Bulletin of the Geological Society of Malaysia, Volume 75, May 2023, pp. 85 - 99
DOI: https://doi.org/10.7186/bgsm75202308

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

Muhamad Shafiq Mohd Ali*, Zuliskandar Ramli, Nur Farriehah Azizan

Laboratory of Archaeology and Archaeometry, Institute of the Malay World and Civilization,
Universiti Kebangsaan Malaysia, 43600 UKM Bangi, Selangor
* Corresponding author email address: muhdshafiq@ukm.edu.my

Abstract: Gua Kelew is one of the newly discovered caves by archaeological researchers in Malaysia. This cave has the potential to become one of Malaysia's prehistoric sites from the findings from site surveys and archaeological excavations. Among the artifacts that were found are stone tools, earthenware, ceramics, snail shells, cave paintings and animal bones. The discovered earthenwares were analyzed for their mineral content in order to determine whether they were made locally around the cave or brought from elsewhere. The outcome of this analysis is vital as the data would provide proof that the community living in the cave or the surrounding area had its own technology for manufacturing the earthenwares. However, if the earthenwares were brought from elsewhere, it is believed that trade or the exchange of goods between communities living inland and near the coast had occurred. Composition of the earthenware samples was obtained by X-Ray Diffraction (XRD) analysis for mineral content and also X-Ray Flouresence (XRF) analysis for major, minor and trace elements. Findings from the XRD analysis show that the mineral content in the earthenware samples are quartz, calcite and anorthoclase, while the XRF analysis shows a high content of silica and aluminium. The analysis also discovered that all of the earthenwares found in Gua Kelew used the same raw materials obtained from the same area. Based on the graph plot analysis, the data exhibits differences in the elements between the earthenware samples and the surrounding area's clay samples. Hence, it is suggested that the earthenwares found in Gua Kelew were not produced in the Hulu Kelantan area. This also indicates that the earthenwares may have been brought from another location to the area through trade deal between the local and foreign communities.

Keywords: Gua Kelew, Nenggiri Valley, Kelantan, earthenware, prehistoric, XRD, XRF

INTRODUCTION

The cave is one of the favoured places for shelter amongst prehistoric people. There is also evidence that caves were still in use until the historical era, particularly by indigenous people. However, the discussion of whether it was occupied for an extended period or seasonal is still debatable. The word 'Gua' was originally from the Sanskrit word 'Guha', while 'Cave' comes from the Latin word 'Cavus', which means hole. Most of the caves found in Malaysia are on limestone hills, in which most of the prehistoric settlements in Malaysia are found.

Since 1939, there have been several studies conducted by foreign and local researchers regarding the limestone caves habited by prehistoric communities. Among the sites that were subjected to research by scholars are Gua Cha (Noone, 1939; Sieveking, 1954, 1954-1955, 1987; Hooijer, 1962; Adi Taha, 1985; Endicott & Bellwood, 1991; Bullbeck, 2005), Gua Musang (Peacock, 1959), Gua Chawas (Adi Taha, 1998 & 2007), Gua Peraling (William-Hunt, 1951; Adi Taha, 1998 & 2007), Gua Tampaq (Peacock & Dunn, 1968), Gua Madu (Tweedie, 1940), Bukit Pulai (Rentse, 1947), Gua Batu Cincin (Zuliskandar, 2019), Gua Chawan (Peacock, 1964; Ahkemal Ismail *et al*., 2018), Gua Jaya (Peacock, 1964: Supian *et al*., 2018), Gua Kecil Batu Tambah (Azhar *et al*., 2018), Gua Gemalah (Muhamad Fazrullah *et al*., 2019) and also a survey of the prehistoric sites and limestones caves around the Nenggiri River by Zuliskandar (2019). Research on limestone or protected caves in Hulu Kelantan is no longer unfamiliar to archaeologists. After several continuous discoveries, the limestone sites inhabited by prehistoric communities has made Hulu Kelantan unique in terms of its history.

Among the new sites that have never been mentioned or surveyed before by researchers is Gua Kelew. Gua Kelew is one of the caves near the Neggiri River in Kelantan. The coordinate of Gua Kelew is 105.03976° N and 101.54564° E. This cave is 62 meters above sea level. It is also situated near Kampung Kledong (Nur Farriehah *et al*., 2019), where the path shares the same route to an aboriginal village, Pos Pulat. The distance between Gua Kelew and the Neggiri River is only 700 meters in a straight line (Figure 2).

0126-6187; 2637-109X / Published by the Geological Society of Malaysia.

Based on the preliminary research and survey conducted by Nur Farriehah *et al.* (2019), they assume that Gua Kelew may have been a settlement or stopover for prehistoric society and lasted until the historical era of society, particularly the indigenous people. This opinion is based on several exciting discoveries in Gua Kelew. Among the findings discovered from surveying the surface around the cave is some artefacts removed from the ground. These include stone tools, earthenware and ceramics. Other than artefacts, cave paintings are plastered on several cave wall panels (Zuliskandar *et al*., 2020).

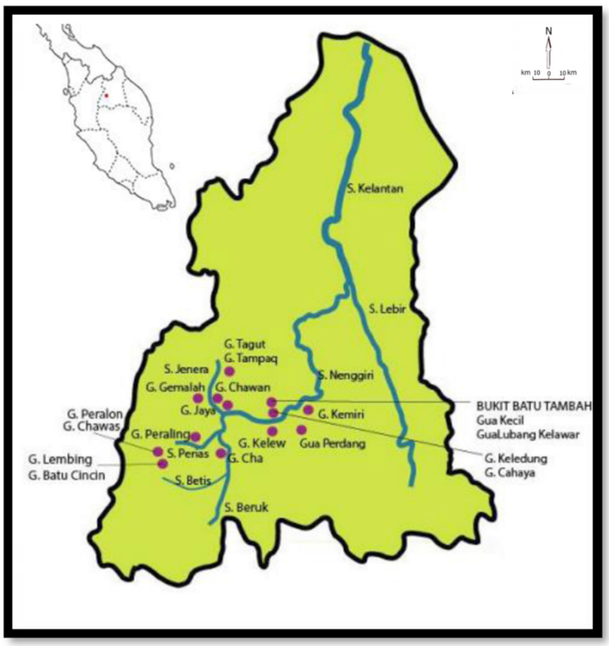

Figure 1: Map showing the location of Gua Kelew. Source: Modified from Zuliskandar (2019).

These findings indirectly prove that prehistoric societies once inhabited the area until historical era societies, especially the indigenous people.

The discovered stone tools consist of hammers, axes and even smoothing stones. These items are tools used by prehistoric societies for various functions ranging from cutting, chopping and hunting (Wan Noor Shamimi *et al.,* 2018). Axes were produced in different shapes and sizes with tapered sides to suit their function and use. In addition, several ceramics were also found on the surface. This includes blue and white bowl fragments from the Ching Dynasty of the 19th century, blue and white ceramic fragments from the Ming Dynasty of 16th century that has a Sanskrit inscription that reads 'sacred symbol'; and a red and white ceramic fragment dated in the 19th century A.D. from the Netherlands.

Besides stone tools and ceramics, several cave drawings can also be seen on the cave walls. The cave drawings inside Gua Kelew are pictographs that used monochromatic black as charcoal was the medium used. The several forms identified by Nur Farriehah *et al*. (2019) and Zuliskandar *et al*. (2020) are of anthropomorphic (human), zoomorphic (animal), geometric and even abstract unidentifiable shapes. The discovery of the cave drawings shows that the people who inhabited Gua Kelew had left marks to indicate their presence. Cave drawings that were drawn using black charcoal as medium are usually made by the indigenous people, as many researchers believe that the prehistoric society preferred using hematite material that has a darker red colour to draw on the cave walls; where hematite is an older drawing medium compared to charcoal (Zuliskandar, 2020). Zuliskandar (2020) also believes that the drawings found in Gua Kelew were drawn by the Temiar indigenous people from the Senoi ethnic group from the nearby areas surrounding the cave.

Figure 2: Map showing the location of Gua Kelew and the Neggiri River. Source: Google Earth (2022).

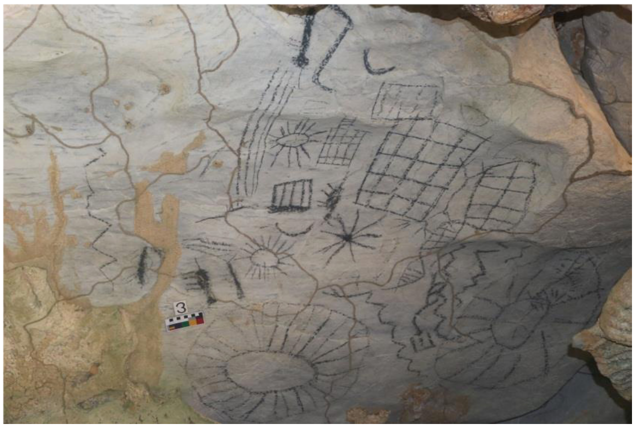

Figure 3: Cave drawings found in Gua Kelew.

EARTHENWARE

Earthenware is among the findings that were obtained during the surface survey. The earthenware found is entirely in the form of fragments as not many complete earthenware was found in the preliminary research and studies conducted by the archaeological teams in Kelantan, Kedah and Pahang. Earthenware is one of the tools used by the prehistoric society in their daily life. In addition, earthenware is also believed to have been first used and made by the Neolithic community. Therefore, it can be said that the remains of earthenware fragments and several complete ones can be found at every Neolithic site in Malaysia. However, according to Rivka (1973), humans had began to recognize the use of clay in the Mesolithic period, about 12,000 years ago, when humans at that time were good at shaping animal sculptures and human bodies using unburned clay.

Asyaari (2002) stated that prehistoric society had begun to treat clay as one of the essential tools in their daily lives as it could be used as food storage containers and tools or utensils to prepare food. Its importance was recognized by prehistoric society when they needed utensils for cooking and storing cooked food. The use of pottery as a daily tool began when prehistoric culture understood the nature of clay which is easy to form when wet and hardens when dry after burning. This knowledge was then further developed with the production of various types of earthenware used as tools in daily life for different purposes such as cooking, storing food and drinking water (Asyaari, 2010).

Several decorative shapes or motifs can be physically seen on the earthenware fragments. Among the patterns or decorative motifs identified on the fragments is the parallel line motif, and a parallel notching motif better known as the comb pattern. The parallel lines or comb patterns are typically created using the incising technique with tools such as sharp wood, bone or animal tooth (Muhammad Afiq, 2017). In addition, a small number of decorative motifs in the form of cord marks were also found, while some other earthenware fragments do not have any pattern or motif. There are also some fragments of the earthenware with parts that can be identified, while some are difficult to identify due to the fragments being too small.

The earthenware found at archaeological sites must be examined to determine whether they were locally made or brought in from elsewhere. If it was made by the local community, the earthenware is then a proof of the local community's wisdom in pottery-making technology in the past. However, if proven otherwise, this indicates that the local community at that time had a relationship with other communities outside of their circle. This can attest to the local wisdom of prehistoric societies of the past. The origin of the earthernware can be known by analyzing the source of the raw material i.e., clay, that was used to make the pottery. Karina (1990) states that a potter will only take clay within a seven kilometre radius from his location. Mohd Kamaruzzaman (1991) noted that the clay in Pulau Kalumpang was taken between 7 and 14 kilometres radius, which considers the distance between Pulau Kalumpang with Kuala Sepetang and Kuala Gula. However, boats were used in Pulau Kalumpang to take clay from a remote area.

In order to identify the source of clay used to make the earthenwares, a chemical analysis can be done to obtain the most critical chemical and morphological content (Mohd Anuar, 1991; Chia, 1997; Ertem & Demirci, 1999; Bishop *et al*., 1982; Moradi *et al*., 2013; Sarhaddi-Dadian *et al*., 2015; 2017; 2021; Ramli, 2011; 2012; 2013a; 2013b; 2014a; 2014b; 2017; 2018; Ali *et al*., 2015). This can be done by comparing the chemical content of the earthenware with the clay around the discovery area. If the chemical content is similar, it can be postulated that the earthenware were made around the area. However, if proven otherwise, it can be ascertained that the earthenware were brought from outside the area, due to an exchange of goods between the coastal and inland communities. Past researchers favoured chemical analysis to determine the mineral or element content of earthenwares.

Among the earthenwares that had been studied are the ones found in Kota Melawati, Selangor (Zuliskandar *et al*., 2011), Pulau Kalumpang (Mohd Kamaruzzaman *et al*., 1991), Gua Angin, Kota Gelanggi, Pahang (Zuliskandar *et al*., 2001), Gua Peraling, Kelantan (Zuliskandar *et al*., 2006), Gua Bukit Chawas (Zuliskandar *et al*., 2007), Gua Cha, Kelantan (Asyaari, 1998; Zuliskandar *et al*., 2006), Bukit Menteri, Selangor (Asyaari, 1998), Kodiang, Kedah (Asyaari, 1998), Gua Harimau, Gua Tukang, Gua Gelok and Gol Bait in Perak (Asyaari, 1998), and also Gua Jaya (Muhamad Shafiq *et al*., 2021). Past researchers on prehistoric earthenwares had done numerous tests, while this study would provide additional information on the chemical content analysis of prehistoric clay earthenware found during excavations and surveys conducted in Gua Kelew, Ulu Kelantan.

RESEARCH METHODS

This study was conducted by taking 15 samples of earthenware found during the survey and archaeological excavations conducted at Gua Kelew. Each sample was named T.G.K. in conjunction with the Gua Kelew pottery, namely TGK1, TGK2, TGK3, TGK4, TGK5, TGK6, TGK7, TGK8, TGK9, TGK10, TGK11, TGK12, TGK13, TGK14 and TGK15, as can be seen in Table 1. Other earthenware finds were only analyzed physically because their size is too small to be taken as a sample. Before a sample is crushed, the physical characteristic i.e., its colour, was recorded. The colour information on the outside, middle and inside of the samples can be seen in Table 1 below. Once crushed, the colour of the sample was again retrieved, as seen in Table 2.

The earthenware samples were washed with clean water and dried under the sun for several days to ensure the samples were completely dehydrated. Two analyses

Table 1: The name and colour of the Gua Kelew earthenware shard samples.

Sample name	Sample colour
 TGK1	Outside: HUE 10YR 8/1 White Middle: HUE 10YR 8/3 Very Pale Brown Inside: HUE 10YR 8/3 Very Pale Brown
 TGK2	Outside: HUE 10YR 3/3 Dark Brown Middle: HUE 10YR 4/3 Dark Brown Inside: HUE 10YR 3/1 Very Dark Gray
 TGK3	Outside: HUE 10YR 3/2 Very Dark Grayish Brown Middle: HUE 10YR 5/2 Grayish Brown Inside: HUE 10YR 3/1 Very Dark Gray

Sample name	Sample colour
 TGK4	Outside: HUE 10YR 8/3 Very Pale Brown Middle: HUE 10YR 8/4 Very Pale Brown Inside: HUE 10YR 6/4 Light Yellowish Brown
 TGK5	Outside: HUE 10YR 3/1 Very Dark Gray Middle: HUE 10YR 3/1 Very Dark Gray Inside: HUE 10YR 3/3 Dark Brown
 TGK6	Outside: HUE 10YR 4/1 Dark Gray Middle: HUE 10YR 5/1 Gray Inside: HUE 10YR 3/2 Very Dark Gray
 TGK7	Outside: HUE 10YR 3/2 Very Dark Grayish Brown Middle: HUE 10YR 4/3 Dark Brown Inside: HUE 10YR 3/2 Very Dark Grayish Brown

Sample name	**Sample colour**
 TGK8	Outside: HUE 10YR 3/2 Dark Brown Middle: HUE 10YR 5/1 Gray Inside: HUE 10YR 3/2 Very Dark Brown
 TGK9	Outside: HUE 10YR 4/1 Dark Gray Middle: HUE 10YR 4/4 Dark Yellowish Brown Inside: HUE 10YR 2/2 Very Dark Brown
 TGK10	Outside: HUE 10YR 4/4 Dark Yellowish Brown Middle: HUE 10YR 3/1 Very Dark Gray Inside: HUE 10YR 4/3 Dark Brown
 TGK11	Outside: HUE 10YR 4/1 Dark Gray Middle: HUE 10YR 4/1 Dark Gray Inside: HUE 10YR 3/1 Very Dark Gray

Sample name	Sample colour
 TGK12	Outside: HUE 10YR 3/1 Very Dark Gray Middle: HUE 10YR 4/3 Brown Inside: HUE 10YR 3/1 Very Dark Gray
 TGK13	Outside: HUE 10YR 6/6 Brownish Yellow Middle: HUE 10YR 6/4 Light Yelowish Brown Inside: HUE 10YR 6/4 Light Yellowish Brown
 TGK14	Outside: HUE 10YR 4/3 Dark Brown Middle: HUE 10YR Inside: HUE 10YR 4/4 Dark Yellowish Brown
 TGK15	Outside: HUE 10YR 3/3 Dark Brown Middle: HUE 10YR 4/1 Dark Gray Inside: HUE 10YR 3/2 Very Dark Grayish Brown

Table 2: Description of the physical color of the samples after crushing.

Sample	Sample description
TGK1	Dark grey coarse powder
TGK2	Light grey coarse powder
TGK3	Dark grey coarse powder
TGK4	Brown coarse powder
TGK5	Dark grey coarse powder
TGK6	Dark grey coarse powder
TGK7	Dark brown coarse powder
TGK8	Brown coarse powder
TGK9	Dark grey coarse powder
TGK10	Brown coarse powder
TGK11	Light grey coarse powder
TGK12	Dark grey coarse powder
TGK13	Brown coarse powder
TGK14	Dark grey coarse powder
TGK15	Dark brown coarse powder

were conducted on the Gua Kelew earthenware samples, physical exmination and chemical content analysis. A physical study of the earthenware samples was conducted to obtain information regarding the colour, decorative motifs, thickness and even the size of the pottery. This physical analysis was performed on all earthenware found during the survey and excavation activities.

On the other hand, the chemical content analysis only involved 15 samples. Each sample represents the earthenware findings in this study. These samples were crushed for chemical content analysis. Each sample was pounded until fine using a ceramic mortar, until it passed a 500 µm filter. This was to ensure that each powder sample is homogeneous and that the data obtained are accurate and precise.

Two chemical content analysis were performed on the samples; the X-Ray Diffraction and X-Ray Fluorescent analysis. The XRD analysis was conducted to obtain the mineral content of each sample, while the XRF analysis was to determine the major and trace elements in each sample.

For the XRD analysis, the apparatus used was a Bruker D8 Advance, available at the Physical Characterization Laboratory, U.K.M. Research Management and Instrumentation Center. The samples in the form of finely pounded powder were placed in a sample holder and flattened several times to ensure that the X-rays applied to the sample are uniform before being inserted into the XRD device. Each sample was subjected to X-ray exposure for 15 minutes. Upon completion, the spectral graph was analysed using the E.V.A. software to obtain the mineral content for each peak visible on the spectral graph for each sample.

As for the XRF analysis, there are two methods for preparing the samples. The samples were powdered and heated up for one hour at a temperature of 105°C. The first method is where the sample was made into a fused glass to obtain major elements, while the second method is to prepare the samples in the form of a pressured pallet to obtain minor elements and trace elements. For the fused glass samples, 0.5 g of the sample was mixed with 5.0 g of spectroflux and baked at 1100 °C for 20 minutes before being placed in a glass mould measuring 32 mm in diameter. These samples were then further analysed to obtain ten main elements using the Axios Max (Holland) XRF PanAlytical spectrometer with standard element preparation. Calibrations was carried out to ensure that the apparatus and methods provide accurate and precise readings. Ten element curve graphs were constructed using 30 high quality international standard reference materials.

The samples were prepared as pressed powder pallets to obtain the minor and trace elements. The pallets measured 32 mm in diameter in the shape of a round disc, using 1 g of sample and 6 g of boric acid as a binder. The sample was placed in the middle with boric acid around the sample as a binder. The samples were pressed with a 15-ton hydraulic press machine for 2 minutes. The XRF analysis was performed by scanning the presence of elements peaks using the Omnian software.

The earthenware samples analysed with the XRD and XRF apparatus then were compared with the clay samples found around the Sungai Nenggiri basin (Zuliskandar, 1999). The purpose of the comparison is to identify the origin of the earthenware; whether the earthenware samples were made using raw materials i.e., clay located in the surrounding area of Gua Kelew or brought from outside of the site.

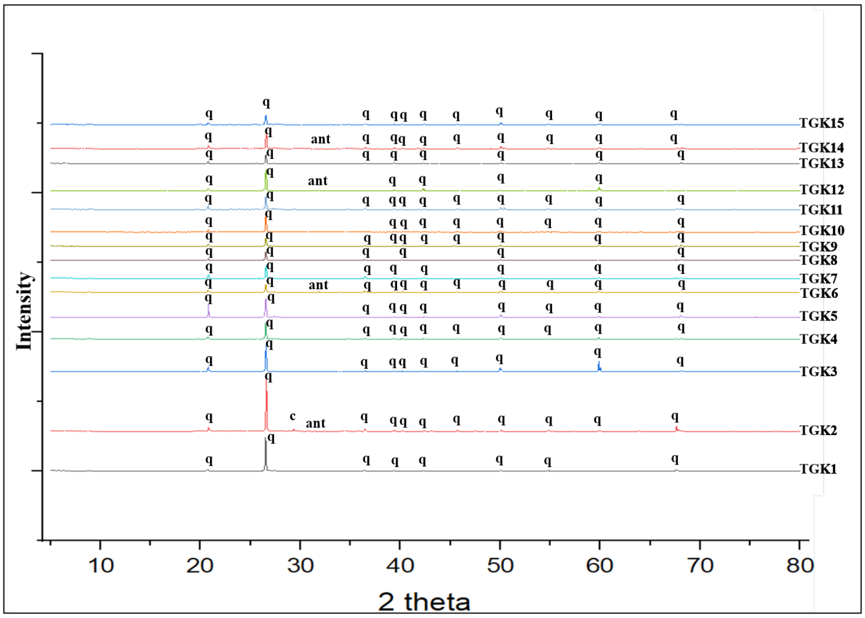

Figure 3: The XRD pattern for earthenware samples in Gua Kelew.
q=quartz; c= calcite; ant=anorthoclase

RESULTS AND DISCUSSION

The XRD test results of the 15 earthenware samples found during the excavation in Gua Kelew are shown in Table 3. The mineral content obtained from the Gua Kelew earthenware shows that the presence of minerals are limited to quartz, anorthoclase and calcite. The XRD patterns of the earthenware samples can be seen in Figure 4. Four earthenware samples, namely TGK2, TGK6, TGK12 and TGK14, show the presence of anorthoclase. TGK2 sample only exhibits the presence of calcite. Furthermore, earthenware samples such as TGK1, TGK3, TGK4, TGK5, TGK7, TGK8, TGK9, TGK10, TGK11, TGK13 and TGK15 only contain quartz, which indicates that these earthenware were fired at a high temperature during their manufacturing process. The high temperature caused most of the minerals to decompose, leaving only quartz. Quartz also probably came from the sand temper used by the potters (Suresh *et al*., 2022).

Comparative analysis was performed on soil sample in Gua Kelew with soil samples studied by Zuliskandar (1999) which were from several areas around Hulu Kelantan, including near Sungai Nenggiri (S.N.), Sungai Betis (S.B.), Sungai Perias (S.S.), Sungai Chai (S.C.), Sungai Jenera (S.J.) and Sungai Peralon (S.P.). The comparison was conducted based on the mineral and main element content found in the earthenware samples and river clay samples around Hulu Kelantan. The mineral content of the clay samples can be seen in Table 4.

Based on the mineral content data of the clay samples taken from rivers in Hulu Kelantan, the data shows that the samples only contain quartz and muscovite, except for the clay samples of Sungai Perias which also contain

Table 3: Mineral content of the earthenware discovered in Gua Kelew.

Sample	Mineral content
TGK1	Quartz, SiO_2
TGK2	Quartz, SiO_2 Calcite, $CaCO_3$ Anorthoclase, $(Na,K)AlSi_3O_8$
TGK3	Quartz, SiO_2
TGK4	Quartz, SiO_2
TGK5	Quartz, SiO_2
TGK6	Quartz, SiO_2 Anorthoclase, $(Na,K)AlSi_3O_8$
TGK7	Quartz, SiO_2
TGK8	Quartz, SiO_2
TGK9	Quartz, SiO_2
TGK10	Quartz, SiO_2
TGK11	Quartz, SiO_2
TGK12	Quartz, SiO_2 Anorthoclase, $(Na,K)AlSi_3O_8$
TGK13	Quartz, SiO_2
TGK14	Quartz, SiO_2 Anorthoclase, $(Na,K)AlSi_3O_8$
TGK15	Quartz, SiO_2

orthoclase besides quartz and muscovite. These clay samples were baked at a temperature of 600°C to 700°C in the laboratory. Therefore, the content of typical clay minerals such as kaolinite, illite and montmorillonite was not present. The data was compared with the mineral content of the earthenware samples; and it is discovered that the mineral content of the earthenware samples from Gua Kelew does not have the same mineral content as the clay samples from the rivers around Hulu Kelantan area. Previous research in Hulu Kelantan show that the earthenwares in the area were brought in by the coastal communities; as they have a more developed life and higher culture (Zuliskandar *et al*., 2011). Trade activities through barter had taken place in the area around Hulu Kelantan, where the coastal community would supply goods from the coast, such as earthenwares or goods from foreign trade to the inland community which in turn supplied forest products consisting of resin, rattan and so forth to be traded by the coastal community.

Table 4: Mineral content of the clay in Hulu Kelantan area.

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	SJ	Quartz Muscovite
Sungai Peralon	SP	Quartz Muscovite

Source: Zuliskandar, 1999

Scientific analysis using the XRF method was conducted on 15 samples of the earthenware obtained from excavations in Gua Kelew. Based on the XRF test, the dry weight content of the main element in the earthenware samples in Gua Kelew can be refered to in Table 5. The analysis results indicate that silica and aluminium are the highest elements, between 60.02% to 72.19% and 12.62% to 17.58%. This shows that the community at that time was skilled in choosing the appropriate raw materials to produce earthenwares, that was mainly clay, to create clay pottery. The iron content was also high in the samples, where the dry weight percentage of iron (Fe_2O_3) ranged from 3.16% to 6.24%. The potassium and calcium content in the earthenware samples ranged from 1.6% to 3.07% and 1.44% to 5.58%, respectively.

The dry weight percentages for sodium, magnesium and titanium elements showed readings ranging from 0.17%, to 0.49%, 0.47% to 1.42% and 0.5% to 0.83%. The phosphorus content in the earthenware samples showed a dry weight percentage between 0.11% to 3.7%. Among the samples that recorded the highest phosphorus reading are TGK6 and TGK12. The high phosphorus content in the two earthenware samples indicates that these pottery utensils were used for cooking, while other samples were for storing food. Phosphorus is a mineral that results from the decomposition of cooked food. These data are in line

Table 5: Dry weight percentage of the main elements found in earthenware samples from Gua Kelew.

Sample	Main element (%)											
	SiO_2	TiO_2	Al_2O_3	Fe_2O_3	MnO	MgO	CaO	Na_2O	K_2O	P_2O_5	LOI	Total
TGK1	60.02	0.66	17.58	6.24	0.18	1.04	2.81	0.20	1.93	0.87	8.45	99.98
TGK2	65.78	0.50	12.71	3.18	0.06	0.49	5.88	0.28	2.15	0.33	8.63	99.99
TGK3	71.91	0.51	12.78	3.16	0.11	0.47	1.85	0.38	2.40	0.14	6.28	99.99
TGK4	67.12	0.64	15.61	5.46	0.11	1.11	1.72	0.23	1.74	0.36	5.88	99.98
TGK5	71.19	0.55	12.79	3.18	0.08	0.55	1.44	0.33	2.87	0.11	6.90	99.99
TGK6	60.22	0.61	16.10	5.54	0.06	1.22	3.18	0.18	1.76	3.70	7.41	99.98
TGK7	63.25	0.66	16.41	5.70	0.08	0.95	2.55	0.18	1.74	0.80	7.65	99.97
TGK8	65.14	0.63	16.61	5.72	0.12	0.94	2.45	0.27	2.02	0.80	5.27	99.97
TGK9	63.78	0.83	16.46	4.87	0.11	0.81	1.97	0.38	2.46	0.24	8.07	99.98
TGK10	63.72	0.63	16.35	5.68	0.10	0.88	2.54	0.21	1.73	0.78	7.37	99.99
TGK11	67.83	0.59	15.99	5.39	0.14	0.99	1.78	0.31	1.83	0.21	4.92	99.98
TGK12	62.47	0.61	16.09	5.43	0.12	1.24	3.37	0.17	1.62	1.85	7.02	99.99
TGK13	63.01	0.60	16.09	5.59	0.10	0.88	2.58	0.20	1.60	0.78	8.56	99.99
TGK14	72.19	0.60	12.62	3.31	0.05	0.55	1.54	0.4	3.07	0.35	5.31	99.99
TGK15	62.20	0.72	17.32	5.74	0.22	1.42	2.12	0.49	2.08	0.59	7.09	99.99

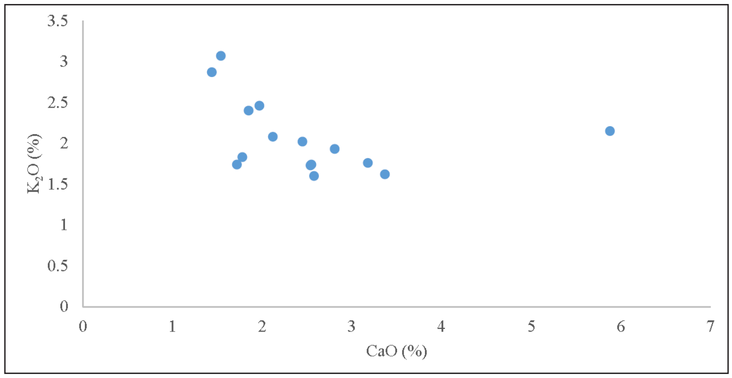

Figure 4: Graph of the percentage distribution of dry weight (%) of potassium and calcium elements in earthenware samples in Gua Kelew, Hulu Kelantan.

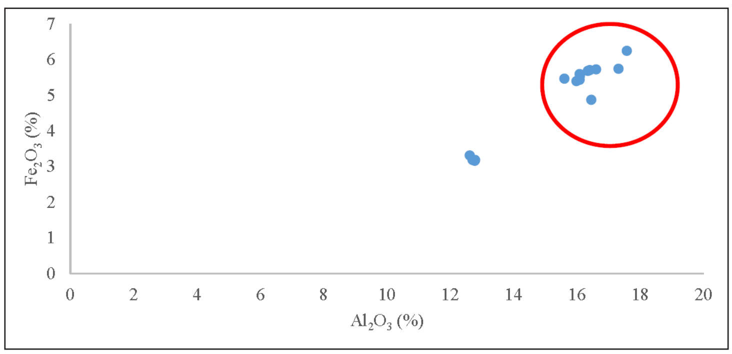

Figure 5: Graph of the percentage distribution of dry weight (%) of aluminium and iron elements in earthenware samples in Gua Kelew, Hulu Kelantan.

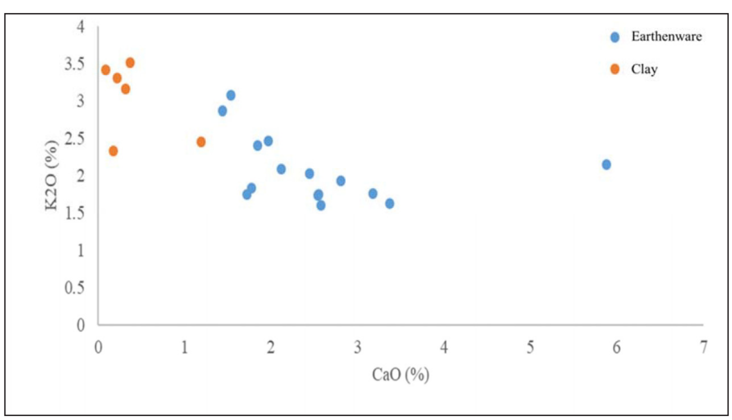

Figure 6: Graph of the distribution of dry weight percentage (%) of potassium and calcium elements in earthenware samples in Gua Kelew and clay samples in Hulu Kelantan.

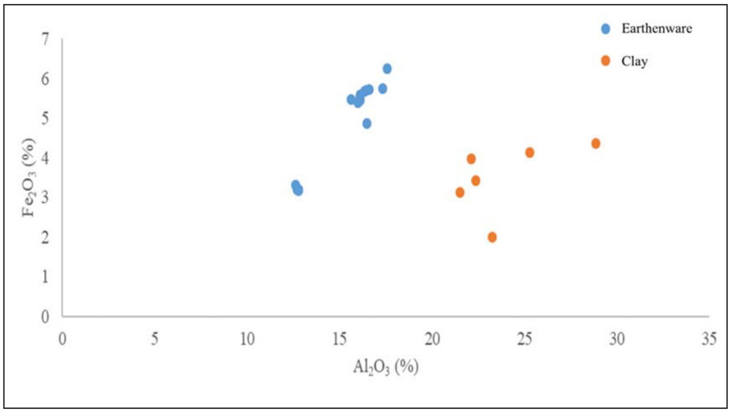

Figure 7: Graph of the percentage distribution of dry weight (%) of the aluminium and iron elements in earthenware samples in Gua Kelew and clay samples in Hulu Kelantan.

Table 6: Dry weight content of minor element and trace elements of earthenware samples from Gua Kelew (µg/g).

	TGK1	TGK2	TGK3	TGK4	TGK5	TGK6	TGK7	TGK8	TGK9	TGK10	TGK11	TGK12	TGK13	TGK14	TGK15
Ba	2292	2523	608	1463	2754	1658	863	1340	2924	1282	BDL	1375	BDL	1450	1590
Br	51	36	31	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	53	BDL
Ce	402	BDL	BDL	433	BDL	494	BDL	512	BDL	446	653	BDL	457	504	BDL
Cl	696	1023	1570	418	1070	618	276	270	443	345	545	389	426	2354	532
Cr	179	BDL	BDL	244	123	276	96	222	230	89	161	BDL	144	150	189
Cu	151	205	103	169	199	165	117	98	152	82	147	132	113	154	BDL
Ni	163	191	51	161	109	BDL	93	125	125	129	105	115	116	150	132
Pb	182	272	118	167	251	197	BDL	121	278	152	145	141	206	345	191
Rb	676	741	377	705	810	452	457	695	606	522	524	507	439	787	653
S	287	458	2547	149	621	481	1327	552	801	468	206	232	242	650	296
Sc	76	BDL	BDL	355	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
Sr	534	627	245	BDL	534	399	346	500	600	495	365	452	407	471	630
Y	319	85	63	251	159	194	206	275	189	263	180	182	219	87	166
Zn	378	299	222	387	269	490	321	294	445	418	293	470	328	321	618
Zr	865	1245	634	949	1864	760	618	751	2990	838	797	891	757	1627	795
Ga	BDL	53	BDL	54	65	72	190	43	40	266	60	BDL	61	38	42
Nb	BDL	80	50	94	112	BDL	78	BDL	113	BDL	BDL	BDL	BDL	149	155
Th	BDL	BDL	90	BDL	309	BDL	BDL	BDL	232	BDL	BDL	BDL	BDL	125	BDL
F	BDL	BDL	BDL	545	BDL	675	BDL	468	BDL	398	BDL	702	BDL	BDL	BDL
Tl	BDL	BDL	BDL	60	BDL	BDL	BDL	BDL	BDL	BDL	BDL	82	BDL	BDL	BDL
Bi	BDL	BDL	BDL	BDL	83	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
Co	BDL	BDL	BDL	BDL	BDL	BDL	1794	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL

with the findings of burning ash in excavation plots which indirectly indicates that the prehistoric communities actively used Gua Kelew as a suitable shelter during the dry or rainy season when river water levels during the monsoon or rainy season are higher.

Figure 4 shows the dry weight percentage distribution of the calcium and potassium elements. Based on the graph, it was found that the composition of calcium and potassium did not show significant differences between the earthenware samples that were analysed. Figure 5 shows a graph of aluminium and iron's dry weight percentage distribution. Based on the graph, it is found that most of these earthenware samples have almost the same composition. Therefore, the findings of each analysis suggests that most of the clay earthenware in Gua Kelew used the same raw material and were obtained from the same area.

Futhermore, Figure 6 displays a graph of the dry weight percentage distribution of the calcium and potassium elements in the clay and earthenware samples around Hulu Kelantan. Based on the graph plots obtained, it is discovered that the dry weight percentage of calcium and potassium for the earthenware and clay samples from Hulu Kelantan area differed from each other, where two groups are present. While the graph in Figure 7 shows the dry weight percentage distribution of aluminium and iron elements contained in the earthenware and clay samples in Hulu Kelantan. Based on the graph plot, it is found that the dry weight percentage of aluminium and iron elements for the earthenware samples and clay samples are also different, where two large groups exist. Concerning this, it is suggested that the earthernwares in Gua Kelew were not produced in the area around Hulu Kelantan.

Other than the main elements, the content of minor and trace elements can also be seen in Table 6. By referring to Table 6, there are several elements found in each sample and there are also some elements that are only found in a few samples. Among the elements that can be seen in each sample are chlorine, rubidium, sulphur, yirium, zinc and even zirconium. In addition to this, there are also elements that are simply not present in one or two samples, that is barium, chromium, copper, nickel, lead, strontium and gallium. Furthermore, there are several elements that can only be seen in a few samples where the value of the content is also low, such as bromine, cerium, scandium, niobium, thorium, fluorine and also thallium. For bismuth and cobalt, these elements are only seen in one sample, that is in TGK5 and TGK7, respectively.

CONCLUSIONS

Analysis of the mineral content, major elements and trace elements found in prehistoric pottery is very important in order to identify the origin of earthenware found in prehistoric sites to determine whether it was originally from the area and made by the local community, or it was brought from outside through trade routes between the interior and the coastal community. Based on the results of mineral content analysis, it is believed that the earthenware found at the prehistoric site of Gua Kelew was burned at high temperature as most of the samples had only quartz mineral, while other minerals were probably lost or destroyed due to the high burning temperature. In addition to this, based on the comparison of the main elements between the earthenware samples with the clay samples found around the Gua Kelew area indicates that there are significant differences between the two groups. This shows that the earthenware found in Gua Kelew was not produced using clay available in the surrounding area, which proves that the earthenwares were brought from elsewhere. The presence of earthenware from other places is likely the result of trade activities and the exchange of goods between the communities living inland with coastal communities. Inland communities needed earthenwares for cooking and storing food, while coastal communities required forest resources for foreign trade.

ACKNOWLEDGEMENTS

We would like to express our highest gratitude to Universiti Kebangsaan Malaysia (U.K.M.) which funded this study through the grant GGPP-2019-005, Chemical Content Study of Prehistoric Earthenwares Artifacts in Ulu Kelantan *(Kajian Kandungan Kimia Artifak Tembikar Tanah Prasejarah di Ulu Kelantan)*. We would also like to extend our sincere appreciation to Universiti Kebangsaan Malaysia through the Physical Characterization Laboratory *(Makmal Pencirian Fizikal)*, iCRIM for the use of the XRD and XRF apparatus. Useful comments and suggestions received from the two anonymous reviewers for GSM helped to improve the paper.

AUTHOR CONTRIBUTIONS

MSMA carried out all scientific analysis and drafted the manuscript, ZR carried out XRD analysis and NFA carried out the physical analysis.

CONFLICT OF INTEREST

We declare that we do not have any conflict of interest with regard to the contents of this paper.

REFERENCES

Adi Taha, 1985. The re-excavation of the rock shelter of Gua Cha, Ulu Kelantan, West Malaysia. Federation Museums Journal, 30(New series), 1-134.

Adi Taha, 1998. Research in the rockshelters of Gua Chawas and Gua Peraling, Ulu Kelantan, Malaysia, Indo-Pacific prehistory: The Melaka papers, Vol.1: Pre-Congress issues, additional abstract. Bulletin of the Indo-Pacific Prehistory Association, 17, 15-16.

Adi Taha, 2007. Archaeology of Ulu Kelantan. Jabatan Muzium Malaysia, Kuala Lumpur. 245 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. https://iaeme.com/Home/article_id/IJMET_09_13_106.

Ali, M.S.M., Ramli, Z., Rahman, N.H.S.N.A., Razman, M.R., & Utomo, B.B., 2015. X-ray fluorescence and X-ray diffraction analysis of ancient bricks from the temple SEG-II (Ubur Lempeng), Batujaya, Indonesia. Journal of Food, Agriculture and Environment, 13(2), 197–200. https://doi.org/10.1234/4.2015.3989.

Asyaari, M., 1998. X-Ray Fluorescence analysis of clay pottery in Perak. Journal of Malaysian Archaeology, 11, 1 – 40.

Asyaari, M., 2002. Development of prehistoric pottery in Malaysia. Journal of Malaysian Archaeology, 15, 29-37.

Asyaari, M., 2010. Ancient ceramics traded in Peninsular 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. https://iaeme.com/Home/article_id/IJMET_09_13_126.

Bishop, R.L., Rands, R.L. & Holley, G.R., 1982. Ceramic compositional analysis in archaeological perspective. Advance in Archaeological Method and Theory. Academic Press Inc, 5, 275 – 327. https://doi.org/10.1016/B978-0-12-003105-4.50012-1.

Bullbeck, D., 2005. The Gua Cha burials. In: Zuraina Majid (Eds.), The Perak Man and Other Prehistoric Skeletons of Malaysia, Universiti Sains Malaysia Publisher, Pulau Pinang. 253-310.

Chia, S., 1997. Prehistoric pottery sources and technology in Peninsular Malaysia, based on compositional and morphological studies. Malaysia Museum Journal, 33, 91 – 125.

Endicott, K. & Bellwood, P., 1991. The possibility of independent foraging in the rain forest of Peninsular Malaysia. Human Ecology, 19(2), 151-185. https://doi.org/10.1007/BF00888744.

Ertem, E. & Demirci, S., 1999. Characteristics of Hittite pottery sherds from sites in the Kilirmak Basin. Journal of Archaeological Science, 26, 1017 – 1023. DOI:10.1006/JASC.1999.0425.

Hooijer, D.A., 1962. Rhinoceros sondaicus Desmarest from the Hoabinhian of Gua Cha Rock Shelter, Kelantan. Federation Museums Journal, 7, 23-24. http://www.rhinoresourcecenter.com/index.php?s=96a8e7804c5e1e97e1911fad57acb130&act=refs&CODE=ref_detail&id=1165237239.

Karina, A., 1990. Social aspects of pottery manufacture in Boera, Papua New Guinea. Paper presented at 14th Congress of Indo Pacific Prehistoric Association, Yogyakarta.

Mohd Anuar, F., 1991. The use of X-Ray diffraction techniques: Identifying mineral content in the study of ancient pottery. Thesis Department of Geology, Faculty of Science, U.K.M.

Mohd Kamaruzzaman, A.R., Mohamad Deraman, Ramli Jaya & Mohd Ali Sufi, 1991. A scientific study of earthenware found in Kalumpang Island, Perak: Preliminary results. Journal of Malaysian Archaeology, 4, 59-73.

Moradi, H. Hossein, Sarhaddi Dadian, H., Zuliskandar, R., & Nik Hassan Shuhaimi Nik Abdul Rahman, 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 Fazrullah, Z., Zuliskandar, R., & Azimah, H., 2019. Finding stone tools and pottery in Gemalah Cave, Hulu Kelantan. In: Suresh Narayanen, Nasha Rodziadi Khaw & Mokhtar Saidin (Eds.), Prehistoric Culture, Ethnography & Orang Asli in Peninsular Malaysia. Centre for Global Archaeological Research, Universiti Sains Malaysia, Penang, 62-75.

Muhamad Shafiq, M.A., Zuliskandar, R., & Nur Sarahah, M.S., 2021. Geochemistry and mineralogy of prehistoric pottery shards found at Gua Jaya, Nenggiri Valley, Kelantan, Malaysia. Bulletin of the Geological Society of Malaysia, 72, 205-213. https://doi.org/10.7186/bgsm72202116.

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.

Noone, H.D., 1939. Report on a new neolithic site in Ulu Kelantan. Journal of Federated Malay States Museums, 15, 170-174.

Nur Farriehah, A., Nuratikah, A.B., & Zuliskandar, R., 2019. Kelew Cave, Ulu Kelantan as remains of prehistoric societies placement evidence based on surface survey. International Journal of Civil Engineering and Technology (IJCIET), 10(8), 233-240. https://ssrn.com/abstract=3450352.

Peacock, B.A.V., 1959. A short description of Malayan prehistoric pottery. Asian Perspective, 3(2), 121-156. https://www.jstor.org/stable/42928913.

Peacock, B.A.V., 1964. Recent archaeological discoveries in Malaya, 1962-1963. Journal of the Malayan Branch of the Royal Asiatic Society, 37(2), 258-260. https://www.jstor.org/stable/41492206.

Peacock, B.A.V., 1965. Recent archaeological discoveries in Malaysia 1964 Malaya. Journal of the Malayan Branch of the Royal Asiatic Society, 38(1), 248-255. https://www.jstor.org/stable/41491845.

Peacock, B.A.V. & Dunn, F.L., 1968. Recent archaeological discoveries in Malaysia, 1967: West Malaysia. Journal of the Malayan Branch of the Royal Asiatic Society, 41(1), 171-179.

Ramli, Z., Rahman, N.H.S.N.A., Jusoh, A., & Sauman, Y., 2011. X-ray diffraction and X-ray fluorescent analyses of prehistoric pottery shards from Ulu Kelantan. American Journal of Applied Sciences, 8(12), 1337–1342. https://doi.org/10.3844/ajassp.2011.1337.1342.

Ramli, Z., Abd. Rahman, N.H.S.N., Jusoh, A., & Hussein, M.Z., 2012. Compositional analysis on ancient bricks from Candi Sungai Mas (Site 32/34), Bujang Valley, Kedah. American Journal of Applied Sciences, 9(2), 196–201. Doi:10.3844/AJASSP.2012.196.201.

Ramli, Z., Rahman, N.H.S.N.A., Samian, A.L., Sauman, Y., & Yatim, O.M., 2013a. Compositional analysis of ancient bricks at Site 2211, Candi Pengkalan Bujang, Kedah. Research Journal of Applied Sciences, Engineering and Technology, 6(16), 3027–3033. Doi:10.19026/RJASET.6.3690.

Ramli, Z., Shuhaimi, N.H., Rahman, N.A., Zakaria, S.Z.S., & Yusof, A.R.M., 2013b. Scientific studies of Candi Pengkalan Bujang (Site 19) ancient bricks: Knowledge of old Kedah community's in usage of local raw materials. Research Journal of Applied Sciences, Engineering and Technology, 6(15), 2859–2864. Doi:10.19026/rjaset.6.3796.

Ramli, Z., Shuhaimi Nik Abdul Rahman, N.H., Samian, A.L., Syed Zakaria, S.Z., & Mohd Yusof, A.R., 2014a. Usage of local raw material in the construction of candi Pengkalan Bujang (Site 18), Bujang Valley, Kedah. Research Journal of Applied Sciences, Engineering and Technology, 7(9), 1779–1786. http://

dx.doi.org/10.19026/rjaset.7.462.

Ramli, Z., Abdul Rahman, N.H.S.N., Samian, A.L., Sauman, Y., & Sarhaddi-Dadian, H., 2014b. 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. http://dx.doi.org/10.19026/rjaset.7.405.

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, 17(2), 117–129. Doi:10.5281/zenodo.800835.

Ramli, Z., Nasir, M.N.M., Samian, A.L., Harun, N.Z., Hadi, M.S.B.A., Bakar, N.A., & Supian, N.S.M., 2018. XRD and XRF Analysis of the ancient bricks from Candi Kampung Baru, Kedah. Planning Malaysia, 16(1), 314–323. https://doi.org/10.21837/pm.v16i5.434.

Rentse, A., 1947. A historical note on the northeast Malay states. Journal of the Malayan Branch of the Royal Asiatic Society, 20(1), 23-40. https://www.jstor.org/stable/41560002.

Rivka, G., 1973. Ancient Pottery. Cassell & Co. Ltd., Britain. 95 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. Doi:10.5281/zenodo.18357.

Sarhaddi-Dadian, H., Moradi, H., Ramli, Z., & Purzarghan, V., 2017. X-ray fluorescence analysis of the pottery shards from dahan-E ghulaman, the achaemenid site in Sistan, east of Iran. Interdisciplinaria Archaeologica, 8(1), 35–42. Doi:10.24916/iansa.2017.1.3.

Sarhaddi-Dadian, H., Ramli, Z., Moradi, H., & Jozi, Z., 2021. Compositional analysis analysis of the pottery shards from kuh-i khawja historical site, sistan, east of Iran. Journal of Ancient History and Archaeology, 8(1), 127–133. Doi:10.19026/RJASET.6.4177.

Sieveking, G. de G., 1954. Gua Cha and the Malayan stone age. Malayan Historical Journal, 1, 111-125.

Sieveking, G. de G., 1954-55. Excavation at Gua Cha, Kelantan 1954. Part 1. Federations Museums Journal, 1-2, 75-143.

Sieveking, G. de G., 1987. Gua Cha in the light of recents research. In: Nik Hassan Shuhaimi Nik Abd. Rahman (Eds.), Early Period Kelantan: Archaeological and Historical Studies in Malaysia. Perbadanan Muzium Negeri Kelantan, Kota Bharu, 73-86.

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), 998–100.

Suresh, N., Nasha, R.K., Ahmad Fadly, J., & Ahmad Syahir, Z., 2022. Tracing the source and origin of earthenware stove in Kelantan State Museum by means of geochemical and mineralogical methods. Bulletin of the Geological Society of Malaysia, 73, 53-65.

Tweedie, M.W.F., 1940. Report on excavation in Kelantan. Journal of the Malayan Branch of the Royal Asiatic Society, 18(2), 1-22. https://www.jstor.org/stable/41559951.

Wan Noor Shamimi, W.A., Zuliskandar, R., Azimah, H., & Ruzairy, A., 2018. Lithic technology and material culture of Haobinhian and Neolithic Settlement in the Lubang Kelawar Batu Tambah Cave. International Journal of Mechanical Engineering and Technology, 9(13), 1244–1252.

William-Hunt, P.D.R., 1951. Recent archaeological discoveries in Malaya (1945-1950). Journal of the Malayan Branch of the Royal Asiatic Society, 24(1), 186-191. https://www.jstor.org/stable/41502989.

Zuliskandar, R., 1999. Chemical studies of artefacts and soils from archaeological sites. Tesis Sarjana Muda, UPM.

Zuliskandar, R., 2019. Recent discoveries of prehistoric sites in the Nenggiri River basin. Journal of Malaysian Archaeology, 32(1), 1-20.

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. Journal of Malaysian Archaeology, 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. Journal of Malaysian Archaeology, 19, 27 – 36.

Zuliskandar, R., Mohd Zobir, H., Asmah, Y., Kamaruddin, Z., & Mahfuz, N., 2007. A study of the chemical composition of kalam semah and clay pottery found in Bukit Chawas Cave. Journal of Malaysian Archaeology, 20, 22 – 65.

Zuliskandar, R., Yunus Sauman & Nik Hassan Shuhaimi Nik Abdul Rahman, 2011. Analysis clay pottery, mortar and bricks composition found in Kota Melawati, Selangor. Journal of Malaysian Archaeology, 24, 93 – 131.

Zuliskandar, R., Nur Farriehah Azizan, Wan Noor Shamimi Wan Azhar, Natasha Edreena Mohamad Nasruddin & Angela Injun, 2020. Cave painting in Kelew Cave, Kelantan as the media communications of the prehistoric community. Asian Journal of Environment, History and Heritage (AJEHH), 4(1), 25-44. https://spaj.ukm.my/ajehh/index.php/ajehh/article/view/146.

Manuscript received 18 August 2022;
Received in revised form 23 March 2023;
Accepted 12 May 2023
Available online 26 May 2023