Geochemical and Mineralogical Characteristics of Andesitic Derived Laterite from the Serian Volcanic, West Sarawak, Malaysia: Potential for Ion-Adsorption-Type Ree Deposit

(Ciri Geokimia dan Mineralogi Laterit Asalan Andesitik daripada Serian Volkanik, Sarawak Barat, Malaysia: Potensi untuk Deposit Ree Jenis Jerapan Ion)

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ABSTRACT

The rare earth elements (REEs) found in the Earth's crust are highly sophisticated minerals that have a significant application in various high-tech industries such as green technology and defense. Due to the limited study of REE derived from volcanic rocks, the main objectives of this research are focusing to determine the geochemical and mineralogical, as well to examine the potential resources of ion-adsorption type deposits in volcanic rock and their derived laterite profile from the Serian Volcanic, West Sarawak, Malaysia. Two lateritic profiles were examined, and the parent volcanic rock is classified as basaltic trachyandesite in composition with moderate REE content of 209 ppm. In both profiles the abundance of the REE is subjected to the deep weathering through CIA (99-100%) that promote the enrichment of REE to the laterite profile up to 1715.98 ppm. The dominance of kaolinite mineral as an absorbent material, coupled with the absence of RE mineral in the laterite samples, indicates the potential for ion-adsorption clay deposit with high (>75%) extracted REY content, as determined by ion-exchangeable analysis. The chondrite-normalized REE pattern for both profiles were almost flat for parent rock, whereas LREE and HREE were equally abundant in the laterite profile. However, the TREE significantly decrease toward the upper part of each profile. This study also demonstrates that laterites with a high TREE and low Ce anomalies have the highest ion-exchangeable (REY) fraction relative to the parent rock content, suggesting that ion-adsorption deposit in this region have the potential to recover REY in terms of resources quantity.

Keywords: Andesite; ion-adsorption clay; Malaysia; rare earth element

ABSTRAK

Unsur nadir bumi (REE) yang terdapat di kerak bumi adalah merupakan mineral termaju yang signifikan kepada pembangunan industri berteknologi tinggi meliputi teknologi hijau dan pertahanan. Disebabkan kurangnya kajian REE yang berasalan daripada batuan volkanik, objektif utama kajian ini memberi fokus untuk mengkaji cirian mineralogi dan geokimia serta mengenal pasti potensi jenis longgokan lempung jerapan ion dalam batuan volkanik dan profil luluhawa laterit daripada Serian Volkanik, Wilayah Barat Sarawak, Malaysia. Dua profil laterit telah dikaji dan pengelasan batuan induk volkanik menunjukkan batuan berada dalam kumpulan trakit-andesit-basalt dengan kepekatan kandungan REE 209 bpsj. Kandungan REE yang tinggi pada kedua-dua profil luluhawa adalah berkait-rapat dengan kadar luluhawa yang tinggi melalui peratusan CIA (99-100%) yang menyumbang kepada pengayaan REE mencecah 1715.98 bpsj. Kewujudan mineral lempung yang dominan dan bertindak sebagai mineral penjerap dengan tiadanya kehadiran mineral RE yang dikesan dalam sampel laterit mencadangkan kawasan kajian mempunyai potensi kewujudan longgokan lempung jerapan ion dengan kadar pengekstrakan REY yang tinggi (>75%) melalui analisis pertukaran-ion. Corak penormalan-kondrit unsur nadir bumi pada kedua-dua profil juga menunjukkan bentuk yang hampir rata untuk batuan dan keberadaan LREE dan HREE hampir sama banyak pada profil laterit yang dikaji. Walau bagaimanapun, kandungan TREE dilihat berkurangan ke arah bahagian atas untuk kedua-dua profil. Kajian yang dijalankan juga menunjukkan laterit yang mengandungi kandungan TREE yang tinggi dengan nilai anomali Ce yang rendah mempunyai kadar pertukaran ion REY yang tinggi berbanding

kandungan yang terdapat dalam batuan induk, mencadangkan longgokan lempung jerapan ion di wilayah ini berpotensi mempunyai sumber REY dari segi kuantiti.

Kata kunci: Andesit; lempung jerapan ion; Malaysia; unsur nadir bumi

INTRODUCTION

The group of elements known as rare earth elements (REEs; the same shall apply hereafter) consists of the lanthanides, which include lutetium and all 14 elements positioned between lanthanum and ytterbium in a single row of the periodic table (Atwood 2012; Henderson 1984). Additionally, scandium and yttrium, which share similar chemical properties with the lanthanides and are often found in the same ore deposits, are also considered part of the rare earths. This group of elements is typically not found in isolation but rather occurs in conjunction with one another within a variety of minerals present in the Earth's crust. REEs are sometimes classified into two group: light REEs (LREEs), which include elements from lanthanum to europium, and heavy REEs (HREEs), which include elements from gadolinium to lutetium (Hoshino, Sanematsu & Watanabe 2016).

The ion adsorption clay deposits represent the sole origin of heavy REEs worldwide, and the majority of the advancement industries in the western region heavily depend on China's REEs reserves. Malaysia's ion adsorption deposits studies are relatively new. Yaraghi, Ariffin and Baharun (2020) and Yusoff, Ngwenya and Parsons (2013) reported on some of the research that was done on the REE-rich weathered granitic crust in West Malaysia, however, little attention has been paid to the volcanic rock, especially in West Sarawak which is the exposed section of the Sunda Shield in Southwest Borneo. Between 2016 and 2020, an exploration survey for REEs was conducted by Minerals and Geoscience Department of Malaysia (JMG Malaysia) in multiple Malaysia states, namely Kedah, Perak, Terengganu, Kelantan, Johor, and Sarawak. A reconnaissance survey conducted by Taniou et al. (2019), showed the possible existence of REEs in the intermediate rocks (andesite) of the Serian Volcanic Formation in Riih Serian, Sarawak. The study shows that the concentration of REEs in the weathered crust of Serian Volcanic Formation, which are of late Triassic age, is the highest in Malaysia, with the exception of weathered granite rocks that are present in various regions of Peninsular Malaysia. Additionally, the andesitic rock is widespread in the western section of Sarawak, encompassing roughly 510 km². Moreover, due to its location on the equatorial line, Sarawak experiences a tropical climate which renders the rock surface prone to weathering. Despite their widespread occurrence, research on the geochemistry of REEs in the weathered crust of andesitic rocks in Malaysia has yet to be undertaken. This study presents novel findings on the geochemistry of REEs in the weathered crusts andesitic rocks of Riih Serian, West Sarawak, Malaysia. Moreover, this study provides valuable information that paves the

way for the discoveries and potential development of REE in Sarawak and Malaysia as a whole, particularly for the high technology industries. In addition to the petrological and geochemical data available from previous studies (Taniou, Basori & Sanematsu 2022), many new geochemical, extraction leaching, spectrographic analyses were performed recently and they collectively constitute the base for this paper.

GEOLOGICAL FRAMEWORK

REGIONAL GEOLOGY

Southeast Asia is characterized by two main plate continents aged Mesozoic: Sundaland, which represents Eurasian plate extension moving to the south-east, and Australian plate extension moving to the south and east Indonesian oceans. Sundaland is the mainland for the Southeast Asia continent, which includes Sumatera, Java, and Borneo, where the craton is a part of the Eurasian plate (Hutchison 2005). Borneo is at present a part of the Sundaland mainland, which is located in the middle of Southeast Asia and is a tectonically complex island. The geological formation in the area was a result of various geological processes such as arc-continent collisions, continent-continent collisions, and subduction-accreation convergence of the India-Australian, Pacific, and Philippine Sea Plates (Metcalfe 2011).

The geographical region under research is situated in the northern section of Borneo Island. Specifically, the north-western portion of the island is occupied by Sarawak (Figure 1(a)). According to Liechti (1960), the geological composition of Sarawak is primarily characterized by three distinct regions, namely west, central, and north Sarawak. The Lupar Line and Bukit Mersing Line separate these regions (Hutchison 2005). The Lupar Line is thought to be a suture zone remnant from a southward-directed subduction system (Hamilton 1979; Tan & Lamy 1990).

West Sarawak is part of the West Borneo Basement, which is the exposed part of Southwest Borneo's Sunda Shield (Haile 1974). The basement was tectonically active throughout the Palaeozoic and Mesozoic periods but has since remained a stable area. The Palaeozoic-Mesozoic geological formations in western Sarawak are concealed by substantial Tertiary strata.

GEOLOGICAL BACKGROUND

The general geology of the Riih area typically includes the Serian Volcanic, the Sadong Formation and quaternary deposits (Figure 1(b)). The Serian Volcanic of Late Triassic comprises andesite, lava basalt, tuff and breccia (Pimm



FIGURE 1. (a) Maps showing the general geology of west Sarawak (after Tan 1986) and the (b) general geology of study area (after Taniou et al. 2019)

1965; Wilford 1965), whereas trachytic and dacite volcanic rocks are distinguished and mapped as the Semabang Trachyte Member (Wilford 1965). The Serian Volcanic sequence is intercalated with the Sadong Formation, which is characterized by interbeds of mudstone, silty shale, sandstone, conglomerate and rarely tuff with moderate to steep bedding dips (Pimm 1965; Wilford 1965).

The Serian Volcanic Formation is a succession of igneous rocks that formed the West Sarawak and West Kalimantan Mountain ranges and is known as the Upper Triassic continental arc. It developed as a result of the subduction of Mesozoic oceanic crusts and gave rise to a sequence of volcanic rocks ranging from basalt to andesite.

MATERIALS

DESCRIPTION OF THE PARENT ROCK OF RIIH VOLCANIC

Through observation from a survey conducted by Taniou et al. (2019), it was found that the exposed outcrops of the volcanic rocks in Riih area are located in the eastern part, which is dominated by Serian Volcanic. The estimated range thickness of the weathering profile found in the study area, carried out on an area of 4 km^2 ranges from 2 to 8 meters. Due to the high rate of weathering in the study area, it is exceedingly difficult to find an ideal weathering profile with parent rock at the base of the profile.

Wilford (1965) was first described Serian Volcanic to represent lavas and pyroclastic rocks. The andesite is non-porphyritic with a dark green colour fine-grained textures dominating the study area. However, in some areas the rock shows colour variations suggesting that it has undergone hydrothermal alteration. In thin section, it typically contains varying amounts of colorless pyroxene and feldspar laths of varying sizes embedded in a matrix of glass (Adi 1994). Chlorite and iron oxide are secondary minerals that form as a result of hydrothermal alteration.

According to Taniou, Basori and Sanematsu (2022), the parent rock ranges from trachy-andesitic to andesitic in composition with a moderate REE content of 322 ppm. The chondrite-normalized REE patterns of the volcanic rocks exhibit fractionation of the LREE in comparison to the HREE. Additionally, there is a noticeable negative Eu anomaly and a weak negative Ce anomaly. Whereas, the weathered crusts exhibit negative Eu and insignificant Ce anomalies, with higher REE concentrations up to 1433 ppm with an average of 613 ppm.

DESCRIPTION OF THE WEATHERING PROFILES

Two weathering profiles, P1 and P2, were studied and underlain by either fresh or moderately weathered andesitic rocks in the Riih, Serian, Sarawak, East Malaysia (Figures 2(a) & 2(b)). The samples were taken at suitable horizons A, B, C and D along the weathering profile until reaching the underlying bedrock. The thickness of the profiles P1 and P2 is 7 meters and 6 meters, respectively.

In general, both weathering profiles exhibit a reddish, brownish to yellowish colour, with a red colour on the oxidized surface. The reddish, which is enriched in organics and humus up to 1 meter in thickness, occurs in the upper part of both profiles. Below this deeply weathered crust is the laterite horizon, which is typically reddish to brownish in colour, and is composed of friable soil and loose material up to 5 meters in thickness.

The saprolite horizon or least weathered original rock at P2 is well-developed than those at P1. The fresh rock sample from profile P1 is greenish in colour, nonporphyritic, fine-grained with minor calcite veinlets crosscut the sample. The slickened-sided rock surfaces have greenish a colouration with non-homogeneous and highly fractured textures. The volcanic rock sample collected from profile P2 is moderately weathered dark grey with fine-grained groundmass.

METHODS

WHOLE-ROCK COMPOSITION

In total, seven samples were collected from the outcrops of weathered crusts and fresh rock at both weathering profiles. All collected samples were processed at the lapidary laboratory of the Department of Minerals and Geosciences of Malaysia, Sarawak. To prevent contamination and labelling mistakes throughout the study, the author processed and prepared samples independently.

All the samples from each horizon were mixed thoroughly and then pulverized to less than 75 microns. Meanwhile, the rock samples were crushed using a jaw crusher into fragments several centimetres in size and ground using a tungsten carbide mill to produce rock powder. An X-ray fluorescence (XRF) analysis on a fused disc is performed to determine the major element (Si, Ti, Al, Fe, Mn, Mg, Ca, Na, K, and P) concentration using an XRF spectrometer (Bruker). REEs (La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Sc and Y) contents were analyzed using laser ablation - inductively coupled plasma spectrometry (ICPMS - NexION-2000) by fusion glass of rock powder with lithium tetraborate, lithium metaborate, and lithium bromide. Both of the instrumental analyses were performed at the Laboratory Service Unit of JMG Technical Service Division, Ipoh, Perak, Malaysia. The analysis result was monitored using a Canada Centre for Mineral and Energy Technology (CANMET) geochemical reference, with a recovery of more than 98%. Analytical

precisions between 1% to 3% for major element and better than 5% to 10% for rare earth elements.

EXTRACTION EXPERIMENTS AND ANALYSIS OF ION EXCHANGEABLE REE

In order to access the ion-exchangeable REY with the overall rock grade, a one-step extraction and solution ICP-MS analysis were conducted to evaluate the concentration of ion-exchangeable elements utilizing an electrolyte solution. The methodology employed was primarily based on the single-step procedure outlined by Sanematsu and Kon (2013).

In a 50 mL polypropylene container 3 g of soil sample powder was soaked in 30 mL of 0.5 mol/L (6.6%) ammonium sulphate [(NH4)2SO4] solution with a pH of 5.7. The container was shaken for 24 h at room temperature so that the solid sample could react well with the solution. The solution was filtered with a membrane filter made of cellulose acetate ($\varphi = 0.45 \ \mu m$), and the membrane filter was rinsed repeatedly with ultra-pure water. The filtered solution was acidified using HNO3 and was kept as 1% HNO₃ equivalent in a polypropylene container. The Agilent Technologies 7500cx ICP-MS determined the solution's extracted element concentration. To increase Ce signals intensity, and reduce oxide production rate (140Ce16O/140Ce, < 0.01), carrier gas flow rates and ICP-MS ion-lens settings were tuned. We monitored 7Li, 23Na, 24Mg, 27 Al, 29Si, 31P, 39K, 42Ca, 45Sc, 47Ti, 51V, 53Cr, $\sum_{55}^{25} Mn, \sum_{57}^{27} Fe, \sum_{59}^{27} Co, \sum_{60}^{60} Ni, \sum_{63}^{63} Cu, \sum_{66}^{60} Zn, \sum_{69}^{69} Ga, \sum_{75}^{7} As, \sum_{85}^{87} Rb, \sum_{88}^{88} Sr, \\ \sum_{107}^{89} Y, \sum_{107}^{107} Ag, \sum_{111}^{111} Cd, \sum_{115}^{111} In, \sum_{121}^{121} In, \sum_{133}^{132} Cs, \sum_{133}^{132} Ba, \sum_{139}^{140} La, \sum_{140}^{140} Ce, \sum_{141}^{141} Pr,$ ⁸⁹ $_{107}$ $_{107}$ $_{107}$ $_{117}$ $_{117}$ $_{115}$ $_{115}$ $_{121}$ $_{137}$ $_{137}$ $_{137}$ $_{137}$ $_{139}$ $_{140}$ $_{140}$ $_{141}$ $_{141}$ $_{140}$ $_{147}$ $_{141}$ $_{141}$ $_{142}$ $_{147}$ $_{141}$ $_{141}$ $_{142}$ $_{147}$ $__{147}$ $__{147}$ $__{147}$ $__$ using XSTC-1, -8 and -15 multi-element standard solutions (SPEX CertiPrep).

In addition, an analysis involving the ICPMS using the laser ablation technique was carried out to access the whole rock content, as it is needed for the comparison to calculate the ion exchangeable percentage from the selected samples relative to the whole rock content. The extraction experiments for REE and subsequent ones were conducted at the Geological Survey of Japan (AIST).

X-RAY DIFFRACTION ANALYSIS (XRD)

The XRD analysis of specific soil and rock samples was performed in the Mineralogy and Petrology Laboratory of the JMG Technical Service Division. More precise results on the types of minerals found in rock and soil samples, as well as assistance for other analyses like geochemical data, depend on XRD analysis. The samples are finely pulverized to a particle size of 75 μ m and placed into a conventional XRD powder samples container. The powder was compacted using a press block to create a flat surface. The entire sample will be put into the instrument, and X-ray diffraction data will be taken at intervals of 0.02/0/1s in the



FIGURE 2. (a) Photograph and columns depicting vertical laterite profile P1 derived from andesitic between 0 and 7 meters in height and (b) photograph and columns depicting vertical laterite profile P2 derived from andesitic between 0 and 6 meters in height

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 2θ range of 5 - 60° , without obstructing the surface of the sample. The Xpert Highscore Plus Panalytical software is utilized in the data processing laboratory to examine XRD data. The raw data is compared using the Ritveld analysis method to identify the most appropriate characteristics for mineral identification.

SCANNING ELECTRON MICROSCOPY – MINERAL LIBERATION ANALYSIS (SEM-MLA)

The MLA system consists of a custom software program and a standard SEM (Bruker) with energy X-ray analysis. It was designed to quantitatively analyze mineral and material samples. MLA is, in principle, very similar to the estimation procedure for mineral model analysis using an optical microscope and/or semi-automated SEM (Fandrich et al. 2007). In addition, MLA offline processing tool converts raw particle image with x-ray data into particle maps and calculates bulk and liberation data from those images (Gu 2003).

To carry out this analysis, a polished section of selected samples was provided to quantify a wide range of mineral characteristics, such as mineral abundance, grain size, and liberation. The polished section was then analyzed using MLA Hardware FEI SEM Platforms (Quanta 650). Meanwhile Bruker Quanta X-flash SDD is used as the EDS equipment. This analysis was carried out at the Geological Survey of Japan, AIST.

RESULT

MAJOR ELEMENT OF ANDESITE AND WEATHERED PROFILES

Results of major element from XRF for all the samples are listed in Tables 1 and 2. For profile P1, the fresh rocks sample (P1 R7m) contains $SiO_2 = 54.20$ wt.%, $Al_2O_3 =$ 13.60 wt.%, $Fe_2O_3 = 11.50$ wt.%, respectively, with loss on ignition content (LOI) 2.70 wt.%. In contrast, the weathering profile of P1 show high contents of TiO₂, Al₂O₃, Fe₂O₃, and LOI values in the soils indicates the laterization to the near surface weathering product. However, the contents of CaO, Na₂O, and K₂O, are lower than the parent rock are seen in this weathering profile can be explained by the element's partitioning to the aqueous phase and subsequent leaching from the system. The chemical index alteration (CIA) as proposed by Nesbitt and Young (1982) for the rock sample is 55, and mostly over 93% for the weathered profile, suggesting weathered product of andesitic rock series of the profile experienced complete weathering.

Similar to weathering profile P1, most of major elements from second weathering profile P2 are significantly increased in the TiO₂, Al₂O₃, Fe₂O₃, and LOI, with decreased in the SiO₂, MnO, MgO, CaO, Na₂O, K₂O, and P₂O₅ contents compared to their parent rock. The rock

sample's CIA is 86%, whereas those for the weathering profile samples range from 98 to 100%.

RARE EARTH ELEMENT OF ANDESITE AND WEATHERED PROFILES

The result of the REE content representative samples from profile P1 and P2 are shown in Tables 1 and 2. The total content of REE (TREE) on profile P1 ranges widely from 209.53 to 1715.98 ppm, whereas the content of light rare earth elements (LREE) and heavy rare earth elements (HREE) ranges from 155.03 to 1008.20 ppm and 34.29 to 707.78 ppm, respectively. Overall, the weathering profile P1 show enrichment pattern of LREE and HREE which reach to 1008.20 ppm (LREE) and 707.78 ppm (HREE) relative to the rock sample (P1 R7m).

All the samples in weathering profile P2 have a similarity with the REE pattern in weathering profile P1. The TREE content on profile P2 ranges from 69.61 to 1422.26 ppm. On the other hand, the content of LREE and HREE ranges from 62.86 to 954.40 ppm and 6.75 to 467.86 ppm, respectively. It also indicates the enrichment of LREE and HREE from the semi-weathered sample into the soil sample, where the concentration up to 954.40 ppm (LREE) and 467.86 ppm (HREE).

C1-chondrite normalized REE patterns (Sun & McDonough 1989) of the weathered samples from profile P1 show values of Ce/Ce* anomalies ranging from 0.19 to 2.87, with one out of the total samples having negative anomaly values. All samples have negative anomaly values ranging from 0.70 to 0.76 for the Eu/Eu* anomaly and ratio values ranging from 6.47 to 11.93 for the La/Yb ratio. The profile P2 samples, on the other hand typically show a positive Ce/Ce* anomaly, except for one sample that showed a negative Ce anomaly. This anomaly has a range of 0.21 to 10.65. However, the Eu/Eu* anomaly for all of the samples showed negative anomalies ranging from 0.75 to 0.83 and ratio values ranging from 1.73 to 12.79 for the La/Yb ratio. Both weathering profiles are characterized by a positive Ce anomaly (>1) at the top of the weathering profile and subsequently by negative Ce anomaly (<1) below it. The explanation is closely related to the depletion of REE in the upper part (except for Ce), whereas the lower part of the profile tends to be enriched in REE.

Generally, the distribution variation of change in the Ce anomaly for both weathering profile value shows a negative change from rock samples to weathered samples in the middle of the profile and then becomes more positive for the sample at the upper part of the weathering profile. Meanwhile, there is no significant amplitude in the Eu anomaly from the rock the sample to the weathered samples throughout both of the weathered profiles. In general, the La/Yb ratio indicates that LREE enrichment is greater in soil samples while HREE enrichment is greater in rock sample for both weathering profiles.

For the single-step in exchangeable experiment, the two samples with the highest concentration were chosen based on the LA-ICPMS analysis results (Tables 1 & 2). Profile samples P1 S4m (TREE = 1715.98 ppm) and P2 S5m (TREE = 1422.26 ppm) are the two samples, respectively. Table 3 summarizes the results of the single-step extraction of LREE, HREE, REY, Th, and U from the selected samples.

The result obtained from the experimental condition involving the utilization of ammonium sulfate solution for single-step extraction indicates a significant concentration level for both samples. The extracted REY from the weathered profiles for samples P1 S4m is 77.86%, equivalent to 1184.7 ppm relative to the whole rock content. In the meantime, it is noteworthy that the sample P2 S5m exhibits a comparatively elevated and significant level of extracted REY, amounting to 83.76% (1754.7 ppm) in relative to the whole rock content. Figure 3(a) and 3(b) provides a brief representation of the outcomes of the concentration extraction through a single-step ion exchangeable analysis for REY, LREE, and HREE in relation to the whole rock composition from the chosen samples.

Furthermore, the two radioactive elements, namely Th and U, along with other REEs are observed through this experiment. The element Th could not be detected (n.d) for both samples involved. However, single-step extraction yields up to 0.01 ppm (0.46%) and 0.03 ppm (0.68%) of the element U for samples P1 S4m and P2 S5m, respectively. The sample's constituent elements exhibit a significantly low extraction value (<1 ppm) in relation to the whole rock content.

The analysis of normal pattern anomaly in the result of the single-step extraction test was conducted using the normalizing factor chondrite value based on Sun and McDonough (1989). The calculated Ce/Ce* and Eu/Eu* anomalies in both samples had negative anomaly values which is less than 1. In the meantime, the La_N /Yb_N ratio for these two samples is 11.82 (P1 S4m) and 6.12 (P2 S5m), respectively.

XRD RESULT

Figure 3(c) and 3(d) shows the representatives powder XRD patterns of the semi-quantitative analysis of two soil samples (P1 S4m and P2 S5m). Both samples were selected for the study to see the pattern of mineral composition variations which contained the highest TREE values. The bulk powder analysis for soil sample P1 S4m shows an X-ray diffraction pattern that consists of the peak and background of minerals from most abundant to least abundant, which are kaolinite, goethite, feldspar, gibbsite and silicate. Meanwhile, the X-ray diffraction pattern of the soil sample P2 S5m indicates the presence of minerals including kaolinite, goethite, illite, silicate, and gibbsite, as

observed from the peaks and backgrounds. Both samples were dominantly kaolinitic shows the advanced phase of the weathering process occurring in the upper horizon, wherein the soil undergoes recurrent cycles of wetting and drying, leading to the transformation of minerals such as kaolinite (Colman 1982; Papoulis, Tsolis-Katagas & Katagas 2004).

MINERAL LIBERATION

For sample P1 S4m, automated scanning electron microscopy-mineral liberation analysis (SEM-MLA) was used to classify and retrieve mineralogical data. The summarized quantitative analysis for the model mineralogy of this sample is shown in Figure 4. Overall, the findings from the polished mount of the sample reported here are based on the analysis of 330,148 individual particles in a single sample. Sample P1 S4m showed the occurrences of major minerals such as kaolinite, silica, goethite, and rutile, while K-feldspar, Mn-Fe oxide, illite, hematite, pyrite, Al-Si oxide, gibbsite, and albite were quantified in minor quantities (Figure 4(a) & 4(b)). This sample represents 49.41 wt.% cumulate-enriched kaolinite (7%Fe) and covers 53.38% of the total area (μ m2) of the framed sample. It can be observed that silica and goethite are the second and third significant minerals, with 17.46 wt.% (18.69% area) and 15.43 wt.% (11.41% area) of the minerals, respectively. The mineral grains that constitute goethite>kaolinite and kaolinite+goethte+rutile mineral particles are also present in the majority of the dispersed mineral distribution in the sample. The two mineral mixtures make up 7.35 wt.% and 5.7 wt.% of the weight percentage and cover 5.9% and 6.16% of the total area, respectively. Whereas, as previously indicated, the occurrences of other minor minerals account for less than 1% of the total area (µm2) and weight percentage (Figure 4(c)). The SEM back-scattered electron images of the minerals related to the sample are shown in Figure 4(d)-4(g).

DISCUSSION

GEOCHEMISTRY OF THE ROCK AND THEIR WEATHERING PRODUCT

In Figure 5(a), the sample suite is categorized using the total alkali-silica diagram of Le Bas et al. (1986). On the basis of this scheme, the rocks for this study and Taniou et al. (2019) are categorized as sodic ($[Na_2O_2] > K_2O$) and vary from mugearite (basaltic trachyandesite) to latite (trachyandesite) andesite. The parent rock trachyandesite basaltic (P1 R7m) in general experienced a strong weathering process, as the chemical index alteration (CIA) proposed by Nesbitt and Young (1982) shows the value nearly reached 100%. Meanwhile, the classification of the rock sample found on the P2 profile cannot be done as the sample was moderately weathered.

Sample#	P1 S0m	P1 S4m	P1 S6m	P1 R7m		
Rock Type	Laterite	Laterite	Laterite	Andesite		
SiO ₂ (wt.%)	32.60	36.20	39.60	54.20		
TiO ₂	2.88	2.87	2.62	1.46		
Al ₂ O ₃	25.20	22.90	21.30	13.60		
Fe ₂ O ₃	19.70	16.70	14.50	11.50		
MnO	0.05	0.19	0.09	0.18		
MgO	0.27	0.76	0.95	3.45		
CaO	0.01	0.01	0.02	5.08		
Na ₂ O	0.04	0.01	0.06	4.24		
K ₂ O	0.09	0.45	1.60	2.01		
P_2O_5	0.11	0.17	0.22	0.23		
LOI	15.10	13.8	17.6	2.70		
Total	96.05	94.06	98.56	98.65		
CIA (%)	99	98	93	55		
La (ppm)	27.20	354.00	35.60	26.40		
Ce	155.00	121.00	84.50	59.40		
Pr	6.43	65.50	7.14	5.20		
Nd	25.90	350.00	36.90	27.70		
Sm	5.13	63.10	8.66	6.09		
Eu	1.22	17.80	2.40	1.54		
Gd	5.61	82.10	41.50	6.88		
Tb	0.76	5.33	11.00	1.03		
Dy	4.50	34.40	1.85	6.20		
Но	0.92	7.12	2.57	1.36		
Er	2.23	18.10	5.87	3.29		
Tm	0.40	2.69	1.02	0.53		
Yb	2.28	15.60	5.50	3.40		
Lu	0.39	2.44	0.93	0.51		
Y	17.20	540	60.50	31.30		
Sc	52.00	36.80	41.50	28.70		
LREE	272.88	1008.20	216.70	155.03		
HREE	34.29	707.78	101.24	54.50		
TREE	307.17	1715.98	317.94	209.53		
Ce/Ce*	2.87	0.19	1.30	1.24		
Eu/Eu*	0.70	0.76	0.75	0.73		
La/Yb	11.93	22.69	6.47	7.76		

TABLE 1. Whole-rock major, trace and rare earth of profile P1 weathered crust and related to parent rock

 $\overline{\text{Ce/Ce}^* = \text{Ce}_N/(\text{La}_N \times \text{Pr}_N)^{1/2}}$, $\text{Eu/Eu}^* = \text{Eu}_N \times (\text{Sm}_N \times \text{Gd}_N)^{1/2}$, where subscript N represents normalization by C1- chondrite (Sun & McDonough 1989)

Sample#	P2 S0m	P2 S5m	P2 R6m
Rock Type	Laterite	Laterite	Weathered Andesite
SiO ₂ (wt.%)	24.60	29.20	37.50
TiO ₂	3.22	2.78	2.20
Al ₂ O ₃	26.50	25.50	19.50
Fe ₂ O ₃	22.90	19.10	15.40
MnO	0.04	0.24	0.21
MgO	0.20	0.99	3.73
CaO	0.01	0.01	1.15
Na ₂ O	0.01	0.17	0.83
K ₂ O	0.02	0.30	1.31
P_2O_5	0.08	0.11	0.19
LOI	18.40	19.10	16.40
Total	95.98	97.5	98.42
CIA (%)	100	98	86
La (ppm)	1.71	298.00	36.30
Ce	35.00	121.00	83.40
Pr	0.38	64.40	7.55
Nd	2.19	353.00	43.90
Sm	0.53	58.80	10.00
Eu	0.15	16.30	2.89
Gd	0.71	61.20	11.70
Tb	0.27	8.76	1.78
Dy	1.76	51.60	10.70
Но	0.37	10.30	2.32
Er	0.92	26.20	5.52
Tm	0.17	3.93	0.88
Yb	0.99	23.30	5.50
Lu	0.17	3.57	0.82
Y	1.39	279.00	54.40
Sc	22.9	42.90	43.10
LREE	62.86	954.40	227.14
HREE	6.75	467.86	93.62
TREE	69.61	1422.26	320.76
Ce/Ce*	10.65	0.21	1.24
Eu/Eu*	0.75	0.83	0.82
La/Yb	1.73	12.79	6.60

TABLE 2. Whole-rock major, trace and rare earth of profile P2 weathered crust and related to parent rock

 $\frac{1}{\text{Ce/Ce}^* = \text{Ce}_N/(\text{La}_N \times \text{Pr}_N)^{1/2}, \text{Eu/Eu}^* = \text{Eu}_N \times (\text{Sm}_N \times \text{Gd}_N)^{1/2}, \text{ where subscript N represents normalization by C1- chondrite (Sun & McDonough 1989)}}$

TABLE 3. The result of a single-step extraction reveals the extracted element concentration (in ppm) and percentage of the whole rock content

Sample #	Fraction	LREE		HREE		REY		Th		U		Ce/	Eu/	La _N /
		ppm	%	ppm	%	ppm	%	ppm	%	ppm	%	Ce*	Eu*	Yb_N
P1 S4m	Ion Exchangeable	777.1	74.88	407.6	84.26	1184.7	77.86	n.d	n.d	0.01	0.46	0.05	0.86	11.82
P2 S5m	(extracted cations by 0.5 mol/L (6.6%)	881.3	79.22	873.4	88.91	1754.7	83.76	n.d	n.d	0.03	0.68	0.06	0.80	6.12
	ammonium sulphate determined by ICPMS													

 $\overline{\text{Ce/Ce}^* = \text{Ce}_N/(\text{La}_N \times \text{Pr}_N)^{1/2}, \text{Eu/Eu}^* = \text{Eu}_N \times (\text{Sm}_N \times \text{Gd}_N)^{1/2}, \text{ where subscript N represents normalization by C1- chondrite}}$ (Sun & McDonough 1989)



FIGURE 3. (a) Percentages of extracted LREE, HREE, REY concentration relative to their whole-rock content; (b) percentage of radioactive elements (Th and U) fraction through single step extraction; (c) XRD diffractogram of soil sample P1 S4m and (d) XRD diffractogram of soil sample P2 S5m



FIGURE 4. (a) Backscattered electron image (BSE) of one frame of an automated scanning electron microscopy-mineral liberation analysis (SEM-MLA) measurement of a polished thin section for sample P1 S4m. The BSE image display a very fine grain sample (<75 micron). (b) Classified grouped and color-coded presentation of the frame in (c) in an automated SEM-MLA measurement, where the legend shows the percentages of mineral detected through the analysis and related to the whole sample area. SEM back-scattered electron images of the sample P1 S4m. Occurrences of (d) kaolinite which is rich in iron (Fe) content. The sample also exhibits a very fine grain (e) goethite where the occurrence is not in single phase. Due to the fine grain of sample, mixture of (f) goethite and kaolinite is encountered with goethite is more dominant in the weathered product of profile P1. Moreover, some parts of the sample shows the occurrences mixture of (g) kaolinite, goethite and rutile as the EDS show the



FIGURE 5. (a) Total alkali-silica diagram of Le Bas et al. (1986), showing the position of the samples from this study and Taniou et al. (2019); (b) Classification of weathering product of the area (after Aleva & Creutzberg 1994) and CIA ternary diagram plotting for (c) profile P1 and (d) profile P2, showing the degree of argillation from the bedrock to the laterite weathering profile (after Fedo, Nesbitt & Young 1995; Nesbitt & Young 1982)

The process of lateritic weathering plays a crucial role in the formation of specific deposits of REE, including the ion adsorption clay deposit located in South China and the Mount Weld deposit in Australia (Sanematsu & Watanabe 2016; Verplanck 2017). The ternary classification by Aleva and Creutzberg (1994) indicates that the majority of the soils from both of the weathering profiles fall in the zone of laterite, whereas one sample fall in the kaolinite zones (Figure 5(b)). Laterization often leads to a decrease in SiO₂ and an increase in Al₂O₃ and Fe₂O₃ concentration (Bárdossy & Aleva 1990). The major oxide composition data indicates an increase in the percentage content of Fe₂O₃ and Al₂O₃ from the bedrock to the near surface weathering product, while the percentage value of SiO, decreased accordingly. This observation is consistent with the established trends in the field.

In general, a reduction in the concentration of MgO, CaO, Na₂O, and K₂O was observed across both weathering profiles as the parent rock transitioned to the weathered layer with an increased weathering process. According to Middelburg, van der Weijden and Woittiez (1988), due to its soluble and immobile nature throughout the weathering process, this element tends to show a decrease in the yield of the weathering product compared to its bedrock. Thus, it suggests the elements are partitioning into the aqueous phase, followed by their subsequent removal from the system through the leaching process. In contrast to the two primary oxide contents, Al₂O₂ which increases from the bedrock to the weathering layer and may be generating other minerals in the secondary phases such as goethite, hematite and various clay minerals (Chengyu, Dianhao & Zhongxun 1990; Verplanck 2017). The statement is confirmed by the result of the SEM-MLA and XRD examination of minerals, wherein the sample (P1 S4m) exhibiting a high concentration of REE indicates the existence of Fe oxyhydroxide minerals that are naturally abundant in the lateritic layer. This also explains why these two oxides have a tendency to become immobile and residual elements during the weathering process, causing their concentration to increase as a result of bedrock weathering. The presence of phosphate minerals carrying REE in a sample can be inferred from the chemical composition of P2O5. Thus, REE accumulation in ion-adsorption clay and REE differentiation in weathering profiles are both significantly influenced by the weathering of phosphate minerals such as apatite, monazite and xenotime (Sanematsu, Kon & Imai 2015). However, from the interpretation of the phosphate oxide data, the concentration in the bedrock is not very different from the amount of this oxide in laterized soil. This suggests that this oxide was only slightly dissolved and moved outward by the process of laterization.

Overall, the CIA is usually higher than 90% in the upper part of the weathered profile and lower than 90% in the lower part for weathered and fresh rock. It is known that feldspar is the most abundant mineral in the upper continental crust. The CIA values for unaltered feldspars such as anorthite, albite and orthoclase are 50%. Furthermore, CIA values of 55% and 86%, respectively, are observed in the two rocks sample for P1 R7m and P2 R6m. According to Sanematsu et al. (2009), kaolinite a common clay mineral in weathered crusts, has a CIA value of 100%. In relation to this, CIA values for the P1 and P2 profiles suggest that most of the weathering laterite profiles of andesitic are highly weathered and enriched in clay minerals. Thus, in this study, the CIA values of the parent rock and the weathered crusts range 50 - 100%, indicating the degree of argillation from feldspar to kaolinite as illustrated in Figure 5(c) and 5(d). In addition, SEM-MLA and XRD analysis have also showed that the kaolinite mineral is the most dominant with gibbsite occurring as a minor mineral in the laterite sample that is rich in REE.

REE DISTRIBUTION IN THE ANDESITIC DERIVE LATERITE OF SERIAN VOLCANIC

The degree of weathering, which is controlled by the depth from the surface, has a substantial impact on the vertical variation in REE concentration in a weathering profile. The graphic illustration in Figure 6(a) and 6(b) illustrates the correlation among the concentrations of Ce, LREE, HREE, and Y in profiles P1 and P2, respectively. The mobilization of REE during the process of weathering is influenced by various factors associated with the mineralogy of the parent rock. These factors include the distribution of REE in the primary bearing minerals, the stability of these minerals during weathering, and their abundance in the parent material, as highlighted by Duddy (1980) and Nesbitt (1979). Thus, the concentration of the REE content demonstrates a rising trend in correspondence with the depth of the weathering profile. Specifically, profile P1 displays a TREE of 1715.98 ppm at a depth of 4 meters, while for profile P2, at a depth of 5 meters, the TREE reaches 1422.26 ppm. However, TREE decreased significantly in both profiles toward the upper part of the profile. The REE enrichment at both depths of the profiles is likely caused by the presence of absorbent materials, such as clay, or the concentration of REE-bearing minerals in the horizons. The findings obtained from the SEM-MLA analysis suggest that the primary components of clay minerals, namely kaolinite (with a 7% Fe content), along with other minerals such as goethite, gibbsite, and illite, function as absorbent materials in the P2 profile with the assumption that REE and Y are adsorbed on variable oxide surfaces, such as hematite and magnetite, which are the abundant Fe oxides and Fe oxyhydroxides in laterite and may adsorb REE (Sanematsu et al. 2011). However, no REE minerals were found through the SEM-MLA and XRD analyses which suggests that most of the minerals in the soil sample are dominated by weathering product minerals such as kaolinite and others. Furthermore, the significant abundance of element Y (HREE) observed in both profiles could potentially be attributed to the existence



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FIGURE 6. Ce, LREE, HREE and Y concentration of (a) profile P1 and (b) profile P2 weathered crusts of andesitic and related parent rock and Chondrite-normalized REE diagram of the weathered crust of andesitic and related parent rock of (c) profile P1 and (d) profile P2. Values of chondrite are form C1-chondrite Sun and McDonough (1989)

of minerals containing Y within the bedrock formation. The aforementioned phenomenon occurs as a result of the process of weathering of the parent rock, whereby the element Y, in its Y^{3+} form, becomes adsorbed onto the surface of the clay, leading to an enrichment of the horizon with this particular element (Chi et al. 2005).

Figure 6(c) and 6(d) illustrates the chondrite normalized diagram of the weathered crust of andesitic and related parent rocks of the profiles P1 and P2, respectively. The CI-chondrite normalized pattern (Sun & McDonough 1989) shows an almost flat pattern of andesitic, implying that the parent rock had equally abundant LREE and HREE, with the exception of the Y element for both profiles. On profile P1, the pattern shows non-significant Ce anomalies throughout the profile, with Ce anomalies ranging from 0.19 to 2.87 and moderately strong negative Eu anomalies ranging from 0.70 to 0.76. On the other hand, profile P2 shows a wide range of Ce anomalies ranging from 0.21 to 10.65 and moderately strong negative Eu anomalies ranging from 0.75 to 0.83. Overall, the laterite profile of weathered andesitic rocks is enriched in REE relative to the parent rock. The La/Yb ratio analysis indicates that the LREE fractionation is more obvious than HREE (except

for the Y element) on the weathering profile, as proven by the higher readings of 22.69 (profile P1) and 12.79 (profile P2).

Profile P1 exhibits a positive Ce anomaly in the upper part of the profile, with Ce/Ce* anomaly of 2.87. Conversely, the lower part of the profile displays a negative anomaly, with a Ce anomaly of 0.19. With the exception of Ce, the upper section of the profiles exhibits a depleted amount of REE, whereas the lower section demonstrates a tendency towards enrichment in REE. In contrast to the enrichment trend observed on profile P2, the upper layer of the soil exhibits predominantly positive anomaly values (Ce/Ce*=10.65) with a negative Ce anomaly value (Ce/ $Ce^*= 0.21$) detected in the weathered profile. The lower part of the weathered profile exhibits a higher concentration of REE in comparison to the upper part. The upper section of both profiles exhibits a notable concentration of Ce relative to the whole rock composition indicating that the layer contributes significantly to the enrichment of LREE. The relationship of Ce with cerianite (CeO_2) is an acceptable explanation for the observed phenomenon, as it is distinguished by its inherent properties, which render it extremely immobile and infrequently extracted from

the leaching zone during the process of weathering. In addition, the genesis of cerianite is linked to the process of weathering, whereby Ce and other REE undergo fractionation, leading to the formation of cerianite in the soil horizon (Abedini, Khosravi & Dill 2020; Sanematsu et al. 2013; Verplanck 2017; Yaraghi, Ariffin & Baharun 2020). Moreover, it has been demonstrated by Bao and Zhao (2008) and Chi et al. (2005) that cerium exhibits dissimilar behavior in comparison to other REE within the weathering profile, leading to positive anomalies in the upper horizon.

Meanwhile, there is no significant amplitude in the Eu anomaly throughout both of the weathered profiles relative to the parent rock. Negative Eu anomaly values have been observed in every sample collected from both profiles. This is consistent with a study conducted by Rudnick and Gao (2003) that indicates an negative Eu anomaly characterizes the upper crust of the earth. Through observation, this constant negative anomaly value indicates that it remains stable even under severe weathering conditions (CIA value = 99 - 100%) in both profiles. In addition, the presence of a negative Eu anomaly suggests that during the process of weathering, a phase containing Eu, such as plagioclase, is selectively dissolved (Abedini, Khosravi & Dill 2020). Thus, the similar Eu anomaly suggests that andesitic rock is the source of the laterite profiles.

REE POTENTIAL IN ANDESITIC DERIVED LATERITE

According to a study conducted by Moldoveanu and Papangelakis (2012), it was observed that NH⁴⁺ facilitates greater exchange of REY as compared to Na⁺ from ion adsorption ores. Consequently, in order to verify the nature of the acquired sample as ion adsorption ores, an analytical assessment of the ion-exchangeable fraction is conducted on the samples exhibiting a significant concentration of REY. According to Sanematsu and Kon (2013), the concentration of extracted REY through single-step ion exchangeable analysis is subject to influence not only by the exchangeable cations such as NH4⁺ and Na⁺, but also by the reagents including sulphate, acetate, and chloride.

Through the analysis, the analytical data for both samples (P1 S4m and P2 S5m) indicate that the majority of REY may be present in the ion-exchangeable fraction. Ion-exchangeable REY for sample P1 S4m is 1184.7 ppm (77.86%) relative to whole rock content. In comparison to the P1 S4m sample, the P2 S5m sample shows a notably higher content of ion-exchangeable REY, quantifying at 1754.7 ppm (83.76%) relative to the whole rock content. In addition, as compared to the P1 S4m, the P2 S5m sample shows a very high ion-exchangeable content in HREE and a very low La_N/Yb_N ratio. The whole rock analysis findings also demonstrate the sample's response to HREE fractionation as well as the laterite profile response to HREE enrichment relative to the whole rock content on the chondrite normalized REE diagram for P2. The depleted Ce anomaly relative to the whole rock also suggests the

area has potential for the occurrences of ion-adsorption ore in the area.

The thorium and uranium were also ion-exchanged by the ammonium sulphate solutions. Through the extraction analysis, thorium was not detected in both samples. Meanwhile, for uranium, they account for 0.46% (0.01 ppm) and 0.68% (0.03 ppm) for samples P1 S4m and P2 S5m, respectively. Furthermore, the two samples stated earlier show extremely low content relative to whole rock composition, with less than one becquerel (Bq/g) for Th (~ 250 ppm) and U (~80 ppm).

Sarawak is a region characterized by a tropical climate that is situated along the equator. The region is characterized by a tropical climate that is accompanied by elevated levels of atmospheric moisture. According to the Sarawak Government Portal Website (http://www. did.sarawak.gov.my), the annual precipitation in the area varies from 3,300 to 4,600 millimetres, while the average daily sunshine duration is approximately five to six hours. This criterion was given priority in promoting the region as a more suitable location for the conservation of ion-adsorption-type rare earth element deposits in tropical climate.

CONCLUSION

The Serian Volcanic of Late Triassic in the Rih Serian, West Sarawak, Malaysia indicates the possibility of REE resources in ion-adsorption clay deposits. Through rock classification, it was found that basaltic trachyandesite rocks are part of the bedrock that underlies the study area. This rock shows a TREE concentration of 209.53 ppm and can be enriched up to 1715.98 ppm in the laterite layer that has been studied.

The REY fractionation that occurred in the study area is subjected to a highly or deep weathering process with CIA values ranging from 98 to 100%. This indirectly increases the adsorption of REY on adsorbent materials such as kaolinite, which dominates the mineral composition of the laterite samples analyzed. Illite and gibbsite are additional minerals that have been identified and could potentially contribute to the adsorption percentage in the weathering profile. The almost flat pattern of chondrite-normalization suggests that the enrichment for LREE and HREE are almost equally abundant from the parent rock into the enriched REY layer for both of the profiles studied. The high yttrium element concentration that contributes to the enrichment of HREE might be influenced by the mineral content of the bedrock.

Through the analysis of single-step extraction, most of the REY is present in the ion-exchangeable fraction. REY extraction percentages exceed 75% relative to the wholerock content for both samples, suggesting that the region has the potential for ion-adsorption ore that preserves the REY in terms of resources. The Serian Volcanic Formation, which occupies an area of nearly 510 km², exhibit promising indications of being a viable source of REE for Sarawak, Malaysia.

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