

## Geochemistry and mineralogy of prehistoric pottery shards found at Gua Jaya, Nenggiri Valley, Kelantan, Malaysia

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**Abstract:** Earthenware pottery is one of the common artefacts found during archaeological excavation works. Earthenware pottery is one of the tools used by prehistoric society as a tool for use in daily life. Earthenware pottery found at archaeological sites should be determined whether it was made by the local community or brought in from outside. Therefore, chemical analysis using X-ray Fluorescence (XRF) and mineralogical analysis using X-ray Diffraction (XRD) methods need to be done to obtain the mineral content and elements of earthenware pottery that can be compared with clay found in the area. This comparison is to ascertain whether the prehistoric pottery was made in the vicinity of the discovery area or brought in from outside. The results of this study found that the pottery discovered during excavations at Gua Jaya was brought in from other areas. Besides, it was also determined that the pottery was burned openly due to the uneven combustion temperature. The content of the pottery element also indicates that the pottery was used as food storage containers and also as appliances for cooking.

**Keywords:** Gua Jaya, Nenggiri Valley, Kelantan, pottery, prehistory, XRD, XRF

### INTRODUCTION

Earthenware pottery is one of the tools used by prehistoric society as a tool for use in everyday life. Earthenware pottery is believed to be first used and made by the Neolithic community. Therefore, it can be said that the remains of pottery fragments can be found at practically every Neolithic site in Malaysia, and also a few complete earthenware potteries. However, according to Rivka (1973), humans have begun to recognize the use of clay since the Mesolithic period, which is about 12,000 years ago when humans at that time were good at shaping animal and human body sculptures using clay which is not burned.

However, since 10,000 years ago, prehistoric communities in rural areas have practiced farming while the coastal communities began to have knowledge on how to catch fish and use forest products. The development of this prehistoric society has changed the function of clay (Gardner, 1978; Weinhold, 1983). Asyaari (2002) stated that the prehistoric society began to regard clay as one of the requirements of essential tools in their daily lives, which are food storage containers and essential tools for preparing food in everyday life. The importance of this was realized by prehistoric societies when they needed utensils to cook and store food that has been cooked. The use of pottery as a tool for daily use began when the prehistoric society had learned the nature of clay, that is it is easy to form when wet and, hard when dry after baking. This knowledge developed with the production of various types of earthenware that were used as tools for daily living with a variety of purposes

such as cooking, storing food, and drinking water (Asyaari, 2010; Moradi *et al.*, 2013; Sarhaddi-Dadian *et al.*, 2015).

Earthenware pottery found at archaeology sites should be determined whether it was made by the local community or brought in from outside. If the pottery was made by the local community, then the wisdom of the local communities in the past in pottery-making technology can be proven. If the pottery was brought in from another area, it can be proven that the local community at that time already had contact with the outside community. This can prove the local wisdom of local communities of the past. The origin of the pottery can be traced by analysing the source of raw materials, specifically clay that was used to make the pottery. Karina (1990) stated that pottery makers will only take clay from within seven kilometres of their settlements. Mohd Kamaruzzaman (1991) stated that the source of clay in Pulau Kalumpang was from areas between 7 and 14 kilometers away which takes into account the distance between Pulau Kalumpang with Kuala Sepetang and Kuala Gula. But in Pulau Kalumpang, a remote place to collect clay can be connected by boat.

To determine the source of clay used to make pottery, chemical analysis can be performed to obtain the chemical substance and morphological. These analysed substances are the most important to be used to determine the origin of the pottery (Bishop *et al.*, 1982; Mohd Anuar, 1991; Chia, 1997; Ertem & Demirci, 1999). This can be done by comparing the chemical substance of the pottery with the clay that is in the vicinity of the discovery area. If

the chemical substance of pottery is almost the same as the clay samples from nearby areas, it can be ascertained that the pottery was created around the area by the local community. If it is not the same, it can be concluded that the pottery was brought in from outside through the exchange of goods between the coastal communities and rural communities. Chemical testing to obtain the substance of minerals or elements contained in pottery has been done by many researchers in the past.

Among the earthenware pottery that had been studied by researchers in the past include pottery in Kota Melawati, Selangor (Zuliskandar *et al.*, 2011a), Pulau Kalumpang (Mohd Kamaruzzaman *et al.*, 1991), Gua Angin, Kota Gelanggi, Pahang (Zuliskandar *et al.*, 2001), Gua Peraling, Kelantan (Zuliskandar *et al.*, 2006, 2011b), Gua Bukit Chawas (Zuliskandar *et al.*, 2007, 2011), Gua Cha, Kelantan (Asyaari, 1998; Zuliskandar *et al.*, 2006), Bukit Menteri, Selangor (Asyaari, 1998), Kodiang, Kedah (Asyaari, 1998), and Gua Harimau, Gua Tukang, Gua Gelok and Gol Bait in Perak (Asyaari, 1998).

### GUA JAYA SITE

Nenggiri Valley area is predominantly consisting of rocks from Gua Musang Formation which was dated from Middle Permian to Middle Triassic. This formation was mapped and named by Yin (1965) who described the sequence as limestone, slate, phyllite, sandstone and minor conglomerate.

In general, most of the limestone at Nenggiri area was metamorphosed to marble. According to NEB Report (1986), the catchment of Nenggiri comprises of a combination of metamorphosed sedimentary and igneous intrusive rocks. Another study conducted by Jinap & Che Abdul Rahman at downstream of Sg. Wias discovered a sequence of slate, schist, and limestone as well as some igneous rocks, sandstone and marble (JMG, 2015a). Igneous intrusions are well exposed on the north and south of Nenggiri. Berangkat Granite batholith is exposed on the north side and is known as the oldest rock unit in Stong Migmatite Complex consisting of granitoid tonalite. There is an intrusion of Senting Granite on the south side of Nenggiri at Bukit Ulu Lalat which was dated Jurassic to Permian. It is comprised of coarse grained porphyritic granite biotite (JMG, 2015a). The limestone hills are exposed at several places in Nenggiri area such as Batu Berangin, Batu Baloh, Bukit Keldong, Gua Jaya, Gua Dala, Gua Cha and Gua Chawan (JMG, 2015b, Figure 1). Geological heritage site was also identified at Gua Cha which is important as a heritage of human civilization. It must be conserved and developed as one of the archaeology attractions. According to JMG (2015b), limestone at Nenggiri area is indicated by low to high hills with very steep faces and pinnacles. The limestone is white and dark greyish, fine to medium grained and its strength is hard / very hard. This limestone is characterised by sharp bedding from bottom to top of the limestone hill. Some limestone blocks can also be found along the Sungai Nenggiri.

Gua Jaya (N: 05°05'39.6"; E: 101°46'20.4") is a limestone cave located on the left bank of Sungai Nenggiri and located nearly 200 meters after the confluence of Sungai Jenera and Sungai Nenggiri (Zulkifli, 2003). This cave has a length of 36 meters and a width of 23 meters. The floor of this cave is in a humid condition due to the thick layer of bat droppings. This cave has relatively good lighting, spacious, and is higher than the water level of the river. Therefore, this feature would have allowed the prehistoric people to choose this cave as either a temporary or a permanent settlement.

In 1963, Lamb and Peacock who were British archaeologists conducted archaeological research and excavation at Gua Jaya. As a result of the excavation, they found 1500 pieces of pottery as thick as 30 cm which have a range of colours, shapes, and even decorations or motifs in the burning area, besides presence of soot stains on the ceiling of the cave (Peacock, 1964). Apart from the earthenware, the British researchers also found remains of food waste such as freshwater snail shells and animal bones. 30 years later, Adi Taha did another research at this cave in 1993. As a result of the research, he found two pieces of dark-coloured pottery fragments on the cave floor (Adi Haji Taha, 2007). He concluded based on the pottery discoveries that there was a settlement of prehistoric society at Gua Jaya, thereby supporting the opinion expressed by Peacock regarding the settlement of the Neolithic community at Gua Jaya.

### RESEARCH METHODOLOGY

This study was conducted by taking 13 samples namely TGJ1, TGJ2, TGJ3, TGJ4, TGJ5, TGJ6, TGJ7, TGJ8, TGJ9,



**Figure 1:** The location of prehistoric caves in Ulu Kelantan. Source: Adi Haji Taha (2007).

TGJ10, TGJ11, TGJ12 and TGJ13 from the findings of excavations carried out at Gua Jaya for scientific analysis, while the remaining pottery was only physically studied. The samples were cleaned and dried under the sun for several days, to ensure that the samples were completely dry. Two types of analysis were performed on the pottery fragmentations, that is physical and also chemical analysis. The physical analysis involves the color, decorative motifs, and the thickness of the pottery as discussed by Nur Sarahah *et al.* (2018) and Supian *et al.* (2018), while chemical analysis involves X-ray Fluorescence (XRF) test to obtain the content of the main elements and trace elements of the pottery. Besides the XRF test, X-ray Diffraction (XRD) testing was also carried out. Both XRF and XRD analysis are commonly used by archaeologists to determine the origin of ancient artifacts such as beads, votive tablet, metal artifacts and ancient bricks (Jusoh *et al.*, 2012; Ramli & Rahman, 2013). Both tests were performed at the Physical Characterization Laboratory, Research and Instrumentation Management Center (CRIM), UKM. The earthenware pottery samples from Gua Jaya were labeled as in Figure 2.



Figure 2: Pottery fragments selected for XRD and XRF analysis.

The samples were physically analyzed in advance before being crushed in preparation for the chemical analysis. In the preparation of the samples for chemical analysis, the samples have to be finely pounded to a size of 500  $\mu\text{m}$  using ceramic mortar to ensure homogeneity and to prevent the reaction of the samples with the mortar material. For the XRD test, the apparatus used is Bruker D8 Advance. The sample in powder form will be inserted in the sample container. The sample has to be securely placed in the container and flattened several times to ensure that it is in a homogeneous state. Upon completion, the given mineral concentration graph was analyzed using EVA software, to obtain the mineral content present in each pottery sample.

As for the XRF test, the method of press pallet was used because it is easier and faster than the fused glass method. The apparatus used for the test is type Bruker S8 TIGER. 5 g of sample was mixed with 10 g boric acid, which is used as a sample holder. The sample then was pressed up to 20 psi using a hydraulic pressurized pressure. Following that, the sample was inserted into the XRF apparatus until the analysis by the apparatus was completed. The graph of  $\text{Al}_2\text{O}_3$  against  $\text{SiO}_2$  was plotted to see the difference between the elements. The elements were also compared to the content of soil samples that were taken from several areas around Ulu Kelantan as reported by Zuliskandar (1999). Comparisons were made based on the mineral content as well as the content of the main elements found in the pottery sample with river clay samples from around Ulu Kelantan.

## RESULTS AND DISCUSSIONS

The study was conducted on 97 fragments of earthenware pottery that were found during excavation at Gua Jaya, Ulu Kelantan. The analysis involved 28 fragments of pottery of the mouth or lips parts, 32 fragments of body parts without decoration, 36 pieces of the body with ornaments, and one carination fragment or pottery fragment from a combination of several parts.

### Physical analysis

#### Color

The color analysis of pottery samples found during excavation at Gua Jaya found that the potteries are only of a few color groups, i.e. brown, blackish brown, black, and brownish-black (Nur Sarahah *et al.*, 2018). Nik Hassan Shuhaimi (1999) stated that the color of pottery depends on the pottery firing temperature as well as the content of minerals found in the raw material i.e. clay that was used to make the pottery.

#### Decorative motifs

Several decorative motifs often found on the earthenware pottery of prehistoric societies can be noted on the pottery discovered during the excavation. According to Nik Hassan Shuhaimi (1999), the decorative motifs on prehistoric earthenware pottery that can be seen include string stamps,



nets, mats, and baskets or geometric decorative motifs such as line dots, circles, and wavy lines. The diversity of the decorative motifs according to Mokhtar (2010) is due to the ability of the Neolithic community to control the temperature during the firing process at the minimum temperature of 600 °C.

Based on the analysis of decorative motifs, the pottery found in Gua Jaya was identified as having motifs such as parallel lines, string marks and zig-zag lines (Figure 3). Most of the decorative motifs on the pottery of Gua Jaya are parallel lines, as well as strap stamps, where the parallel incision method is also known as a bunch of patterns (Nur Sarahah *et al.*, 2018). These patterns are also present on potteries found at other caves in Ulu Kelantan, such as Gua Menteri (Noone, 1939), Gua Musang (Tweedie, 1940), Gua Chawan (Ahkemal Ismail *et al.*, 2018) and Gua Lubang Kelawar (Azhar *et al.*, 2018). Muhammad Afiq (2017) stated

that prehistoric societies used wood or animal sharp teeth to create parallel lines on the pottery.

### Thickness

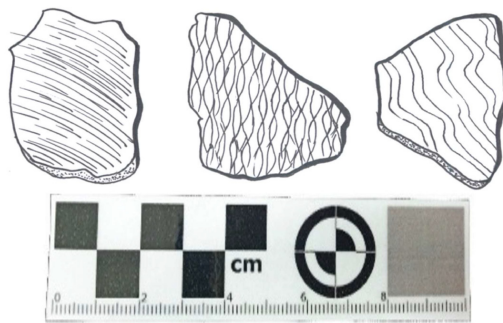
The thickness of pottery is measured to obtain the relevant information on the function of the pottery used by prehistoric societies. Chia (1997) stated that thick earthenware pottery is believed to be used as an appliance to store food or cook food or as water storage utensils, while thinner pottery was used as serving food utensils. This opinion is also been supported by Zuliskandar *et al.* (2001; 2011).

There are three sets of thickness that have been standardized by Chia (1997), that is thin (less than 6 mm), medium-thin (6 mm to 10 mm) and thick (over 10 mm). Based on the analysis of thickness of pottery done by Nur Sarahah (2018), 10.3% (10 pieces) are in the thin group, 76.3% (74 pieces) are in the medium-thin group and the remaining 13.4% (13 pieces) are in the thick group.

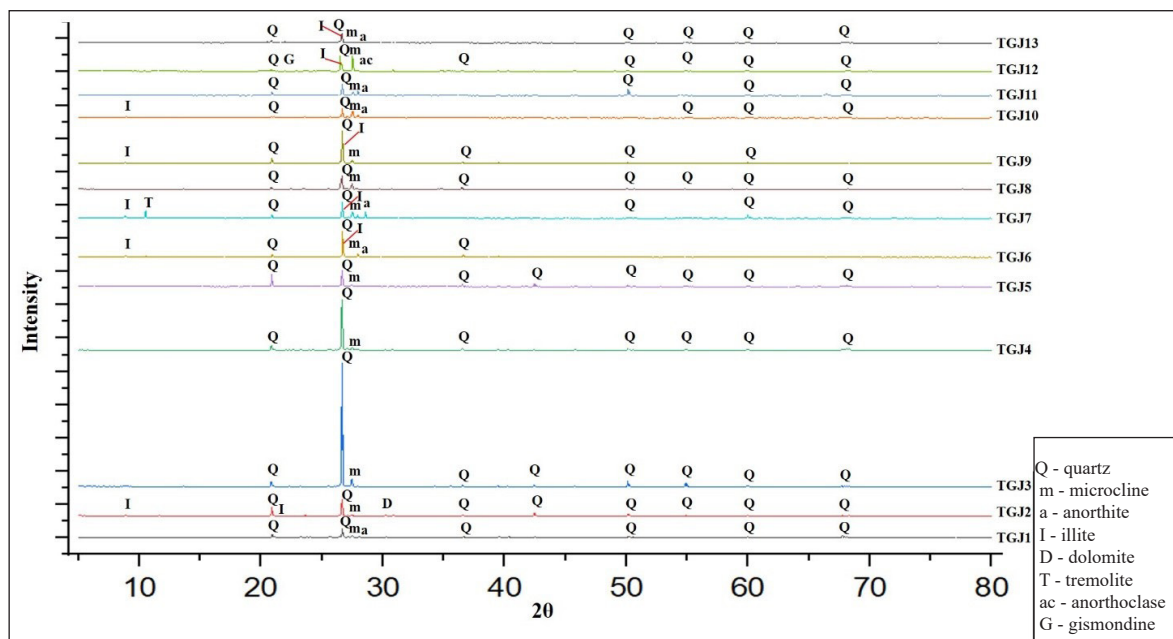
### Mineralogy

The results of the XRD test which is information on the mineral content of 13 earthenware pottery samples found during the excavation at Gua Jaya can be seen in Table 1. The mineral composition of pottery obtained from Gua Jaya includes minerals such as quartz, microcline, anorthite, dolomite, illite, lantinit, tremolite, anorthoclase, gismondin, and calcite. The XRD diffraction pattern of the pottery sample can be seen in Figure 4.

Based on the mineral content shown in Table 1, there are two groups of earthenware pottery, i.e. potteries that contain quartz and microcline only, and also potteries



**Figure 3:** Decorative motifs of parallel lines, strap marks and zig-zag lines. Source: Muhamad Shafiq *et al.* (2018).



**Figure 4:** XRD diffraction pattern of Gua Jaya earthenware pottery.

**Table 1:** Mineral content of earthenware pottery discovered at Gua Jaya.

Sample	Mineral content
TGJ1	Quartz, $\text{SiO}_2$ Microcline, $\text{KAlSi}_3\text{O}_8$ Anorthite, $\text{CaAl}_2\text{Si}_2\text{O}_8$
TGJ2	Quartz, $\text{SiO}_2$ Microcline, $\text{KAlSi}_3\text{O}_8$ Dolomite, $\text{CaMg}(\text{CO}_3)_2$ Illite, $\text{KAl}_2(\text{Si}_3\text{AlO}_{10})(\text{OH})_2$ Lanthinite
TGJ3	Quartz, $\text{SiO}_2$ Microcline, $\text{KAlSi}_3\text{O}_8$
TGJ4	Quartz, $\text{SiO}_2$ Microcline, $\text{KAlSi}_3\text{O}_8$
TGJ5	Quartz, $\text{SiO}_2$ Microcline, $\text{KAlSi}_3\text{O}_8$
TGJ6	Quartz, $\text{SiO}_2$ Microcline, $\text{KAlSi}_3\text{O}_8$ Anorthite, $\text{CaAl}_2\text{Si}_2\text{O}_8$ Illite, $\text{KAl}_2(\text{Si}_3\text{AlO}_{10})(\text{OH})_2$
TGJ17	Quartz, $\text{SiO}_2$ Microcline, $\text{KAlSi}_3\text{O}_8$ Anorthite, $\text{CaAl}_2\text{Si}_2\text{O}_8$ Illite, $\text{KAl}_2(\text{Si}_3\text{AlO}_{10})(\text{OH})_2$ Tremolite
TGJ8	Quartz, $\text{SiO}_2$ Microcline, $\text{KAlSi}_3\text{O}_8$
TGJ9	Quartz, $\text{SiO}_2$ Microcline, $\text{KAlSi}_3\text{O}_8$ Illite, $\text{KAl}_2(\text{Si}_3\text{AlO}_{10})(\text{OH})_2$
TGJ10	Quartz, $\text{SiO}_2$ Microcline, $\text{KAlSi}_3\text{O}_8$ Anorthite, $\text{CaAl}_2\text{Si}_2\text{O}_8$ Illite, $\text{KAl}_2(\text{Si}_3\text{AlO}_{10})(\text{OH})_2$
TGJ11	Quartz, $\text{SiO}_2$ Microcline, $\text{KAlSi}_3\text{O}_8$ Anorthite, $\text{CaAl}_2\text{Si}_2\text{O}_8$
TGJ12	Quartz, $\text{SiO}_2$ Microcline, $\text{KAlSi}_3\text{O}_8$ Anorthoclase, $((\text{Na,K})\text{AlSi}_3\text{O}_8)$ Illite, $\text{KAl}_2(\text{Si}_3\text{AlO}_{10})(\text{OH})_2$ Gismondine
TGJ13	Quartz, $\text{SiO}_2$ Microcline, $\text{KAlSi}_3\text{O}_8$ Anorthite, $\text{CaAl}_2\text{Si}_2\text{O}_8$ Illite, $\text{KAl}_2(\text{Si}_3\text{AlO}_{10})(\text{OH})_2$

that contain quartz, microcline, and other minerals as well. Samples TGJ3, TGJ4, TGJ5, and TGJ8 contain quartz and microcline only, while the other samples have additional minerals other than quartz and microcline. At this point, an assumption can be made that there were two different areas for the consumption of raw materials (clay) to produce pottery. This raw material i.e. clay can be found at Gua Jaya.

Besides that, the mineral content of anorthite found in TGJ1 and TGJ11 samples shows that this pottery sample had been baked at temperatures above 850 °C. Anorthite is a mineral formed when a carbonatic mixture is heated to a high temperature. Anorthite is a mineral formed into a new phase of calcium silicate when illite and calcite react at high temperatures above 850 °C after the formation of gehlenite at a combustion temperature of 800 °C (Cardiano *et al.*, 2004). The illite mineral content found in the TGJ2, TGJ9, and TGJ12 samples shows that the pottery was baked at temperatures below 850 °C.

However, there are some samples such as TGJ6, TGJ7, TGJ10, and TGJ13 which have both anorthite and illite. The presence of these two minerals proves that the pottery samples had underwent firing under threshold temperature conditions where it is possible that an exchange phase between illite and anorthite took place and, the insufficient temperature was not enough to convert all illite into anorthite. Based on this situation, it can be proven that this pottery sample was baked using the open burning method without being baked in a kiln. This is because the visible effects can be seen on the pottery baked at uneven temperatures. Mohd Kamaruzzaman (1991) stated that such a situation may occur because pottery makers used firewood as fuel placed in a wood oven.

Mohd Kamaruzzaman (1991) also noted that the prehistoric society at that time had its own manufacturing methods including the acquisition of raw materials. He mentioned that the community had been able to ensure that the pottery produced have uniform natural properties, at least in terms of mineral content. In addition to this, they also knew how to choose suitable clay for pottery making, at least in terms of their ability to choose the raw materials from the same category that can produce various types of pottery that have the same or almost the same minerals.

### Geochemistry

The results of the XRF analysis, which is information on the main elements of the 13 earthenware pottery samples found during excavation at Gua Jaya can be seen in Table 2. The dry weight of all trace elements is in the form of percentages. The dry weight content of the silica element is in the range of 51.35 to 69.57 %. Aluminium also has a dry weight ranging between 17.83 and 22.23 %. On the other hand, the dry weight content of iron is in the range of 2.33 to 13.19 %. The dry weight content of potassium

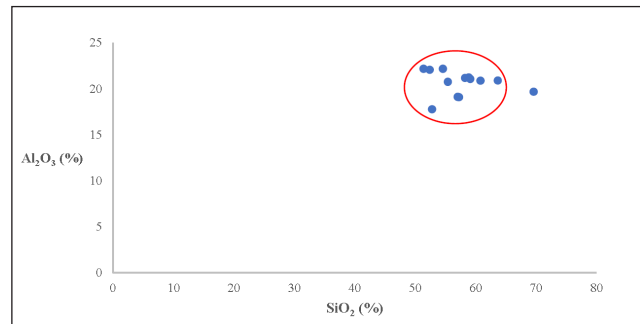
**Table 2:** Composition of key elements in earthenware pottery of Gua Jaya.

Sample	Main Elements (%)										
	MgO	Al <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	P <sub>2</sub> O <sub>5</sub>	SO <sub>3</sub>	K <sub>2</sub> O	CaO	TiO <sub>2</sub>	MnO	Fe <sub>2</sub> O <sub>3</sub>	SrO
TGJ1	1.21	21.24	58.87	1.18	0.21	4.10	3.28	1.46	0.09	8.01	0.01
TGJ2	0.69	19.09	57.28	7.49	1.82	3.48	2.31	1.31	0.07	6.51	0.01
TGJ3	0.62	21.08	59.12	8.41	0.21	2.69	2.78	0.51	0.03	3.98	0.02
TGJ4	0.59	19.69	69.61	0.19	0.20	4.18	1.48	0.72	0.09	2.28	0.01
TGJ5	1.49	20.91	63.68	0.32	0.10	3.60	2.29	0.15	0.14	6.58	0.01
TGJ6	1.59	22.18	54.59	0.32	0.18	2.22	2.67	1.89	0.11	13.23	0.01
TGJ7	1.74	22.07	52.41	4.23	0.52	2.64	3.28	1.36	0.03	11.08	0.02
TGJ8	0.91	21.19	58.24	4.69	0.10	5.01	2.13	1.10	0.03	6.14	0.01
TGJ9	0.59	20.76	55.41	6.24	0.14	5.73	1.64	1.45	0.03	6.78	0.02
TGJ10	1.27	22.19	51.39	6.79	0.72	1.68	2.62	2.08	0.03	10.15	0.01
TGJ11	2.61	17.78	52.79	7.57	0.29	2.18	3.34	0.94	0.05	11.68	0.02
TGJ12	0.68	19.12	57.05	8.38	2.52	3.39	0.97	1.24	0.02	6.30	0.01
TGJ13	1.23	20.89	60.82	0.38	0.10	6.27	2.48	1.01	0.01	5.80	0.03

and calcium is in the range of 1.72 to 6.27 % and 0.99 to 3.37 %, respectively. The percentage of the dry weight of sodium and magnesium is in the range of 0.17 to 1.07% and 0.64 to 2.64 %, respectively. While the dry weight content of titanium and phosphorus is in the range of 0.12 to 2.10 % and 8.46 to 12.23 %, respectively.

The high content of silica and aluminum in the samples indicates that pottery makers have learned to choose suitable raw material for making pottery (Supian *et al.*, 2018). Supian *et al.* (2018) also stated that high phosphorus content in samples TGJ2, TGJ3, TGJ7, TGJ8, TGJ9, TGJ10, TGJ11, and TGJ12 shows that the pottery had been used as cooking utensils while other samples were used as containers for storing food. She also added that the pottery used as cooking utensils is in line with the discovery of burning ash in the excavated compartment. Cooking utensils and even burning ashes remains of the prehistoric society that inhabited Gua Jaya show that this cave had been used as a convenient place to stay during the rainy and dry seasons because physically, this cave is located higher than the level of Sungai Nenggiri.

The graph of the dry weight percentage of aluminium elements against silicon is shown in Figure 5. From the graph, it is found that earthenware pottery has a high silica content. Based on the graph, it can also be seen that there is a sample of clusters and there are also scattered samples. The grouping of these samples shows that some pottery samples have used raw materials from the same area or nearby areas. Meanwhile, the scattered or non-clustered samples indicate that the raw materials used were taken from different areas. The variety of places for the intake of raw materials to make pottery at Gua Jaya is consistent with the findings from the XRD analysis regarding the same minerals found in some samples while other samples have different mineral contents.


**Figure 5:** Percentage of dry weight content of Al<sub>2</sub>O<sub>3</sub> against SiO<sub>2</sub> of earthenware pottery samples from Gua Jaya.

### Comparative analysis

A comparative analysis between the earthenware pottery samples with soil samples taken from several areas around Ulu Kelantan such as Sungai Nenggiri (SN), Sungai Betis (SB), Sungai Perias (SS), Sungai Chai (Sc), Sungai Jenera (Si) and Sungai Peralon by Zuliskandar (1999) was made. Comparisons were made based on the mineral content and the content of the main elements of the pottery samples with river clay samples from around Ulu Kelantan. The mineral content of clay samples can be seen in Table 3. Most of the clay samples contain only quartz and muscovite except sample SS which has quartz, muscovite, and orthoclase. Muscovite will decompose into orthoclase, corundum, and water at a temperature of 600 °C to 700 °C (Mason & Berry, 1968). When compared to the mineral content of the pottery samples, there is no similarity in the mineral content between these two samples. This proves that the source of clay for making pottery at Gua Jaya was not from around Ulu Kelantan.

Besides, the content of the main element of pottery samples was also compared to the content of the main

element of river clay samples. The content of the main elements of clay samples from around Ulu Kelantan can be seen in Table 4. Based on the percentage of element content already known, the comparison graph of Al element against Si was plotted between the clay of pottery samples from Gua Jaya and the river clay samples. This graph can be seen in Figure 6. Based on the comparison graph, it can be seen that although the content of the main elements of pottery samples and clay samples are quite close, nothing overlaps with each other. This means that the raw material of the

pottery samples was not taken near Ulu Kelantan. Therefore, we suggest that most of the pottery shards have different geochemical contents when compared to the geochemistry of clay samples from areas in the vicinity of Gua Jaya.

The distribution of elements and minerals that were obtained are different between pottery samples and clay samples. This shows that the clay used to make pottery at Gua Jaya was not taken from the areas around Ulu Kelantan. This proves that the pottery was brought from outside the area. We suggest that the potteries used by the Gua Jaya settlers were bought from the coastal communities which were more advanced in culture and technology. Exchange activities have taken place in the area around Ulu Kelantan where the coastal community will supply goods from the coast such as pottery or goods from foreign traders to the rural community who supply forest products such as resin, bamboo and so on (Zuliskandar *et al.*, 2007) to be traded by the coastal community.

The distribution of elements and minerals that are different between pottery from Gua Jaya and river clay samples from around Ulu Kelantan is also consistent with studies that have been done on earthenware pottery discovered at Gua Bukit Chawas (Zuliskandar *et al.*, 2007), Gua Cha and Gua Peraling (Zuliskandar *et al.*, 2006).

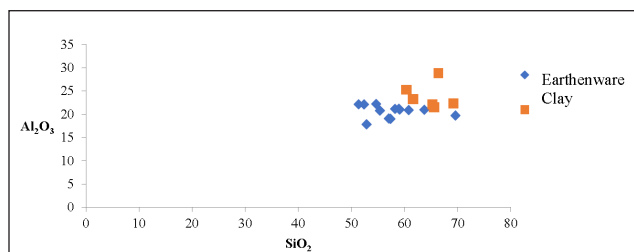
## CONCLUSION

The analysis of mineral content and the content of prehistoric pottery elements is very important to determine the origin of the pottery. It is to determine whether the potteries were made by the local community using raw materials from nearby or they were brought in from outside in the course of exchange of goods between the coastal and inland communities. Based on the analysis of mineral content and content elements, it can be ascertained that the pottery found at Gua Jaya was not made in the area around Gua Jaya. This proves that the pottery was brought in from outside through trade arrangements. Also, the pottery was fired by the method of open burning because the pottery was baked at an uneven temperature. Besides that, there is an amount of pottery that is believed to be food storage containers and also pottery used for cooking.

**Table 3:** Mineral content of river clay samples from around Ulu Kelantan.

Location	Sample	Mineral
Sungai Nenggiri	SN	Quartz Muscovite
Sungai Betis	SB	Quartz Muscovite
Sungai Perias	SS	Quartz Muscovite Orthoclase
Sungai Chai	SC	Quartz Muscovite
Sungai Jenera	Si	Quartz Muscovite
Sungai Peralon	SP	Quartz Muscovite

Source: Zuliskandar Ramli (1999)



**Figure 6:** The percentage of dry weight (%)  $\text{Al}_2\text{O}_3$  against  $\text{SiO}_2$  of the earthenware pottery samples from Gua Jaya and clay river samples from around Ulu Kelantan.

**Table 4:** Content of dry weight (%) of the main elements of river clay samples from around Ulu Kelantan.

Sample	Dry Weight (%)							
	Al	K	Ca	Fe	Mg	Ti	Na	Si
Sc	21.52	3.31	0.22	3.13	1.58	0.87	0.22	65.53
SS	25.29	3.16	0.32	4.13	0.91	0.89	0.27	6.35
SP	28.87	3.42	0.09	4.35	1.56	1.01	0.34	66.35
Si	22.13	2.45	1.19	3.96	1.31	0.98	0.31	65.24
SB	23.27	2.33	0.18	1.99	0.89	0.81	0.24	61.59
SN	22.35	3.52	0.37	3.41	1.29	0.95	0.24	69.20

Source: Zuliskandar (1999)



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## AUTHOR CONTRIBUTIONS

The main content of the paper is based on the research and analysis by MSMA and ZR. Archaeological data input was contributed by ZR and NSMS.

## CONFLICT OF INTEREST

The authors have no conflicts of interest to declare that are relevant to the content of this article.

## REFERENCES

- Adi Haji Taha, 2007. Archaeology of Ulu Kelantan. Jabatan Muzium Malaysia, Kuala Lumpur. 278 p.
- Ahkemal Ismail, N.N., Ramli, Z., Mohd Supian, N.S., Hussin, A. & Duli, A., 2018. Hoabinhian's lithic technology in Chawan cave, Hulu Kelantan. *International Journal of Mechanical Engineering and Technology*, 9(13), 1007-1015.
- Asyaari, M., 1998. Analisis X-Ray Fluorescence tembikar tanah di Perak. *Jurnal Arkeologi Malaysia*, 11, 1 – 40.
- Asyaari, M., 2002. Perkembangan tembikar prasejarah di Malaysia. *Jurnal Arkeologi Malaysia*, 15, 29-37.
- Asyaari, M., 2010. Seramik purba yang diperdagangkan di Semenanjung Malaysia. *SARI-International Journal of the Malay World*, 28, 3 – 40.
- Azhar, W.N.S.W., Ramli, Z., Hussin, A. & Arbi, R., 2018. Lithic technology and material culture of Hoabinhian and Neolithic settlement in the Lubang Kelawar Batu Tambah Cave. *International Journal of Mechanical Engineering and Technology*, 9(13), 1244-1252.
- Bishop, R.L., Rands, R.L. & Holley, G.R., 1982. Ceramic compositional analysis in archaeological perspective. *Advance in Archaeological Method and Theory*, 5, 275 – 327.
- Cardiano, P., Ioppolo S., Stefano C.D., Pettignano A., Sergi S., & Piraino, P., 2004. Study and characterization of the ancient bricks of monastery of "San Filippo di Fragala" in Frazzano (Sicily). *Analytica Chimica Acta*, 519, 103-111.
- Chia, S., 1997. Prehistoric pottery sources and technology in Peninsular Malaysia, based on compositional and morphological studies. *Malaysia Museum Journal*, 33, 91 – 125.
- Ertem, E. & Demirci, S., 1999. Characteristics of Hittite pottery sherds from sites in the Kilirmak Basin. *Journal of Archaeological Science*, 26, 1017 – 1023.
- Gardner, E.J., 1978. The Pottery Technology of the Neolithic Period in Southeastern Europe. Tesis PhD, University of California, USA.
- Jabatan Mineral dan Geosains Malaysia, 2015a. Kajian Potensi Sumber Mineral Kawasan Cadangan Projek Hidroelektrik Nenggiri, Kelantan. Report No JMG.KLT (MBL) 05/2015.
- Jabatan Mineral dan Geosains Malaysia, 2015b. Kajian Potensi Sumber Batu Kapur Tapak Cadangan Projek Hidroelektrik Nenggiri, Kelantan. Report No JMG.KLT (MPI) 02/2015.
- Jusoh, A., Sauman, Y., Abdul Rahman, N.H.S.N. & Ramli, Z., 2012. Scientific analysis of samples of some artefacts metal age in Malaysia. *Social Sciences (Pakistan)*, 7(6), 772-777.
- Karina, A., 1990. Social aspects of pottery manufacture in Boera, Papua New Guinea. Kertas kerja dibentangkan di 14<sup>th</sup> Congress of Indo Pacific Prehistoric Association, Yogyakarta.
- Mason, B. & Berry, L.G., 1968. *Elements of Mineralogy*. W.H. Freeman & Company, San Francisco & London. 570 p.
- Mohd Anuar, F., 1991. Penggunaan teknik pembelauan Pancaran-X: Mengenal pasti kandungan mineral dalam kajian tembikar purba. Tesis Jabatan Geologi, Fakulti Sains, UKM.
- Mohd Kamaruzzaman, A. R., Mohamad Deraman, Ramli Jaya & Mohd Ali Sufi, 1991. Kajian sains terhadap jumpaan tembikar tanah di Pulau Kalumpang, Perak: Keputusan Awal. *Jurnal Arkeologi Malaysia*, 4, 59 – 73.
- Mokhtar, S., 2010. *Bukti Prasejarah di Malaysia*. Dewan Bahasa & Pustaka, Kuala Lumpur. 109 p.
- Moradi, H., Hossein Sarhaddi Dadian, Ramli, Z. & Rahman, N.H.S.N.A., 2013. Compositional analysis of the pottery shards of Shahr-I Sokhta, South Eastern Iran. *Research Journal of Applied Sciences, Engineering and Technology*, 6(4), 654-659.
- Muhamad Shafiq, M. A., Zuliskandar, R. & Nur Sarahah, M. S., 2018. Analisis kandungan kimia tembikar prasejarah: Jumpaan di Gua Jaya. Paper presented at Seminar Kelangsungan Pemikiran, Jati Diri & Budaya Melayu 2018, organized by Institute of Malay World & Civilization, UKM on 18 December.
- Muhammad Afiq, O., 2017. Kajian Arkeologi di Tok Sik, Gunung Pulai dan Gua Baling, Bukit Baling, Baling, Kedah. Tesis Sarjana, Institut Alam & Tamadun Melayu, Universiti Kebangsaan Malaysia.
- Nik Hassan Shuhaimi, N. A. R., 1999. Tembikar tanah daripada Kuala Selinsing, Pulau Kalumpang, Perak. *Jurnal Arkeologi Malaysia*, 12, 24 – 59.
- Noone, H.D., 1939. Report on a Neolithic Site in Ulu Kelantan. *Journal of the Federated Malay States Museum*, 15, 170 – 174.
- Nur Sarahah, M. S., Zuliskandar, R. & Azimah, H., 2018. Kebudayaan Hoabinhian dan Neolitik di Gua Jaya, Hulu Kelantan. *Jurnal Arkeologi Malaysia*, 31(1), 49 – 62.
- Peacock, B.A.V., 1964. Recent archaeological discoveries 1962 – 1963 Malaya. *Journal of the Royal Asiatic Society Malaysian Branch*, 37(2), 201 – 206.
- Ramli, Z. & Rahman, N.H.S.N.A., 2013. Composition analysis of ancient Bricks, Candi Bukit Kechil, Bujang Valley, Kedah. *Research Journal of Applied Sciences, Engineering and Technology*, 6(5), 924-930.
- Ramli, Z., Abdul Rahman, N.H.S.N., Samian, A.L., Razman, M.R., Zakaria, S.Z.S., Sauman, Y. & Sarhaddi-Dadian, H., 2014. X-Ray Diffraction (XRD) and X-Ray Fluorescence (XRF) analysis of proto-historic votive tablets from Chawas cave, Hulu Kelantan, Malaysia. *Research Journal of Applied Sciences, Engineering and Technology*, 7(7), 1195-1201.
- Ramli, Z., Abdul Rahman, N.Q., Hussin, A., Hasan, S.N.I.S. & Dali, A.M., 2017. Compositional analysis of sungai mas, kuala selinsing and santubong glass beads. *Mediterranean Archaeology and Archaeometry*, 7(2), 117-129.
- Richardson, J.A., 1939. *The Geology and Mineral Resources of the*



- Neighbourhood of Raub, Pahang, Federated Malay States. Geological Survey Department Federation of Malaya, Ipoh. 25 p.
- Richardson, J.A., 1946. Facies Changes and Lithological Variation in the Permo-Carboniferous Formation of Northwest Pahang and Southeast Kelantan, Malaya. Geological Magazine, 84, 281-288.
- Richardson, J.A., 1950. The Geology and Mineral Resources of the Neighbourhood of Chegar Perah and Merapoh, Pahang, Federated Malay States. Geological Survey Department Federation of Malaya, Ipoh. 162 p.
- Rivka, G., 1973. Ancient Pottery. Cassell & Co. Ltd., London. 100 p.
- Sarhaddi-Dadian, H., Ramli, Z., Abdul Rahman, N.H.S.N. & Mehrafarin, R., 2015. X-ray diffraction and X-ray fluorescence analysis of pottery shards from new archaeological survey in south region of Sistan, Iran. Mediterranean Archaeology and Archaeometry, 15(3), 45-56.
- Scrivenor, J.B., 1911. The Geology and Mining Industries of Ulu Pahang. Government Printing Office, Kuala Lumpur. 61 p.
- Supian, N.S.M., Ramli, Z., Ismail, N.N.A., Hussin, A. & Duli, A., 2018. The study of earthenware at Jaya cave prehistoric site, Hulu Kelantan. International Journal of Mechanical Engineering and Technology, 9(13), 998-1006.
- The National Electricity Board of Malaysia (NEB), 1986. Nenggiri Dam Project: Feasibility Study.
- Tweedie, M.W.F., 1940. Report on Excavation in Kelantan (Plates I – XVI). Journal of the Malayan Branch of the Royal Asiatic Society, 18(2), 1 – 22.
- Weinhold, R., 1983. The Many Faces of Clay. Druckerei Fortschritt Erfurt, Germany. 238 p.
- Yin, E.H., 1965. Provisional Draft Report on the Geology and Mineral Resources of the Gua Musang Area, Sheet 45, South Kelantan. Geological Survey of Malaysia. 49 p (unpublished).
- Zulkifli, J., 2003. Gua-gua Batu Kapur di Semenanjung Malaysia dalam Perspektif Arkeologi. Jabatan Muzium Malaysia, Kuala Lumpur. 232 p.
- Zuliskandar, R., 1999. Chemical studies of artefacts and soils from archaeological sites. Bachelor Science Thesis, UPM.
- Zuliskandar, R., Zobir, H., Asmah, Y. & Zulkifli, J., 2001. Chemical analysis of prehistory pottery sherds found at Gua Angin, Kota Gelanggi Complex, Jerantut, Pahang, Malaysia. Jurnal Arkeologi Malaysia, 14, 1 – 12.
- Zuliskandar, R., Mohd Zobir, H., Asmah, Y. & Kamaruddin, Z., 2006. Preliminary analysis of prehistoric pottery sherds excavated at Gua Peraling and Gua Cha, Ulu Kelantan, Malaysia. Jurnal Arkeologi Malaysia, 19, 27 – 36.
- Zuliskandar, R., Mohd Zobir Hussein, Asmah, Y., Kamaruddin, Z. & Mahfuz, N., 2007. Kajian komposisi kimia kalam semah dan tembikar tanah yang ditemui di Gua Bukit Chawas. Jurnal Arkeologi Malaysia, 20, 22 – 65.
- Zuliskandar, R., Yunus, S. & Nik Hassan Shuhaimi, N.A.R., 2011a. Analisis komposisi tembikar tanah, mortar dan bata yang terdapat di Kota Melawati, Selangor. Jurnal Arkeologi Malaysia, 24, 93 – 131.
- Zuliskandar, R., Nik Hassan Shuhaimi, N. A. R., Adnan, J. & Yunus, S., 2011b. X-Ray Diffraction and X-Ray Fluorescent Analyses of Prehistoric Pottery Shards from Ulu Kelantan. American Journal of Applied Sciences, 8(12), 1337-1342.

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