013	0.21	3.82	1.42	91.72	0.28	1.17	0.87	0.02	0.8	21.3
4	HT- 1015	0.26	1.28	6.25	67.91	1.13	19.84	1.61	0.05	1.6	045.4
5	J- 2009	0.36	1.11	4.78	48.13	1.65	4.10	9.34	25.06	67.4	560.3
6	J- 2502	0.38	4.07	4.43	76.60	2.24	3.53	5.42	1.41	22.9	27.6
7	J- 2001	0.44	0.70	2.42	89.36	1.35	2.77	1.31	0.62	172.7	64.5
8	J- 2003	0.47	1.12	2.05	89.47	1.02	3.11	1.28	0.31	216.4	891.2
9	J- 2002	0.49	0.96	5.70	72.09	2.18	2.90	4.30	8.89	32.0	845.9
10	J-2009	0.52	1.72	5.10	53.22	2.90	2.88	15.61	14.66	59.1	124.7
11	J- 7009	0.66	0.51	3.62	79.17	2.34	7.78	1.65	0.42	13171.8	5247.1
12	HT- 1629	0.68	0.99	5.00	83.34	2.99	2.18	3.71	0.13	1.7	72.0
13	QA- 3001	0.79	0.46	0.49	79.71	0.38	2.87	0.21	3.07	82399.6	105.1
14	HT- 1016	0.87	4.62	4.07	73.46	3.09	5.14	2.26	2.67	18.8	577.1
15	QA- 3002	0.92	0.58	0.68	79.92	0.16	3.20	0.26	4.81	76698.3	82.2
16	HT- 1005	0.92	3.35	4.31	80.58	2.98	1.28	1.96	2.82	8.5	590.1
17	HT- 1001	0.95	5.81	4.47	73.71	2.64	6.51	2.17	1.88	26.2	809.7
18	HT- 1005	1.17	1.19	5.07	82.34	3.41	1.65	2.07	1.11	13.4	599.0
19	HT- 1017	1.17	2.06	7.73	57.04	1.69	6.43	3.28	7.25	994.2	9851.6
20	HT- 1004	1.22	4.87	2.65	71.42	1.30	3.74	0.83	11.98	37.0	41.1
21	HT- 1628	1.60	7.37	7.94	75.34	1.89	2.08	1.85	0.01	6.4	212.7
22	J- 2008	1.99	4.03	5.80	66.51	2.32	6.84	3.90	7.12	65.3	104.3
23	HT- 1011	3.55	2.64	3.53	74.74	6.23	5.49	1.20	0.89	16.2	763.9
24	J- 2004	4.41	5.26	11.46	50.94	3.29	13.19	6.00	0.29	8.7	117.0
25	HT- 2006	4.82	5.16	8.03	61.55	4.68	8.53	4.92	0.56	590.1	219.3
26	J- 2501	4.94	4.22	12.52	58.85	3.55	8.84	4.81	0.50	10.7	60.4
27	CZT- 4014	13.68	3.42	0.73	67.18	3.74	8.47	1.12	0.42	111.5	106.6
28	CZT- 4004	7.63	2.02	0.59	79.36	2.40	4.32	1.60	0.91	5.6	14.4
29	CZT- 4003	9.01	2.05	1.01	76.78	2.60	5.31	0.96	1.22	13.4	76.5
30	CZT- 4005	7.38	2.69	1.11	77.65	2.13	5.06	1.56	1.54	6.8	37.0
31	CZT- 4002	10.32	2.16	0.62	75.67	3.24	5.89	0.62	0.34	6.2	118.3
32	CZT- 4006	13.59	2.93	0.48	69.76	3.55	5.98	0.83	1.39	0.7	4.5

33	CZT- 4007	6.55	4.15	1.08	77.00	2.48	5.33	0.97	1.14	2.0	3.4
34	CZT- 4009	5.88	6.80	6.68	59.10	4.34	11.00	4.52	0.26	1.1	9.6
35	CZT- 4012	12.81	3.21	1.06	68.80	3.58	6.62	1.79	0.70	4.8	22.2
36	CZT- 4013	11.55	2.59	0.95	74.94	2.42	5.54	0.53	0.01	1.7	2.3
37	CZT- 4010	8.43	2.01	0.56	78.30	2.86	4.76	0.88	1.30	9.1	74.8
38	CZT- 4001	12.57	2.94	0.77	70.96	3.21	7.05	0.92	0.50	3.6	11.3
39	HT- 1005	11.75	3.20	1.70	66.22	7.06	6.97	0.70	1.05	11.2	589.1
40	HT- 1006	8.13	2.58	2.92	63.77	5.00	7.47	1.14	1.03	51.3	9131.7
41	HT- 1006	7.67	0.00	0.00	41.78	0.00	11.63	2.89	4.53	0	9822.0
42	HT- 1008	7.10	2.66	2.91	62.22	4.29	7.17	1.17	0.98	87.7	2248.5
43	HT- 1009	6.86	7.63	10.32	51.96	4.21	9.67	6.51	1.13	12.9	55.0
44	HT- 1015	11.36	3.73	2.76	70.06	3.77	5.93	0.73	0.08	16.5	335.6
45	HT- 1017	9.85	2.41	2.29	61.36	4.97	6.90	0.95	4.08	620.3	3327.0
46	HT- 1018	9.02	2.79	4.21	64.70	4.49	7.99	1.73	2.50	69.2	560.0
47	HT- 1860	17.51	4.38	0.61	65.16	3.15	6.25	0.40	1.11	12.4	31.4
48	HT- 1901	9.88	3.55	2.72	65.11	4.01	10.45	2.49	0.11	3.7	449.1
49	HT- 1992	9.71	3.83	2.81	63.41	6.41	9.44	1.03	1.52	112.9	299.0
50	J- 2005	10.25	3.70	0.98	67.29	5.78	7.94	1.34	0.87	12.4	338.5
51	J- 2008	6.17	4.65	10.04	57.67	4.17	8.53	3.98	3.26	12.5	77.3
52	J- 2501	8.34	2.06	10.08	62.35	6.00	3.94	4.01	1.66	54.0	121.1
53	J- 2585	9.43	2.26	9.14	60.38	4.97	4.91	4.77	2.78	14.5	14.1
54	J- 7008	5.13	3.26	2.61	70.71	7.41	6.53	1.15	1.21	12.5	166.4

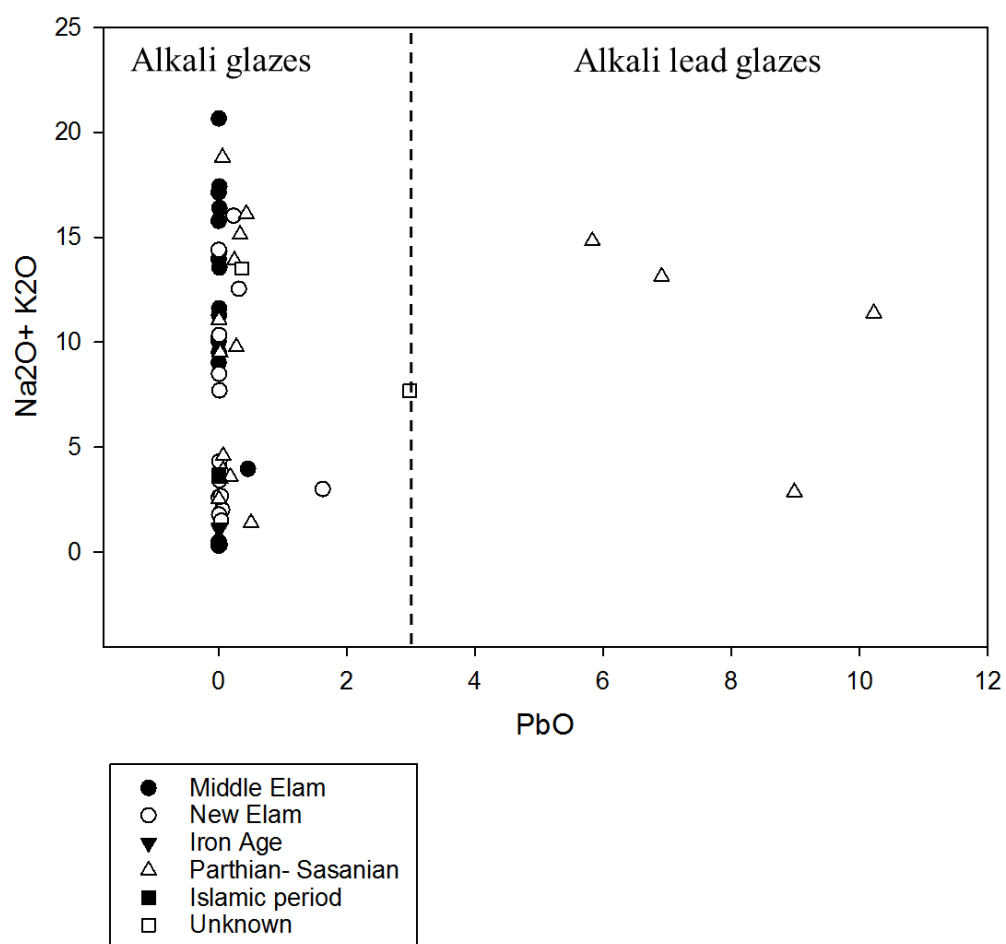


Figure 5.3- The main composition of the glazes

Figure 5.4 shows the optical microscopic images of the altered parts of Elamite glaze samples and altered layers. Therefore, the percentage of alkaline elements is low in the samples with altered layers. Also in the Achaemenid sites, Susa and Persepolis alteration layers in the glazes are reported which experienced a complete loss of alkali elements (Raith et al., 2022).

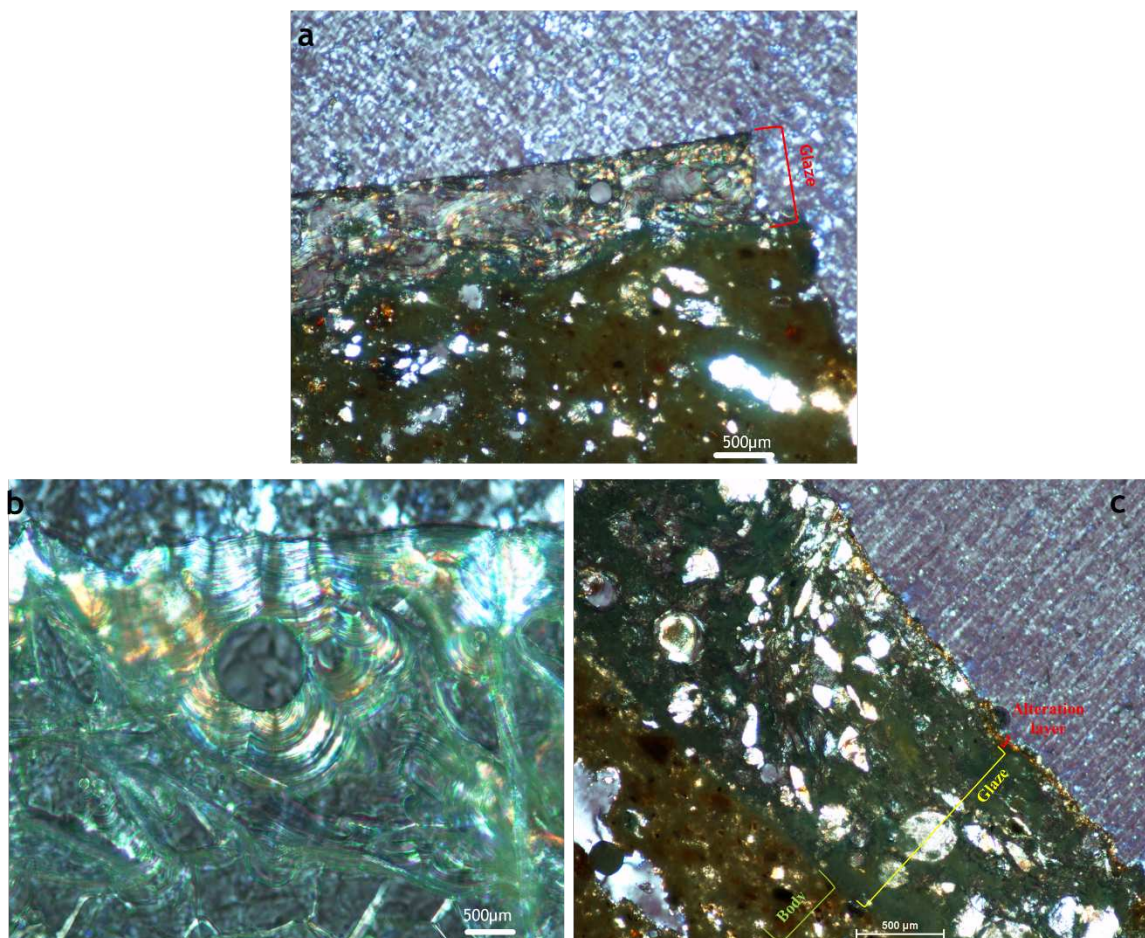


Figure 5.4- Optical microscopy of altered Middle Elamite glazes; a) HT- 1016, b) CZT- 4004, and c) CZT- 4011

In addition, Raman spectra analysis is available to classify the glassy silicates as a function of their composition types (Figure 5.15). The glass is a more or less polymerized SiO_4 network, with each tetrahedron sharing or not their oxygenion, and the Raman spectrum of an amorphous silicate is composed of two wide bands resulting from vibrations of the SiO_4 tetrahedron, a strong covalent entity and vibrationally well defined: in the middle frequencies the band centred around 500 cm^{-1} corresponds to the symmetrical deformation vibrations ($\delta \text{ SiO}_4$); the second band around 1000 cm^{-1} corresponds to the symmetric stretching vibrations of the tetrahedron ($\nu \text{ SiO}_4$) (Fischbach et al., 2016). According to the Si-O stretching peak maximum and the polymerization index, it was possible to classify glazes from different periods (Colomban et al., 2006). In Table 5.3, the main Raman parameters of 10 samples (which were selected according to the LA-ICP-MS

result by paying attention to the content of the fluxes and color agents) were presented, and, in Figure 5.5, all the information used in Table 5.3, is introduced. According to Figure 5.5, one of the Haft Tepe samples from an unknown period (HT-1018) was selected for Raman classification so that it may be possible to make some hypotheses about the period of this sample. Finally, as it is known, this method seems to be useful for dating ancient glazes, and the sample HT-1018 from an unknown period (according to the problems mentioned earlier about Haft Tepe) is localized among the Parthian-Sasanian samples (Figure 5.5) and has a significant distance from the samples of Middle Elam. Therefore, it can be said that this sample probably belonged to the Parthian-Sasanian period. By paying attention to the I_p ⁹ (between 0.78 – 1.1) of the samples, all of them can be classified in the category of ancient glazes with a high percentage of alkali according to a previous study (Colomban et al., 2006).

⁹ The polymerisation index I_p is defined as $I_p = A_{500}/A_{1000}$ with A being the area under the Raman band.

Table 5.3- Main Raman parameters of selected materials; I_p , index of polymerisation, νQ_n , centre of gravity wavenumber (cm^{-1}) of the Si-O stretching Q_n component, A_{Q_n} , component area (% of all the peak area) and ratio, based on (Colomban et al., 2006)

Name of the sample	I_p	Q0	Q1	Q2	Q3	Q4	A_Q0	A_Q1	A_Q2	A_Q3	A_Q4	A_Q2/A_Q3	A_Q2/A_Q1
QA-3001	1.05	825	950	1009	1046	1132	0.86	0.86	0.87	0.85	0.84	1.02	1.01
HT-1017	1.09	792	950	990	1092	1114	0.62	0.76	0.85	0.98	0.92	0.86	1.11
HT-1018	1.07	801	945	994	1099	1131	0.80	0.85	0.91	0.99	0.91	0.91	1.07
HT-1860	1	804	949	993	1016	-	0.75	0.81	0.82	0.82	-	1	1.01
HT-1992	0.95	789	944	996	1104	1127	0.72	0.83	0.89	0.99	0.92	0.89	1.07
CZT-4014	1.1	802	950	995	1104	-	0.62	0.70	0.71	0.96	-	0.73	1.01
CZT-4013	0.90	800	947	1013	1101	1146	0.87	0.90	0.92	0.93	0.94	0.98	1.02
CZT-4012	0.78	788	950	1010	1109	1133	0.64	0.78	0.81	0.98	0.90	0.83	1.03
CZT-4006	1.08	810	942	997	1106	1144	0.88	0.89	0.90	0.92	0.89	0.97	1.01
HT-1016	0.84	814	906	982	1106	1157	0.77	0.83	0.85	0.92	0.92	0.92	1.02

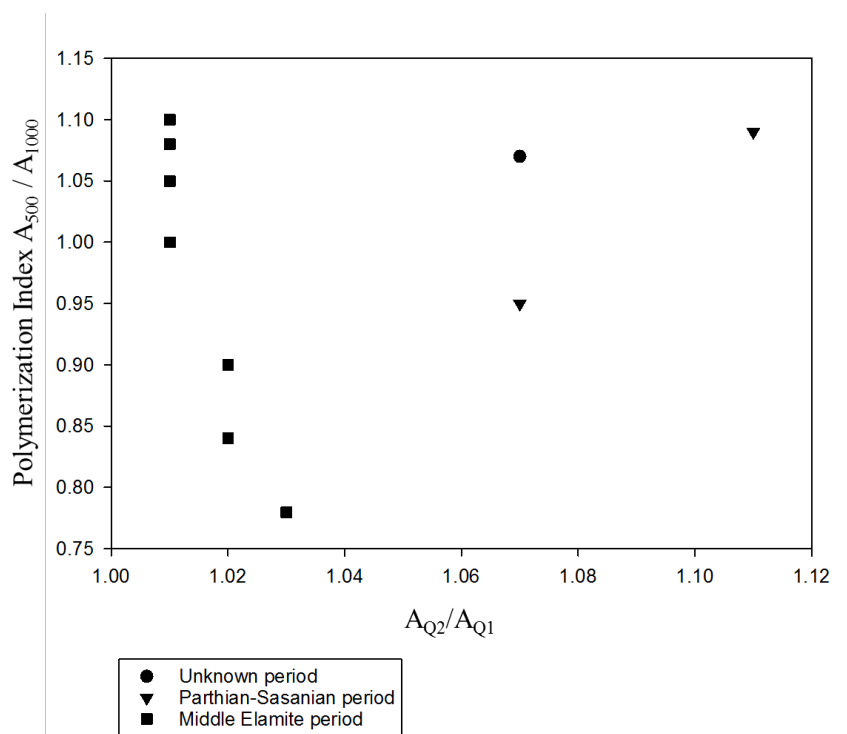


Figure 5.5. The Q2/Q1 area ratio

5.3.2-Color agent and opacifier of the glazes

According to Figure 5.6, which shows the UV-visible spectra of the studied glazes, one of the most widely used coloring agents in the studied glazes is copper (II) ions that has a strong absorption band centered at $12,000\text{ cm}^{-1}$. Especially, Cu^{2+} is a coloring agent in turquoise colors that is one of the most frequent colors in the Elamite period. By paying attention to Figure 5.6, the bands related to Cu^{2+} all have two peaks in the range around 5000 and 7000 cm^{-1} , and these two peaks could be related to molecular water and hydroxyl groups may be seen in the near infrared at about 5200 , 4500 , and 4050 cm^{-1} , superimposed on the tail of the fundamental OH stretching vibration band at 3570 cm^{-1} . These bands have been widely investigated in natural and synthetic glasses to get information on the speciation of OH and H_2O in magmas (Galoisy & Calas, 2021). Therefore, it can be said that the presence of two peaks around 5000 and 7000 cm^{-1} indicates the alteration of the glaze.

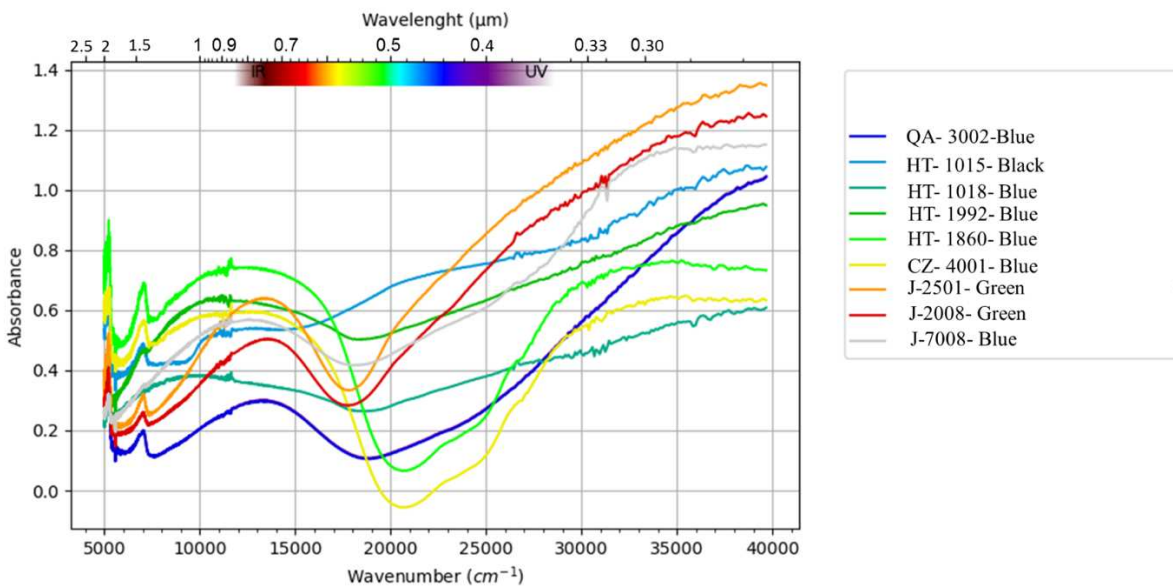


Figure 5.6- Glaze colorants. UV–Vis spectra of the most common colorants.

To more accurately identify the coloring agent and opacifier in the glaze, the PCA (Principal Component Analysis) was used to interpret the large dataset obtained by LA-ICP-MS. PCA shows, the opacifier and color agents that correlated to each element. Also, through this plot, the dispersion of the samples can be seen (Figures 5.7, 5.8). The PCA plot is presented according to Table 5.2. According to this plot, it seems that the blue glazes that have been heavily altered are mostly white. This issue can be mentioned for samples 2, 14, and 17 (CZT- 4002, HT- 1016, 1001), though they have blue (turquoise) points in some parts which seem to be their primary color. The whitish coloration is the result of alteration. These samples in the PCA plot are located in the area between blue and white glazes (Figure 5.8). In addition, the two blue (turquoise) samples placed among the white samples are Iron Age samples numbered 13 and 15 (QA- 3002, 3001). The percentage of antimony in these two samples is very high, and it seems that it could be related to different opacifiers for these glazes, which is more similar to white glazes. For this purpose, SEM-EDS analysis was performed on these two samples and it was found that sodium antimonate nanoparticles along with calcium antimonate in these two samples were used as an opacifier (Figure 5.9 and, Table 5.4). Also, rather than a unique opacifier being used in an individual glaze, a mixture of opacifiers might have been used (Holakooei, 2013; Holakooei, Ahmadi, et al., 2017). By paying attention to Table 5.4, EDS results indicate that the quantities of Calcium in the

composition of nanoparticles are significant too (2.28 wt%). It means that, in Qalaichi glazes, a mixture of Sodium and Calcium antimonate have been used as an opacifier. Also, Raman analysis of these two samples shows the use of sodium antimonate as an opacifier in Qalaichi samples (Figure 5.15). Sodium antimonate has four index peaks at 156, 230, 614, and 660 cm^{-1} , which were also identified in the Raman spectra of these two samples from Qalaichi (Figure 5.15a, f). In addition, in the center of these nanoparticles, calcium carbonate (CaCO_3) is located (Figure 9, and Table 5.4). CaCO_3 can migrate into the clay body or crystallize over the glaze surface during drying. This produces scum forming on the glaze during firing, and thus the glaze appears powdery with patches of glaze mixed with regions showing undissolved glaze components (Pradell & Molera, 2020). Of course, it should be noted that the possibility of losing calcite during the firing of the glaze is very high, hence the presence of secondary calcite in ancient glazes has been reported as a consequence of burial conditions (Li et al., 2020). Also, according to Figure 5.10 and Table 5.4 (spectrum number 4), which are related to SEM-EDS analysis of the sample (J- 2001), it can be suggested that the calcium carbonate in this sample should be of the secondary type and formed because of burial conditions.

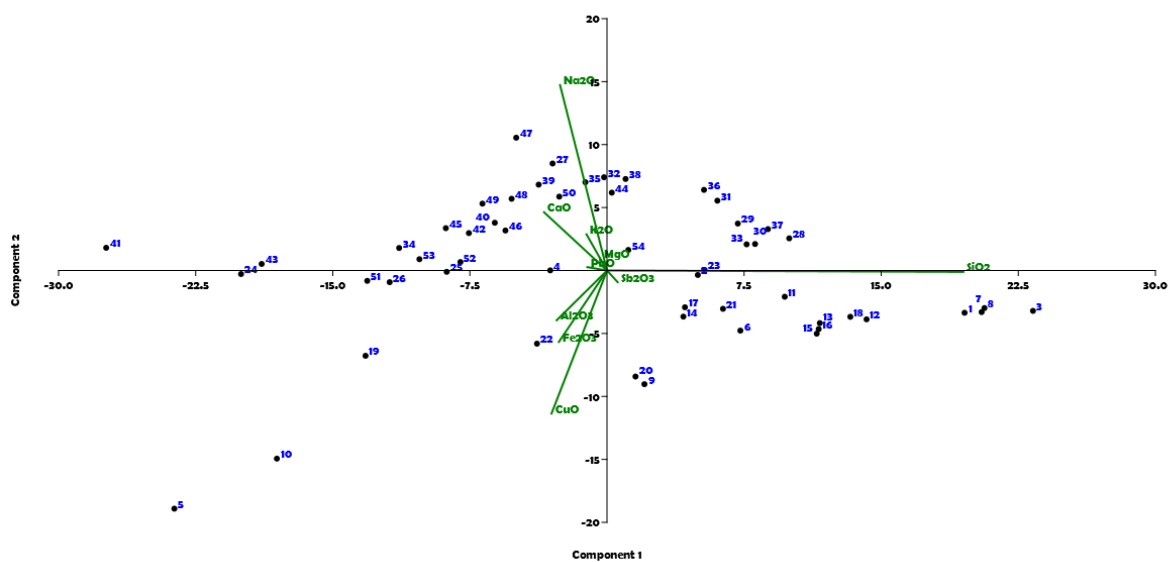


Figure 5.7- PCA (Bi-plot) of the glazes (Variance PC1= 64.75%, Variance PC2= 15.96%)

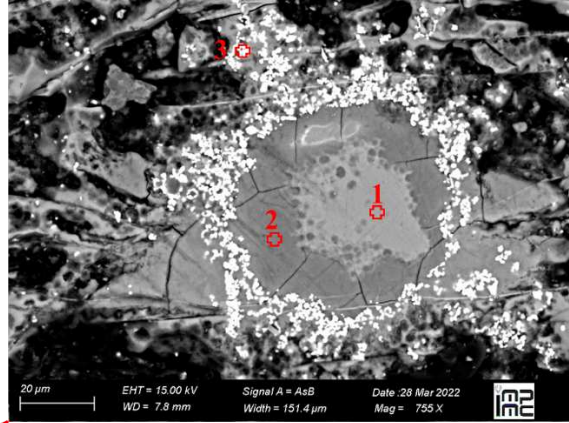
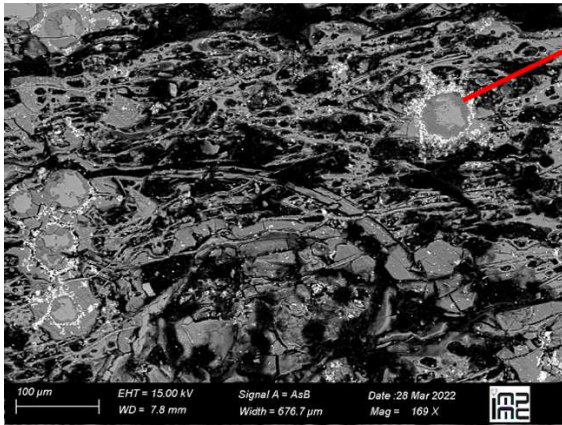


Figure 5.9- SEM images of the glazed sample: QA- 3001

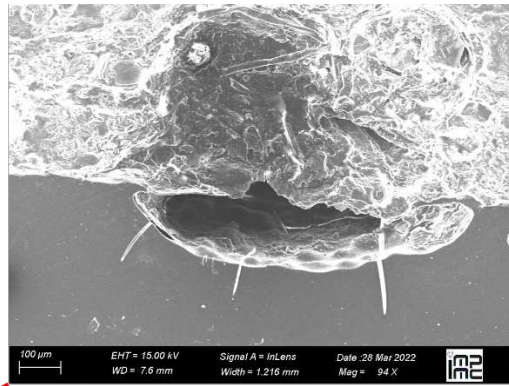
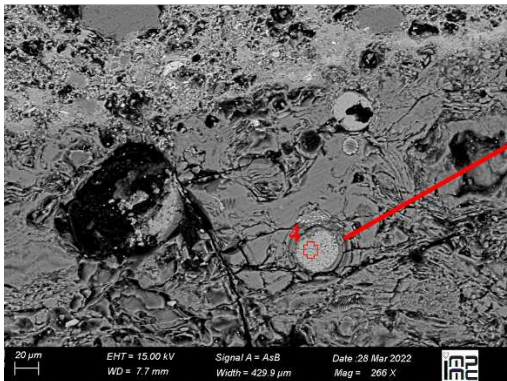


Figure 5.10-SEM images of the glazed sample: J- 2001

Table 5.4- EDS result of figures 5.9, and 5.10 (Wt%)

Spectrum number	Oxygen	Carbone	Antimony	Sodium	silicon	Calcium	Copper	Aluminum	Potassium	Lead
1	14.51	4.33	-	-	-	80.44	-	-	-	-
2	39.86	11.45	-	-	-	47.88	-	-	-	-
3	20.6	8.14	46.95	8.75	10.47	2.28	2.33	0.27	0.12	0.03
4	45.25	12.24	-	-	1.57	39.83	-	-	-	-

In addition, it should be said that sodium antimonate was seen as an opacifier only in Qalaichi samples and calcium antimonate can be mentioned as the main opacifier of Elamite samples. As shown in Figure 5.15c, 5.15e, and 5.15g, the use of calcium antimonate as an opacifier was common in the Middle Elamite, Neo Elamite, and Parthian-Sasanian periods. But sodium antimonate has not been used as an opacifier in these periods. The non-use of sodium antimonate in Neo Elamite glazes and the use of calcium antimonate as an opacifier shaped the assumption that the Neo Elamite glazes continue the tradition of the Middle Elamite glazes. It should be noted that in the Neo Elamite glazes there are metallic nanoparticles in glazes (Figure 5.14) that are similar to those observed in Qalaichi glazes, while there are no such nanoparticles in the Middle Elam glazes. This issue is important because the formation of nanoparticles in glazes can be related to the firing conditions. In this way, controlled combined reducing/oxidizing sequences atmospheres is achieved to allow the formation of metal nanoparticles (Colomban, 2009, 2021). However, according to the SEM images obtained on Middle Elam samples (Figure 5.11), what is clear is the absence of nanoparticles in the glazes of this period (Raith et al., 2022: Fig. 16, 15, 7). In other words, globule and elongated structures have been seen in Middle Elamite glazes. In addition, the elongated shape of the bigger particles and globular particles in the glazes are related to the anisotropic and homogenous firing temperature of the glazes (Colomban, 2009). So, it is possible to talk about a different technique in making Neo Elamite glazes compared to the Middle Elam and, possibly, a firing technique that is more similar to Qalaichi. However, the opacifier of Neo Elamite glazes is also different from Qalaichi glazes. Nanoparticles have also been seen in the glazes after the Elamite period (Parthian and Sasanian periods) in Khuzestan (Figure 5.12). This means that during the Parthian and Sassanid periods in Khuzestan, the glaze-making technique is more similar to the glazes of Neo Elam and not Middle Elam. Regarding the presence of nanoparticles in the glazes of Neo Elam and its similarity with the glazes of the Iron Age in the northwest of Iran, it should be said that the amount of boron element in the samples of Neo Elam from Jubaji and Qalaichi samples is very high (Figure 5.13). Glass with high boron percentage was mentioned earlier in the Byzantine period, which can indicate their origin (R. Brill, 2002; Schibille, 2011; Swan et al., 2018). In other words, high boron (more than 1600 ppm) in the glaze could be related to the source of materials that were used for making the glazes, because boron is thought to enter the glass largely via the fluxing agent (Devulder et al., 2014). Boron deposits in Iran were reported in the northwest of Iran, which are related to hot springs (Assadpour et al., 2017), and

there is a Boron mine in Zanjan province (northwest of Iran) including Mg-Ca-borate (Rahimpour & Kazemi, 2003). In this way, it can even be said that the origin of some of the Neo Elamite Jubaji glazes is more similar to Qalaichi samples and different from the Middle Elamite samples in Haft Tepe and Chogha Zanbil.

Another issue is the black color in the studied glazes. By paying attention, to the PCA plot, it seems that black glazes have a very similar structure to blue (turquoise) glazes. Therefore, it is difficult to discuss the coloring agents in these glazes using the PCA plot. However, according to the Raman spectra, it seems that iron sulfide was used as the color agent of black color in the Middle Elamite glaze of Chogha Zanbil (CZT- 4013). The presence of two Raman peaks at 415 and 375 cm^{-1} can be an indicator to identify the black color resulting from iron sulfide (Figure 15 b). Raman peak at 415 cm^{-1} originated from the Fe-S chromophore (Prinsloo & Colombari, 2008), and the Raman peak around 420 cm^{-1} and its shoulder are related to the Fe^{3+} - S^{2-} chromophores which appear in spectra of numerous black glazes of Assur and Babylon (Alloteau et al., 2022). However, in Assur and Babylon black glazes the copper sulfide nanoparticles are identified along with Fe-S. In addition, in the Parthian-Sasanian black glaze from Haft Tepe (HT-1901), copper sulfide nanoparticles were also identified by SEM, indicating that it was also used as a black color agent (Figure 12), which can be considered a continuation of the same technique of Assyrian and Babylonian black glazes.

With regards to the PCA plot (Figures 5.7, and 5.8), the yellow glazes are located at the left side of the plot, and sample number 41 (HT- 1006) is far from other samples (as mentioned this sample could be related to the Islamic period). According to the SEM-EDS elemental mapping (Figure 14), lead antimonate nanoparticles could be considered as the yellow opacifier in Neo Elamite yellow glazes. Lead antimonate is present together with calcium antimonate. Yellow color has not been observed among the Middle Elamite glazes. Raman spectra bands also determined that lead antimonate was used in the green color of the Parthian- Sasanian glaze (Figure 15 d). The use of lead antimonate as an opacifier in green and yellow glazes from the Iron Age and the Achaemenid periods has also been reported in Tepe Rabat and Persepolis (Holakooei, Ahmadi, et al., 2017). However, in the green glaze of Neo Elam, according to Table 2 the amount of copper and iron is reported to be very high (25.06 wt% Cu, and 9.34 wt% Fe), which can be due to the

strongly colored colloform domains identified in a previous investigation (Raith et al., 2022). A point that is clear according to the PCA plot is that the two green samples 5 and 10 (J- 2009) are more related to iron, aluminum, and copper.

According to the optical microscopy image of the Chogha Zanbil glazes and Raman spectra, there are quartz grains in the white glazes (Figures 5.4, and 5.15h). Some studies have mentioned quartz as one of the white opacifiers in glaze (Maltoni & Silvestri, 2018; Matin, 2018). However, this possibility should be kept in mind that quartz can be formed in the glaze during the condition of sample burial (Holakooei, Ahmadi, et al., 2017). Since coarse grains of quartz can be seen in abundance only in white glazes, therefore, the hypothesis that considers quartz as a white opacifier can be considered more probable. In addition, the Persian Islamic texts mentioned quartz in the white opaque glazes recipe (Afshar & Daryāgasht, 2003; Kashani, 1966). Furthermore, the use of calcium antimonate as the main white opacifier in the vitreous materials is documented from the 3rd millennium BC in the Near East (Tite et al., 2002). In addition, the Raman bands at 239, 325, 340, 521, and 670 cm^{-1} indicate the presence of calcium antimonate (CaSb_2O_6) (Gedzevičiūtė et al., 2009), Raman bands, shown in Figure 15g, are related to the Elamite white glaze that indicates the presence of calcium antimonate in the Elamite glazes. Caubet also mentioned the use of calcium antimonate as an opacifier in the Middle Elam glazes of Chogha Zanbil (Caubet & Bouquillon, 2007b). Considering Figure 15c, calcium antimonate Raman bands have been detected as an opacifier also in the blue glazes during the Neo Elamite period. Also, according to figures 11, 12, and 14, it can be pointed out that the presence of calcium antimony in Khuzestan glazes in different colors can be detected from the Middle Elamite period to the Parthian-Sasanian period.

Calcium antimonate does not exist in nature. It is attested in the form of hexagonal (CaSb_2O_6) or cubic ($\text{Ca}_2\text{Sb}_2\text{O}_7$) crystals (Boschetti et al., 2020). It should also be noted that the cubic form of calcium antimonate exhibited Raman bands at 318, 367, 472, 624, 781, and 821 cm^{-1} (Gedzevičiūtė et al., 2009), which was not detected in any of the Elamite samples. Calcium antimonate formed in the glass (or glaze) during the cooling phase, and it seems the presence of MgO appears to contribute to the segregation of CaO, which in turn may promote the formation of $\text{Ca}_2\text{Sb}_2\text{O}_7$ (Boschetti et al., 2020), and CaSb_2O_6 crystals dissolve in the vitreous matrices when the temperature is about 1225°C (Lahlil et al., 2010). This means that the firing temperature of Elamite

glazes is not higher than 1225°C. In the following, an attempt has been made to introduce a method through which it is possible to accurately discuss the firing temperature of ancient glazes.

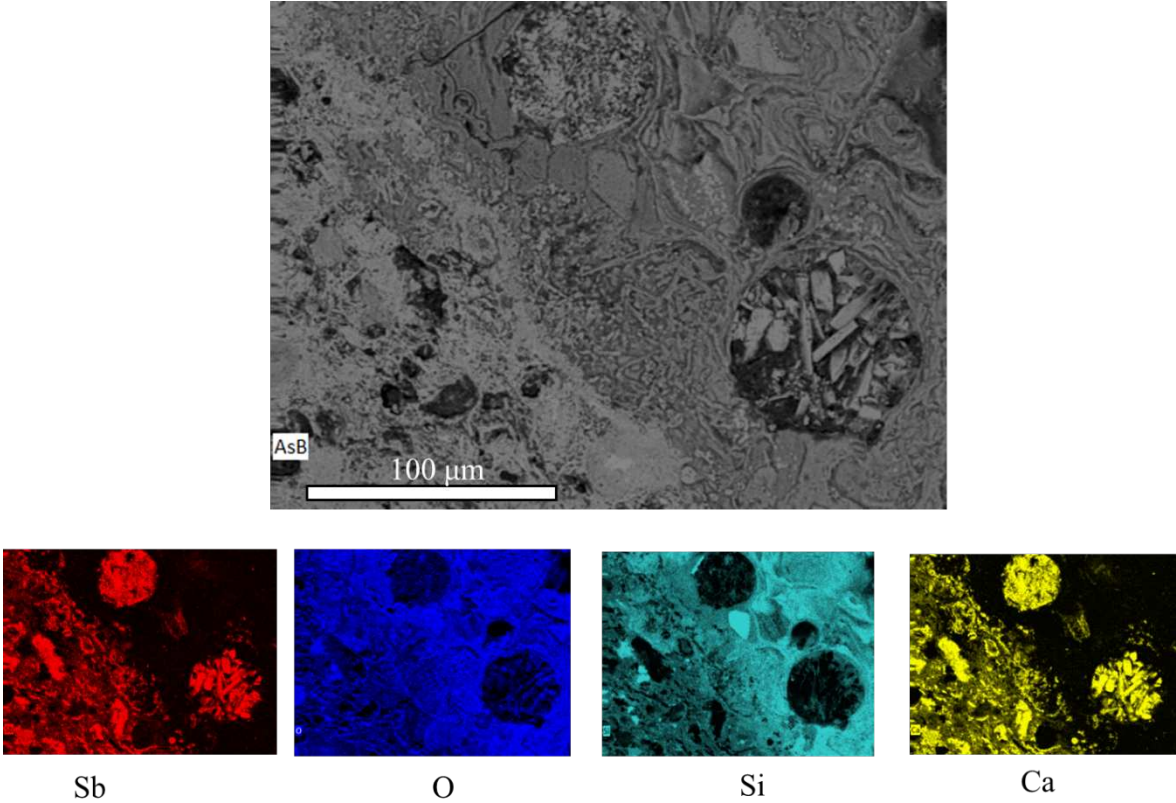


Figure 5.11- SEM-EDS image and mapping of: CZT- 4011

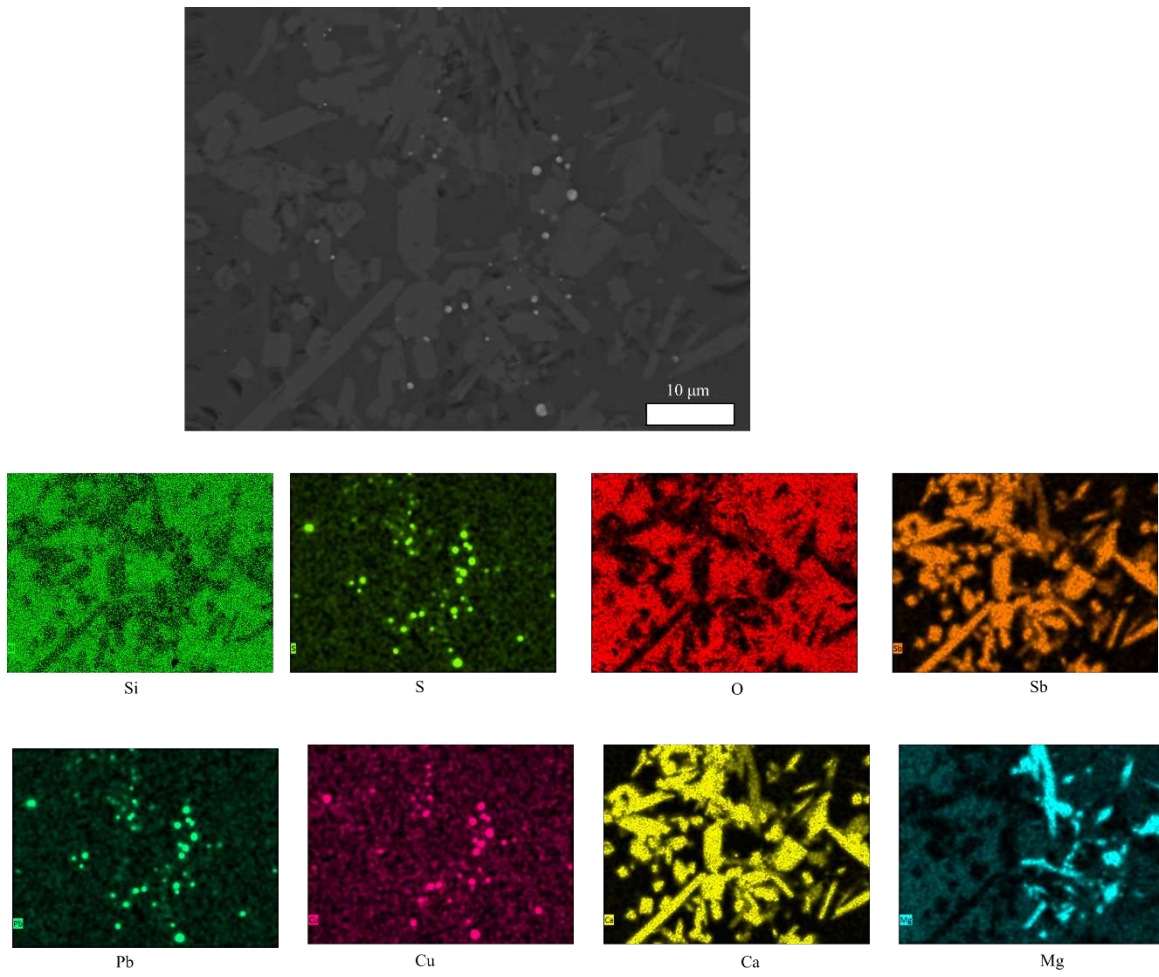


Figure 5.12- SEM-EDS image and mapping of: HT- 1901

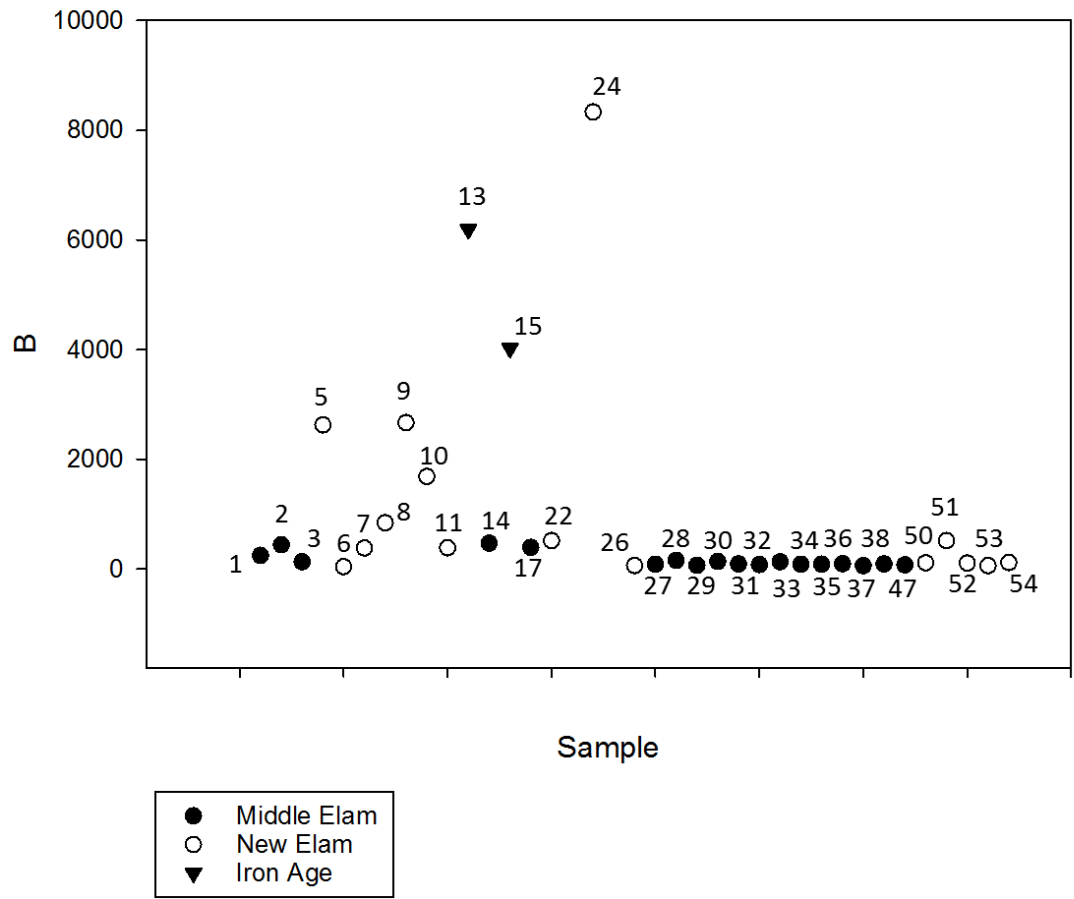


Figure 5.13- Boron content in Elamite glazes (ppm)

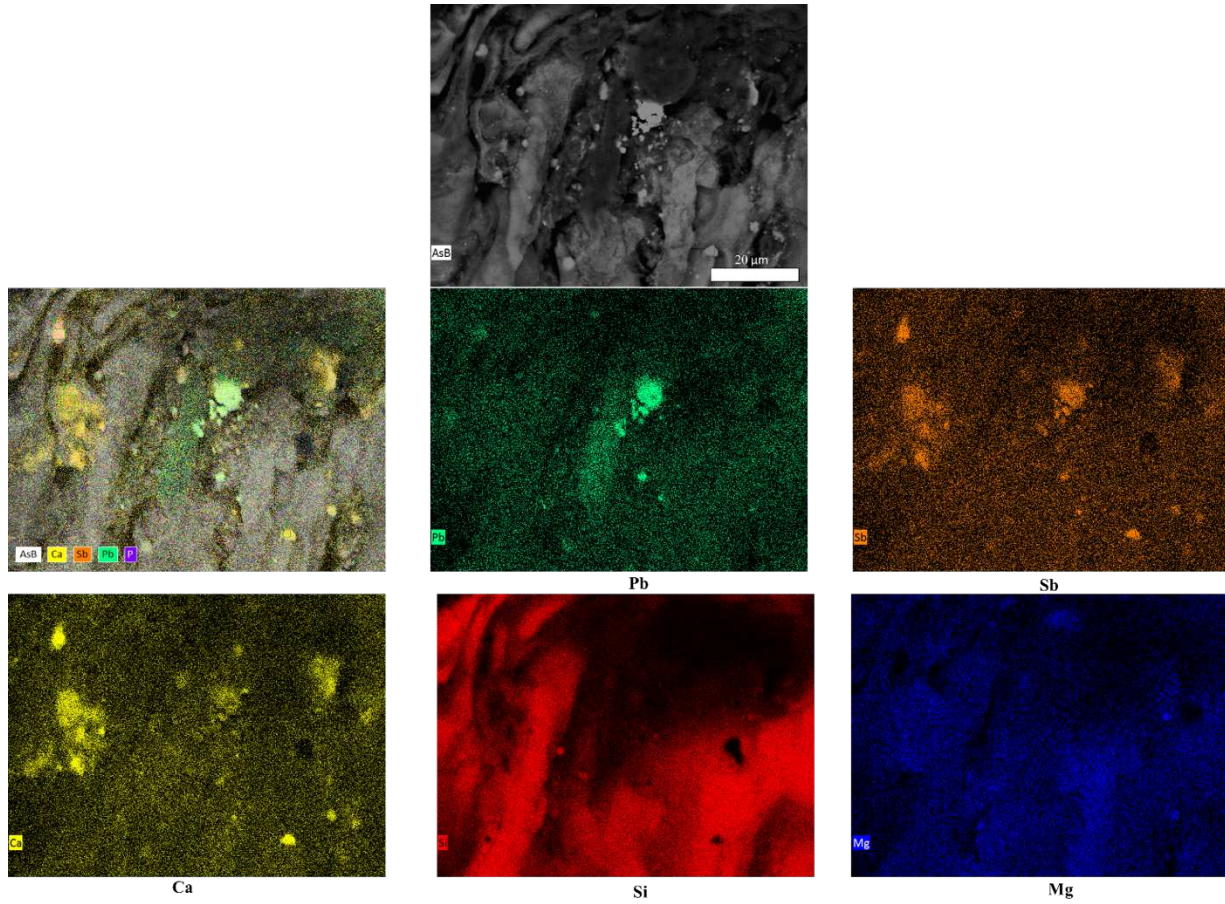


Figure 5.14- SEM-EDS image and mapping of: J- 7009

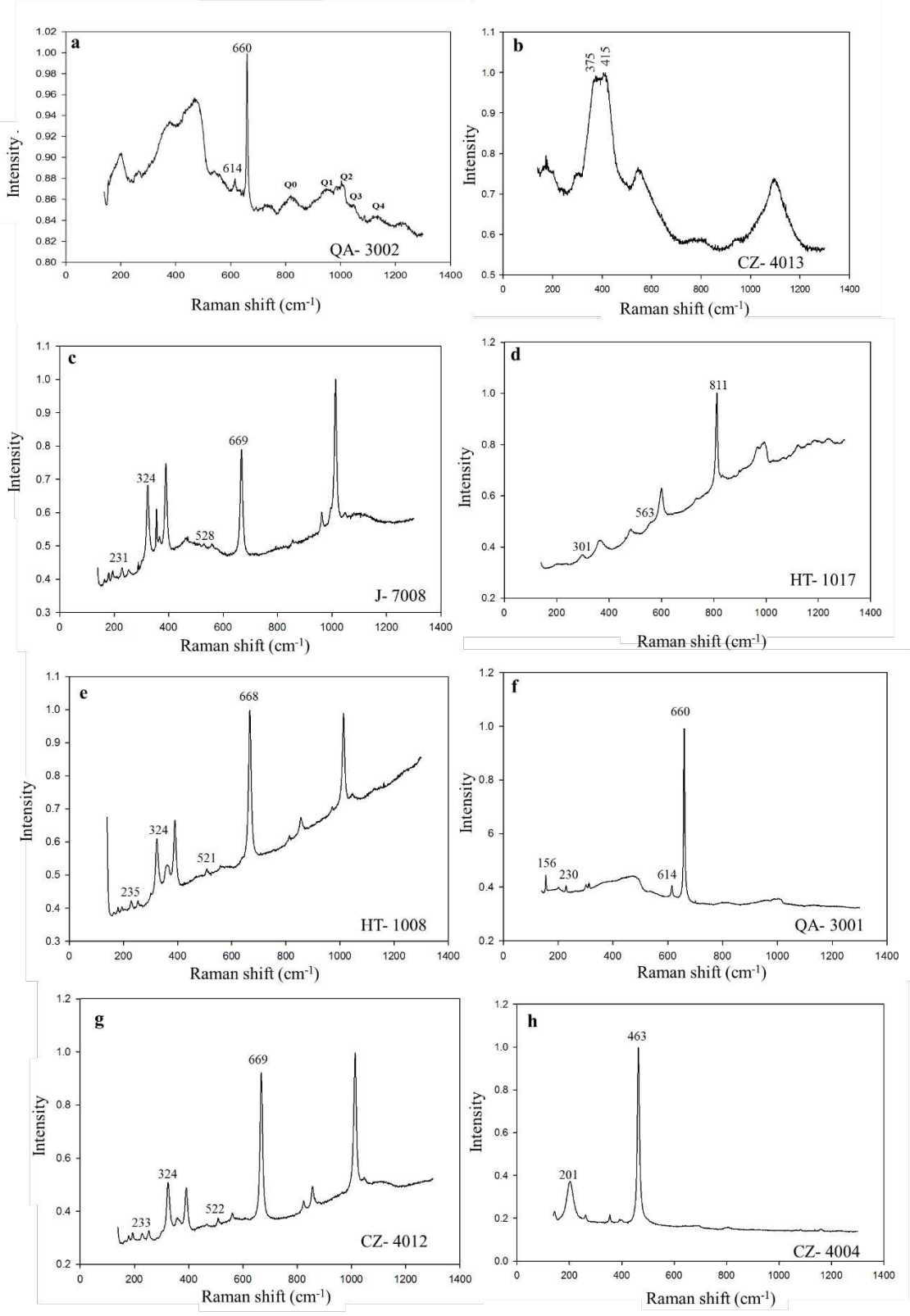


Figure 5.15- The Raman bands of a) Sodium antimonate, and Q₀-Q₄ bands of table 3 b) Iron sulfate c, e, g) Calcium antimonate f) Sodium antimonate h) Quartz

5.3.3-Processing temperature of the glazes

The processing temperatures of all glazed samples are calculated using the approach developed by Lakatos et al. (1972). The calculation of Lakatos et al. (1972) is chosen because it integrates considerable contents of lead in contrast to modern glass calculation methods (Gradmann, 2016). The viscosity could, in principle, be described by an Arrhenius-type expression. However, as the structures of most glasses vary with temperature, the activation energies also vary, and thus one cannot use an Arrhenian expression to calculate the viscosity. Exceptions have only been reported for very few network-glass forming oxides, such as SiO₂, GeO₂, and P₂O₅ glass, for which the structure is temperature-independent. Hence, the viscosity versus temperature curves of most glasses are usually described by an empirical expression of the Vogel–Fulcher–Tamman (VFT) type (Zanotto, 1998):

$$T = T_0 + \frac{B}{\log \eta + A}$$

A reasonable viscosity to form a glassy material as a glaze layer is assumed to correspond to a value of $\eta = 10^3$ Pa.s (Gradmann, 2016). From this viscosity value, it is thus possible to determine a temperature that is considered to be the firing temperature. In addition, A, B, and T₀ are empirical fitting parameters that depend solely on the glass composition. In the present work, these parameters are chosen according to the Lakatos viscosity model (Lakatos et al., 1972). To have reliable results, we have considered less altered samples, and thus with higher percentages of alkaline elements (Table 5.5). As mentioned above, all the Elamite glazes are alkali glazes, so the calculated firing temperatures of Elamite glazes are presented in Table 5. In this table, according to the Lakatos model, the amount of elements sodium, magnesium, calcium, potassium, and aluminum should be in mole percent, so the percentage of these 5 elements has been converted from weight percent to mole percent. To check the rate of change of glaze firing temperature based

on the Lakatos model, points were selected from different parts of a glaze (for samples CZT- 4009 and J- 2004). Finally, it was determined that the estimated firing temperature based on the Lakatos model from different parts of a glaze will be very close to each other.

As shown in figure 5.16, the average firing temperatures for Jubaji samples is higher than that for Chogha Zanbil samples. The standard deviation is also higher in Jubaji samples. Perhaps the reason for that is more color variation among Neo Elamite (Jubaji) glazes than that of Middle Elam (Chogha Zanbil) glazes (Standard deviation of Chogha Zanbil glazes: 32.33, and for Jubaji glazes: 44.47).

Also, according to Figure 5.17, it is clear that there is an inverse relationship between the molar percentage of alkali elements ($\text{Na}_2\text{O} + \text{K}_2\text{O}$) and the firing temperature of glazes. This observation reflects well the fluxing role of these elements. The three samples that are farthest from the regression line in this figure are the green glazes of Middle Elam and Neo Elam. It seems that the main reason for the deviation of these three green samples from the regression line is the high amount of aluminum in these samples. In other words, the high amount of aluminum in the green samples caused an increase in the viscosity of these samples and, as a result, their firing temperatures also increase. Probably, the reason for the high percentage of aluminum in the green samples is due to the high iron content in these samples as one of the factors of the green color in the glaze. Indeed, it can be proposed that the iron ore used for the green glazes is probably containing a high percentage of aluminum oxide.

Table 5.5- Firing temperature and elements of Elamite glazes (Mole%)

Sample/Color	MgO	CaO	Na ₂ O	K ₂ O	SiO ₂	Al ₂ O ₃	Firing temperature (°C)
CZT -4012/ Blue	5.02	7.49	13.32	2.35	71.2 4	0.58	898.9
J-7008/Blue	5.18	7.46	5.30	5.04	75.3 9	1.64	977.2
CZT-4009/ Greenish blue (Point1)	10.8 5	12.6 2	6.10	2.96	63.2 6	4.21	979.5
CZT/4009 Greenish blue (Point2)	13.6 0	7.75	5.48	2.90	68.0 6	2.20	975.5
J-2008/ Green	7.85	10.3 5	6.77	3.01	65.3 2	6.70	1013
CZT- 4013/ Black	3.94	6.05	11.42	1.57	76.4 4	0.57	940.4
J-2005/ Blue	5.42	8.33	10.51	3.97	71.0 1	0.75	913.2
CZT-4014/Blue	5.02	9.09	13.63	2.50	69.3 5	0.40	882.2
CZT-4004/ Blue	3.13	4.81	7.68	1.59	82.4 3	0.36	988.2
J-2501/ Green	3.51	4.82	9.24	4.37	71.2 6	6.79	1003
J-2585/ Blue	3.89	6.07	10.54	3.66	69.6 4	6.21	982.5
CZT- 4003/ Blue	3.17	5.89	9.05	1.72	79.5 5	0.62	969.1
CZT- 4005/ Blue	4.52	6.18	9.25	1.72	77.4 8	0.84	969.8
CZT-4002- blue	3.30	6.46	10.25	2.12	77.5 0	0.37	946.8
CZT- 4006/ Blue	4.19	6.33	11.90	2.15	75.2 1	0.23	924
CZT- 4007/White	6.79	8.01	8.96	1.83	72.2 9	2.12	965
J-2004/ Yellow (Point 1)	8.23	11.5 8	5.26	2.64	65.8 9	6.39	1026.2
J- 2004/ Yellow (Point 2)	6.31	8.70	4.25	3.10	71.8 7	5.77	1040
CZT- 4010/ Blue	3.10	5.27	8.45	1.89	80.9 5	0.34	974.4
CZT-4001- Blue	4.67	7.84	13.21	2.10	71.5 1	0.66	903.3

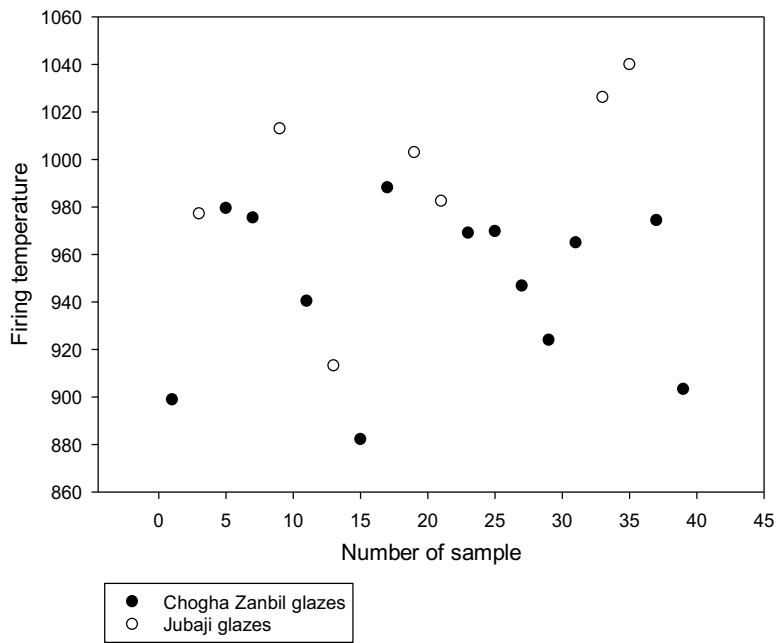


Figure 5.16- Firing temperature of New and Middle Elamite glazes

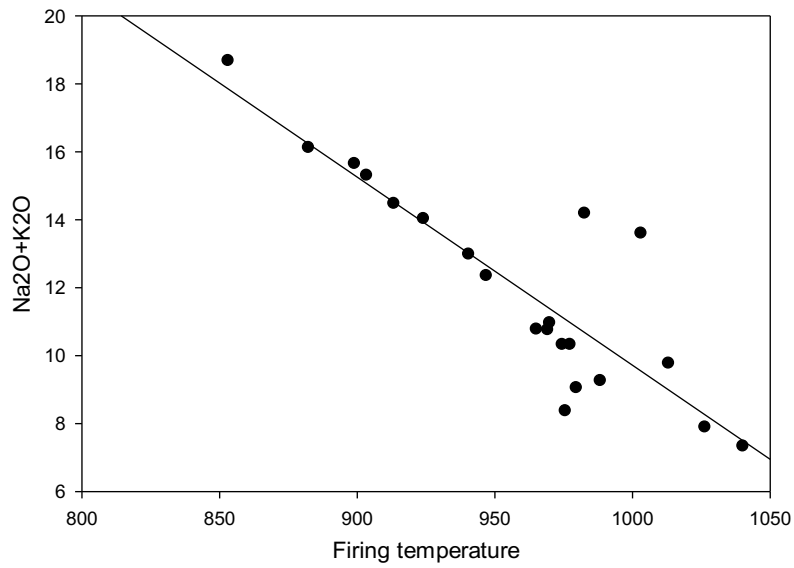


Figure 5.17- Regression line of alkali elements and firing temperature of the glazes

5.3.4- Firing atmosphere of the glazes

Another topic that can be useful in the study of ancient glazes is the firing conditions of the glazes in the kiln. Of course, little research has been done on the firing conditions of ancient glazes. In the meantime, it can be referring to the lustre glazes of the Islamic period, which are among the few ancient glazes that have been discussed in many types of research related to the firing conditions of these glazes. In this way, even the strength of the reduction condition can effect the abundance of metal nanoparticles in the glaze. (Pérez-Arategui et al., 2001; Pradell et al., 2013). As mentioned, metal nanoparticles were not observed in Middle Elamite glazes (Haft Tepe and Chogha Zanbil) (Figure 5.18). Not only during this study, metal nanoparticles were not found in middle Elamite glazes, but also metal nanoparticles were not found in another study that was conducted on Chogha Zanbil glazes (Raith et al., 2022). However, in the iron age glazes of northwestern Iran, there is a significant abundance of metal nanoparticles (Figure 5.9). But another way through which we can discuss the firing atmosphere of the samples is ceramic bodies. In other words, a higher amount of magnetite represents a reducing atmosphere and a higher amount of hematite indicates an oxidizing atmosphere (Seetha & Velraj, 2016). According to the fourth chapter, what was determined by the XRD analysis is the presence of magnetite in the ceramic bodies of Qalaichi (Figure 5.19). In other words, Qalaichi ceramic bodies, like their glazes, confirm the existence of reduction firing conditions in the Iron Age samples of northwest Iran. Gray pottery in the Iron Age in northern Iran is one of the indicators of the use of reduction kilns in the production of pottery. Such a way that many researchers consider the existence of this pottery (gray pottery) to be related to the arrival of new people from the northeast or northwest to Iran (Fahimi, 2020; Kuz'mina, 2007). It seems that the newly arrived people in Iran have brought with them the new technique of firing pottery (reduction kilns), and the use of the new technique has led to changes in the glaze-making in the northwest of Iran. The reason for the absence of metal nanoparticles in the glazes of Middle Elam is that the southwest of Iran still does not have gray pottery (reduction firing) during the second millennium and the beginning of the first millennium BC. In other words, the predominant pottery in southwestern Iran in these periods is buff ware. Unlike the north of Iran and even the center of Iran, where the gray pottery, which probably belonged to the new people, has the predominant abundance. And even the presence of black core in some pottery of southwestern Iran probably is more related to both the addition of organic matter (such as straw) to the clayey material and an incomplete oxidizing atmosphere in the kiln (Oudbashi, Naseri, et al., 2021).

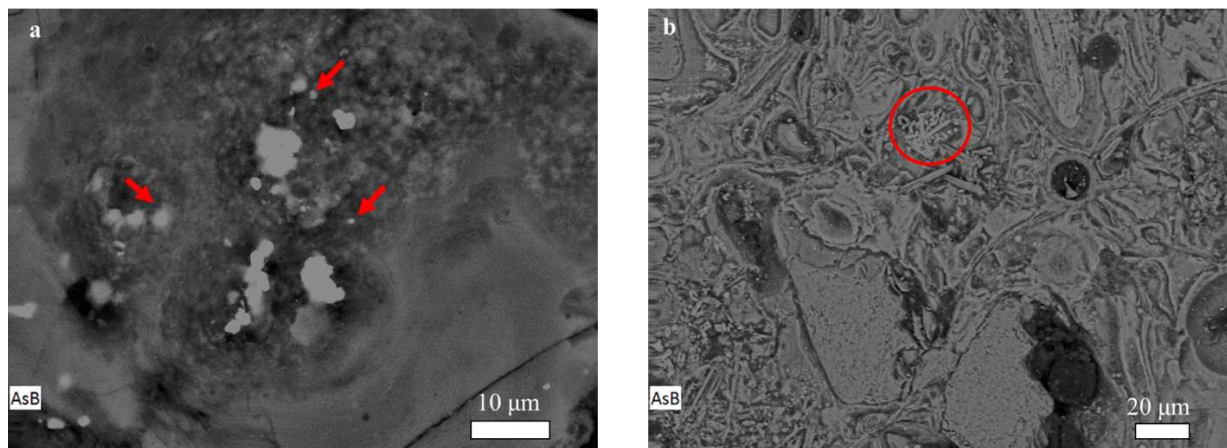


Figure 5.18- The difference between calcium antimonate in the glaze of the Neo Elam (nanoparticles) (a) and the glaze of Middle Elam (b)

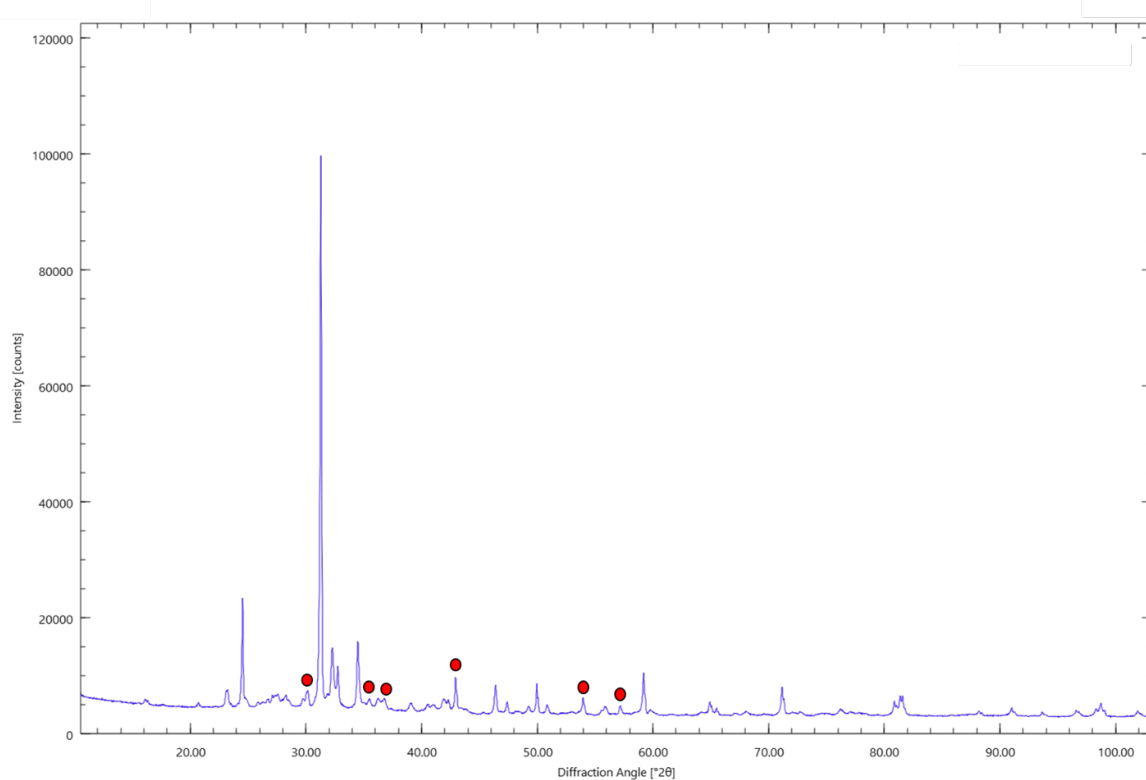


Figure 5.19- The presense of magnetite in sample Qalaichi- 3001, according to the XRD pattern

5.4-Conclusion

According to the present study, the selection of Haft Tepe Elamite glazes is associated with the problems of stratigraphy of the Elamite layers. But such problems are not unique to the Haft Tepe site and similar situations can be encountered at other ancient sites. Therefore, several Parthian-Sasanian and Islamic glazes were also selected to get a better understanding of the glaze production process after the Elam period in Khuzestan and to make a more accurate comparison with Elamite glazes. What was found is that, in the southwest of Iran (Khuzestan province), most of the glazes from the Elam era to the Islamic era are alkaline glazes, and the addition of lead with an amount of more than 3 percent to the glaze is only specific to the Parthian-Sasanian era and in the Elamite period no such feature was observed.

The use of sodium antimonate as an opacifier was observed only in Iron Age glazes northwest of Iran, and this is the main distinguishing feature of Iron Age and Elamite period glazes. However, it should be noted that the amount of boron element is very high (more than 4000 ppm) in Iron Age glazes, a phenomenon that was also observed in 3 samples of Neo Elamite glazes. Such a feature can indicate that some of the Neo Elamite glazes can be imported to Khuzestan (southwest of Iran) one of the centers of Elam. This matter is important because it investigates the trade relations of Elam to find the toponyms related to the supply of metal resources of Elamite centers. One of the areas that have always been considered as the lands supplying Elam's metal resources is the northern neighbors of Elam. The existence of possible trade relations between Elam and its northern neighbors in the period of Neo Elam can be a sign of trade relations between Elam and its northern neighbors in the periods of Middle Elam and Old Elam too. The periods that, according to the Elamite and Mesopotamian texts, trade-in metal resources were of special importance. But to find out the presence or absence of trade relations between Elam and its northern neighbors during the Middle Elamite period, it is necessary to conduct such a study on the iron age glazes of northwestern Iran, and especially on the boron element (an element whose abundance in the North West of Iran has been fully confirmed).

However, with the research that has been carried out so far on the glazes of Middle Elam, it was found that mostly nanoparticles were not observed in the glazes of this period. This phenomenon is related to the glaze firing technique and can be seen in Iron Age, Neo Elam, and Parthian-Sasanian glazes. This issue itself can be a sign that probably the Neo Elamite glazes were formed under the influence of iron age glazes, although accepting such a hypothesis requires a study with a focus on the Iron Age glazes of northwestern Iran with such a view.

It was also found that the Lakatos method can be used in the study of ancient alkaline glazes that are less altered, and the firing temperature of the glazes can be estimated with this method. Based on the current research, it can be said that the average firing temperature in Neo Elamite glazes was higher than in Middle Elam glazes. It was also found that the changes in the glaze in the first millennium BC in Iran could be due to the arrival of new people in Iran and the widespread use of reduction kilns by them.

Chapter SIX: Final remarks and conclusions

This thesis represented one of the first studies on glazes that can be considered Elamite with high certainty. In addition, it became clear that the first glazes of Iran should be considered in Haft Tepe and not in Chogha Zanbil. Also, Haft Tepe glazes can be one of the first glazes on ceramics in the ancient world. Therefore, they are of great importance. But one of the problems in Haft Tepe is related to the stratigraphy of its Elamite layers. Of course, such problems exist for almost all ancient sites that have several settlement periods. Therefore, it was tried to find patterns for dating ancient glazes using their compositions and chemical characteristics. However, without a doubt, the first step in studying glazes would be to find texts that discuss the method of glaze-making. Unfortunately, no text was found on how to make ancient glazes before Islam. It can only be referred to one text from the Neo-Assyrian period, which is related to glass-making (not glaze-making) and does not have many details (R. H. Brill, 1972). But there are texts from the Islamic era that have mentioned in great detail how to make the glaze in the Islamic era. The importance of Islamic texts in the present study was because they were able to form a more comprehensive view of the traditional (pre-modern) glazing of Iran. It was also found that the version that Iraj Afshar used to correct Neyshabouri's treatise has many defects, but Porter found a version in Tashkent that has fewer defects and can be more useful in studying the method of making glaze (Porter, 2004).

In addition, one of the mistakes that have been common for years was related to not combining the two plants Shura and Ushnan, which Allan refers to base on Kashani's treatise (Allan, 1973). But it was found that he had a misunderstanding of the Kashani text, and the combination of the two plants Shura and Ushnan will also lead to glaze. Also, the comparison of the analysis of Islamic glazes with the texts of that period showed that those texts can be considered reliable documents in the study of ancient glazes.

Regarding the words related to different colors during the Elamite period, it was found that the Elamites used stones, minerals, and colored materials to express different colors (for example, gold as yellow/golden color). In other words, they did not have specific words to express colors. They attributed color to something that was naturally colored. So the Elamites did not have words

like red or blue to represent a color. Regarding glazes, one of the common ways to understand that glaze is mentioned in the Elamite text is the use of colored materials after words related to brick.

One of these colored materials that is repeated in Elamite and Mesopotamian texts is Marhashi or Duise stone. According to Elamite and Mesopotamian texts, we know that Marhashi and Shimashki are two of the most important toponyms that supply various colored materials (minerals and precious stones) to Elam and Mesopotamia. In addition, the name of Marhashi has been associated with Shimashki in late third millennium BC texts as two important toponyms (Lamberg-Karlovsky, 2013). Therefore, the location of these two toponyms could be important in this thesis. In this way, an attempt was made to locate Shimashki with the available evidence. Linear Elamite writing is a new solution that can be used to help place Shimashki. In this way, in the end, it is more likely that the eastern region of Iran can be suggested as the place of Shimashki. As one of the toponyms that supplied color materials to Elam and made the world of Elam colorful.

However, it is still not possible to discuss definitively the location of Shimashki. Perhaps the best way to find the place of toponyms like Shimashki and Marhashi, who supplied metal materials to Elam and Mesopotamia is Isotope analysis. So far, lead isotope analyzes have been carried out on the metals of Elam and its neighboring areas to find the mines supplying the metals (Oudbashi, Rademakers, et al., 2021; Oudbashi, Renson, et al., 2021). But it seems that metals are not very suitable for such studies. Because, after multiple melting and recycling, it is very difficult to identify the mine from which the metal has been extracted and this would affect the final analysis. But it seems that this problem is less important for glazes, while metal oxides are also used in glazes.

In the present study, LA-ICP-MS analysis was performed on the studied glazes. Since it is possible to identify trace elements in this analysis, it can also be used to investigate the origin of glazes. In this way, the amount of boron element in studied glazes was taken into consideration. Because the amount of boron in 5 glaze samples was more than 1600 ppm. The high amount of boron in these samples can indicate their different origin. Among the glazes studied, there were two Qalaichi glazes and 3 Jubaji glazes, which had high boron content. Boron mines and high levels of boron have been reported in northwest Iran. In this way, it is possible to raise the possibility that during the Neo-Elamite period, several glazes were produced outside of Jubaji and probably in northwestern Iran (Qalaichi is also located in northwestern Iran). Also, analyses such

as petrography, XRD, and EPMA were performed on the ceramic bodies of the samples. According to the study of ceramic bodies, it seems that the samples with high boron content are different from the rest of the samples in terms of the ceramic body, and in the PCA dendrogram, these ceramic bodies are different from the rest of the samples. This issue can also be a confirmation of their different origin. Besides, it seems that, according to the formation of cristobalite in the ceramic bodies of these samples, it can be said that the firing conditions of some Jubaji samples are different from other ceramic bodies, and it seems that some of the Jubaji samples were probably fired in the kiln twice. It means that the body is fired once and the body and glaze were fired together the second time. Raman analysis was also performed on the glazes, and finally, calcium antimonate, lead antimonate, and sodium antimonate were used as opacifiers in the glazes. But the interesting point was that sodium antimonate was observed only in Qalaichi glazes. Also, regarding the coloring agents in glazes, it was found that copper was used more than other elements, and we should not forget the alteration of the glaze and its color change. In addition, all glazes of Middle Elam, Neo Elam, and Iron Age can be considered alkaline glazes. However, in the Parthian-Sasanian periods, the amount of lead in the glazes increased significantly.

Also, another topic that reminds us of the different firing conditions of some samples is the study of SEM-EDS on glazes. In other words, the glazes in which nanoparticles are formed have different firing conditions from samples without nanoparticles. We know that nanoparticles in glazes are formed in controlled combined reducing/oxidizing sequences atmospheres. Nanoparticles were not observed in the Middle Elamite samples (middle and late second millennium BC), while they were significantly abundant in the Qalaichi samples (early first millennium BC). This issue becomes interesting when a comprehensive view is taken of Iranian pottery in the second and first millennium BC. It seems that in this period, it is possible to point out the abundance of gray pottery in the center and north of the Iranian plateau. Gray pottery is therefore important in the present study that the firing method of gray pottery is mainly reduction firing. Meanwhile, in the southwest of Iran, especially in Khuzestan, gray pottery is very limited. The presence of gray ware in Khuzestan can be traced at least from the Sukkalmah period (1900-1600 BC), for example, we can refer to Elamite gray flasks (Carter, 1979). However, similar gray vessels have also been recovered from the Diyala, Central Zagros, southern Mesopotamia, and Susa suggesting that it was a widely traded ware at this time (D. T. Potts, 1999). So it seems that

they are not pottery produced in Khuzestan but imported to Khuzestan from another region. In this way, it can be said that reduction firing did not exist among Khuzestan pottery in the second millennium and the first half of the first millennium BC, and buff ware forms the majority of Khuzestan potteries. In other words, the firing of pottery in Khuzestan is oxidizing and not a reduction. However, the abundance of gray pottery with reduction firing in the northern half of Iran raised a hypothesis that people from the northeast or northwest of Iran migrated to this land (Kuz'mina, 2007). The presence of gray pottery with revival firing in the northern neighbors of Iran such as Georgia and Central Asia has been reported since the fourth millennium BC, while such pottery appears in the northern half of Iran since the second half of the fourth millennium BC, and at the beginning of the third millennium BC (Bessenay-Prolonge & Vallet, 2020; Kakhiani et al., 2013). However, it should be noted that gray pottery was not only a new form of pottery but a new pottery firing and production technique. I am going to propose here a hypothesis that is still in its early stages. I present this hypothesis in the form of an answer to a question:

Why did gray pottery and reduction firing become important in the current research?

Because the absence of nanoparticles in the glazes of Middle Elam in Khuzestan is due to the lack of familiarity or the lack of use of reduction firing atmosphere in that region. The arrival of newly arrived people in the northern parts of Iran led to the production of glazes with nanoparticles in the northwest of Iran and the Qalaichi during the early Iron Age, in which nanoparticles were very abundant. Because in the northwest of Iran, due to the arrival of new people, reduction kilns have been well-known and used. Finally, although it is possible to identify the beginning of making glaze on ceramic bodies in Iran with Haft Tepe in Khuzestan, the familiarity of the northern regions of Iran with ceramic reduction firing atmosphere led to the production of a new type of glaze that was used until the Islamic era, later, this technique was used a lot in Islamic lustre glazes. Kashani in his treatise called lustre glazes "*Do Atashe*", which means the glazes with two firing regimes. We know that the main reason for the shine of lustre glazes are these metal nanoparticles (Pradell, 2016). In this way, it seems that the firing conditions of these glazes are so special that the names of these glazes were given based on their firing conditions in ancient times.

Among the colored glazes of Elam, only the possibility of the presence of nanoparticles in the black glaze of sample CZT-4013 can be considered. Because in this glaze based on Raman studies, we can consider iron sulfide as the coloring agent. The presence of sulfate in this glaze can indicate the reduction firing of the glaze. But it should be kept in mind that it was also possible to use raw

materials with iron sulfide. Also, the black glaze is very limited in the Middle Elamite period, and most of the glazes of Middle Elam are in white and turquoise colors. The black sample of CZT-4013 is also heavily altered, which made it very difficult to carry out more detailed studies.

Also, another point that can strengthen the proposed hypothesis is the answer to the question that Alloteau et al., raised recently (Alloteau et al., 2022). They ask why before the 8th century BC, the use of manganese as a black color agent was widespread, but suddenly in the 8th century BC, copper sulfide nanoparticles became the black color agent in Assyrian glazes? According to the proposed hypothesis, it should be said that in the 8th century BC, in the west and northwest of Iran, we see sites such as Ziwiye, Hasanlu, Qalaichi and, Rabat. Areas whose prosperity can be related to the arrival of new people (Razmjou, 2005). These people have used gray pottery (reduction technique) a lot. Due to Assyria's extensive connections with the west and northwest of Iran in the 8th century BC, the Assyrians became familiar with the reduction firing used by their eastern neighbors and were able to use copper sulfide nanoparticles as the main agent of black color in glazes, and they abandoned the use of manganese. This is the answer that can be considered for this question if the hypothesis is accepted. Also, in the glass found from Hasanlu in the 9th century BC, the use of metal nanoparticles is mentioned (Stapleton, 2011), which can be another evidence to strengthen this hypothesis. But it should be noted that despite the similarities between glaze and glass, there are also important differences between these two cultural materials. Therefore, the existence of nanoparticles in ancient glasses is a little far from the debate raised on glazed ceramic bodies. Of course, accepting or rejecting this hypothesis requires another study with the same point of view on the iron age glazes of northwestern Iran. To achieve a complete comparison between the glazes of Middle Elam and the Iron Age.

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Abstract

Iran has always been an important place in the history of studying glaze on ceramic bodies. According to some scholars, the glazes on ceramic bodies from the Chogha Zanbil site (1250 BC) can be considered as the first glazes on ceramics in the ancient world. However, the present research has shown that the beginning of making glaze on ceramic bodies in Iran should be known a little earlier. Indeed, we have proposed that the process of making glazes on ceramic bodies in Iran should be considered from the Haft Tepe site (1500 BC). In addition, to investigate ancient glazes, what can be helpful as the most important research prerequisite are ancient texts that concern the methods of making glazes. In this regard, it should be said that there is no pre-Islamic text about making glazes. However, the Islamic texts in Iran have dealt with the production of glaze in the pre-modern era in details and we have compiled and studied these different sources dealing with elaboration of traditional glazes. An important topic provided by the investigation of manuscripts and ancient texts concerns the terminology of colors during the Elamite period. Indeed, without a doubt, the color will be one of the main factors in the study of Elamite glazes. In our research we have seen that Elamites do not have special words to express different colors and the colors used on glazes. Alternatively, they use known color materials to express colors.

During this dissertation, analyses such as XRD, EPMA, and petrography were performed on glazed ceramic bodies, which led to the identification of the firing temperature of the samples. We have classified the samples based on their ceramic compositions. In this way, it was determined that the variety of ceramic bodies in the Neo-Elamite period is more important than during the Middle-Elamite period. It is also possible to consider the possibility of the variety of firing conditions in the Neo-Elamite period. Finally, various analyses, such as LA-ICP-MS, Raman, UV-Visible, and SEM-EDS, were performed on Elamite glazes. By performing these analyses, the coloring agents in the glazes were determined, and the opacifiers were also identified. In addition, we did not observe nanoparticles in the glazes of Middle-Elam, while nanoparticles can be identified in the glazes of the Iron Age from Qalaichi site and the neo-Elamite glazes from Jubaji site. These nanoparticles can be related to the firing atmosphere of the glazes.

Keywords: Elam; Glaze; Ceramic; Color agent; Opacifier.

Résumé

L'Iran a toujours été un lieu important pour l'histoire de l'étude des glaçures sur les corps en céramique. Selon certains chercheurs, les glaçures sur céramiques du site de Chogha Zanbil (1250 avant notre ère) peuvent être considérées comme les premières glaçures sur céramique dans le monde antique. Cependant, la présente recherche a montré que le début de la fabrication de glaçure sur les corps en céramique en Iran devrait être connu un peu plus tôt. En effet, nous avons proposé que le processus de fabrication de glaçures sur les corps en céramique en Iran soit envisagé à partir du site de Haft Tepe (1500 avant notre ère). En outre, pour étudier les glaçures anciennes, ce qui peut être utile comme le plus important préalable en recherche sont les textes anciens qui concernent les méthodes de fabrication des glaçures. A cet égard, il faut dire qu'il n'y a pas de texte pré-islamique sur la fabrication de glaçures. Cependant, les textes islamiques en Iran ont traité de la production de glaçure dans l'ère pré-moderne en détails et nous avons compilé et étudié ces différentes sources traitant de l'élaboration de glaçures traditionnelles. Un sujet important fourni par l'enquête des manuscrits et des textes anciens concerne la terminologie des couleurs pendant la période élamite. En effet, sans aucun doute, la couleur sera l'un des principaux facteurs dans l'étude des glaçures Elamite. Dans notre recherche, nous avons vu que les élamites n'ont pas de mots spéciaux pour exprimer les différentes couleurs et les couleurs utilisées sur les glaçures. Alternativement, ils utilisent des matériaux de couleur connus pour exprimer les couleurs.

Au cours de cette thèse, des analyses telles que XRD, EPMA, et la pétrographie ont été effectuées sur des corps en céramique glaçurée, ce qui a conduit à l'identification de la température de cuisson des échantillons. Nous avons classé les échantillons en fonction de la composition de la céramique. De cette façon, il a été déterminé que la variété des corps en céramique dans la période néo-Elamite est plus importante que pendant la période moyen-Elamite. Il est également possible d'envisager la possibilité de la variété des conditions de fusion dans la période néo-Elamite. Enfin, des analyses variées (LA-ICP-MS, Raman, UV-Visible et SEM-EDS) ont été réalisées sur les glaçures Elamite. Ces analyses ont permis de déterminer les agents colorants dans les glaçures et d'identifier les opacifiants. En outre, nous n'avons pas observé de nanoparticules dans les glaçures Moyen-Elamite, tandis que des nanoparticules peuvent être identifiées dans les glaçures de l'âge du fer du site de Qalaichi et les glaçures Neo-Elamite du site de Jubaji. Ces nanoparticules peuvent être liées à l'atmosphère de cuisson des glaçures.

Mots-clefs: Elam; Glaçure; Céramique; Agent colorant; Opacifiant.