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IN-SITU COMPOSITIONAL ANALYSIS AND PROVENANCE STUDY OF THE HISTORIC TERENGGANU STONE (THE INSCRIBED STONE "BATU BERSURAT") USING NEUTRON-INDUCED PROMPT GAMMA-RAY TECHNIQUES (NIPGAT)

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ABSTRACT

"Batu Bersurat Terengganu (inscribed stone)" is the oldest artifact with Jawi writing on it. The artifact proves that the Kingdom of Terengganu exist earlier than 1326 or 1386. To date, a lot of studies on the content of the inscription have been carried out by historians and archaeologists, but no scientific investigation about the material composition and its provenance has been performed. This paper focuses on the study of the origin of the Batu Bersurat Terengganu using Neutron-Induced Prompt Gamma-Ray Techniques (NIPGAT). Portable NIPGAT system has been designed and developed based on volumetric measurement methods and it will be considered as a non-destructive testing. The system uses low activity of californium-252 (Cf-252) neutron radioactive sources, gamma ray spectroscopy and special computer software to carry out the investigation. The study found that the Batu Bersurat Terengganu is made of dolerite based on the elemental composition of the stone. Although most of the scientific data for the study of the origin are already obtained, but further research is still ongoing to complete the scope of this study.

Keywords: Californium-252, irradiation, neutron-induced prompt gamma, non-destructive testing, Terengganu Inscribed Stone

INTRODUCTION

The stone on which the inscription was chiseled is unpolished. It is 84 cm in height and weighs 214.8 kg. The stone was probably chosen because of its convenient shape. It is slightly wider at the top with flat faces on the front, back, and sides. This gave sufficient space for the edict. The top part has long since been broken off (most writers assume that there are writings on the missing part), thus affecting the inscriptions on the lateral sides (Figure 1). These appear to run up one side, over the (missing) top, and down the opposite side. Until now, efforts to locate the missing part of the stone have been unsuccessful.

The stone itself is still shrouded in historical mysteries and there is no scientific data or reports available in opened literatures up to now. Due to high historical values, the stone is now gazetted as a world heritage by the UNESCO and it is been display in a cubicle at Terengganu State Museum, Malaysia (Figure 2). A lot of methods are being developed by scientists, archaeologists and historians to study different aspects of the inscribed stone. Among others, a scientific project is to develop an advanced non-intrusive technique based on nuclear techniques. Neutron activation analysis is a well established nuclear technique, which is ideally suited to investigate the microstructural or elemental composition and can be applied to studies of a large variety of samples. Indeed, it may be used to address the above-mentioned questions. Neutrons are useful as probes for non-destructive examination of extended media because neutrons can travel relatively long distances before interacting with the nuclei of the media. It is for this portable NIPGAT being developed at the Malaysian Nuclear Agency for in-situ quantitative elemental analysis and finally for provenance study of the Terengganu's Inscribed Stone (Abdullah et al., 2004).

Figure 1: Terengganu Inscribed Stone

Figure 2: Terengganu Inscribed Stone displayed at Terengganu State Museum

EXPERIMENTAL AND METHOD

Instead of using thermal neutron from nuclear reactor, isotopic neutron source from californium-252 (Cf-252) with an activity of around 2 mCi has been used in this project. The size of this neutron source is 0.5 cm in diameter and 5 cm in length and its mass is about 4.7 microgram. During irradiation, neutron source is placed inside neutron howitzer which is made of paraffin wax. Size of the howitzer is 40 cm in diameter, 40 cm in height and it has

10 cm deep hole. However for irradiation purpose, the neutron source is arranged to be placed at only 2 cm away from the howitzer opening window. High purity germanium detector with 1.2 liter liquid nitrogen dewar was used to detect prompt gamma that emit from the elements inside the sample. Irradiation time is set at one hour. Large dolerite and granite stone samples which are around 40 kg to 50 kg have been collected from three major areas Figure 3. The areas are Panchor, Lawit and Pangsun. The stone sample then been irradiated in such arrangement of detector and neutron source as shown in Figure 4. The distance between detector and source is 15 cm in 90 degrees arrangement. Lead sheet of 6 mm thickness is placed in front of the detector and source collimator window.

Figure 3: Some of collected dolerite and granite stone samples

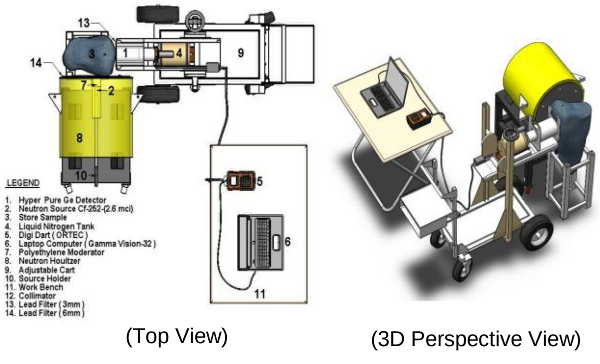

Figure 4: Arrangement of detector and neutron source inside howitzer in 90 degrees

The study is divided into three stages. First stage is carrying out several expeditions to collect samples and making geological observations at selected areas in peninsular Malaysia. In the second stage NIPGAT system is designed, developed and tested in the laboratory and then in the third stage, the system has been taken out from the laboratory for the use in field.

Sampling and Geological Observatory

- Observations at location of Batu Bersurat Terengganu discovery

The study began with the observation of external physical of Batu Bersurat Terengganu in Terengganu State Museum. Then, it is followed by a visit to the location where the Batu Bersurat Terengganu was found, in Alur Tara, Kampung Buloh, Kuala Berang. The discovery of ancient graves in the vicinity show that the tombstones in the cemetery has the same type of rock if compare to Batu Bersurat Terengganu. According to sources of the villagers, this tombstone comes from Panchor area. Indirectly, it gives an early indication of the original location of Batu Bersurat Terengganu.

- Observations at Panchor

The area is located about 20 km to the west of Kampung Buluh and is close to the entrance area of the Kenyir Dam. Tributaries that form the waterfall flowing on the east side of the road and are connected to the main groove of Sungai Hulu Terengganu. Kenyir Dam, completed in 1986, is located only about 300 m from the waterfall area. Around the cliffs and ravines there are many granite rock pile and circular dolerite boulders in various sizes.

- Observations at Hulu Sungai Lawit

In this area, dolerite boulders which located on the ground are mossy and have peeling structure. This indirectly suggests that Batu Bersurat Terengganu is not taken on the ground but rather taken from the rivers where the surface is smooth and clean. The interesting discovery is that when a boulder of dolerite which is really identical with Batu Bersurat Terengganu was found. It was same type of rock, dark gray, tabular-shaped with four faces relatively flat, tapered at the bottom and wide at the top (Figure 5).

Figure 5: Dolerite boulder that looks identical to Batu Bersurat Terengganu with smooth surface

- Observation at the Sungai Terengganu

Observations were start from Kuala Terengganu and ends at the Panchor waterfall near the Kenyir Dam, which is about 55 km from Kuala Terengganu. Along the river, sampling was done at certain places. From the observation, it was found that the samples do not have the characteristics of dolerite. Among the rock types found are from sedimentary rock, granite and igneous rocks.

- Observations at Pangsun

Pangsun Valley is one of the areas in Hulu Langat, Selangor. It is an area of streams that flow from Nuang Mountain. Along the path of the valley, there are a lot of dolerite pile and granite. Dolerite grained at this place is quite coarse and it is clearly visible. Most of Granite boulder which is found at this area is weighing between 30 kg and 50 kg, and they are chunk of debris from the explosion process. Most likely this is not locally granite but rather taken from other areas to reduce erosion on the banks of the groove in this area.

Sample Irradiation at Malaysian Nuclear Agency

Rock samples were collected from three selected areas namely Panchor and Lawit (Kuala Berang, Terengganu) and Pangsun (Hulu Langat, Selangor). The sample is then irradiated with a neutron source where the distance between the neutron source and radiation detectors set at 15 cm in orientation 90^o (Figure 6).

Figure 6: In-situ irradiation of samples at Malaysian Nuclear Agency

In-situ Irradiation Work

- Irradiation at Hulu Sungai Lawit, Tasik Kenyir

NIPGAT system was brought to Hulu Sungai Lawit, Tasik Kenyir. This area was chosen because a few chunks of boulder that is identical to the Batu Bersurat Terengganu were found earlier. Figure 7 shows the in-situ irradiation of dolerite rock samples that have been carried out in Hulu Sungai Lawit.

Figure 7: The process for in-situ irradiation of dolerite rock samples in Hulu Sungai Lawit, Tasik Kenyir

- Irradiation at the Terengganu State Museum

The results of the observations made in Terengganu Museum found that Batu Bersurat Terengganu composed of fine-grained igneous rock that is homogeneous and dark gray-green in colour. It gives the initial impression that the stone are dolerite. Batu Bersurat Terengganu is circular with an external surface and edges are relatively smooth and are consistent with the geological interpretation that Batu Bersurat Terengganu is made of river rock. Figure 8 shows the irradiation towards on original Batu Bersurat Terengganu at Terengganu State Museum.

Figure 8: Irradiation is carried out on Batu Bersurat Terengganu in Terengganu State Museum

RESULTS AND DISCUSSION

Surface of rock samples collected are cleaned without using any special treatment. This is to maintain the original sample. During irradiation with neutrons, gamma ray spectrums are recorded. From these data, the peak energy for elements such as hydrogen (H), aluminum (Al), silicon (Si), sodium (Na), titanium (Ti) and Iron (Fe) can be clearly detected. Other elements can also be detected, however not taken into account in this experiment because its error is too large compared with the dominant elements. In all measurements using NIPGAT, gamma ray of Al is the dominant compared with other elements which it gave the highest reading on the graph. However, the Al content in the sample cannot be compared directly because the large number of gamma ray is likely to influence by collimator on HPGe detector which is also made with Al. All results were recorded by taking into account the average result of six measurements for each sample. This provides more consistent results with high accuracy and can represent the entire volume of the sample.

Comparison between Inscribed Stone with Granite

Graph profile (Figure 9) for the granite rock samples obtained from Panchor and Kenyir Dam is very different from the Inscribed Stone. The total number of gamma rays for Si (3539 keV) and Si (4934 keV) for the granite rock is higher (2 times) than the inscribed stone, which shows that the Si content is generally higher in granite. However, the total number of prompt gamma ray for the Ti (6418 keV) and Ti (6760 keV) for the granite rock is very low compared to Inscribed Stone and not be detected directly. Two elements are compared for, Si is the highest element (72%) and Ti is the lowest element content (0.3%) in the granite rock. Significant differences can be seen if these granite rocks compared to the inscribed stone. The total number of gamma rays to Fe (7631 keV) and Fe (7645 keV) for these granite rocks were also low when compared with the inscribed stone. This difference clearly indicates that the granite rock is not from the same rock type of the inscribed stone. This strongly supports the conclusion made in previous geological studies (Jamaluddin et al., 2010) in denying that this inscribed stone is made of granite as indicated in the previous study (Peterson, 1924).

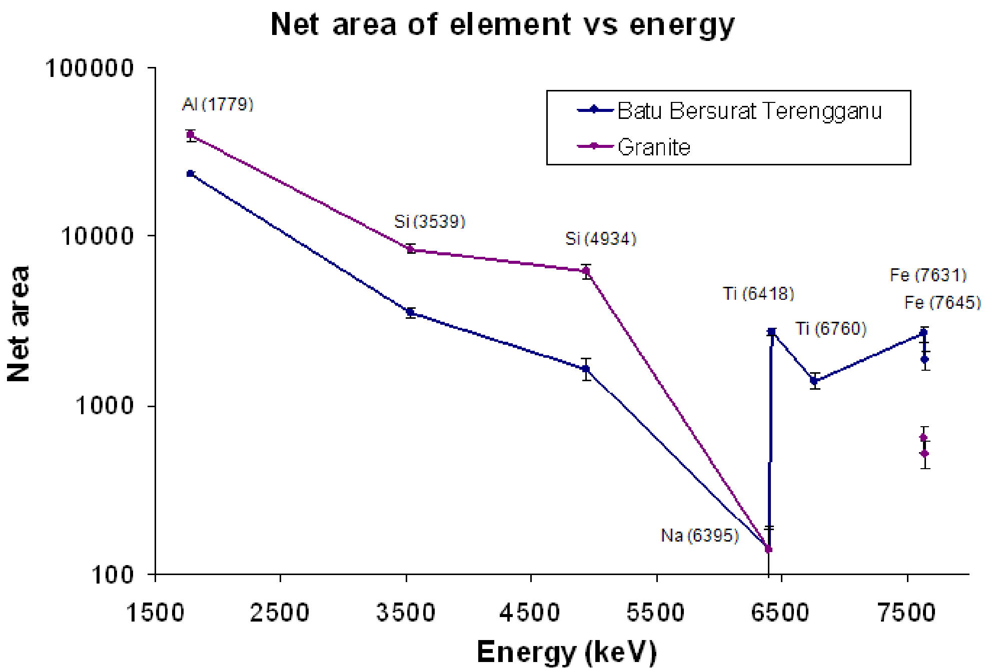

Figure 9: Comparison of prompt gamma between the inscribed stone with granite

Comparison of the Inscribed Stone with Dolerite

In theory, the number of gamma rays induced by fast neutrons is dependent on water content (moisture) of a sample, because the hydrogen atoms in water molecules are the main agents that can slow down the high-energy neutrons to low energy neutron or thermal neutron (Alfassi and Chung, 1995; Khelifi et al., 1999). Thermal neutron is great potential for activating element in the sample to produce prompt gamma. Profile of the graph (Figure 10) for dolerite rock samples from the upper river of Lawit and Panchor is similar to the inscribed

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stone. However, the total prompt gamma ray for Si (3539 keV), Si (4934 keV), Na (6395 keV), Ti (6418 keV), Ti (6760 keV), Fe (7631 keV) and Fe (7645 keV) is slightly higher compared to readings obtained from the inscribed stone. This is because the inscribed stone has been exposed to a dry environment where it is placed in a controlled area air-conditioned and illuminated with infrared light, resulting in lower moisture content. When the hydrogen content is low, the number of elements can be activated is less due to the lack of thermal neutrons present in the samples. This situation is very different from the dolerite rock samples obtained from the aqueous environment of the area exposed to the rain, which typically have high moisture content. This answered the question why the Si content is low in the inscribed stone over dolerite derived from the upper river of Lawit and Panchor.

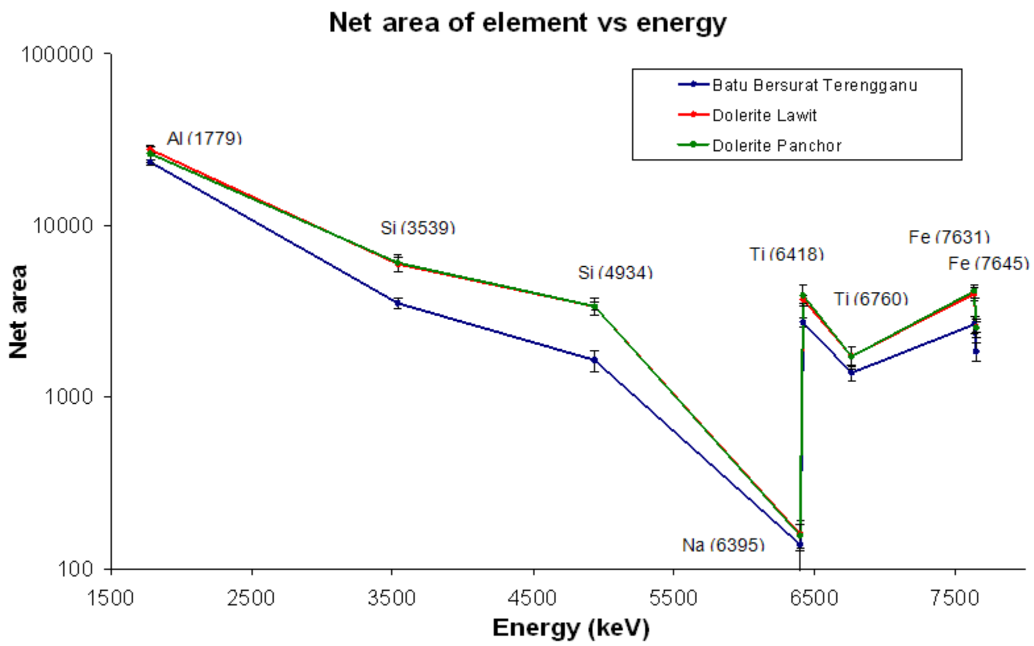

Figure 10: Comparison between prompt gamma from the Inscribed Stone with the Panchor and Lawit dolerite

According to the geological study, dolerite rocks can also be found in several areas in peninsular Malaysia, including at the Pangsun in Selangor and Lembah Bujang at Kedah (Schwartz, 1995). To support the results obtained, a number of dolerite samples from outside the Hulu Terengganu were analyzed using NIPGAT system. For this purpose, several samples of dolerite from Pangsun have been taken for comparison in terms of the composition of the dominant element. Results showed that samples obtained from Pangsun has almost a same number of prompt gamma rays from Hulu Sungai Lawit and Panchor (Figure 11) for all the dominant elements of Si (3539 keV), Si (4934 keV), Na (6395 keV), Fe (7631 keV) and Fe (7645 keV), except for the elements Ti (6418 keV) and Ti (6760 keV), which is very different from the inscribed stone. These results can conclude that the composition of dolerite elements is different between the one at Terengganu and Selangor.

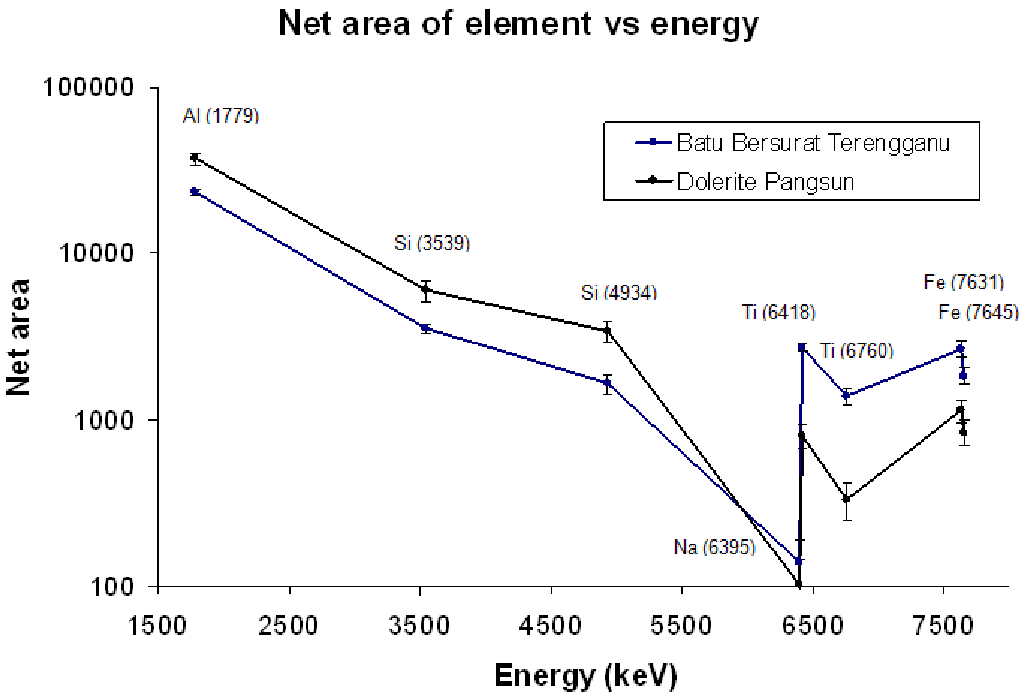

Figure 11: Comparison between prompt gamma from the inscribed stone with Pangsun dolerite

Comparison between the Inscribed Stone with All Samples

Referring to Figure 12, the total prompt gamma ray of inscribed stone for Si (3539 keV) and Si (4934 keV) is almost the same for dolerite rock samples from the Panchor, Lawit and Pangsun. This clearly shows that they belong to the same family as the Si is the dominant element in the composition of these rocks. However, for the prompt gamma ray of Ti (6418 keV), Ti (6760 keV) and Na (6395 keV), there are different from dolerite samples in Pangsun compare to Lawit and Panchor. Ti and Na is a minor element in the dolerite rocks and its contents are quite different from one area to another. This is visible on the graph profile in Figure 12. If the moisture in all samples is equal, then the graph of Figure 12 for the inscribed stone should have a profile that most closely matches the graph profile of the Dolerite from Panchor and Lawit.

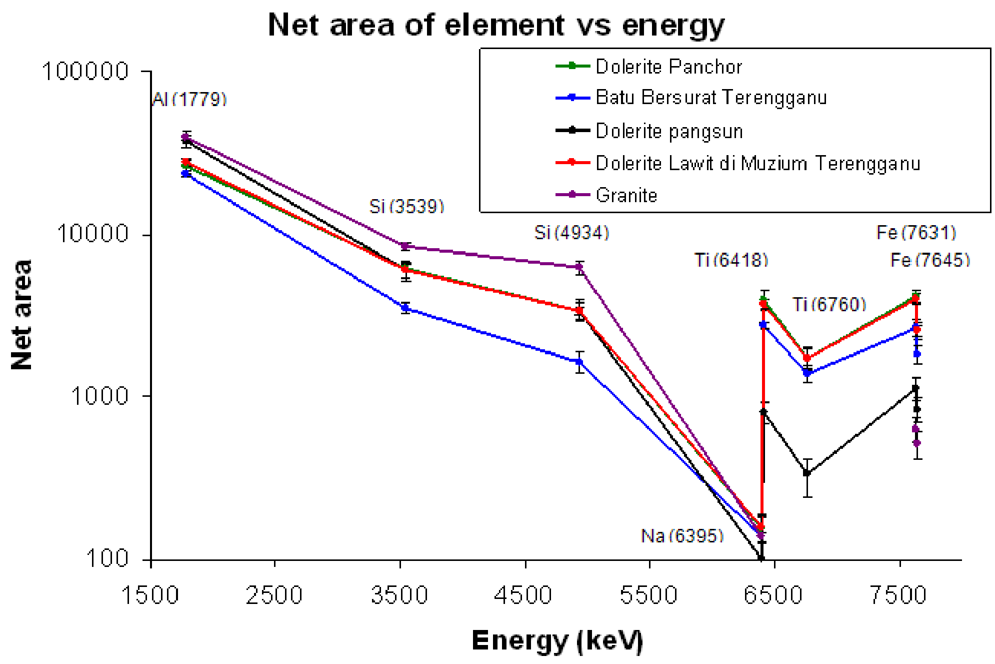

Figure 12: Comparison between prompt gamma of the inscribed stone with all dolerite samples from Panchor, Lawit, and Pangsun together with granite

CONCLUSION

Based on these results, a portable system based on neutron-induced prompt gamma-ray technique has been successfully developed. This will allow for the stone to be investigated non-destructively in a volumetric manner on site. For now, inscribed stone seems similar to dolerite stone from Lawit and Panchor. More samples from other places were needed to determine the provenance of this inscribed stone. The inscribed stone is not granite as reported in the history and it is more similar to dolerite stone. The scientific data or technical evidence obtained from this project will be of great benefits not only to curators and archaeologists but also to scientists, historians and many others in the future.

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