Bulletin of the Geological Society of Malaysia, Volume 69, May 2020, pp. 149 - 156
DOI: https://doi.org/10.7186/bgsm69202013

Mineralogical and geochemical properties of bricks from Sungai Batu monuments

Ratnah Wati Mohd Rapi*, Ahmad Fadly Jusoh, Mokhtar Saidin

Centre for Global Archaeological Research (PPAG), Universiti Sains Malaysia, 11800, Pulau Pinang, Malaysia
* Corresponding author email address: ratnah1986@gmail.com

Abstract: Evidence of Kedah Tua Kingdom since 6th Century BC was discovered by USM through excavation in Sungai Batu, Bujang Valley, Kedah. The evidence are namely the sites of iron smelting workshops and monuments. There are 53 excavated sites, and 33 of them are monuments which functioned as ritual sites, jetties and administration buildings. The monuments show evidence of floor, stair, wall and corridor structures that was entirely built from brick material. A total of 24 brick samples from eight monument sites were analysed by XRD and XRF to determine their composition. The X-ray Diffraction (XRD) and X-ray Fluorescence (XRF) techniques can determine the mineral contents and trace the elements in qualitative and quantitative values, and Thermogravimetric Analysis (TGA) to clarify the range temperature. The results will provide information whether the ancient inhabitants had used materials obtained locally or conversely. The samples were taken from site SB1B (ritual site), SB1H, SB2B, SB1A (jetty sites) and SB1M, SB1N, SB1R, SB1U,SB1S, SB1V, SB1X, SB2ZY (administrative building sites). The results obtained showed that the people used local material for brick production. Therefore, this verified that the ancient people of Sungai Batu Archaeological Complex already had the skills and knowledge to find the locality and quality raw materials to make their bricks.

Keywords: Sungai Batu Archaeological Complex, Bricks, XRD, XRF and TGA Technique

INTRODUCTION

Archaeology is a multidisciplinary field involving the various application of science and art. Apart from written sources such as Arabic, Chinese, Western and Indian, the existence of Ancient Kedah civilization was obtained from the archaeological discoveries. Ancient Kedah has been known as a civilization centre and research had already began in the 1840s, almost 170 years ago.

An early record stated that Ancient Kedah consists of an area of about 400 km² covering the northern part of Bukit Choras, and bordered at the south by Sungai Muda, west by the Straits of Malacca and east by the North-South Expressway (Adi, 1998). Study of the ancient sea level of ancient Kedah showed that during the first to third Centuries CE, Sungai Merbok estuary was located near the Sungai Batu Archaeological Complex (Mokhtar *et al.,* 2011).

An archaeological survey conducted in the year 2007 found bricks on the surface layer in an oil palm plantation between the Semeling – Merbok new road (Mokhtar *et al.,* 2011). The area lies at latitude 5°41.6' North and longitude 100°27.1 East, with an altitude of 12 m above the sea level and located near the upstream of Sungai Merbok. The survey has mapped 97 potential sites with small mound hills with an area of approximately four (4) km².

Excavations began in 2009 and to date has revealed 54 sites of iron smelting and monument structures dating from the 6th century BCE (Naizatul & Mokhtar, 2018) (Figure 1). All monuments at the Sungai Batu Archaeological Complex were built using bricks.

BRICK MATERIAL

Bricks are products produced from clay and water (El- Gohary, M.A. & Al-Naddaf, M.M., 2009; Dalkılıç & Adnan Nabikoğlu, 2017). The rectangular bricks made of clay and baked was used for construction purposes (Maria *et al*., 2015). Iklil (2014) classified bricks into four types namely kiln burned bricks, adobe (sunbeds), concrete bricks and limestone bricks. This classification was based on its raw material and stiffening techniques.

The use of clay brick is one of the oldest and most durable manufacturing techniques applied by human (Fernandes *et al.,* 2010; Bardan, 1990). The base material has been used since the Mesopotamia, Egypt and Roman era. Soil is an inexpensive primary material and can be easily obtained in large quantity. It could be shaped easily and is lighter than rocks (Paula *et al.,* 2010).

The selection of this material is due to its high compressive strength, durability and thermal insulation. The burned brick is much better than the sun-dried brick. This is because the unburned brick easily crumbles when in contact with water. A combustion temperature of 500°C and above will strengthen the brick. If the combustion temperature reaches up to 870°C, the process of vitrification will occur that turns mineral into glass. This formed substance can bind the clay elements to make the brick even stronger. (Beamish & Donovan, 1993; Alaa A. Shakir & Ali Ahmed Mohammed, 2013). The traditional brick making often produced bricks in an unequal shapes and size, messy and with a rough texture. That is caused by the deficiency of manpower and tools (Haryono, 2013).

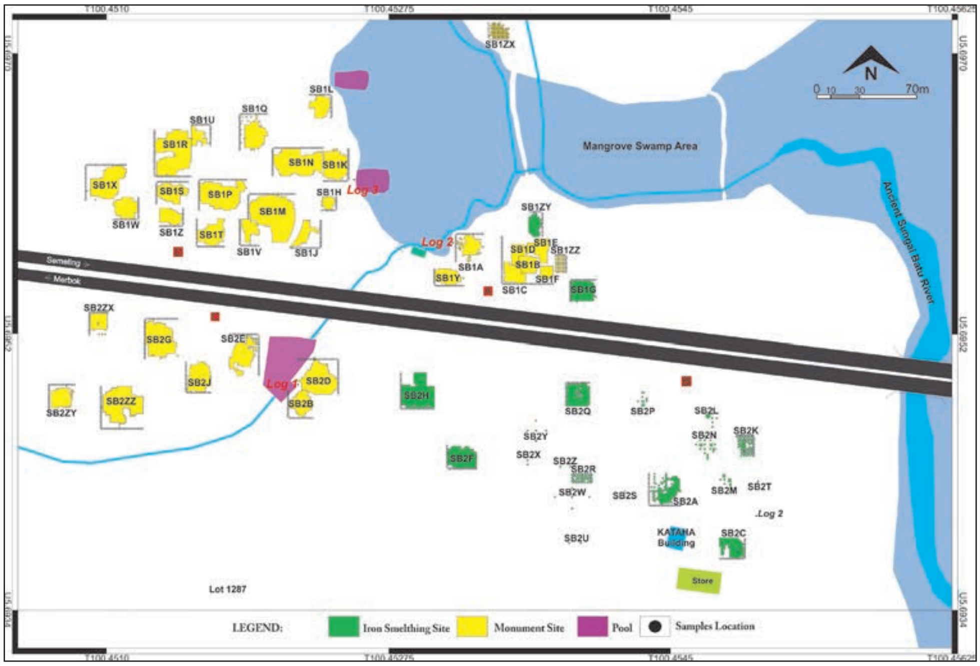

Figure 1: Distribution of iron smelting site, monument structures and location of soil samples at Sungai Batu, Kedah.

The creation of bricks is one of the ideas that led to a great paradigm of the human civilization history in architectural knowledge. This development proves the mental stage of the community. Their ability to identify the source and mixing it with other materials in producing bricks proves their community had a high geological knowledge and adapted it in their daily life activities. Their knowledge can be measured in the production of quality, durable, hard and not easily damaged bricks despite weather changes.

The earliest records of brick usage in construction showed it started between 10,000 – 8,000 BC in Jerico, and is still used throughout the world (Campbell & Pryce, 2003). Due to the advancement of technological knowledge and facilities to obtain the necessary materials for brick making, this suggests that it has become the main choice of society. It is not surprising therefore that the main material used in the monument constructions at Sungai Batu Archaeological Complex was bricks.

METHODOLOGY

This study uses a scientific approach in determining the mineral content, trace elements and major elements of bricks used in the construction of monuments at the Sungai Batu Archaeological Complex. It aims to verify the use of local raw materials or materials from elsewhere. 25 brick samples were taken from 12 sites, together with 3 soil samples (Log 1, Log 2 and Log 3) which represented clayey materials around the ancient river remains (Figure 1). The samples include the brick samples from representing ritual site (SB1B), jetties (SB1H, SB2B and SB1A) and administration areas (SB1M, SB1N, SB1R, SB1S, SB1U, SB1V, SB1X and SB2ZY) (Table 1).

All these samples were analysed using X-Ray Diffraction (XRD) and X-Ray Fluorescence (XRF) for mineralogy and elemental composition. Selected samples underwent another analysis that is Thermogravimetric Analysis (TGA) to clarify the range of temperature. The samples were crushed and pulverized to a standardize grain size between 20 – 30µm using Retch PM100 Milling Machine. The crushed samples was then dried for 2 hours in Memmert oven at 105°C with half fresh air flow. All samples were labelled and sorted according to their site locations and functions (refer Table 1).

The XRF analysis was performed by preparing fuse beads for major element analysis and pressed pallet embedded in boric acid for trace element analysis. All prepared samples were analysed with Panalytical Axios Max XRF. Powdered samples was loaded to 25mm PMMA sample holder and analysed with Bruker D8 Advance XRD with a scanning parameter 10-70° 2θ (2theta), step size 0.02s and continuous scanning mode with Cu X-Ray tube. Selected brick samples was subjected to Thermogravimetric Analysis (TGA), using model Perkin Elmer SDTA 6000. 6.0 mg of sample was heated at a temperature range of 20 – 900°C, with heating rate 20°C/minute in dynamic air (Cayme *et al.*, 2016). The percentage by weight of sample relative to the change in temperature was recorded.

RESULTS AND DISCUSSION

Cayme *et al.* (2016) found ancient brick contained clay and silicate with variable amount of metallic element including iron, calcium and magnesium. From

Table 1: The 25 samples representing the dating and the function of the monuments at Sungai Batu Archaeological Complex.

Site Name & Dating	Function	Samples
SB1B (2 AD)	Ritual	T10, X16
SB2B (5 AD)	Jetty	G18
SB1A (3 AD)	Jetty	F13, J10
SB1H (1 BC)	Jetty	T9, P8
SB1M (1 AD)	Port Administration	S33
SB1N (3 BC)	Port Administration	E14, Z9, R10, Y12, D11, J14
SB1X (2 AD)	Port Administration	S1, A8
SB1R (5 BC)	Port Administration	T26
SB1S (5 BC)	Port Administration	H20
SB1U (2 BC)	Port Administration	C6
SB1V (3 AD)	Port Administration	G14
SB2ZY (5 BC)	Port Administration	K10, E12, F9, B5, L11

the XRD analysis, it was found that the bricks consist of several minerals such as quartz (SiO_2), montmorillonite ($AlSi_2O_6(OH)_2$), microcline ($K_{0.96}Na_{0\cdot04}AlSi_3O_8$), feldspar and metals. The list of minerals is recorded in Table 2.

The X-Ray Diffractogram was analysed using Bruker DIFFRAC.eva software by referring to the International Centre for Diffraction Data ICDD PDF-2 database. It shows that all the samples contain quartz (SiO_2) which indicate that the ancient people in Sungai Batu had mixed clay soil with sand to enhance the durability and hardness of the bricks. Two types of clay minerals found in the Sungai Batu bricks are montmorillonite and kaolinite. Clay could be found easily near the sites because most of the sites were located near river banks. Clay is very sensitive to heat; several bricks were found to be absent of clay minerals (Samples 1NR10, 1NJ14, 1ND11, 1HP8). This shows that the minerals of clay, feldspar and mica have transformed / restructured to another mineral due to burning at high temperature.

Zuliskandar *et al.* (2012) mentioned that the bricks containing quartz only burns at a temperature over 550˚C. At this temperature, clay mineral such as kaolinite and montmorillonite will transform to another mineral called Meta-Kaolin that is an amorphous material which will not show at the peak in the XRD diffractogram. This therefore suggests that the Sungai Batu bricks were dried and burnt in an open flame at low temperature because the clayey minerals are still present in the brick samples. When the brick makers arranged the bricks to burn, bricks at the lowest and furthest position were exposed to low temperature compare to those nearer to the fire.

Bricks that was burnt at high temperature are more solid and hard compared to bricks produced in low temperature. The samples was further analysed using the Thermogravimetric Analysis (TGA), the results confirming the degradation temperature of the Sungai Batu bricks. On the other side, comparison of the different brick functions with the mineral content shows that the bricks used for jetty structures are more robust compared to bricks used in other structures or at other sites. In the jetty bricks samples, kaolinite may also be present due to the sedimentation process that occurred along the river banks. The XRD method is the best way to identify the minerals in the bricks samples and give much more information about the technology of ancient brick making.

The XRF Analysis of major elements is recorded in Table 3. Silica (Si) showed the highest dry weight percentage in all samples in the range of 59.89 – 80.28%. The dry weight of alumina (Al) recorded second higher concentration in a range of 8.41 – 22.48%, while titanium (Ti) is between 0.5 – 6.65%, ferum (Fe) shows 1.35 – 7.74%, while manganase (Mn) only showed less than 0.01%. Magnesium (Mg) and calcium (Ca) contains dry weight 0.11 – 0.89% and 0.05 – 0.43%, respectively. Natrium (Na), potassium (K) and phosphorus (P) are in small amount in the Sungai Batu bricks samples.

To clarify the origin of the material of the bricks, the comparison of dry weight between silicate (SiO_2) and alumina (Al_2O_3) of Sungai Batu bricks and soil samples taken from around the site near the ancient river banks was made (Figure 2). Figure 2 showed the close relativity of the soil samples and the Sungai Batu bricks, in the range of 60 – 80% dry weight of silica and 10 – 25% dry weight of alumina. This suggests that the bricks were locally made and used the soil sourced from around the sites.

The content of trace element in Sungai Batu ancient bricks in part per million (ppm) is showed in Table 4, 5 and 6. Almost 21 trace elements have been reported by using XRF Omnion Analysis. Chlorine (Cl) recorded the highest concertation followed by sulphur (S), tungsten (W), zircon (Zr) and zink (Zn). Other elements are present in very low concentration; below the instrument detection limit.

All of these trace elements are commonly found in soils especially clayey soil. Comparison of trace element between zircon (Zr) and zink (Zn) was made to identify the similarity of the Sungai Batu bricks and the soil samples. The graph in Figure 3 shows the relation between these

Table 2: The list of minerals in Sungai Batu ancient bricks.

Site	Sample	Mineral
1B	T10	Quartz (SiO_2), Kaolinite-1A ($Al_2Si_2O_5(OH)_4$, Montmorillonite-22A ($Na_{0.3}(Al,Mg)_2Si_4O_{10}(OH)_2.8H_2O$)
	X16	Quartz (SiO_2), Kaolinite-1A ($Al_2Si_2O_5(OH)_4$), Montmorillonite-22A (($Na_{0.3}(Al,Mg)_2Si_4O_{10}(OH)_{2.}8H_2O$), Diaspore (AlO(OH))
2B	G18	Quartz (SiO_2), Montmorillonite ($Na_x(Al,Mg)_2Si_4O_{10}(OH)_2.zH_2O$), Lepidrocrocite ($Fe_2O_3.H_2O$)
1A	P12	Quartz (SiO_2), Kaolinite-1A ($Al_2Si_2o_5(OH)_4$), Montmorillonite-22A (($Na_{0.3}(Al,Mg)_2Si_4O_{10}(OH)_2.8H_2O$)
	J10	Quartz (SiO_2), Kalsilite ($KAlSiO_4$)
1H	T9	Quartz (SiO_2), Montmorillonite-15A ($Ca_{0.2}(Al,Mg)_2\ Si_4O_{10}(OH)_2.4H_20$), Mullite (Al4.44Si1.56O9.78), Cristobalite (SiO_2)
	P8	Quartz (SiO_2)
1M	S33	Quartz (SiO_2), Kalsilite (K Al SiO_4), Microcline ($K_{0.5}Na_{0.5}AlSi_3O_8$)
1N	E14	Quartz (SiO_2), Microcline ($KAlSi_3O_8$)
	Z9	Quartz (SiO_2), Microcline ($KAlSi_3O_8$)
	R10	Quartz (SiO_2)
	Y12	Quartz (SiO_2), Montmorillonite ($Na_{0.3}(AlMg)_2Si_4O_{10}OH_2.6H_2O$)
	D11	Quartz (SiO_2)
	J14	Quartz (SiO_2)
1X	S1	Quartz (SiO_2), Montmorillonite ($Na_{0.3}(AlMg)_2Si_4O_{10}OH_2.6H_2O$)
	A8	Quartz (SiO_2), Montmorillonite ($AlSi_2O_6(OH)_2$)
1R	T26	Quartz (SiO_2), Montmorillonite-18A ($Na_{0.3}(AlMg)_2Si_4O_{10}OH_2.6H_2O$)
1S	H20	Quartz (SiO_2), Montmorillonite ($AlSi_2O_6$)
1U	C6	Quartz (SiO_2), Montmorillonite ($(Ca,Na)_{0.3}Al_2(Si,Al)_4O_{10}(OH)_2.xH_20$), Anatase ($Ti_{0.72}O_2$), Lepidocrocite FeO(OH), Maghemite (Fe_2O_3)
1V	G14	Quartz (SiO_2), Kalsilite ($K_{0.985}(AlSiO_4$)
2ZY	K10	Quartz (SiO_2), Montmorillonite-18A ($Na_{0.3}(AlMg)_2Si_4O_{10}OH_2.6H_2O$)
	E12	Quartz (SiO_2), Montmorillonite-18A ($Na_{0.3}(AlMg)_2Si_4O_{10})H_2.6H_2O$), Magnetite ($Fe_{0.8}Ni_{0.2}Mg_{0.6})O_4$)
	F9	Quartz (SiO_2), Montmorillonite-22A ($Na_{0.3}(Al,Mg)_2Si_4O_{10}(OH)_2.8H_2O$), Microcline ($K_{0.96}Na_{0.04}AlSi_3O_8$)
	B5	Quartz (SiO_2), Montmorillonite-18A ($Na_{0.3}(Al,Mg)_2Si_4O_{10}OH2.6H_2O$), Microcline ($K_{0.96}AlSi_3O_8$)
	L11	Quartz (SiO_2), Montmorillonite-18A ($Na_{0.3}(Al,Mg)_2Si_4O_{10}OH_2.6H_2O$)

Table 3: The list of major elements with quantitative value for Sungai Batu ancient bricks.

Sample	Dry Weight (%)									
	Si	Ti	Al	Fe	Mn	Mg	Ca	Na	K	P
1BT10	75.58	0.76	14.55	3.54	0.01	0.37	0.18	0.16	0.37	0.05
1BX16	68.24	0.75	14.96	2.79	bdl	0.29	0.06	0.04	0.61	0.04
2BG18	71.94	0.62	11.62	2.27	0.01	0.14	0.07	0.73	0.43	0.02
1AF13	61.56	0.96	22.48	7.74	0.01	0.89	0.11	0.17	0.92	0.04
1AJ10	72.03	0.72	13.27	2.34	bdl	0.09	0.07	0.03	0.40	0.03
1H T9	67.03	0.83	16.51	1.79	bdl	0.13	0.08	bdl	0.33	0.03
1HP8	72.98	0.67	12.48	2.02	bdl	0.19	0.17	0.02	0.29	0.02
1M S33	79.28	0.52	9.23	1.51	bdl	0.12	0.06	bdl	0.19	0.01
1N E14	80.28	0.56	8.74	1.34	bdl	0.11	0.06	0.15	0.25	0.02
1NZ9	79.12	0.65	10.09	1.61	bdl	0.12	0.06	bdl	0.27	0.02
1NR10	76.81	0.67	11.62	1.95	bdl	0.17	0.05	bdl	0.20	0.02
1NY12	73.12	0.85	13.38	1.34	bdl	0.11	0.06	0.15	0.25	0.02
1ND11	77.04	6.65	11.33	1.72	bdl	0.13	0.07	bdl	0.26	0.02
1NJ14	78.80	0.61	10.03	1.59	bdl	0.14	0.05	0.01	0.22	0.02
1X S1	71.42	0.62	10.93	2.88	bdl	0.15	0.09	bdl	0.42	0.02
1AX8	76.10	0.62	8.81	1.52	bdl	0.10	0.06	bdl	0.19	0.02
1RT26	69.37	0.76	12.88	1.76	bdl	0.15	0.08	bdl	0.32	0.02
1SH20	59.89	0.50	8.41	1.35	bdl	0.13	0.05	1.00	0.02	0.01
1UC6	73.34	0.62	10.16	1.46	bdl	0.11	0.08	bdl	0.22	0.16
1V G14	72.68	0.58	8.55	1.48	bdl	0.10	0.06	bdl	0.15	0.02
2ZYK10	73.09	0.61	12.78	2.39	bdl	0.12	0.05	bdl	0.54	0.02
2ZYE12	71.33	0.67	14.20	2.47	bdl	0.41	0.43	bdl	0.46	0.02
2ZYF9	73.43	0.69	13.18	2.06	bdl	0.12	0.08	bdl	0.40	0.02
2ZYB5	72.71	0.68	14.29	2.38	bdl	0.13	0.05	bdl	0.53	0.02
2ZYL11	72.47	0.72	13.91	2.28	bdl	0.13	0.05	bdl	0.31	0.02

*bdl = below detection limit

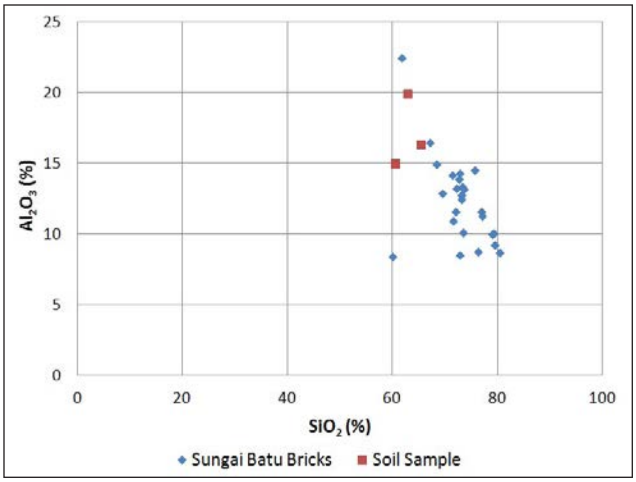

Figure 2: Comparison dry weight (%) of silicate and alumina in Sungai Batu bricks and soil samples.

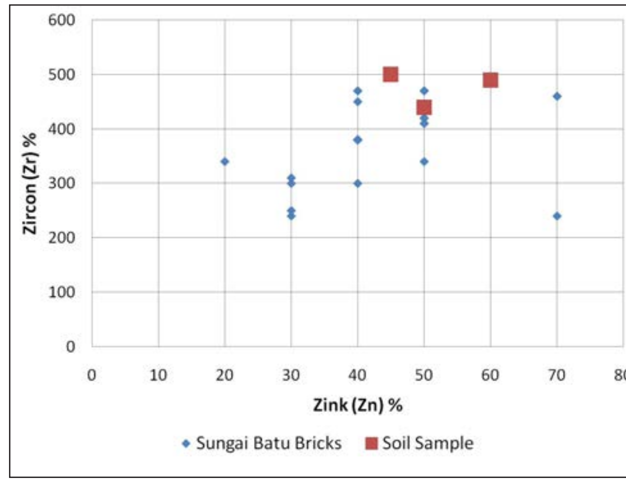

Figure 3: Comparison dry weight (%) of zircon (Zr) and zinc (Zn) in Sungai Batu bricks and soil samples.

Table 4: The list of trace element with quantitative value.

Element (ppm)	Dry Weight (%)							
	1BT10	1BX16	2BG18	1AF13	1AJ10	1HT9	1HP8	1MS33
As	bdl	bdl	bdl	60	bdl	bdl	bdl	bdl
Br	bdl	20	10	bdl	30	20	50	bdl
Cl	1360	2220	2940	2090	7740	640	2090	170
Co	2140	bdl	bdl	1930	bdl	bdl	bdl	280
Cr	bdl	110	90	130	190	130	60	80
Cu	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl
F	bdl	bdl	470	bdl	780	bdl	bdl	bdl
Ga	30	30	330	30	20	30	20	200
Hf	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl
Nb	30	30	20	30	bdl	20	40	30
Nd	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl
Ni	80	bdl	bdl	70	bdl	bdl	50	bdl
Pb	bdl	50	50	80	bdl	50	50	bdl
Rb	40	60	30	90	30	30	bdl	20
S	150	2280	1120	230	2050	4290	4080	1750
Sr	40	20	10	50	bdl	20	20	bdl
Ti	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl
W	2250	280	bdl	830	380	550	730	980
Y	20	20	20	40	40	30	30	20
Zn	30	30	30	70	50	50	40	40
Zr	310	300	250	240	340	420	450	380

*bdl = below detection limit

Table 5: The list of trace element with quantitative value for Sungai Batu Ancient bricks.

Element (ppm)	Dry Weight (%)							
	1NE14	1NZ9	1NR10	1NY12	1ND11	1NJ14	1XS1	1XA8
As	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl
Br	bdl	bdl	bdl	bdl	bdl	bdl	10	bdl
Cl	620	280	310	590	4930	1980	450	390
Co	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl
Cr	80	70	90	bdl	110	130	120	70
Cu	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl
F	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl
Ga	bdl	30	290	40	290	250	290	20
Hf	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl
Nb	20	20	20	30	20	30	20	20
Nd	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl
Ni	bdl	70	70	bdl	bdl	bdl	bdl	bdl
Pb	bdl	bdl	bdl	bdl	bdl	bdl	bdl	40
Rb	20	30	30	30	20	20	30	10
S	860	1350	1350	1080	860	2300	730	4890
Sr	bdl	bdl	bdl	bdl	bdl	bdl	10	bdl
Ti	bdl	bdl	bdl	bdl	bdl	240	20	bdl
W	490	510	460	340	490	410	bdl	bdl
Y	20	20	30	30	20	bdl	20	20
Zn	50	50	70	40	50	50	30	20
Zr	440	470	460	470	440	410	240	340

*bdl = below detection limit

Table 6: The list of trace element with quantitative value for Sungai Batu Ancient bricks.

Element (ppm)	Dry Weight (%)								
	1RT26	1SH20	1UC6	1VG14	2ZYK10	2ZYE12	2ZYF9	2ZYB5	2ZYL11
As	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl
Bi	bdl	bdl	bdl	bdl	bdl	bdl	40	bdl	bdl
Br	bdl	bdl	bdl	bdl	bdl	bdl	bdl	20	20
Cl	620	280	310	590	4930	1980	220	210	670
Co	bdl	bdl	bdl	bdl	bdl	bdl	bdl	1400	bdl
Cr	80	70	90	bdl	110	130	90	60	bdl
Cu	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl
F	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl
Ga	bdl	30	290	40	290	250	220	250	250
Hf	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl
Nb	20	20	20	30	20	30	30	20	20
Nd	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl
Ni	bdl	70	70	bdl	bdl	bdl	bdl	bdl	bdl
Pb	bdl	bdl	bdl	bdl	bdl	bdl	50	bdl	bdl
Rb	20	30	30	30	20	20	20	30	30
S	860	1350	1350	1080	860	2300	1930	1540	1540
Sr	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl
Ti	bdl	bdl	bdl	bdl	bdl	240	bdl	bdl	bdl
W	490	510	460	340	490	410	590	590	590
Y	20	20	30	30	20	bdl	30	30	30
Zn	50	50	70	40	50	50	40	50	50
Zr	440	470	460	470	440	410	300	420	420

*bdl = below detection limit

two samples. Both Sungai Batu bricks and the soil samples have the same range of concentration of zircon and zinc. It can be used to support the similarity of the major element discussed above.

In Figure 4, a distinct endothermic peak and loss of mass corresponding to the dihydroxylation of mineral in the bricks are highlighted by the red circle.

Figure 4 shows the degradation of the mineral around 200˚C – 800˚C. Between the temperature 80 – 100˚C, the moisture in the sample was removed. As the temperature increased, the organic material and the water contained in the bricks microstructure was combusted at temperature between 200˚C – 500˚C. A drop of sample weight was detected at 438˚C which is due to dihydroxylation of kaolinite mineral in the Sungai Batu bricks to form another type of mineral called meta-kaolin. Meta-kaolin is amorphous and is not shown in the XRD diffractogram (Samples 1NR10, 1NJ14, 1ND11, 1HP8). The TGA technique can be used to determine the temperature of mineral degradation and provide information regarding the burning process in brick making.

CONCLUSION

XRD is the best analysis to apply in mineral characterization. The highly sensitive X-Rays will give the accurate composition of artefact in the archaeological study. Sungai Batu bricks contain a large number of silicates and aluminates. The presence and absence of some minerals like kaolinite and montmorillonite will give a good explanation of how the bricks were made. The XRF analysis found that the major and trace elements can be used as a comparison of artifacts and its material origin. With the quantitative measurement, it can trace the similarity of specific elements in the soil samples and artifacts.

Silicate (SiO_2) and alumina (Al_2O_3) with zircon (Zr) and zinc (Zn) can be a fingerprint to verify this similarity (Figures 1 and 2). TGA can be applied to confirm the temperature of burning process in brick making. At 530˚C, clay mineral appeared to degrade, suggesting that the burning temperature for Sungai Batu bricks was over 438˚C in open firing. This is concluded as there are absence of some clay minerals in the Sungai Batu bricks.

Therefore, the ancient people in Sungai Batu not only knew how to make bricks, but also had the knowledge to select the best source of clay to produce quality bricks in a large scale. In the different era from before century (BCE) to a century (CE), bricks productions had evolved in technology, from dried to burning in open firing. The people had choosen the best local source of soil to make bricks by mixing sand (quartz) with clay to obtain a hard and durable material.

ACKNOWLEDGEMENTS

Thank you for the grant "Kajian Arkeologi Sungai Batu (1001/PARKEO/870007)" for funding this research. Not forgetting all the lecturers, staff, Sungai Batu teams and all who were involved directly and indirectly in the success of this project.

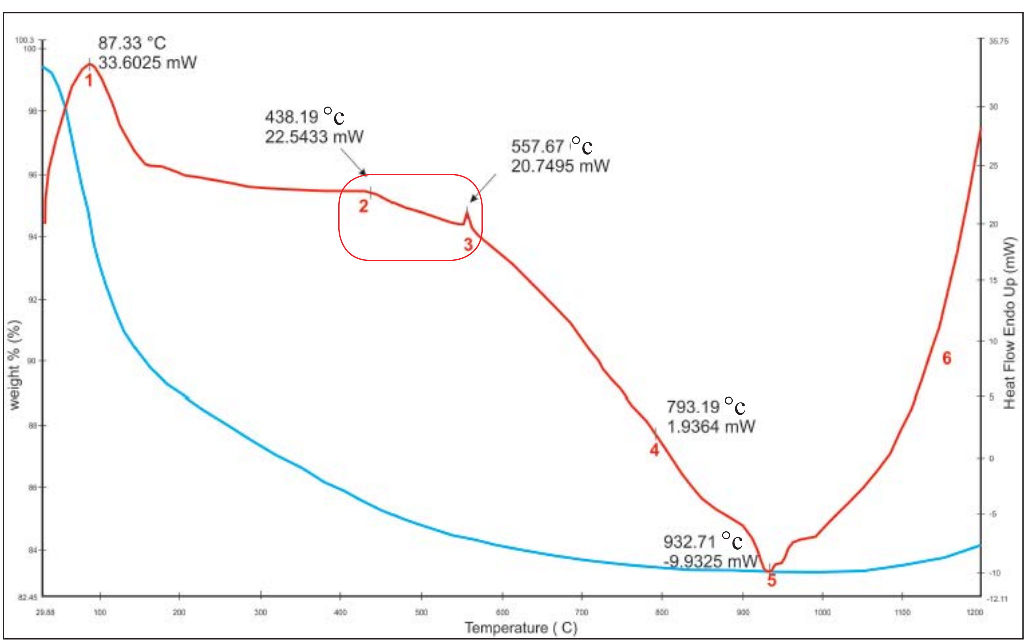

Figure 4: Thermogravimetric Analysis (TGA) graph of Sungai Batu bricks.

REFERENCES

Adi Hj Taha, 1998. Bujang Valley National Historical Park. Malaysia Musuem Journal, 34, 1-16.

Alaa A. Shakir & Ali Ahmed Mohammed, 2013. Manufacturing of Bricks in the Past, in the Present and in the Future: A State of the Art Review. International Journal of Advances in Applied Sciences (IJAAS), 2(3), 145-156.

Bardan, B., 1990. A New Restoration Material for Adobe Structure. 6th International Conference of the Conservation of Earthen Architecture. Getty Conservation Institute, Los Angeles, USA.

Beamish, A. & W. Donovan, 1993. Village Level Brickmaking. Deutsches Zentrum ftir Entwicklungstechnologian.

Campbell, J.W.P. & Pryce, W., 2003. Brick: A World History. Thames & Hudson, London. 320 p.

Cayme, J.M. & Asor, A., Jr., Alano, M.K.A. & Miranda, E., 2016. Chemical Characterization of historical Brickwork of the church Convento in Pagsanjan, Laguna. KAMIKA , 27(1), 30-41.

El-Gohary, M.A. & Al-Naddaf, M.M., 2009. Characterization of Bricks Used in The External Casing of Roman Bath Walls "Gadara- Jordan". Mediterranean Archaeology and Archaemetry, 9(2), 29-46.

Francisco, M. Fernandes, Paulo B. Lourenco & Fernando Castro, 2010. Ancient Clay Bricks: Manufacture and Properties. In: M. Bostenaru Dan *et al.,* (Eds.), Material, Techonologies and Practies in Historic Heritage Structure. DOI 10. 1-20.

Haryono, 2013. Analisis Perbandingan Metode Pelaksanaan, Mutu dan Biaya Konstruksi Gedung Menggunakan Bata Press dan Bata Konversional di Atmi Surakarta. Universitas Muhammadiyah Surakarta. Thesis.

Iklil Izzati Zakaria, 2014. Kajian Arkelogi di Tapak SB2B dan SB2D, Kompleks Sungai Batu, Lembah Bujang. Thesis MA, Universiti Sains Malaysia (Not Published).

Maria Stefanidou, Ioanna Papayianni & Vasiliki Pacha, 2015. Analysis and Characterization of Roman and Byzantine Fired Brick from Greece. In: Material and Structures. https://doi.org/10.1617/s11527-014-0306-7. Ms 1-10

Mokhtar Saidin, Jefrey Abdullah, Jalil Osman &Azman Abdullah, 2011. Issue and Problem of Previous Study in Bujang Valley and Discovery of Sungai Batu. In: Chia, S. & Andaya, B. (Eds.), Bujang Valley and Early Civilizations in Southeast Asia. USM, Penang, 15-26.

Mokhtar Saidin, Jeffery Abdullah & Jalil Osman, 2011. Masa Depan USM and USU dalam Penyelidikan Arkeologi Serantai. In: Mokhatr Saidin & Suprayinto (Eds.), Mengungkap Peradaban Asia Tenggara melalui Tapak Padang Lawas, Sumatera dan Tapak Sungai Batu, Kedah. USM, Penang, 1-12.

Naizatul Akma & Mokhtar Saidin, 2018. Sumbangan Lembah Bujang Terhadap Industri Besi di Malaysia. Jurnal Antarabangsa Dunia Melayu, 11(2), 183-200.

Neslihan Dalkılıç & Adnan Nabikoğlu, 2017. Traditional Manufacturing of Clay Brick used in The Historical Building of Diyarbakir (Turkey). Frontiers of Architectural Research 6, Faculty of Architecture, Department of Architecture, Dicle University, Diyarbakir, Turkey. 346-359.

Paula B. Lourenco, Francisco M. Fernandes & Fernando Castro, 2010. Handmade Clay Bricks: Chemical, Physical and Mechanical Properties. International Journal of Architectural Heritage: Conservation, Analysis and Restoration, 4, 38-58.

Zuliskandar Ramli, Nik Hassan Shuhaimi, Adnan Jusoh & Mohd Zobir Hussein, 2012. Compositional Analysis on Ancient Bricks from Candi Sungai Mas (Site32/34), Bujang Valley, Kedah. American Journal of Applied Science, 9(2), 196-201.

Manuscript received 8 November 2018
Revised manuscript received 25 May 2019
Manuscript accepted 27 December 2019