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REE and Th potential from placer deposits: a reconnaissance study of monazite and xenotime from Jerai pluton, Kedah, Malaysia

Fakhruddin Afif Fauzi[1*], Arda Anasha Jamil[2], Abdul Hadi Abdul Rahman[3], Mahat Hj. Sibon[3], Mohamad Sari Hasan[4], Muhammad Falah Zahri[5], Hamdan Ariffin[1], Abdullah Sulaiman[1]

[1]*Department of Mineral and Geoscience Kedah / Perlis / Pulau Pinang, Alor Setar, Malaysia*
[2]*Department of Mineral and Geoscience Johor, Johor Bahru, Malaysia*
[3]*Department of Mineral and Geoscience Selangor / W.P., Shah Alam, Malaysia*
[4]*Department of Mineral and Geoscience Pahang, Kuantan, Malaysia*
[5]*Technical Service Division, Department of Mineral and Geoscience, Ipoh, Malaysia*

*Corresponding author: afif@jmg.gov.my

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Abstract

Thorium (Th), rare earth elements (REE), yttrium (Y) and scandium (Sc) are among crucial elements in minerals that have a very high worldwide demand for green energy generation and high technology manufacturing industries. The current principal ore minerals for these elements are monazite, bastnäsite and xenotime. A reconnaissance study on monazite and xenotime minerals was conducted in southern part of Jerai peak area, which consists of mostly pegmatites and granite bedrocks and alluvial plains. Heavy mineral concentrate samples were obtained from various origins including gravelly layer from stream banks, flowing stream beds and seasonal stream beds for recent fluvial environment. Samples were also taken from weathered bedrocks of pegmatites and granites and different subsurface profiles from 12 pits dug in the alluvial plain area. Monazite and xenotime contents from stream bed samples are higher (8.43% and 6.05%) compared to other origins in recent fluvial environment and higher in weathered pegmatites (13.70% and 1.45%) compared to weathered granites. The monazite and xenotime content are also higher in eastern side of the alluvial plain, up to 3.16% and 2.91% respectively, but lower than samples from recent fluvial environment. The Th, REE and Y contents are very high up to 1,530 ppm, 21,031 ppm and 7,604 ppm respectively in samples containing monazite and/or xenotime. The Sc content, however, is very low which is up to 87.8 ppm in all samples although it shows positive correlation with monazite and/or xenotime contents. Both REE containing minerals could be economically potential if mined as placer suites together with garnet, tourmaline and other industrially beneficial minerals.

Keywords: Jerai, Kedah, monazite, REE, thorium, xenotime

1. Introduction

1.1 Background

The importance of rare earth elements (REE), thorium (Th), yttrium (Y) and scandium (Sc) has become worldwide emergence in electronics and high technology industries. REE, Y and Sc are crucial for production of magnets in computer drives and defence applications, metal alloys including batteries and superalloys, phosphors such as LED and optical sensors, additive in ceramics and glass polishing and also used as catalyst in various chemical processes (Jha, 2014). Th, as ThO_2 is well-known for better energy generating purpose than uranium (U) as the former does not easily oxidized and resistant to ionic radiation (IAEA, 2005).

REE is defined as a set of 17 chemical elements in the periodic table, comprising 15 lanthanides which are cerium (Ce), dysprosium (Dy), erbium (Er), europium (Eu), gadolinium (Gd), holmium (Ho), lanthanum (La), lutetium (Lu), neodymium (Nd), praseodymium (Pr), promethium (Pm), samarium (Sm), terbium (Tb), thulium (Tm) and ytterbium (Yb), as well as yttrium (Y) and scandium (Sc) (Connelly and Damhus, 2005).

The history of REE dated back in 1788 when Johan Gadolin discovered a rare pitch--black rock in Ytterby, Sweden. The rock samples were taken across European countries where their scientists competed to distinguish elements that exist as well as to complete the Lanthanide Series of Periodic Table of Elements. In fact, the names of yttrium, erbium, terbium and ytterbium derived from the origin of the sample. Th, on the other hand, was discovered by Jöns Jacob Berzelius, a Swedish scientist in 1828.

Placers are defined as mineral deposits formed by the mechanical concentration of minerals from weathered debris, such as beaches and streams, by which the economic mineral deposits have high density but are very resistant to chemical and physical breakdown (Sengupta and Van Gosen, 2016). Among these minerals are monazite, xenotime, bastnäsite and loparite which are considered as the most important placer rare earth minerals (REM) in the world (Zhou et al., 2017). The former 2 minerals are common by-products of alluvial tin mining in Malay Peninsular since early 1900s (Willbourn, 1925). The placer REM are practically mined together with other minerals including garnet, zircon, cassiterite, and Ti-bearing minerals like rutile and ilmenite as mineral suite or co-products before separation processes (Sengupta and Van Gosen, 2016).

Placer REM currently represent the third most important global REE source of production after Bayan Obo carbonate rocks in inner Mongolia and Mountain Pass carbonatites in California, which come from the monazite and xenotime dispersed in Neogene to Quaternary beach sand in Australia (McLennan and Taylor, 2012). In comparison, current and previous productive placer deposits contain 6% to 7% of heavy minerals including REM as reported in Eneabba district, Australia and Xun Jiang district, China (Shepherd, 1990; Jackson and Christiansen, 1993).

In 1980s, the xenotime-bearing alluvial placer deposits in Malaysia were once the largest source of Y in the world (Castor and Hendrick, 2006). JMG (2019) stated that the production of both monazite and xenotime in Malaysia increased from 25 tonnes in 2009 to 1,654 tonnes in 2018, which were obtained from alluvial tailings in Ipoh, Perak. USGS (2019) estimated that Malaysia has 30,000 tonnes of rare earth oxides (REO) in 2018.

Monazite is a phosphate mineral consisting REE (Ce, La, Nd), Th and U. Monazite is a common accessory mineral in peraluminous granites, syenitic and granitic pegmatites, quartz veins and carbonatites but lesser in charnockites, migmatites and paragneisses (Rapp and Watson, 1986). In peraluminous granites, monazite constitutes a major host of LREE, excluding Eu, Th and U, with minor amount of Y and HREE (Hinton and Paterson, 1994; Bea et al., 1994; Bea, 1996). The monazite stability in silicate melts depends on SiO_2, CaO and P_2O_5, including oxygen fugacity, peraluminous content and content ratios of lanthanides and actinides (Cuney and Friedrich, 1987; Casilas et al., 1995). Felsic differentiation towards granite plutons strongly depletes LREE and Th due to monazite fractionation (Ward et al., 1992; Wark and Miller, 1993; Zhao and Cooper, 1993). According to Che Zainol Bahri et al. (2018), the Th content in Malaysian monazite ranges between 2,525 ppm to 40,868 ppm while Willbourn (1925) mentioned that the mineral contains 3.5% to 8.38% of ThO_2. *Atomic Energy Licencing Act 1984* stated that if radionuclide of Naturally Occurring Radioactive Materials (NORM) of Th-232 exceeds 1 Bq/g, it is considered as radioactive. 1 Bq/g of Th-232 is equivalent to 246 ppm, assuming the chain is in equilibrium.

Xenotime is also a phosphate mineral but abundant particularly in Ca-poor peraluminous granites, which accounts for huge fraction of Y and HREE contents and variable portion of substituted U (Wark and Miller, 1993; Bea, 1996). In xenotime-bearing peraluminous granites, the Y and HREE fractions contained in xenotime vary from 30% to 50%, that is closely related to xenotime-apatite-zircon concomitance during plutonic facies differentiation (Bea, 1996; Wark and Miller, 1993; Förster and Tischendorf, 1994). Xenotime also contains minor amounts of Th and LREE, particularly Nd and Sm (Förster, 1998b).

Hence, this conducted study is to determine the potential of Th and REE based on monazite and xenotime distributions in placer environments within Jerai Pluton area.

1.2 Previous studies

Studies on the occurrences of monazite and xenotime in Malaysia were commenced as parts of regional mapping and regional geochemical surveys by Geological Survey of Malaysia since 1925, continued by Department of Mineral and Geoscience Malaysia (JMG) from 2000 until present.

The Malaysian monazite generally has moderately well-rounded discrete or as some parts with colour varies from clear, deep canary yellow through cream coloured with resinous lustre due to progressive oxidation of REE (Flinter et al., 1963; Wan Hassan, 1989). The Jerai monazite, however, has unusual elongated rolled grains of flattened form with deep green colour (Wan Hassan, 1989; Flinter et al., 1963). Wan Hassan (1989) distinguished the xenotime crystal habit to have squat tetragonal bipyramid in Malaysian Tin Belt and prism with double pyramidal terminations in Malaysian Eastern Belt.

Studies on radioactive minerals on Malaysian Central Belt then were conducted in 1993 to 1995 (Mohd Hasan et al. 1993, 1995; Zakaria et al. 1994). Academy of Sciences Malaysia (ASM) later initiated a study on REE associated with ion adsorption clay in 2013 (ASM, 2014).

In 2018, JMG conducted a reconnaissance study on REE, Th and Sc potentials in Malaysia. The preliminary study comprised their potential in clayey horizons in weathered granites and placer deposits, including Jerai Pluton (Abdul Rahman et al. 2018a, b). Hence, this paper is aimed to deliver the outcomes of the study commenced in the pluton area.

1.3 Study area

The study area is situated at the south of Mount Jerai, west of Kedah state with a total of 40 km2. The area is mostly covered with paddy fields with freshwater for drainage sourced from streams flowing from the peak. The study area lies in the National Jerai Geopark (Fig.1).

2. Geological Setting

2.1 Regional geology

The Jerai Pluton lies within the western tin granites of Peninsular which is part of the Southeast Asian Magmatic Arc that was triggered during Late Permian to Triassic subduction-collision event due to the closure of Palaeo-Tethyan Ocean beneath the Southeast Asian crust (Robb, 2019) (Fig.1).

The tin granites of western Peninsular Malaysia have been considered as products of partial melting of the metamorphic basement during the collision of Sibumasu and East Malaya blocks (Liu et al., 2020; Ng et al., 2015), based on their ilmenite-series, peraluminous character (Ishihara et al., 1979), and characteristic of S-type granites (Chappell and White, 2001). More recently, the genetic link between the tin mineralization and the granites has been constrained, by which the age of tin ores and their granitic hosted rocks in the western Peninsular Malaysia is within the range of the 230 to 210 Ma (Yang et al., 2020).

2.2 Local geology

The study area consists of granitic bedrock, metasedimentary bedrocks and unconsolidated deposits.

The granitic bedrock covered the northern part of the study area. It is from the Jerai Pluton that intruded during Late Triassic and shaped the mountain (Fig.1). Jamil et al. (2006) divided the pluton into 3 facies according to mineralogical criteria, which are biotite – muscovite granite, tourmaline granite and pegmatite. The pegmatite facies which occurred as veins ranging from few centimetres to several meters, are the primary source of tin associated with Nb-Ta (Bradford, 1972). Khoo (1977) proposed that the pegmatites also occurred as sync-plutonic dykes that filled the ductile cracks in granites during intrusion. Apart of tourmalines, garnets, particularly pinkish variety, are the common accessory minerals in Jerai Granite found in this study.

The metasedimentary facies within the study area are quartzites and schists from Cambrian Jerai Formation. Both facies are mineralized with magnetite and/or hematite (Bean and Hill, 1969; Bradford, 1972) and regarded as source of iron ores since 535 B.C. (Mokhtar and Saidin, 2018).

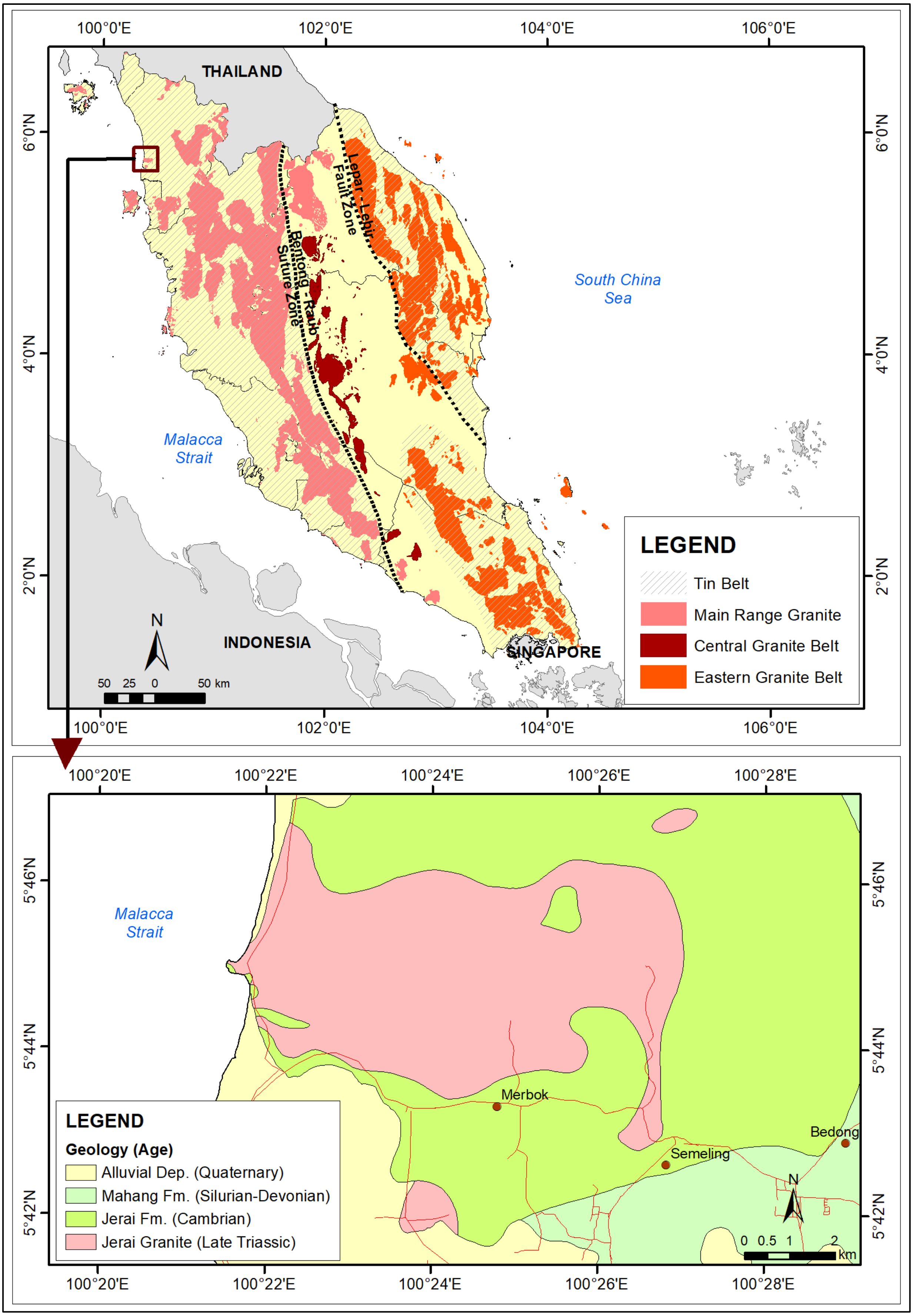

Fig. 1: Maps showing the Granite Belts and tin belts in Peninsular Malaysia (above, after Hosking, 1973) and the general geology of study area (below, after JMG, 2014).

The unconsolidated deposits cover the southern part of the study area. They mostly consist of clay, mud and silt originated from marine influence during Quaternary period (JMG, 2014).

3. Materials and Methodology

Heavy concentrate samples were collected from current and seasonal stream beds, stream banks and gravelly layers on stream banks; weathered granites and pegmatites (Figs. 1, 2, 3, 4 & 5) and distinguishable layers from dug pits with depth between 3.2 metres up to 4.0 metres (Figs. 6 & 7) All sampling locations are shown in Fig. 8.

The samples were obtained through panning using 5 litre sized wooden pan. All samples then underwent removal of light minerals (i.e. SG ≤ 2.85) including quartz using Bromoform solution before thoroughly rinsed with spirit methyl, followed by distilled water. Once dried, the samples were weighted and magnetically separated using hand magnet and Frantz Isodynamic Magnetic Separator Model L-1 into 4 different magnetism; 0.4 Ampere, 0.7 Ampere, 1.0 Ampere and non-magnetic.

Samples of different magnetism were then taken for quantitative mineral estimation (QME) analysis under stereo microscope aided by mineral lists according to magnetism (Table 1) and descriptions by Wan Hassan (1989) and Devismes (1978).

Selected concentrate samples were analysed through LA-ICP-MS method in Laboratory Branch, JMG Technical Service Division in order to determine their REE, Th, Y and Sc contents. The U content is also analysed for all selected samples. Apart from individual content, the REE results are also calculated according to total light REE (TLREE), total heavy REE (THREE) and total REE (TREE).

4. Results

A total of 43 samples including 3 samples from weathered bedrocks and 28 samples from pits were successfully obtained in this study.

4.1 QME analysis

The monazite content in samples from recent fluvial environment (Table 2) is from none to 8.43% and the xenotime content in samples from the same environment is from none to 6.05%. Sample KC56 which originated from stream bed contains the most monazite and xenotime. Samples KC41 and KC59 also contain monazite more than 4% and sample KC41 has xenotime content more than 4%. Garnet, particularly pinkish variety is common in all samples while allanite and zircon exist in few samples. Other minerals identified include mostly magnetite, hematite and tourmaline (Figs. 9 & 10). The average content of monazite and xenotime in recent fluvial environment samples is 5.14% and 2.82% respectively, with an average sum of 7.96%.

While for weathered bedrock samples, the highest monazite and xenotime content is in pegmatite, which is 13.70% and 1.45% respectively, compared to weathered granite.

In general, sandy layers in pits have considerably greater amount of heavy concentrate minerals compared to silty and clayey layers. The monazite and xenotime content in samples from pits (Table 3) is ranging from none to 3.16% and none to 2.91%, respectively. The highest content of both monazite and xenotime originated from pit C but in different layers, by which the deeper layer (KCC003) has higher content of monazite 3.16% while shallower layer (KCC001) has greater content of xenotime (3.91%). Similar to samples from recent fluvial environments, pinkish garnet is also common in all samples while allanite and zircon appear in few samples. Other minerals identified are magnetite, hematite, tourmaline and ilmenite. No mineral concentrates exist from upper layer of pit I (KIC001) and pit K as these pits mainly consist of greyish mud – clay with organic materials. The average content of monazite and xenotime in Quaternary deposit environment samples is only 1.66% and 1.10% respectively, with an average sum of 2.76%.

4.2 LA-ICP-MS analysis

A total of 10 concentrate samples from recent fluvial environment were analysed by LA-ICP-MS (Table 4). The Th content of the

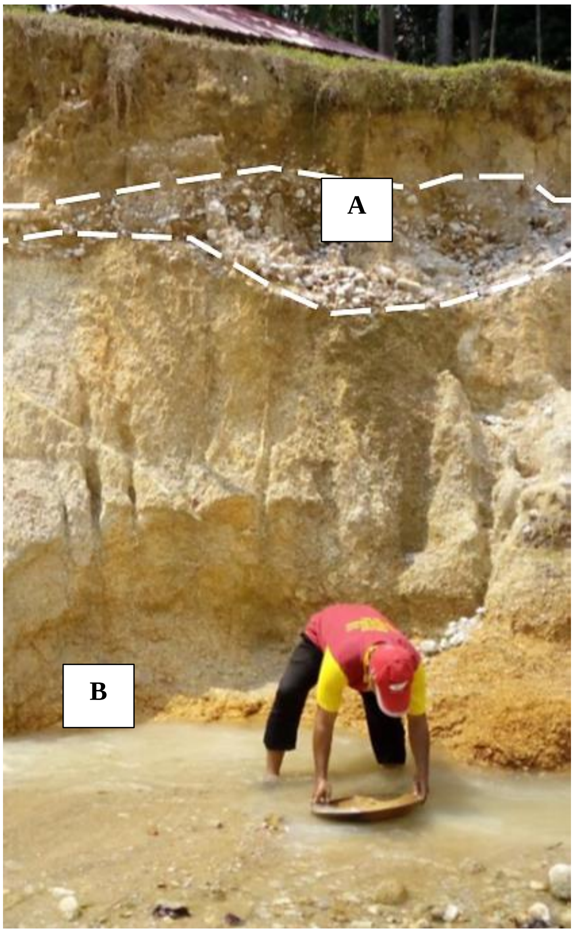

Fig. 2: Origins of the samples in recent fluvial environment include gravelly layer (A) and stream bed (B).

Fig. 3: The occurrence of eluvial bed near S. Batu Pahat.

Fig. 4: Sampling of weathered bedrock.

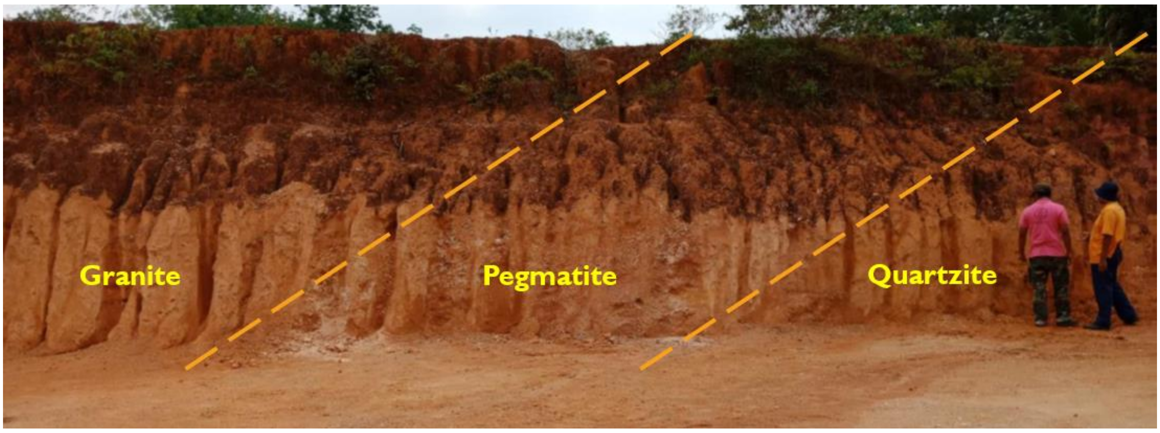

Fig. 5: Outcrops of weathered granite and pegmatite bedrocks from Jerai Pluton in contact with quartzite of Jerai Formation.

Fig. 6: Pit is dug using JCB excavator.

Fig. 7: One of the dug pits.

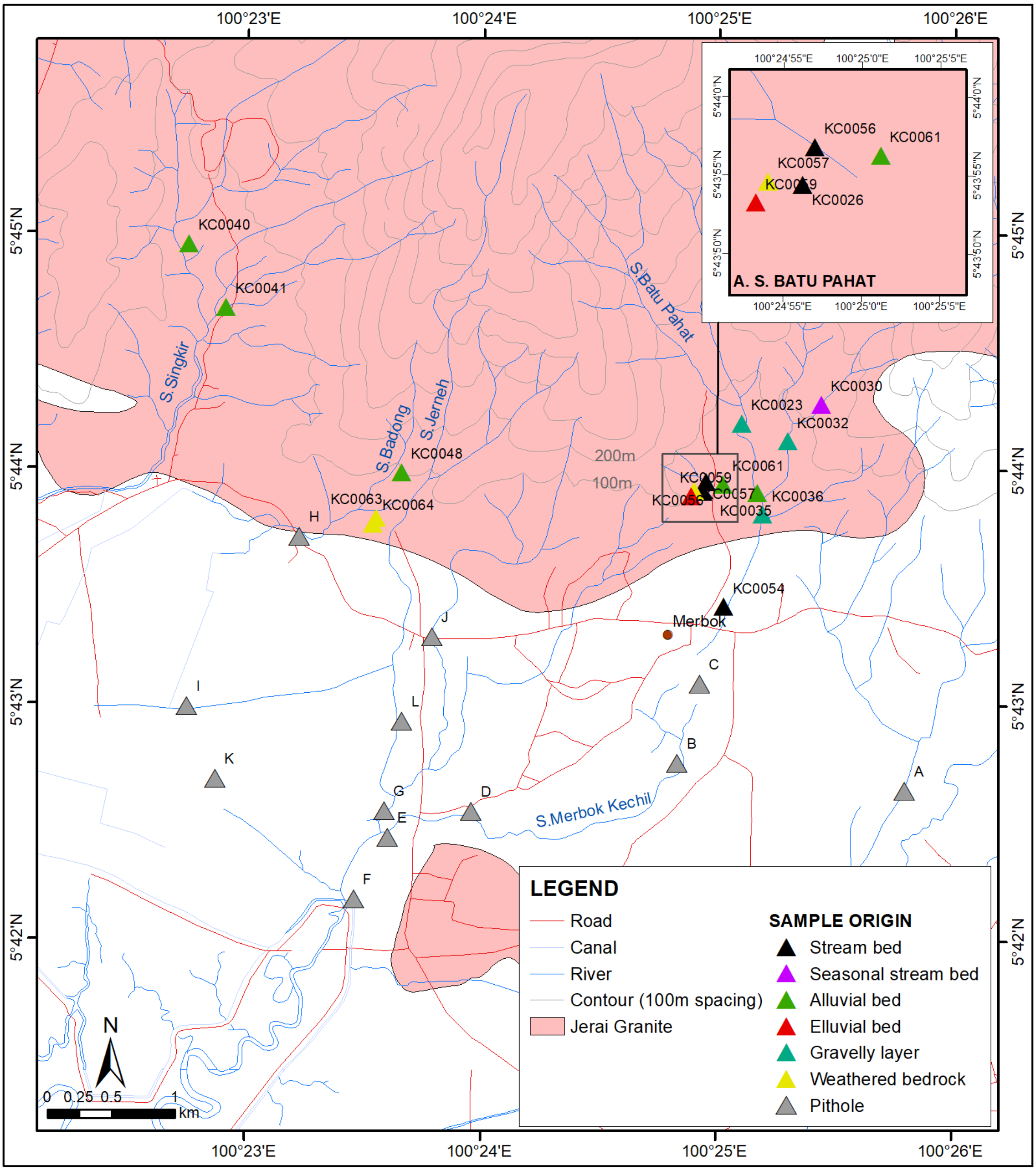

Fig. 8: Sampling locations in the study area.

samples ranges from 74.6 ppm to 1,530.0 ppm. The U content ranges from 48.5 ppm to 1,553.0 ppm. The Sc content ranges from 16.5 ppm to 87.8 ppm while the Y content ranges from 266.0 ppm to 7,604.0 ppm.

The TREE content ranges from 928 ppm to21,031 ppm. The THREE content ranges from 411 ppm to 11,435 while ther TLREE content ranges from 517 ppm to 9,596 ppm All samples have higher TLREE than THREE content except KC56 and KC59.

Sample KC56 has the highest content of U, Y, La, Pr, Nd, Sm, Gd, Tb, Dy, Ho, Er, Tm, Yb and Lu and also TREE, HREE and LREE. Sample KC41 has the greatest content of Th and Ce while sample KC59 has the greatest content of Sc. Sample KC23 has the lowest content of Th, U, Y, TREE, THREE and TLREE.

4.3 Statistical relationships

Comparisons of QME and LA-ICP-MS results

Table 1: List of common minerals separated using Bromoform, hand magnet and magnetic separator according to magnetism (Che Harun et al. 2009).

Light minerals (separated with Bromoform)	Magnetic (hand magnet)	Magnetic Separator (Frantz Isodynamic Model L-1			
		0.4 Ampere	0.7 Ampere	1.0 Ampere	Non-Magnetic
Quartz	Magnetite	Ilmenite	Ilmenite	Tourmaline	Pyrite
	Hematite	Iron Oxide	Tourmaline	Hydroilmenite	Quartz
	Iron Oxide	Garnet	Staurolite	Monazite	Cassiterite
	Pyrrhotite	Siderite	Pyrite	Pyrite	Rutile
			Epidote	Quartz	Leucoxene
			Garnet	Epidote	Corundum
			Hydroilmenite	Staurolite	Tourmaline
			Xenotime	Allanite	Topaz
			Seiderite	Columbite	Zircon
			Allanite	Garnet	Gold
			Columbite	Xenotime	Anatase
			Rutile	Rutile	
			Wolframite	Struverite	

are presented as crossplots of selected elements and minerals in Fig. 11. They show no perfect linear relationship (R2 > 0.95) between the element and mineral content. However, Th and REE have generally strong, positive relationship with monazite. Similar positive histogram patterns for Ce-Nd-La vs monazite are observed. The Sm vs monazite histogram pattern is similar to the REE vs monazite histogram pattern.

The Y element has strong, positive relationship with both monazite and xenotime. Similar pattern of Y vs xenotime histogram is shown by the Yb vs xenotime histogram.

The Sc element, however, has better positive relationship with monazite compared to garnet while U has strong relationship with monazite.

5. Discussions

5.1 Monazite and xenotime magnetism

In this study, the Jerai monazite is not only observed in 0.7-Amp fraction but also appeared but less in 1.0-Amp fraction. This is acceptable as monazite mineral is always slightly magnetised compared to quartz but very much weaker than hematite (Willbourn, 1925; Abaka-Wood et al., 2016).

The xenotime mineral observed in both 0.4- and 0.7-Amp fractions are in concordance with the mineral magnetism guide (Table 1) and the fact that it has slightly higher magnetism than ilmenites (Kim and Jeong, 2019).

5.2 Monazite and xenotime distribution

The higher content of monazite and xenotime in weathered pegmatite compared to weathered granites indicates that pegmatite facies in Jerai Pluton is the primary source of both rare earth minerals.

The humid, tropical climate with high precipitation allows granitic bedrock to decompose to kaolinite and lateritic soils. This permits highly resistant minerals including monazite and xenotime to be transported and deposited either into eluvial environments and nearby river systems before reaching alluvial plain further downstream to become placer deposits (Ghani et al., 2019). The occurrences of pegmatites close or at the upstream of sampling points in recent fluvial environment could be the main factor of different content of the minerals regardless of the sample origin. This is shown by the fact that samples KC56 and KC41, which were taken from stream bed and alluvial bed respectively, have relatively higher monazite and xenotime contents as both sampling points are in the same S. Batu Pahat basin while pegmatite dykes occurred at the upstream of the river basin (Bradford, 1972). Sample KC30 which also contains considerable amount of monazite and xenotime could have pegmatite bedrock at

Table 2: Mineral contents (%) in samples obtained from recent fluvial environment. The symbol '-' denotes not detected and 'TR' denotes trace.

Sample No.	K023	KC30	KC32	KC36	KC40	KC41	KC54	KC56	KC59	KC57	KC63	KC64
Origin	Gravelly layer	Seasonal stream	Gravelly layer	Gravelly layer	Alluvial bed	Alluvial bed	Stream bed	Stream bed	Eluvial bed	Weathered pegmatite	Weathered pegmatite	Weathered granite
Allanite	TR	TR	TR	-	-	-	1.38	-	-	-	-	-
Garnet	32.52	6.18	25.25	39.37	23.75	6.83	47.59	40.56	17.06	0.76	5.61	0.06
Monazite	TR	3.30	-	0.96	-	4.34	0.81	8.43	4.82	4.72	13.70	-
Xenotime	-	3.10	-	TR	TR	4.41	TR	6.05	1.76	0.17	1.45	TR
Zircon	-	4.59	0.27	-	-	0.72	TR	-	0.33	-	0.31	TR
Other Minerals	67.48	82.82	74.49	59.67	76.25	83.69	50.22	44.97	76.02	94.36	78.93	99.94
Total Weight Analyzed (g)	4.53	5.66	2.83	2.76	2.92	2.21	2.53	2.87	2.55	2.11	2.89	2.34

Table 3: Mineral contents (%) in samples obtained from Quaternary alluvial environment. The symbol '-' denotes not detected and 'TR' denotes trace.

Pit	A			B				C			D		
Sample No.	KAC 001	KAC 002	KAC 003	KBC 002	KBC 003	KBC 004	KBC 005	KCC 001	KCC 002	KCC 003	KDC 001	KDC 002	KDC 003
Depth (m)	1.2 – 2.1	2.1 – 2.8	2.8 – 3.6	0.4 – 1.0	1.0 – 1.8	1.8 – 2.8	2.8 – 3.6	0.5 – 0.6	1.2 – 2.3	2.7 – 2.7	0.7 – 1.2	1.2 – 2.4	2.5 – 3.4
Allanite	-	-	0.63	TR	-	-	-	-	-	-	-	-	-
Garnet	7.08	5.34	8.61	11.01	7.64	4.85	8.63	61.28	52.14	59.28	4.05	11.57	4.39
Monazite	-	1.35	-	TR	-	TR	TR	1.47	TR	3.16	-	-	TR
Xenotime	-	-	-	-	TR	-	-	2.91	-	0.79	TR	TR	0.83
Zircon	2.67	0.22	1.42	TR	0.27	0.13	-	TR	0.42	-	0.24	-	0.33
Other Minerals	90.25	93.10	89.34	88.99	92.09	95.02	91.37	34.33	47.44	36.78	95.71	88.43	94.45
Total Weight Analysed (g)	3.05	2.97	3.02	2.98	2.75	2.97	3.06	2.78	2.85	3.04	2.96	3.72	3.01

Table 3 (cont.): Mineral contents (%) in samples obtained from Quaternary alluvial environment. The symbol '-' denotes not detected and 'TR' denotes trace.

Pit	E			F		G		H			I	
Sample No.	**KEC 001**	**KEC 002**	**KEC 003**	**KFC 001**	**KFC 002**	**KGC 001**	**KGC 002**	**KHC 001**	**KHC 002**	**KHC 003**	**KIC 001**	**KIC 002**
Depth (m)	0.7 – 1.6	1.7 – 2.4	2.5 – 3.2	1.0 – 1.2	2.7 – 3.2	0.6 – 1.6	1.6 – 3.2	0.6 – 1.0	3.0 – 3.2	3.2 – 3.7	1.6 – 2.5	2.8 – 3.6
Allanite	-	-	-	-	-	-	-	-	-	-	(no concentrate exists)	-
Garnet	3.25	1.36	3.79	9.90	3.09	5.95	9.20	6.00	2.20	6.20		10.08
Monazite	TR	-	0.75	TR	1.62	TR	TR	TR	-	-		TR
Xenotime	1.54	TR	0.75	0.00	TR	-	-	1.14	-	-		0.86
Zircon	0.32	0.36	-	-	-	-	0.71	-	0.43	TR		0.65
Other Minerals	94.88	98.28	94.71	90.10	95.29	94.05	90.08	92.86	97.36	93.80		88.40
Total Weight Analysed (g)	2.95	2.64	3.06	3.09	3.11	2.69	3.08	2.80	2.54	3.03		2.38

Table 3 (cont.): Mineral contents (%) in samples obtained from Quaternary alluvial environment. The symbol '-' denotes not detected and 'TR' denotes trace.

Pit	J		K		L	
Sample No.	**KJC 001**	**KJC 002**	**KKC 001**	**KKC 002**	**KLC 001**	**KLC 002**
Depth (m)	0.7 – 2.1	3.1 – 3.6			0.7 – 2.0	2.0 – 3.3
Allanite	-	-	(no concentrate obtained)	(no concentrate obtainde)	-	-
Garnet	5.97	9.90			5.76	1.06
Monazite	TR	TR			1.61	TR
Xenotime	-	-			TR	TR
Zircon	0.51	-			0.64	0.15
Other Minerals	93.52	90.10			92.00	98.80
Total Weight Analysed (g)	2.84	3.09			2.18	3.03

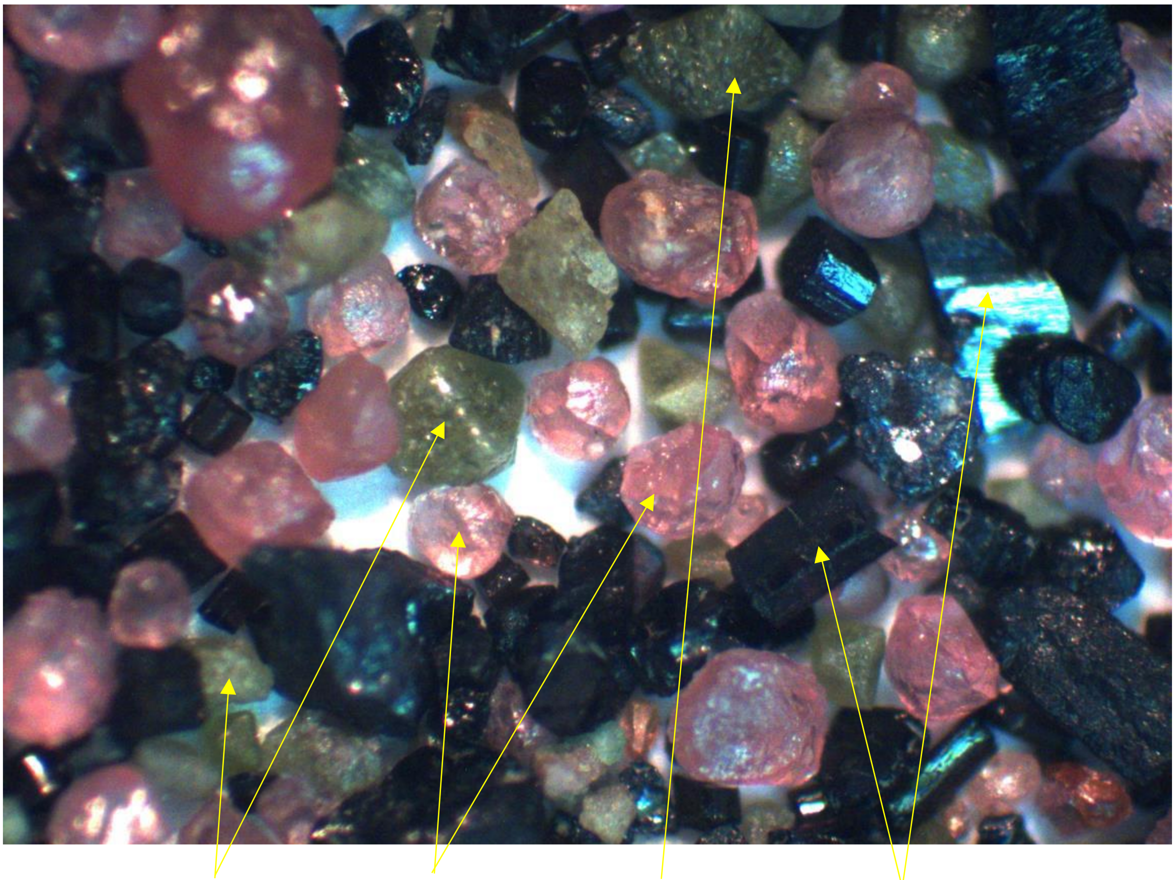

Fig. 9: Minerals observed in 0.7 Amp sample fraction of KC56 under stereo microscope. Total magnification is 40x.

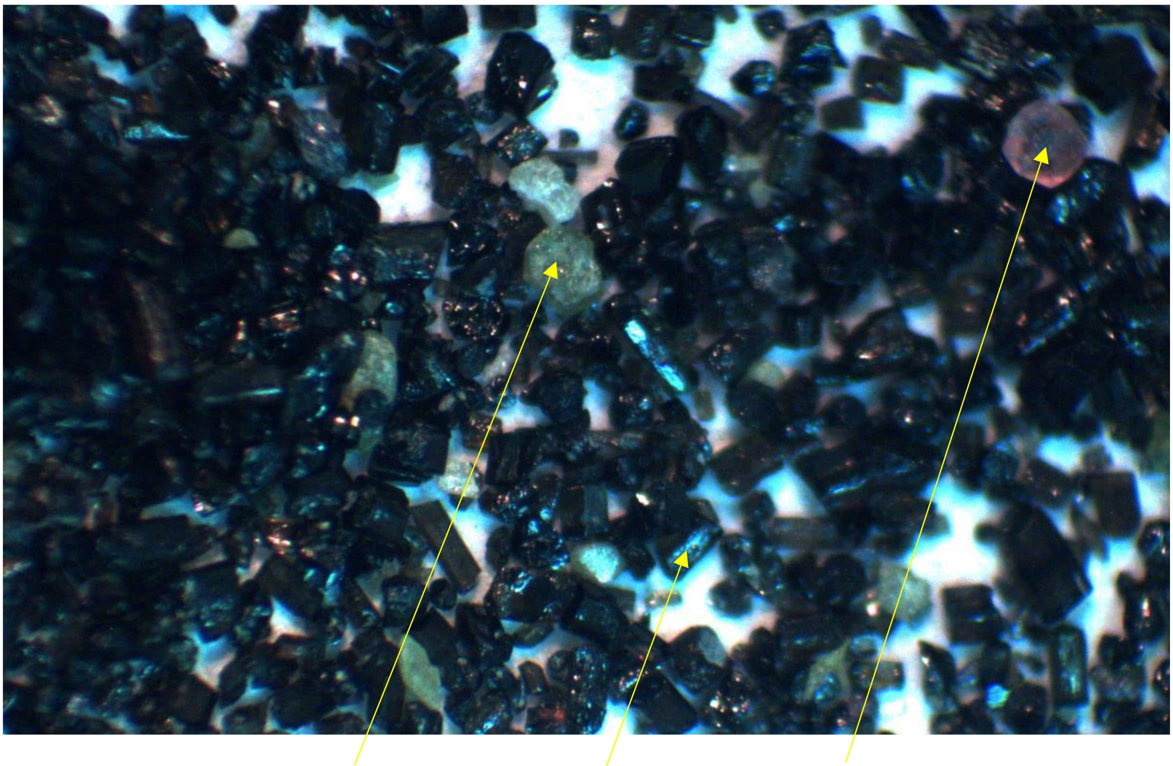

Fig. 10: Minerals observed in 1.0 Amp sample fraction of KCC003 under stereo microscope. Total magnification is 35x.

Table 4: Th, U, Sc, Y and REE content (in ppm) in selected concentrate samples.

Sample No.	Th	U	Sc	Y	LREE						HREE								TREE +Y	THREE +Y	TLREE
					La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu			
KC23	74.6	48.5	22.5	266.0	114.0	255.0	25.6	98.8	23.6	0.3	20.5	4.6	32.9	6.9	23.3	5.2	44.4	7.2	928.4	411.0	517.3
KC30	198.0	255.0	35.2	750.0	312.0	752.0	83.7	297.0	94.1	0.2	81.4	18.1	107.0	19.5	60.0	12.5	102.0	16.5	2706.0	1167.0	1539.0
KC32	210.0	272.0	44.2	1088.0	317.0	750.0	82.0	301.0	97.6	0.8	96.4	23.3	149.0	27.8	83.5	17.8	145.0	23.0	3202.2	1653.8	1548.4
KC36	319.0	201.0	35.7	825.0	441.0	1013.0	112.0	395.0	104.0	2.0	88.1	18.0	112.0	21.8	66.7	13.5	110.0	17.3	3339.5	1272.4	2067.1
KC40	168.0	104.0	16.5	372.0	249.0	565.0	58.9	207.0	47.2	1.0	37.7	7.6	49.3	9.8	30.4	6.2	49.4	7.6	1698.0	569.9	1128.1
KC41	1530.0	639.0	48.9	3650.0	1979.0	4537.0	494.0	1769.0	438.0	10.8	390.0	77.6	482.0	94.8	288.0	55.6	405.0	61.5	14732.3	5504.5	9227.8
KC54	252.0	290.0	48.2	951.0	405.0	905.0	101.0	359.0	99.0	1.9	89.3	20.2	129.0	24.7	76.1	16.5	133.0	20.7	3331.4	1460.5	1870.9
KC56	1383.0	1553.0	70.1	7604.0	2011.0	4531.0	529.0	1911.0	608.0	6.3	644.0	154.0	1014.0	196.0	590.0	124.0	962.0	147.0	21031.3	11435.0	9596.3
KC57	312.0	414.0	36.4	1546.0	497.0	1174.0	133.0	472.0	156.0	0.4	154.0	35.1	221.0	40.3	121.0	24.9	203.0	31.4	4809.1	2376.7	2432.4
KC59	704.0	1097.0	87.8	5214.0	1071.0	2550.0	300.0	1077.0	390.0	1.7	445.0	110.0	717.0	135.0	402.0	85.7	684.0	107.0	13289.4	7899.7	5389.7
Min.	74.6	48.5	16.5	266.0	114.0	255.0	25.6	98.8	23.6	0.2	20.5	4.6	32.9	6.9	23.3	5.2	44.4	7.2	928.4	411.0	517.3
Max.	1530.0	1553.0	87.8	7604.0	2011.0	4537.0	529.0	1911.0	608.0	10.8	644.0	154.0	1014.0	196.0	590.0	124.0	962.0	147.0	21031.3	11435.0	9596.3
Ave.	515.1	487.4	47.7	2226.6	739.6	1703.2	191.9	688.7	205.8	2.5	204.6	46.9	301.3	57.7	174.1	36.2	283.8	43.9	6906.8	3375.1	3531.7

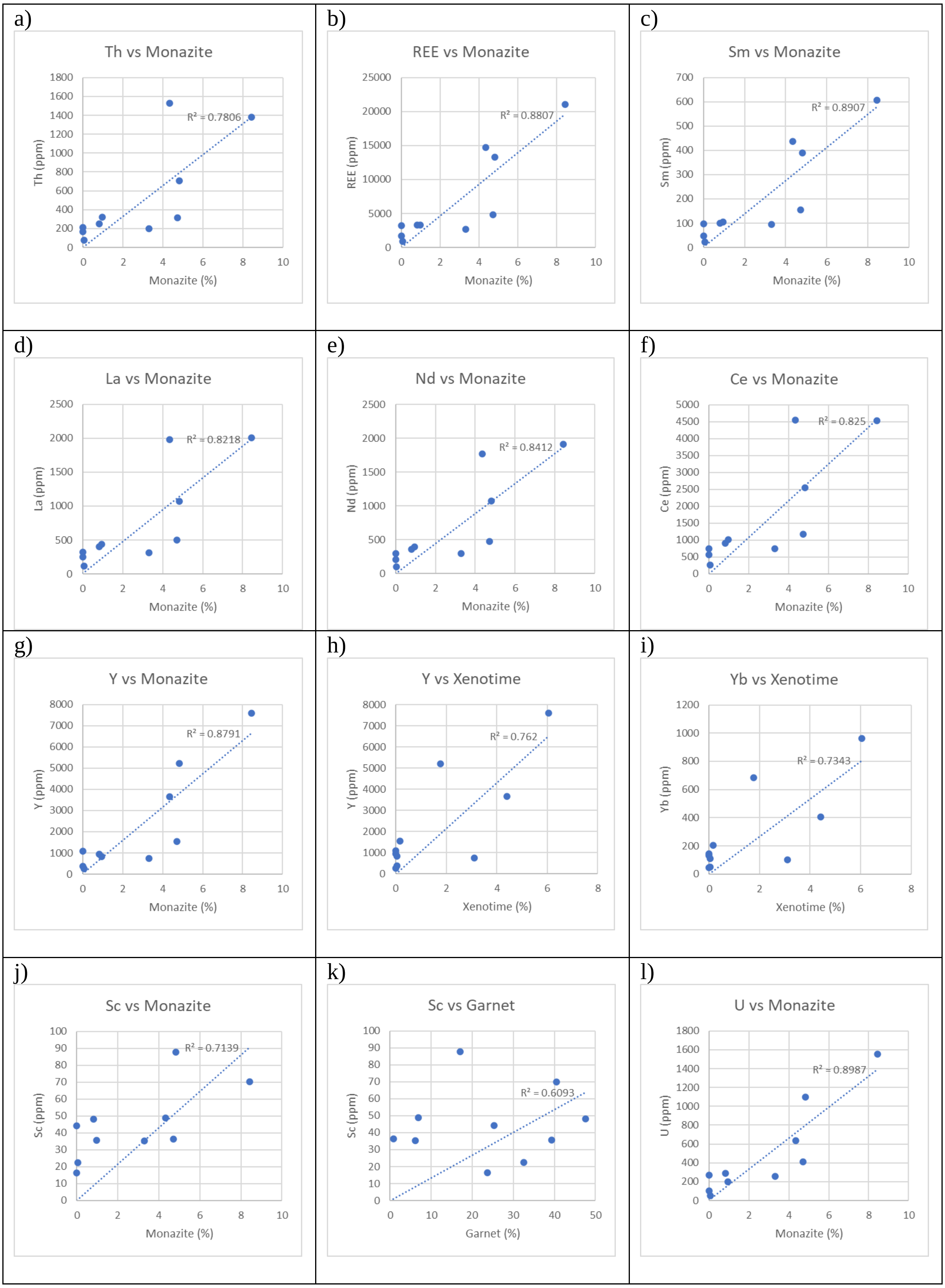

Fig. 11: Plots of selected elements vs minerals with goodness-of-fit values (R^2).

upstream while sample KC54, although collected from stream bed of S.Ayer Jerneh, contains only traces of both minerals. This is due to fact that pegmatites do not occur much at its upstream (Bradford, 1972).

The gravelly layers at recent stream banks are regarded as the paleo stream beds of high energy water flow (Harraz, 2013). However, very low monazite and xenotime content in the layers is either due to dispersion of the minerals during deposition by high energy stream flow or the parent rocks did not completely weather yet to permit both minerals to deposit. In the alluvial plain, the lower content of monazite and xenotime than in the recent fluvial environment is expected as heavy minerals, including both rare earth minerals were widespread when reaching the relatively flat plain area under lower energy regime (Gupta and Krishnamurthy, 2005).

In comparison, the monazite and xenotime contents between sampling points in the alluvial plains show relatively higher at eastern side (downstream S. Batu Pahat) compared to western side (downstream S. Jerneh and S. Badong). The main factor of different distribution pattern is the influence of marine sedimentation during Pleistocene period at the western plain area (Khoo, 1996; Allen, 2000). This is also supported by the occurrence of clayey or muddy layers in pits in that area, suggestive of Gula Formation which were deposited in paleo tidal flat or swamp environment during following Holocene period (Hassan, 1990) (Fig. 12).

5.3 Elements and mineral relationships

A positive relationship is shown by Th and monazite, indicating that monazite is the chief mineral containing the radioactive element. This is also shown by strong relationship between U and monazite. Monazite is one of the minerals searched for its Th content, although the Th content is lower than in thorianite (ThO_2) and thorite ($(Th,U)SiO_4$) (Voncken, 2016). The U occurs as U^{4+} only as accessory in selected minerals like apatite, zircon and monazite that is concentrated into residual melts (Robb, 2005). However, thorianite, thorite and apatite are not observed during QME analysis while zircon occurs in very lesser amount compared to monazite but low Th and U content may also be contributed by dark Nb-Ta minerals that could exist in placer deposits (Bradford, 1972; Zhang et al., 2002).

Apart from radioactive elements, the monazite also has strong relationships with REE, Y and Sc, indicating that the Jerai monazite contains many elements in its crystal lattices. Monazite generally contains higher amount of HREE while xenotime tends to contain higher amount of LREE (Förster, 1998a, b). All samples in this study, excluding KC41, KC564 and KC57, fit the different LREE-HREE concentrations.

The similarities of La, Ce and Nd vs monazite plots could represent uniform concentration ratios of these element in one monazite mineral. In fact, the highest content of Ce compared to La and Nd would suggest the monazite-Ce species occurs the most in Jerai area (Mindat.org, 2021).

The Y content has stronger relationship with monazite compared to xenotime. Although Y is the main element in xenotime, Flinter et al. (1963) studied that the Jerai monazite also contains considerably high amount of Y with value ranges 2.5% to 5.9%. The variation of Y content could also be the main factor of samples containing higher monazite than xenotime, but has higher HREE concentration because Y is considered as HREE in this study.

Similarities of Y and Yb plots are due to fact that both HREE also have uniform concentration ratios and tend to occur together (Förster, 1998b).

Sc is likely contributed by monazite compared to garnet, although its content is considerably very low. Sc is more common as trace amounts in iron and magnesium rich rocks (Gupta and Krishnamurthy, 2005).

5.4 Mining potential

The maximum total content of monazite and xenotime per sample is 14.48% in stream bed of S. Batu Pahat (KC56) for recent fluvial environment and 4.38% in Quaternary alluvial plain environment KCC001 (KCC001). These contents

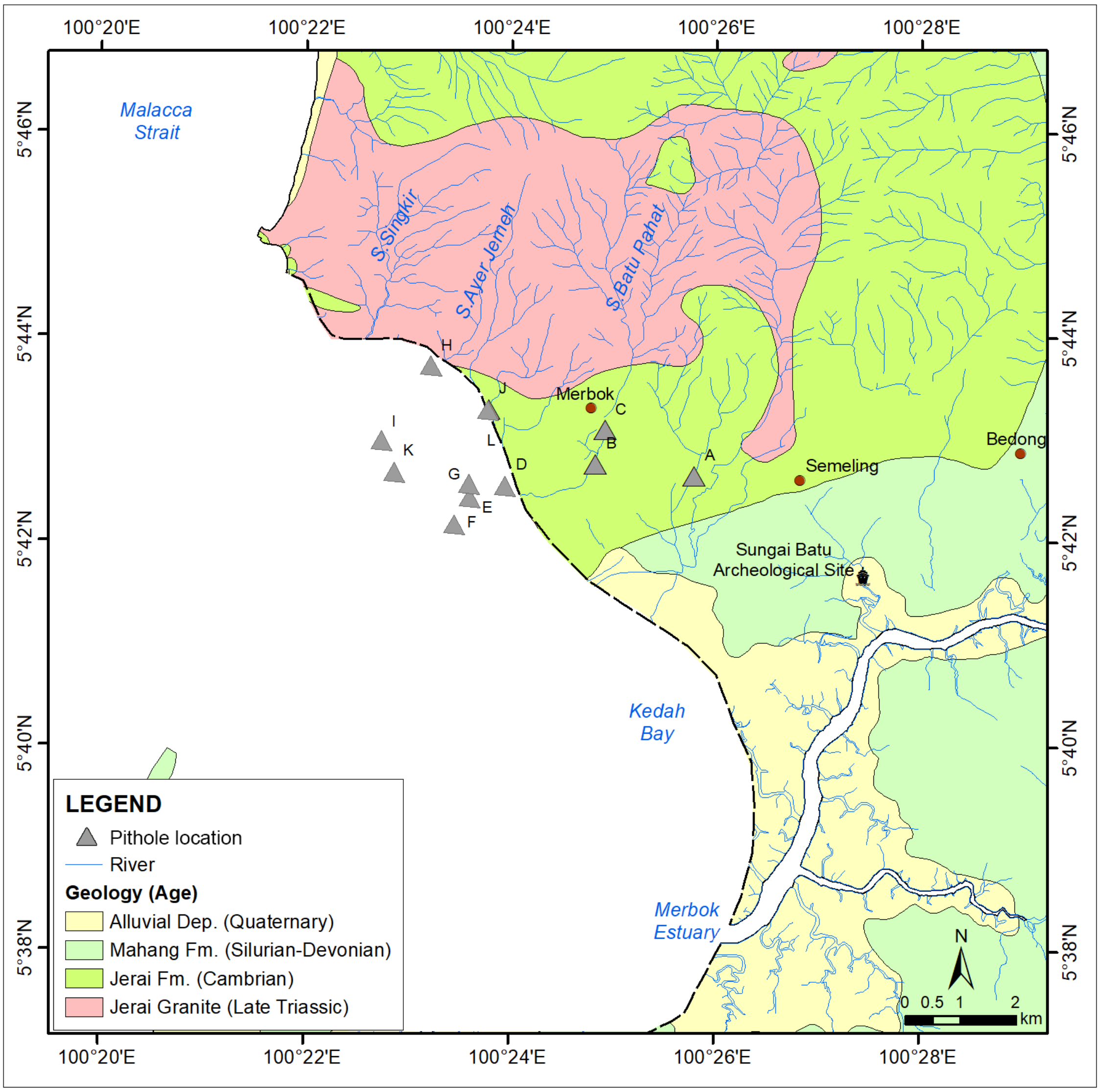

Fig. 12: Simplified geological map showing the studied pithole locations with respect to Kedah Bay and Merbok Estuary area and coastal line before 15th Century (modified from Khoo, 1996). The sea level may be higher during that time according to the discovery of ancient jetty near Sungai Batu Archaeological Site (Zakaria et al., 2011).

are exceptionally higher than those in productive placer deposits in Eneabba (Australia) and Xun Jiang (China) districts (Shepherd, 1990; Jackson and Christiansen, 1993). In comparison of sum average of both minerals, fluvial deposit has relatively more potential.

Rather than mining the REM alone, exploiting all existing heavy mineral suits could be more potential yet economic to practice. Garnets are the most occurring minerals in all samples for both fluvial and alluvial environments. Garnets are used to make abrasives to clean compacted mud and silt from well casings in petroleum industry and to polish optical lenses and metal (Rock&Gem, 2019). Tourmaline, particularly black, schorl species are also abundant in all samples. It is widely used based on its pyroelectricity and emission of far infrared radiation (Lameiras et al., 2010). Other minerals that exist abundantly in placer deposits in Jerai area include hematite, magnetite, Nb-Ta minerals and lesser cassiterite (Bradford, 1972) still has industrial demands.

6. Conclusion and recommendation

The monazite and xenotime contents are relatively higher in recent fluvial deposits than Quaternary alluvial deposits. Higher content of both minerals was shown in both fluvial and downstream, alluvial plains of S. Batu Pahat basin. The occurrences of pegmatite as chief host rock for monazite and xenotime at upstream could be the factor of higher content of the minerals in S. Batu Pahat than S. Ayer Jerneh and S. Singkir. The lower content of mineral samples including monazite and xenotime in western side of Quaternary plain is due to influence of marine sedimentation during Quaternary period. The higher Th and LREE content are contributed by monazite while Y and HREE are chiefly contributed by xenotime. The Sc content is also contributed by monazite although it is considerably low.

The monazite and xenotime in Jerai area are economically potential as placer deposits if mining method considers of harvesting other minerals like garnet, tourmaline, iron ores and Nb-Ta minerals which could be industrially beneficial. Other analysis techniques such as XRD, FESEM and EPMA are also recommended to be done in order to detail the mineral contents in the placer deposits.

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References

Abaka-Wood, G.B., Addai-Mensah, J., & Skinner, W. (2016). Magnetic separation of monazite from mixed minerals. Chemeca.

Abdul Rahman, A.H., Sibon, M., & Hasan, M.S. (2018a). Reconnaissance study on thorium element from monazite source in southern Kedah. Putrajaya, Malaysia: Department of Mineral and Geoscience Malaysia.

Abdul Rahman, A.H., Sibon, M., & Hasan, M.S. (2018b). Reconnaissance study on rare earth elements in southern Kedah. Putrajaya, Malaysia: Department of Mineral and Geoscience Malaysia.

Allen, J. (2000). In support of trade: Coastal site location and environmental transformation in early historical period Malaysia and Thailand. *Bulletin Indo Pacific Prehistory Association*, *20*, 62–78.

ASM (2014). Blueprint for the establishment of rare earth-based industries in Malaysia: summary for policy maker. Kuala Lumpur, Malaysia: Akademi Sains Malaysia.

Bea, F. (1996). Residence of REE, Y, Th and U in granites and crustal protoliths; Implications for the chemistry of crustal melts. *Journal of Petrology, 37(3)*, 521–552. doi: 10.1093/petrology/37.6.1601.

Bea, F., Pereira, M.D., Corretge, L.G., & Fershtater, G.B. (1994). Differentiation of strongly peraluminous, perphosphorous granites: The Pedrobernardo pluton, central Spain. *Geochimica et Cosmochimica Acta*, *58*(12), 2609– 2627. doi: 10.1016/0016-7037(94)90132-5.

Bean, J.H., & Hill, J.H. (1969). The iron-ore deposits of West Malaysia, Vol.2. Kuala Lumpur, Malaysia: Geological Survey of Malaysia.

Bradford, E.F. (1972). The geology and mineral resources of the Gunung Jerai area, Kedah. District Memoir 13. Kuala Lumpur, Malaysia: Geological Survey of Malaysia.

Casillas, R., Nagy, G., Panto´, G., Bra¨ndle, J., & Fo´rizs, I. (1995). Occurrence of Th, U, Y, Zr, and REE- bearing accessory minerals in late Variscan granitic rocks from the Sierra de Guadarrama (Spain). *European Journal of Mineralogy, 7*(4), 989–1006.

Castor, S.B., & Hedrick, J.B. (2006). Rare earth elements. In J.E. Kogel, N.C. Trivedi, J.M. Barker, and S.T. Krukowski (Ed.), Industrial minerals and rocks—commodities, markets, and uses (pp.769-792). Colorado, USA: Society for Mining, Metallurgy, and Exploration, Inc.

Chappell, B.W., & White, A.J.R. (2001). Two contrasting granite types: 25 years later. *Australian Journal of Earth Science, 48*(4), 489–499. doi: 10.1046/j.1440-0952.2001.00882.x.

Che Harun, H., Ismail, A., Che Wan, R., Sulaiman, Z.A., Ash'ari, M.A., Husin, Z., Abdul Ghani, M.Z., Che Kasim, M.Z., Ismail, Y., Abu Hasan, M., Daril, K., Md Slar, K., Hamzah, A.B., & Zakri, H. (Ed.) (2009). Placer tin exploration guidelines (in Malay). JMG.GP.03. Kuala Lumpur, Malaysia: Department of Mineral and Geoscience Malaysia.

Che Zainol Bahri, C.N.A., Ismail, A.F., Ab Majid, A., Mohd Ruf, M.I.F., & Al-Areqi, W.M. (2018). Extraction and purification of thorium oxide from monazite mineral. *Sains Malaysiana, 47*(8), 1873–1882. doi: 10.17576/jsm-2018-4708-28.

Connelly, N.G., & Damhus, T. (2005). Nomenclature of inorganic chemistry: IUPAC recommendations (p. 51). London, England: RSC Publishing.

Cuney, M., & Friedrich, M. (1987). Physicochemical and crystal-chemical controls on accessory mineral paragenesis in granitoids: Implications for uranium metallogenesis. *Bulletin Mine´ralogie, 110*, 235–247.

Devismes, P. (1978). Atlas photographique des mineraux d'alluvions. Paris, France: *Mem. Bur. Rech. Geol. Minières.*

Flinter, B.H., Butler, J.R., & Harral, G.M. (1963). A study of alluvial monazite from Malaya. *The American Mineralogist, 48*, 1210–1226.

Förster, H-J. (1998a). The chemical composition of REE-Y-Th-U-rich accessory minerals in peraluminous granites of the Erzgebirge-Fichtelgebirge region, Germany. Part I: the monazite-(Ce)-brabantite solid solution series. *American Mineralogist, 83*, 259–272.

Förster, H-J. (1998b). The chemical composition of REE-Y-Th-U-rich accessory minerals in peraluminous granites of the Erzgebirge-Fichtelgebirge region, Germany. Part II: Xenotime. American Mineralogist, 83, 1302–1315.

Förster, H.-J., & Tischendorf, G. (1994). The western Erzgebirge-Vogtland granites: implications to the Hercynian magmatism in the Erzgebirge-Fichtelgebirge anticlinorium. In R. Seltmann, H. Ka¨mpf, and P. Mo¨ller (Ed.), Metallogeny of collisional orogens (pp. 35-48). Prague, Czech Republic: Czech Geological Survey.

Ghani, A.A., Shahjamal, M., Ng, T.F., Ismail, N.E.H., Mohamad Zulkifley, M.T., Islami, N., Quek, L.Z., Abu Bakar, A.F., Amir Hassan, M.H., Abdul Aziz, J.H., & Masor, A.F. (2019). Ce anomaly in I-type granitic soil from Kuantan, Peninsular Malaysia: Retention of zircon in the weathering product. *Sains Malaysiana, 482)*, 309–315. doi: 10.17576/jsm-2019-4802-06.

Gupta C.K., & Krishnamurthy, N. (2005). Extractive metallurgy of the rare earths (p. 484). Florida, USA: CRC Press.

Hassan, K. (1990). A summary of the Quaternary geology investigations in Seberang Prai, Pulau Pinang and Kuala Kurau. *Geol. Soc. Malaysia Bulletin, 26*, 47–53.

Harraz, H.Z. (2013). Topic 7: Placer mineral deposits. A short series lectures, Tanta University.

Hinton, R.W., & Paterson, B.A. (1994). Crystallisation history of granitic magma: Evidence from trace element zoning. *Mineralogical Magazine, 58A*, 416–417.

Hosking, K.F.G. (1973). The primary tin mineralization patterns of West Malaysia. *Geol. Soc. Malaysia Bulletin, 6*, 297–308.

IAEA (2005). Thorium fuel cycle – potential benefits and challenges. Report no. IAEA-TECDOC-1450 (p. 90) Vienna, Austria: Author.

Ishihara, S., Sawata, H., Arpornsuwan, S., Busaracome, P., & Bungbrakearti, P. (1979). The magnetite-series and ilmenite-series granitoids and their bearing on tin mineralization, particularly of the Malay Peninsula region. *Geol. Soc. Malaysia Bulletin, 11*, 103–110.

Jackson, W.D., & Christiansen, G. (1993). International strategic minerals inventory summary report—rare-earth oxides: USGS Circular 930-N (p. 68p). Virginia, USA: USGS.

Jamil, A., Ghani, Azman A., Zaw, K., Osman, S., & Quek, L.X. (2016). Origin and tectonic implications of the ~200 Ma, collision-related Jerai Pluton of the Western Granite Belt, Peninsular Malaysia. *Journal of Asian Earth Sciences, 127*(2016), 32–46. doi: 10.1016/j.jseaes.2016.06.004.

Jha, A.R. (2014). Rare earth materials: Properties and applications. Florida, USA: CRC Press.

JMG (2014). Geological map of Kedah. Kuala Lumpur, Malaysia: Author.

JMG (2019). Mining industries in Malaysia 2018. Putrajaya, Malaysia: Author.

Kim, K., & Jeong, S. (2019). Separation of monazite from placer deposit by magnetic separation. *Minerals, 2019*(9), 149–160. doi: 10.3390/min903 0149.

Khoo, T.T. (1977). Occurrence and implication of syncplutonic pegmatite dykes in the Gunung Jerai granite, Kedah. *Geol. Soc. Malaysia Bulletin*, 8, 117–120.

Khoo T.T. (1996). Geomorphological evolution of the Merbok estuary area and its impact on the early state of Kedah, northwest Peninsular Malaysia. *J. SE Asian Earth Sci, 13*(3-5), 347–371. doi: 10.1016/07 43-9547(96)00042-6.

Lameiras, F.S., Nunes, E.H.M., & Maria-Leal, J. (2010). Backgrounds for the industrial use of black tourmaline based on Its crystal structure characteristics. *Ferroelectrics, 377*(1), 107–119. doi: 10.10 800150190802523719.

Liu, L., Hu, R., Zhong, H., Yang, J., Kang, L., Zhang, X., Fu, Y., Mao, W., & Tang, Y. (2020). Petrogenesis of multistage S-type granite from the Malay Peninsula in the Southeast Asian tin belt and their relationship to Tethyan evolution. *Gondwana Research, 84*, 20–37. doi: 10.1016/j.gr.2020.02.013.

Atomic Energy Licensing Act 1984 (Malaysia).

McLennan, S.M., & Taylor, S.R. (2012). Geology, geochemistry and natural abundances of the rare earth elements. In A.D. Atwood (Ed), The rare earth elements: fundamentals and applications (p. 8). West Sussex, United Kingdom: Wiley.

Mindat (2011). Monazite-(Ce). https://www.mindat.org/min-2751.html. Accessed 20 January 2021.

Mohd Hasan, M.Z., Zakaria, M.R., Lee, A.K., & Chong, F.S. (1993). Transect 2 radioactive mineral exploration, Peninsular Malaysia. Kuala Lumpur, Malaysia: Geological Survey of Malaysia.

Mohd Hasan, M.Z., Zakaria, M.R., & Ong, W.S. (1995). Transect 1 radioactive mineral exploration, Peninsular Malaysia. Kuala Lumpur, Malaysia: Geological Survey of Malaysia.

Mohd Mokhtar, N.A., & Saidin, M. (2018). The contributions of Lembah Bujang to the iron industry in Malaysia (in Malay). Melayu: *Jurnal Antarabangsa Dunia Melayu*, *11*(2), 182–199.

Ng, S.W.P., Chung, S.L., Robb, L.J., Searle, M.P., Ghani A.A., Whitehouse, M.J., Oliver, G.J.H., Sone, M., Gardiner, N.J., & Roselee, M.H. (2015). Petrogenesis of Malaysian granitoids in the Southeast Asian tin belt: Part 2. U-Pb zircon geochronology and tectonic model. *Geological Society of America Bulletin*, *127*, 1238–1258. doi:10.1130/B31214.1.

Rapp, R.P., & Watson, E.B. (1986). Monazite solubility and dissolution kinetics: Implications for the thorium and light rare earth chemistry of felsic magmas. *Contr. to Mineral. and Petrol.*, *94*, 304-316. doi:10.1007/BF00371439.

Robb, L. (2005). Introduction to ore-forming processes. New Jersey, USA: Blackwell Science Ltd.

Robb, L.J. (2019). The geology of tin deposits with special reference to tin mineralisation in Peninsular Malaysia. Proceedings of the National Tin Conference 2019. Kuala Lumpur: Malaysia: Tin Board.

Rock & Gem (2019). Rietreved from https://www.rockngem.com/garnet-an-industrial-commodity/#:~:text=Garnet%20Applied%20Benefits&text=The%20petroleum%20industry%20uses%20huge,for%20water%20and%20industrial%20liquids.

Sengupta, D., & Van Gosen, B.S. (2016). Chapter 4: Placer-type rare earth element deposits. In P.L. Verplanck, & M.W. Hitzman (Ed.), Rare earth and critical elements in ore deposits. Reviews in Economic Geology (pp. 81–100). Virginia, USA: SEG.

Shepherd, M.S. (1990). Eneabba heavy mineral sand placers. In F.E. Hughes, (Ed.), Geology and mineral deposits of Australia and Papua New Guinea (p.1591). London, United Kingdom: Institute of Mining and Metallurgy.

USGS (2019). Mineral commodity summary 2019. Virginia, USA: Author.

Voncken, J.H.L. (2016). The rare earth elements: An introduction. London, United Kingdom: Springer Nature.

Wan Hassan, W.F. (1989). Some characteristics of the heavy detrital minerals from Peninsular Malaysia. *Geology Society of Malaysia Bulletin*, *24*, 1–12.

Ward, C.D., McArthur, J.M., & Walsh, J.N. (1992). Rare earth element behaviour during evolution and alteration of the Dartmoor granite, SW England. *Journal of Petrology*, *33*(4), 785–815. doi:10.1093/petrology/33.4.785.

Wark, D.A., & Miller, C.F. (1993). Accessory mineral behaviour during differentiation of a granite suite: Monazite, xenotime, and zircon in the Sweetwater wash pluton, southeastern California, U.S.A. *Chemical Geology*, *110*(3), 49–67. doi: 0009-2541(93)90247-G.

Willbourn, E.S. (1925). A list of minerals found in British Malaya together with a description of their properties, composition, occurrences and uses. *Journal of the Malayan Branch of the Royal Asiatic Society*, *3*(3), 57–100.

Yang, J., Zhou, M., Zhong, H., Williams-Jones, A.E., Liu, L., Zhang, X., Fu, Y., & Mao, W. (2020). Granite-related tin metallogenic events and key controlling factors in Peninsular Malaysia, Southeast Asia: New insights from cassiterite U-Pb dating and zircon geochemistry. *Economic Geology*, *115*(3), 581–601. doi:10.5382/econgeo.4736.

Zakaria, I. I., Saidin, M., & Abdullah, J. (2011). Ancient jetty at Sungai Batu Complex, Bujang Valley, Kedah. Postgraduate Student Forum. Retrieved from http:// www.cuhk.edu.hk/ant/PostgraduateForum2011/Arch/IklilIzzatiZAKARIA.pdf.

Zakaria, M.R., Mohd Hasan, M.Z., Lee, A.K., Ong, W.S., & Teoh, L.H. (1994). Transect 3 radioactive mineral exploration, Peninsular Malaysia. Kuala Lumpur, Malaysia: Geological Survey of Malaysia.

Zhang, P., Tao, K., Yang, Z., Yang, X., & Song, R. (2002). Rare earths, niobium and tantalum minerals in Bayan Obo ore deposit and discussion on their genesis. *Journal of Rare Earths*, *20*(2), 81–86.

Zhao, J.-X., & Cooper, J.A. (1993). Fractionation of monazite in the development of V-shaped REE patterns in leucogranite systems: Evidence from a muscovite leucogranite body in central Australia. *Lithos*, *30*(1), 23–32. doi: 10.1016/0024-4937(93)90003-U.

Zhou, B., Li, Z., & Chen, C. (2017). Global potential of rare earth resources and rare earth demand from clean technologies. *Minerals*, *2017*(7), 203–217. doi:10.3390/min7110203.