

EGYPTOLOGICAL INFORMATION FROM CHEMICAL ANALYSES: THE PROVENANCE OF OBSIDIAN AND GLASS

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Some materials found in archaeological sites have chemical or mineralogical characteristics which differ from one geological site to another. Comparing the characteristics of the archaeological artifact with the characteristics of different geological environments might allow the identification of the original source of the material. This work presents chemical composition provenance studies on glass (natural and artificial) based on trace elements concentration and lead isotope composition.

On the one hand, obsidian—a natural volcanic vitreous material—found in some Upper Egyptian tombs from the Naqada period seems to originate from the oriental African volcanos, probably from Ethiopia, or from the Arabian volcanos (in western Yemen), according to the uranium, thorium and tantalum concentrations.

On the other hand, the chemical analysis of some Egyptian glasses indicates that, during the 18th dynasty, glass was manufactured in Egypt with Egyptian materials (instead of being a Mesopotamian import) and some of them were colored also with Egyptian materials (e.g. galena from the Gebel Zeit mines). The lanthanum and chrome concentrations clearly differ between glasses made in Egypt and in Mesopotamia, allowing the determination of the Egyptian provenance of glasses used in the Mycenaean world.

1. Introduction

The chemical analyses of different ancient materials might provide information on their provenance (Pollard et al. 2007). In these cases, the comparison between the chemical compositions of the material from a quarry or a mine and the material from an archaeological site allows the identification of the most likely sources of the archaeological material. Provenance studies provide knowledge on the size of territories, interactions between different cultures or civilizations and likely commercial routes in antiquity (Tykot 2004).

The provenance methods that use the analysis of the chemical composition are based on the existence of chemical or mineralogical characteristics which depend on the original geographical location of the material, that is to say, they differ from one geological site to another. The chemical characteristics might be the concentration of major and minor components of the material but also of trace components. In the case of rocks, what might change from one location to another is the mineralogical composition, i.e. the proportion of different minerals which compose the rock. In some cases, the total concentration of one element in a material is independent of the geological site but its isotopic composition depends on the location. In this sense, the determination of the lead isotopes ratios (LIA, Lead Isotopes Analysis) has been profusely applied for the provenance of a number of archaeological objects (Stos-Gale 1992).

The main procedure to carry out provenance studies based on chemical analyses might be summarized in the following steps:

1st: Chemical or mineralogical analysis of the material extracted from different ancient geological sources.

2nd: Determination of the chemical or mineralogical parameters (elements, isotopes or minerals) that characterize each geological source.

3rd: Chemical or mineralogical analysis of the material found in an archaeological site.

4th: Mathematical or statistical comparison between the characteristics of the different sources and the characteristics of the archaeological object, and elucidation of the likely source(s) of the material.

Provenance studies based on the determination of the chemical composition of the materials have also been carried out for materials used in ancient Egypt. In the present work,

two materials have been chosen because of their different nature (both are glasses but one is natural and the other synthetic), the different methodologies used for the determination of their provenance, and the Egyptological information obtained from their provenance. The main objectives of this work are: (1) to show the different methodologies employed for provenance studies of Egyptian obsidian and glass; (2) to describe the Egyptological information that might be obtained from provenance studies; and (3) to highlight the advantages of determining the chemical composition of the materials used by the ancient Egyptians.

2. Contacts with the South: The Provenance of Predynastic Egyptian Obsidian

Obsidian is a volcanic rock with a relatively high content of silicon which has some advantageous characteristics for provenance studies. On the one hand, it was widely used in antiquity and there are many archaeological samples. Data from Table I show the distance between archaeological sites and likely sources of obsidian around the world and illustrate the unquestionable interest in antiquity for obsidian. On the other hand, there is a limited number of obsidian mines worked in antiquity, very localized geographically, so that a priori there must not be a high scattering of the chemical data, favoring the delimitation of the chemical values which define each mine. The high number of archaeological obsidian artifacts and the low number of likely mines generated in the last century the development of chemical methods to determine the provenance of the obsidian samples, the first methods being

based on the concentration of barium and zirconium (Renfrew et al. 1966), where different Ba-Zr ratios corresponded to different sources.

However, the classification of the obsidians based on barium and zirconium concentrations did not allow distinguishing between some sources with similar Ba-Zr ratios. In particular, more studies were necessary to establish the provenance of obsidian samples of different volcanos in Eastern Africa (especially the obsidians from Ethiopia, Kenya and Eritrea) and in the south of the Arabian Peninsula (Yemen and Saudi Arabia). For this reason, provenance studies based on the chemical analysis of obsidian have been extended to include other elements that could distinguish between the two volcanic systems that dominate the obsidian availability in the ancient Near East. The first volcanic system corresponds to the volcanos in Cappadocia, Anatolia and Armenia, and the second one corresponds to the volcanos of the Rift Valley, from Ethiopia to Saudi Arabia. Obsidians from both systems have chemical differences because they are the result of different geological processes: tectonic subduction in the 'Anatolian' system, and intraplateau eruption in the Rift Valley. The different processes of formation induced differences in some chemical elements such as thorium, uranium, tantalum and niobium. For this reason, the Th/Ta and U/Ta ratios might be used as an indicator of the provenance of obsidians from Anatolia-Armenia or East Africa-Arabian Peninsula (Bavay et al. 2000).

In ancient Egypt, the use of obsidian during the Predynastic and the first dynasties is very rare (much rarer than in Mesopotamia) and was mostly used as a precious stone in some objects, mainly jewelry, found in high-status tombs. For example, from the

Site (Eastern Mediterranean)	Distance site-source (km)
Nahal Lavan (Israel)	800
Ali Kosh (Iran)	900
Çatal Höyük (Turkey)	200
Beidha (Turkey)	900
Tel e-Malyan (Syria)	1300
Site (other regions of the world)	Distance site-source (km)
Primorye (Russia)	200-700
Korea	400
Arizona (USA)	200
Malaysia	3500
Fiji Islands	3300
Transylvania (Romania)	> 1500

Table 1. Some examples of distances between Neolithic archaeological sites and likely sources of obsidian. Data from Moutsiou 2011.

2200 tombs excavated in Naqada, only five tombs contained obsidian (Bavay et al. 2000). Probably due to this scarcity, there were almost no studies of Egyptian obsidian composition before the study carried out by Bavay et al. (2000), who analyzed different Upper Egypt obsidians, namely:

- Fragments of vessels in Djer's tomb in Umm el-Qaab, Abydos (Naqada III C1 period).
- Fragments of vessels in the U-j tomb in Umm el-Qaab, Abydos (Naqada III A1 period).
- Obsidian bladelet in a necklace found in tomb 1629, cemetery 23, in Qaw el-Kebir (Naqada II C period).
- Obsidian necklace from tomb 499 and knife blade from tomb 743 in Naqada (Naqada II D2 period).
- Obsidian beads found in the beads workshop in Nekhen, Hierakonpolis (Early Dynastic period).

The laser ablation inductively coupled plasma mass spectrometry (LA-ICPMS) technique was used by Bavay et al. to determine the concentration of different elements such as thorium, uranium and tantalum in the Egyptian obsidians and in different geological samples from Anatolia, Ethiopia and Yemen. The results obtained in terms of the Th/Ta and U/Ta ratios are shown in Fig. 1. As it can be seen, Anatolian obsidians have Th/Ta ratios much higher and much more variable than Egyptian obsidians. Yemen obsidians have Th/Ta ratios higher than the Egyptians obsidians while Ethiopian and Egyptian obsidians seem to have very similar

ratios. The main conclusion drawn from these data is that the Predynastic or Early Dynastic obsidian used in Upper Egypt comes from Ethiopia, although it should be noted that only a small number of Egyptian obsidians were analyzed.

Some authors claim that the Ethiopian obsidian could have arrived to Egypt via a maritime route through the Red Sea, and that the obsidian commerce or transport could be related with the commerce with Punt, although there is no textual reference to this rock in the Egyptian lists of products from Punt (Zarins 1996). The traffic of obsidian in the south of the Red Sea is attested at least from the 5th millennium BC, and African obsidian was found in different archaeological sites in the Tihamah coast in Yemen (Khalidi et al. 2010). The path that obsidian followed between the Ethiopian volcanos and the Red Sea is not known yet. One of the potential ports where obsidian could have been shipped is located in the Buri Peninsula and the Gulf of Zula (in Eritrea), where obsidian samples were found in different archaeological sites from the Neolithic to the 4th millennium BC (Beyin 2011). In addition, one of the volcanos known as Kusrale was mined in antiquity for obsidian. Samples from the sites and from the volcano were analyzed by Beyin (2009), including thorium, uranium and tantalum. Fig. 2 shows the Th/Ta and U/Ta for these samples together with the values determined for Egyptian archaeological obsidians and Ethiopian geological obsidians.

As it can be seen, Eritrean samples show relatively low Th/Ta ratios, characteristic of the Rift Valley obsidians, but their Th/U ratios are always lower than the ones corres-

Fig 1.

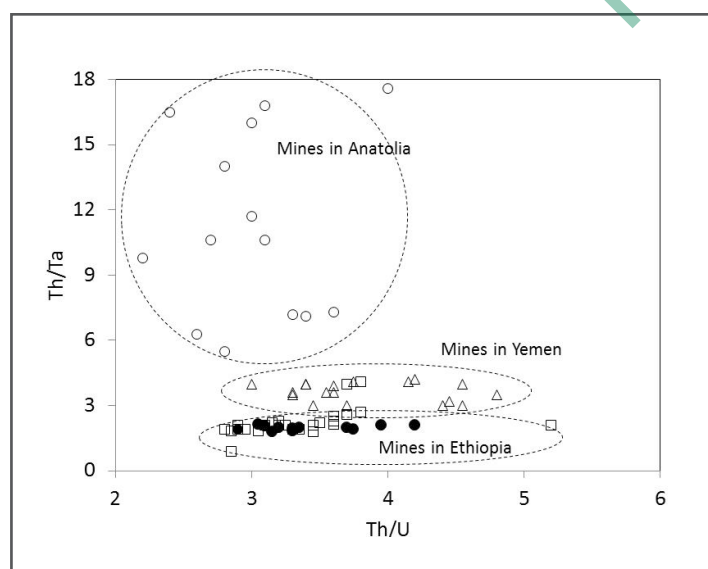


Figure 1. Experimental ratios obtained from different obsidian samples: ○ Mines from Anatolia; △ Mines from West Yemen; □ Mines from Ethiopia; ● Predynastic obsidians. Data from Bavay et al. 2000.

Fig 2.

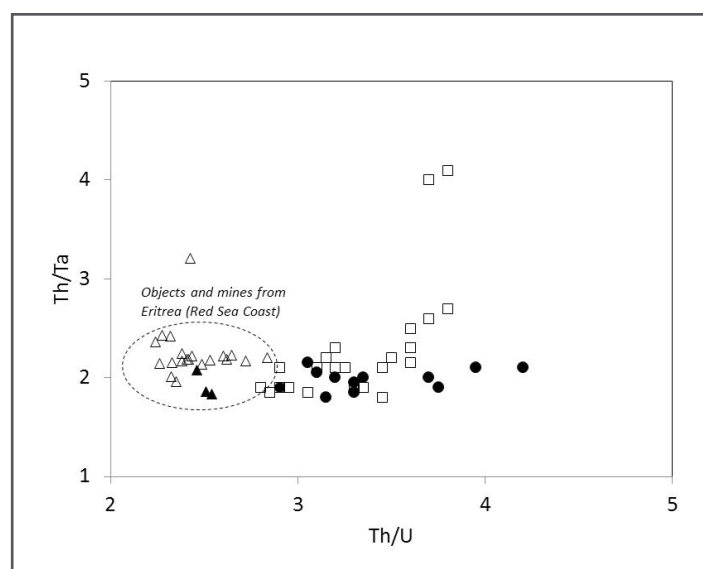


Figure 2. Th/Ta and Th/U ratios from different obsidians: △ Archaeological objects from the Gulf of Zula; ▲ Geological samples from Kusrale volcano; □ Geological samples from Ethiopia; ● Archaeological samples from Upper Egypt. Data from Bavay et al. 2000; Beyin 2009; 2011.

ponding to the Egyptian archaeological samples, suggesting that Egyptian obsidian did not come from this region of Eritrea. These results do not preclude a maritime route through the Red Sea, but indicate that the Gulf of Zula was probably not a part of this route. Even though the route through the Red Sea is generally accepted (Zarins 1989; 1996; Tykot 1996), the possibility of a terrestrial/riverine route should not be precluded. In the Mahal Teglinos settlement from the Gush culture, near the modern city of Kasala in inland Sudan, a number of objects made of Ethiopian obsidian were found together with Egyptian objects (Fattovich 1997). These findings could indicate that Mahal Teglinos was one station in a commercial route that transported obsidian from Ethiopia to the Nile River and through the Nile River to the settlements in Upper Egypt. Mahal Teglinos is currently only an indicator of a possible 'second' obsidian route to Egypt and more data are necessary to confirm or preclude its existence.

Unfortunately, the number of analyzed Predynastic obsidian samples from Lower Egypt is still lower than those from Upper Egypt and only the chemical composition of two samples was published: one sample from Tell el-Fara'in-Buto and another from el-Tell el-Iswid (Bavay et al. 2004). The chemical composition of the samples was determined by LA-ICPMS and the concentration of thorium, uranium and tantalum resulted in Th/Ta ratios of 7.10 and 6.38 for the Tell el-Fara'in-Buto and el-Tell el-Iswid samples, respectively. As it can be seen in Fig. 2, these relatively high ratios would correspond to the chemical composition of the obsidian from Anatolia and are very far from the results that characterize African obsidian sources.

If these results are considered significant, in spite of the very low number of samples analyzed, the main conclusion is that obsidian used in Upper and Lower Egypt came during the Predynastic from different sources. Although no obsidian was found in Predynastic times in Syria-Palestine, commercial routes that connected Lower Egypt with distant zones as Mesopotamia or Anatolia existed. For example, already in Naqada II lapis lazuli arrived to Lower Egypt (and also to Upper Egypt) from Afghanistan through Mesopotamia and was shipped in one of the Mediterranean ports in the Levantine coast, probably Ras Shamra or Byblos (Aubert 2013).

Although in this work the provenance of the obsidian is only based on the concentration of trace elements such as Th, Ta and U, other chemical elements could indicate the provenance of the obsidian samples. Actually, a statistical study of the concentrations of trace elements in obsidians from different volcanic regions conducted at the Universitat Politècnica de Catalunya showed that other discriminating elements could be zirconium, niobium and zinc (Alva Howes 2014).

3. Contacts with the North: The Provenance of Egyptian Glass during the 18th Dynasty

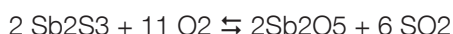
Vitreous synthetic materials were first used in Egypt probably in the Badarian period, when glazed steatite was prepared for the fabrication of necklace beads (Tite and Bimson 1989). During the Predynastic, the synthesis of faience was developed in Egypt and differed from glazed steatite in the nucleus of the object (steatite in glazed steatite and quartz or sand in faience). Glass technology started during the 15th century BC in Mesopotamia and Egypt, perhaps some years earlier in Mesopotamia than in Egypt. Recent studies on the localization of the glassmaking workshops in the Near East showed that secondary glass manufacture workshops existed in Amarna and probably the most ancient primary glass workshop discovered (Smirniou and Rehren 2011). For this reason, the provenance of glass objects from Amarna and from the reigns of the pharaohs before Akhenaten is of interest in order to establish when Egyptians started to fabricate glass in Egypt and what was the provenance of the raw materials.

Glass was fabricated by fusing together three different compounds and cooling slowly the product in order to create a supercooled liquid insoluble in water, transparent, translucent and bright. The three reactants were: (1) the principal component of the glass (quartz or sand), (2) the flux (usually plant ashes or natron), and (3) the stabilizer, which increases the durability of the glass (calcite, CaCO_3). As it can be seen, glass is a synthetic material obtained from a mixture of other materials; as a consequence, provenance studies might give inconclusive results especially if they are based on chemical compositions. The chemical composition of the glass will be the consequence of the different chemical compositions of the reactants (which might have different provenances) but also of the different quantities of each reactant added to the mixture. There are two different fields of glass provenance studies that are yielding robust results on the difference between Egyptian and Mesopotamian glasses: LIA and La-Cr plots.

3.1. The First Manufacture of Glass in Egypt: LIA Analyses

As it was mentioned above, one of the most applied methodologies for the determination of the provenance of Pb-containing materials is LIA. This procedure can be applied to the study of the provenance of some Egyptian glasses because they were colored by using a lead compound, lead antimonate ($\text{Pb}_2\text{Sb}_2\text{O}_7$), which was employed in ancient Egypt in the fabrication of yellow or green glasses (Duckworth et al. 2012). Although there is not a general agreement on the process of lead antimonate incorporation to the glass (an ex situ or an in situ synthesis), it seems that a mixture of two minerals was used: galena (PbS) and stibnite (Sb_2S_3) (Mass et al. 2002), which provided lead and antimony, respectively. The mixture of the minerals was heated to 800°C in open furnaces to obtain the antimonate through a two-step mechanism:

1st step - Sulfides oxidation by the oxygen of the air:



2nd step - Antimonate formation:



While stibnite is believed not to come from Egypt (or at least there are not known stibnite mines in Egypt), there are different mines of galena that were known in antiquity. Shortland et al. (2000) determined the lead isotopic composition of different glasses and other lead-containing Egyptian objects and compared the results obtained with the isotopic compositions of the Egyptian galena mines and of the Mesopotamian galena mines also known in antiquity.

The results of the isotopic compositions of the mines and glasses are shown in Fig. 3. As it can be seen in this figure, there is a difference between the isotopic composition of the glasses from Amarna and the glasses from Thutmose III's reign. Actually, Amarna glasses have isotopic compositions similar to the ones determined for the galena mines in Gebel Zeit, which were mined by the ancient Egyptians at least from the Middle Kingdom onward (Castel and Soukiassian 1985; 1988). On the contrary, the glasses from Thutmose III's reign (from his tomb in the Valley of the Kings and from the tomb of

his 'Syrian' wives in the Wadi Qubbanet el-Qirud) have isotopic compositions similar to the lead of the Mesopotamian galena mines.

Therefore, it is probable that the glasses found in Amarna were fabricated in Amarna using Egyptian materials. During the 19th dynasty, in Qantir Piramesses, glass was made from Egyptian raw materials in workshops that were different from the workshops where glass objects were manufactured (Rehren and Pusch 2005). The existence of separated workshops in Amarna for the synthesis of glass and the manufacture of glass objects was postulated recently (Smirniou and Rehren 2011); this would indicate that during Akhenaten's reign glass was already synthesized and fabricated in Egypt. However, the primary synthesis of glass in Egypt does not imply that the import of Mesopotamian glass had ceased. Mesopotamian glass still arrived to Egypt as it is said in some of the Amarna letters (Shortland 2007), perhaps because it was a product that the king requested as a high-level tribute or simply because it was considered as a glass of better quality.

3.2. The Import of Glass from Egypt: La-Cr Analyses of the Uluburun Cargo

The Uluburun ship sunk in the south coast of Turkey in the 13th century BC (Pulak 2008). The ship carried different materials such as unworked blue glass, blocks of "Egyptian Blue" pigment and ox-hide copper ingots (Gestoso Singer

Fig 3.

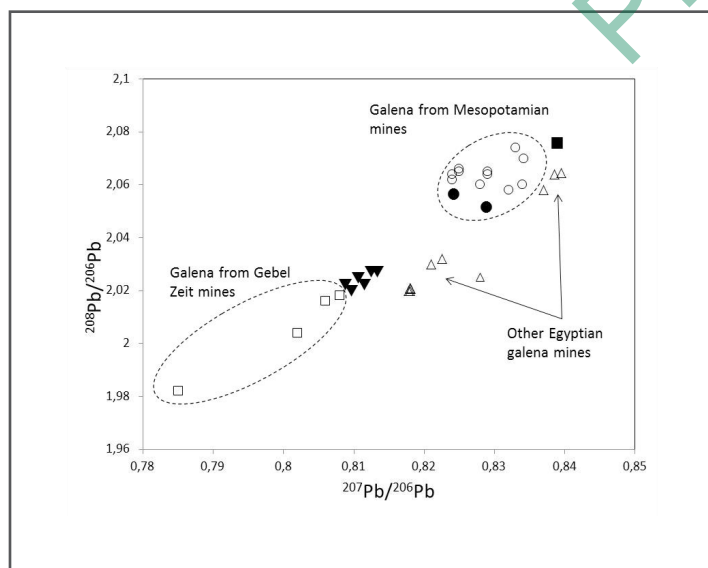


Fig 4.

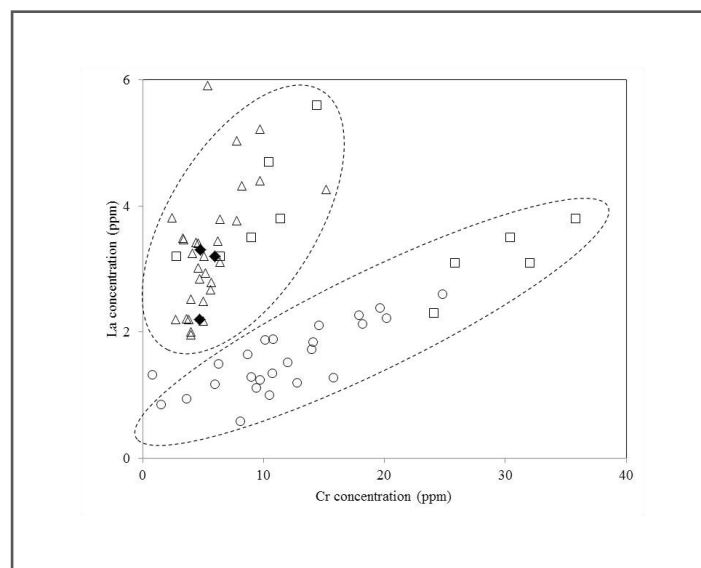


Figure 3. Lead isotopic composition of geological samples from galena mines in Egypt (□ and △) and in Mesopotamia (○) together with the values of the New Kingdom glasses: ▼ Amarna glasses; ● Thutmose III's reign glasses (Wadi Qubbanet el-Qirud); ■ Thutmose III's reign glasses (Tomb of Thutmose III, KV34, Valley of the Kings). Data from Shortland et al. 2000.

Figure 4. Cr and La concentration in different glass samples: ○ Mesopotamian glasses (Shortland et al. 2007); △ Egyptian glasses (Shortland et al. 2007); □ Mycenaean glasses (Walton et al. 2009); ◆ Glass ingots from the Uluburun ship (Jackson and Nicholson 2010).

2011). The glass from the Uluburun shipwreck was chemically analyzed in order to establish if it came from Mesopotamia or Egypt. The provenance methodology used was not based on the colorants but on the differences in the concentration of some trace elements. Shortland et al. (2007) investigated different trace elements as potential discriminants in the provenance of the glasses and concluded that Mesopotamian glasses had relatively high Cr concentrations while Egyptian glasses had higher concentrations of La, Ti and Zr.

Fig. 4 shows a plot of La and Cr concentrations determined for Mesopotamian glasses (from Nuzi and Tell Brak); Egyptian glasses (from Malkata and Amarna); Mycenaean glasses (Walton et al. 2009) and glasses found in the Uluburun shipwreck (Jackson and Nicholson 2010). As it can be seen in the figure, there are two main areas of concentration which correspond to the composition of the Egyptian glasses and the Mesopotamian glasses, respectively. Mycenaean glasses fall into the 'Egyptian composition area', probably indicating that they came from Egypt. The composition of three glass ingots found in the Uluburun shipwreck is included in the figure and falls into the 'Egyptian' area of concentration, once more pointing to an Egyptian origin of such glasses which could be corroborated by other materials found in the shipwreck such as Egyptian Blue and objects from the Amarna period (Gestoso Singer 2008). These results indicate that during the New Kingdom glass was not only fabricated in Egypt but also exported to Mycenae through a maritime commercial route which traversed the Mediterranean.

4. Conclusions

The main objective of this work was to illustrate how the application of analytical chemistry techniques to archaeological objects might provide Egyptological information. This was done through the description of the results on the provenance determination of the natural volcanic glass obsidian and the synthetic glass used in ancient Egypt.

The determination of the chemical composition of the obsidian used in the Predynastic helps locating the connecting routes between Upper Egypt and some regions of Ethiopia. It is probable that a maritime route through the Red Sea existed already during the Predynastic period, although it is possible that an additional terrestrial/fluvial route operated as well. On the other hand, in Predynastic Lower Egypt the obsidian supply seems to depend on connections with Mesopotamia and the Levantine coast (with the primary source located in the Anatolian volcanos), although the number of obsidian samples analyzed is admittedly small.

The results obtained using different glass provenance chemical procedures (isotopes or trace elements) point to the development of glass technology in Egypt between the reigns of Thutmose III and Akhenaten. At that time, the primary manu-

facture of glass from raw materials (and not only the fabrication of glass objects from glass ingots) seems to be already established. Raw materials such as galena are demonstrated to be of Egyptian origin and Egyptian glasses were later exported to the Mycenaean world.

The data and the conclusions presented in this work highlight the importance of the chemical analyses of Egyptian archaeological materials, because they provide information on the provenance of the materials and on ancient Egyptian interconnections. Obsidian and glass chemical studies are only two examples of the potential role of chemistry in Egyptology, and there are other materials such as basalt (Greenough et al. 2001), granite (Williams-Thorpe 1996), pottery (Tite 2008), turquoise (Hull et al. 2008) and lapis lazuli (Re et al. 2011) that are being chemically—or even geochemically and mineralogically—analyzed in order to acquire provenance information ■

5. Acknowledgments

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