

REE Comparison Between Muncung Granite Samples and their Weathering Products, Lingga Regency, Riau Islands

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Abstract - The increasing demand for Rare Earth Elements (REE) is related to the continous development of technology, and these elements are used in modern equipments. REE can occur in igneous and sedimentary rocks in significant amounts as primary deposits, whereas the secondary REE deposit can be produced by intensive lateritic weathering of bedrocks under the tropical or subtropical climate. Lateritic process can increase REE concentration from sub-economic levels in host rocks to be more valuable. Muncung Granite is located in a tropical area of Lingga Regency, Riau Islands Province. REE occurs in the Muncung Granite and in weathered layers (saprolite, laterite, and soil). ICP-MS was applied to measure the REE content in all samples of this study. The average REE content of the Muncung Granite is 265 ppm with Eu anomaly in REE's spider diagrams. Lateritization process has increased REE content by more than four times compared to that in the Muncung Granite. Ce and Eu anomalies in weathered layers can be associated with weathering process and initial REE contents in the host rock. Ce anomaly in a laterite layer is found to have a negative correlation to REE total enrichment. The REE level in the Muncung Granite is higher than the content in the soil and saprolite layers, but lower than that in the laterite.

Keywords: rare earth elements, Muncung Granite, weathered layers, Ce and Eu anomalies

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How to cite this article:

Irzon, R., Syafri, I., Hutanarat, J., and Sendjaja, P., 2016. REE Comparison Between Muncung Granite Samples and their Weathering Products, Lingga Regency, Riau Islands. *Indonesian Journal on Geoscience*, 3 (3), p.149-161. DOI: 10.17014/ijog.3.3.149-161

INTRODUCTION

Background

Rare earth elements (REE) deposits can broadly be divided into two categories: primary and secondary deposits. Primary deposits are associated with igneous rocks which are relatively rich in Light-REE (LREE), whilst secondary deposits are concentrated by weathering processes, also known as lateritization, which contain more Heavy-REE (HREE) (Bao and Zhao, 2008; Bongaerts and Liu, 2013). Primary minerals containing REE in pri-

Accredited by: - LIPI, valid August 2016 - August 2021 - RISTEKDIKTI, valid May 2016 - May 2021 mary deposits may include bastnäsite, monazite, xenotime, eudialyte, allanite, zircon, apatite, and a variety of more uncommon minerals (Cocker, 2012). Laterite, the soil-like layer formed during intense weathering of bedrock under tropical or sub-tropical (*i.e.* hot and humid) conditions, now becomes important sources of viable economic mineral deposits. Lateritic weathering results in the breakdown of primary rocks and minerals, and concentration of metals such as aluminum, iron, manganese, nickel, gold, phosphorous, and niobium (Cocker, 2012).

Lateritic weathering, also known as supergenetype deposit, increases concentration of REE from above background or sub-economic levels to values that are considered to be economic. Aluminous micas are altered to bauxite while olivines are altered to nickel-rich smectite (Retallack, 2010; Cocker, 2012). Several authors have described the lateritization relation to REE elevation especially in Asia (Bao and Zhao, 2008; Sanematsu et al., 2009; Bongaerts and Liu, 2013). HREE that have been extracted from granites lateritization in South China are strongly demanded for magnet market in recent years and used in energy conservation (Ishihara, 2008). Primary minerals can be decomposed by chemical weathering of rocks in the continent and many elements can be activated. Some elements become leached (e.g. Mg, Ca, and Si), others either are secondarily enriched (e.g. Mn, Co, Zn, Y, and Ni) or residually concentrated (e.g. Fe, Cr, Al, Ti, and Zr) within laterite profiles during weathering (Sagapoa et al., 2011). It was difficult to extract rare earth elements from laterites, but recent metallurgical methods allow economic recovery of these elements to become possible. Lateritic deposits are now being explored for and developed at a faster pace than primary hard rock REE deposits.

Riau Islands Province consists of thousands of islands, located in east of Sumatra mainland and northwest of Bangka Island. Lingga is a regency in Riau Islands Province, Indonesia, with Daik City as its capital. This area is affected by dry and rainy seasons. Lingga, Singkep, and Selayar is the three main islands in the regency. The range of yearly rainfall in Riau Islands Province is between 1,000 to 3,000 mm. The average air temperature in the region is 28° C with maximum and minimum temperature are 36°C and 21°C respectively. The S-type Muncung Granite and the I-type Tanjungbuku Granite are the two granitoid units in Lingga Regency. The S-type granite of the studied area is related to tin resources of the regency (Barber et al., 2005; Suprapto, 2008; Jaenuddin et al., 2011). The latest study revealed that Muncung Granite could be divided into A and B facies based on geochemistry data: SiO₂ versus other major oxides, and REE's pattern. Irzon (2015) stated that the B facies is more peraluminous than

A facies, confirming the more S-type character. Mineral existence in Singkep Island has attracted many investors where thirteen mining companies are still active there (Mamengko, 2013).

The objective of this paper is to compare REE concentrations in the Muncung granitoid host rocks to the content in weathered layers. A couple of anomalies draw deeper about petrogenesis of the plutons and their weathering process. This work would add more information for the assessment of REE resource potentials in Indonesia.

Geological Setting

The general geology of this region consists of five lithological units (Sutisna et al., 1994; Irzon, 2015): (1) Permian Persing Complex and Duabelas Bukit Quarzite; (2) Triassic granite and granodiorite of Muncung Granite; (3) Jurassic Tanjungbuku Granite (comprises granite and granodiorite), and Tanjungdatuk Formation (low grade methamorphic rock composed of slate and quartz veinlets); (4) Tengkis Formation, Pancur Formation, and Semarung Formation formed in Cretaceous age; (5) Quaternary Alluvium and Swamp Deposits. The S-type Muncung Granite spreads in the southwestern area of Lingga Island, west part of Selayar Island, and in the central region of Singkep Island. The I-type Tanjungbuku Granite is located in the southwestern part of Singkep Island (Cobbing et al., 1992). The geological map and sampling points are presented in Figure 1.

Sample Description Granitic Rock Samples

From three islands of Lingga Regency: Lingga, Singkep, and Selayar, a total of seven granitoids as host rock samples were collected. All granitoids in this study constitute Triassic Muncung Granite which comprises granite and diorite (Sutisna *et al.*, 2004). All samples are composed of quartz, K-feldspar, plagioclase, hornblende, biotite, and or muscovite with small amounts of accessory minerals. Plagioclase and biotite may partly be sericitized and chloritized, respectively, by alteration in most of the samples. The studied area is an important tin resource of Indonesia although PT Timah industrial mining

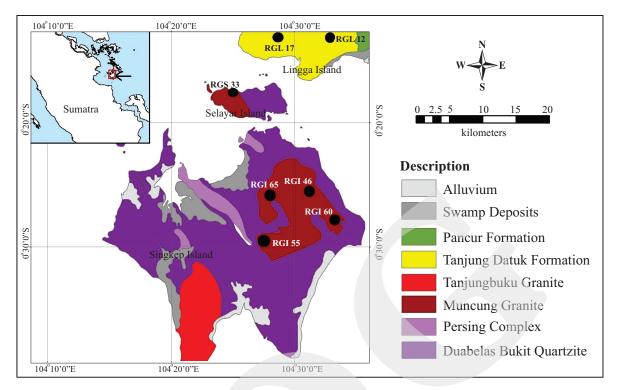


Figure 1. Geological map of studied area and sampling points in Lingga Regency (modified from Sutisna et al., 1994).

activity in Lingga Regency was stopped in 1993 (Suprapto, 2008).

Two and one granitoid samples were collected from Lingga Island and Selayar Island, respectively. Two granitoids from Kelume (RGL 12 and RGL 17), northeastern Lingga Island, are holocrystalline, phaneric medium-grained. Besides quartz, K-feldspar, plagioclase, and hornblende, RGL 12 also contains biotite, while RGL 17 contains muscovite. The sample from Selayar was a holocrystalline, medium-grained, and phaneritic granitoid boulder without hornblende (RGS 33).

Singkep Island pluton samples were obtained from Singkep Timur (RGI 46), Bukit Tumang (RGI 65), and Maroktua (RGI 55 and RGI 60). All samples comprise quartz, K-feldspar, plagioclase, biotite, and or muscovite. Note that no hornblende was found in these four samples. Petrology data confirm that RGI 60 and RGI 65 consist of xenolith of 2% and 5%, respectively. Another interesting fact is that RGI 65 contains muscovite of 12%.

Weathered Rock Samples

The ideal outcrop consists of soil, laterite, saprolite, and host rock layers (A, B, C horizons

in addition to host rock) as illustrated in Figure 2. It was not easy to find this kind of outcrop. Only soil layer and granitoid samples could be obtained in RGS 33, RGI 55, and RGI 60 because the host rocks were revealed just near the surface. Granitoid and other two weathered layers were

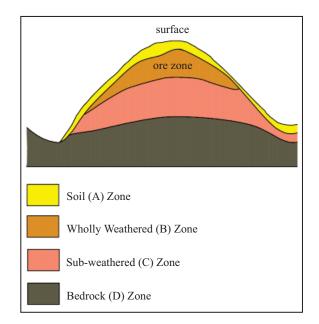


Figure 2. An idealized weathering profile for REE deposits. (Adapted from Cocker, 2012).

observed in RGL 15 (soil and laterite) and RGL 17 (saprolite and laterite) along a new road construction in Kelume. Eventhough soil, laterite, and granitoid can be observed in RGL 12, it was not easy to sample a clear saprolite layer. Actually, RGI 46 outcrop is just near the top layer but saprolite layer can still be distinguished from the host rock. RGI 65 posseses the thickest laterite layer compared to others, almost 3 m thick.

Generally, the soil layers (A horizon) are dark brown to reddish dark brown soil, have the thickness ranging from 10 to 50 cm. This top layer is enriched in organic matters and clay minerals, whereas primary rock-forming minerals are not recognized. The laterite layers (B horizon) are light brown, brown or reddish brown in colour and show no granitic texture in appearance, with biotites are scarcely present. RGL 17 shows a different colour for laterite, *i.e.* greyish brown. The B horizon is enriched in clay minerals with less organic matter. The saprolite layers (C horizon) are slightly weathered but fragile exhibiting gray, light brown or brown in colour. Granitic texture and rock-forming minerals are readily recognized although they are apparently altered in this saprolite layer. Outcrops in some locations are presented in Figure 3.

Analytical Method

The most important work after collecting the samples in the field is to separate the weathered rock samples from the relatively fresh one. After being dried at a room temperature, the samples were crushed using jaw crusher to 200 mesh and were grounded by a mill. The whole rock analysis was chosen here besides mineral analysis and the results are presented in Tabel 2. The REE contents in each samples were measured using the



Figure 3. a) A new road construction exposes granitoid and weathered layers; b) Saprolite (C horizon) of RGI 46; c) RGI 55: granitoid from Maroktua; d) A horizon of RGI 60 which contains roots as organic matter.

X-Series Thermo Inductively Coupled Plasma Mass Spectrometry (ICP-MS). Before the ICP-MS measurement, rock samples were dissolved with three acid leaching: nitric acid (ultrapure grade), formic acid (ultrapure grade), and perchloric acid (pro-analysis grade). AGV-2 and GBW 7110 are used as standard reference materials. Sample preparation, ICP-MS set up procedure, and certified reference evaluation are based on the previous study by Irzon and Permanadewi (2010). Both the preparation and instrumental analyses were done at Pusat Survei Geologi (Centre for Geology Survey of Indonesia) in Bandung.

Result and Discussion

Petrography of Granitoids

Six of seven petrography data of selected granitoid samples have been previously featured by Irzon (2015), except RGI 65. The granites comprise 30-42% quartz, 27-43% K-feldspar, 6-22% plagioclase, <3% hornblende, <12% muscovite, <7% biotite, and minor of opaque mineral as phenocryst (Table 1). Various amounts of secondary minerals of sericite and chlorite are present as biotite replacement. In samples from Lingga and Selayar Islands (RGL 12, RGL 17, and RGS 33), quartz occurs as anhedral crystals and grew together with K-feldspar (orthoclase and sanidine) to form a micrographic texture. Most plagioclase from these two islands are coarse grains and both albite and oligoclase are present. Note that RGL 17, RGI 46, and RGI 55 contain the most sericite, plagioclase, and quartz than any other granitoids, respectively. The major difference of samples from Singkep Island is the presence of xenoliths. The xenoliths consist of schist and gneiss in RGI 60 (2%) and RGI 65 (5%), which could become an important clue to the source of the granitoids. These petrography variations would add more contrasts about two facies of Muncung Granite in addition to geochemistry differences as discussed by Irzon (2015). Microphotograps of selected samples are shown in Figure 4. After all, the samples could be categorized as fresh granites although minor alteration are detected.

REE Content in Granitoids and the Weathered Layers

Polewali and Mamasa Granites (Maulana et al., 2012) are chosen for comparisons of REE

Table 1. Petrographic Data of Granitoid Samples. All Data except RGI 65 are from Irzon (2015). RGL 12 and RGL 17 were Taken from Lingga Island. RGS 33 was taken from Selayar Island. RGI 46, RGI 55, RGI 60 and RGI 65 were from Singkep Island. *= Data from the Previous Study of Irzon (2015)

		RGL 12	RGL 17	RGS 33	RGI 46	RGI 55	RGI 60	RGI 65
Phenocryst								
Quartz	%	33	29	31	40	41	37	30
K-Feldspar	%	43	29	36	32	28	34	42
Plagioclase	%	12	22	18	18	12	12	6
Hornblende	%	1	3	-	-	-	-	-
Muscovite	%	1	2	2	-	-	-	12
Biotite	%	-	1	1.5	2	7	6	-
Ore mineral	%	-	2	2	0.5	0.5	1	1
Alteration Mineral		•••••••••••••••••••••••••••••••••••••••	•••••••••••••••••••••••••••••••••••••••		•	•	•	••••
Sericite	%	7	9	8	6	6	6	2
Chlorite	%	1	2	0.5	0.5	0.5	0.5	1
Secondary quartz	%	1	-	-	-	4	1	-
Porosity	%	1	1	1	1	1	0.5	1
Xenolith	%	•••••••	•		•	•	2	5
Rock Name		Granite	Granite	Granite	Granite	Granite	Granite	Granite

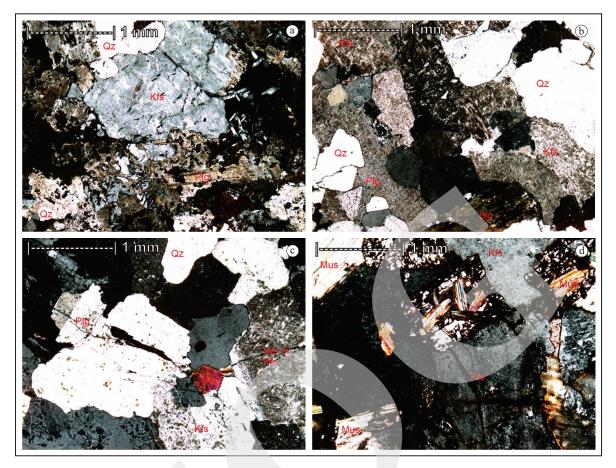


Figure 4. Photomicrographs of Muncung Granite Samples: a) RGL 17; b) RGS 33; c) RGI 46; d) RGI 65. Qz = quartz, Plg = plagioclase, Kfs = K-feldspar, Bio = biotite, Mus = muscovite.

contents to other granitoids in Indonesia. The weathering process on Longnan Granite raised the REE content of 300 - 400 ppm in the host Longnan Granite to more than three times of 1,000 - 1,500ppm. The weathered crust of Longnan Granite is a significant HREE deposit in China and has been compared to Sn-bearing granitoid rock from southern Peninsula Thailand (Imai et al., 2008). Muncung Granite is located in Lingga Regency as one of the three main Sn resource locations in Indonesia besides Bangka and Belitung (Irzon, 2015) which make Longnan Granite becomes a suitable comparison for this study. The total REE in the seven granitoid samples in this study ranges from 128 to 596 ppm, with the average of 265.49 ppm (Table 2). This value is relatively higher than the granitoid from Polewali and Mamasa (191 to 279 ppm), but just below the range of host granitoids from Longnan (300 to 400 ppm). Moreover, the average REE in three samples from

Lingga and Selayar Islands (374 ppm in average) is higher than four granitoids from Singkep Island (183 ppm in average) with RGL 33 posesses the bigest amount of REE (>590 ppm).

Enrichment or depletion of REE occurs because these elements are mobile and they are fractionated during weathering (Gong et al., 2011). The relative enrichment of REE in the weathering products in relation to bedrock is pronounced in laterite layer (B horizon) which could be up to 110 (Cocker et al., 2012). In Muncung Granite, the REE content in laterite of RGI 63 increases massively to more than four times than the granitoid layer, i.e. from 190 to 810 ppm. The REE enrichment in laterite layers in RGL 12 and RGL 17 are below two times with respect to their host rocks, *i.e.* from 266 to 440 ppm and from 260 to 294 ppm, respectively. Saprolite and soil layers show REE depletion compared to granitoid host rocks in the studied area. Saprolite's REE

Table 2. REE Content of Granitoids and Weathered Layers form Muncung Granite Domain. H is Granitoid as Host Rock. A, B, and C are Soil, Laterite, and Saprolite Layer, Respectively
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ppm 10.99 7.18 2.35 10.11 2.84 3.23 19.64 1.89 7.26 4.11 1.66 3.06 1.12 8.88 0.34 ppm 1.66 0.85 0.35 0.35 1.45 0.38 0.28 2.75 0.25 1.39 0.61 0.21 0.52 0.18 1.50 0.08 ppm 10.53 5.03 2.15 8.42 2.15 1.67 1.675 0.28 0.77 4.00 1.13 2.74 0.94 8.10 0.54 ppm 2.12 1.00 0.46 1.59 0.45 0.34 3.05 0.36 0.21 0.76 0.28 1.67 0.14 ppm 6.06 2.86 1.45 1.12 8.52 1.09 6.11 2.67 0.22 0.13 1.67 0.16 0.16 0.22 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 0.16 </th <th>Eu</th> <td>bpm</td> <td>1.70</td> <td>1.01</td> <td>0.26</td> <td>2.04</td> <td>0.42</td> <td>0.29</td> <td>0.56</td> <td>0.08</td> <td>0.18</td> <td>0.37</td> <td>0.04</td> <td>0.33</td> <td>0.07</td> <td>0.25</td> <td>0.01</td> <td>0.25</td> <td>0.15</td> <td>0.16</td>	Eu	bpm	1.70	1.01	0.26	2.04	0.42	0.29	0.56	0.08	0.18	0.37	0.04	0.33	0.07	0.25	0.01	0.25	0.15	0.16
ppm1.66 0.85 0.35 1.45 0.38 0.28 2.75 0.25 1.30 0.61 0.21 0.52 0.18 1.50 0.08 ppm 10.53 5.03 2.15 8.42 2.15 1.67 16.05 1.68 9.77 4.00 1.13 2.74 0.94 8.10 0.54 ppm 2.12 1.00 0.46 1.59 0.45 0.34 3.05 0.36 0.23 0.66 0.21 1.67 0.14 ppm 6.06 2.86 1.45 4.12 1.44 1.12 8.52 1.09 6.11 2.67 0.73 1.46 0.55 4.08 0.36 ppm 0.91 0.43 0.25 0.65 0.24 0.17 1.24 0.19 0.91 0.72 0.13 1.67 0.73 0.63 0.63 0.63 ppm 0.91 0.43 0.25 0.65 0.24 0.17 1.24 0.94 0.91 0.72 0.13 0.63 0.63 ppm 0.91 0.43 0.25 0.66 0.24 0.14 0.12 0.14 0.26 0.91 0.61 0.72 0.13 0.63 0.63 ppm 0.92 0.40 0.25 0.66 0.22 0.16 0.12 0.12 0.14 0.26 0.91 0.75 0.12 0.13 0.63 0.66 0.25 0.13 0.26 0.92 0.92 0.92 0.92 <th>Gd</th> <td>mqq</td> <td>10.99</td> <td>7.18</td> <td>2.35</td> <td>10.11</td> <td>2.84</td> <td>3.23</td> <td>19.64</td> <td>1.89</td> <td>7.26</td> <td>4.11</td> <td>1.66</td> <td>3.06</td> <td>1.12</td> <td>8.88</td> <td>0.34</td> <td>5.72</td> <td>24.23</td> <td>4.34</td>	Gd	mqq	10.99	7.18	2.35	10.11	2.84	3.23	19.64	1.89	7.26	4.11	1.66	3.06	1.12	8.88	0.34	5.72	24.23	4.34
ppm 10.53 5.03 2.15 8.42 2.15 1.67 16.05 1.68 9.77 4.00 1.13 2.74 0.94 8.10 0.54 ppm 2.12 1.00 0.46 1.59 0.45 0.34 3.05 0.36 0.23 0.56 0.21 1.67 0.14 ppm 6.06 2.86 1.45 4.12 1.12 8.52 1.09 6.11 2.67 0.73 1.46 0.55 4.08 0.36 ppm 0.91 0.43 0.25 0.65 0.24 0.17 1.24 0.19 0.94 0.43 0.12 0.63 0.63 0.63 ppm 0.91 0.43 0.25 0.65 0.24 0.17 1.24 0.19 0.94 0.12 0.12 0.63 0.63 0.63 0.91 0.91 0.43 0.25 0.65 0.24 0.17 1.24 0.19 0.12 0.12 0.13 0.63 0.63 0.91 0.91 0.43 0.25 0.66 0.23 0.26 0.91 0.72 0.13 0.63 0.63 0.63 0.82 0.40 0.26 0.59 0.24 0.12 0.12 0.12 0.12 0.12 0.12 0.12 0.12 0.12 0.13 0.65 0.06 0.06 0.92 0.40 0.26 0.29 0.21 0.12 0.21 0.12 0.12 0.12 0.12 0.12	Tb	mqq	1.66	0.85	0.35	1.45	0.38	0.28	2.75	0.25	1.39	0.61	0.21	0.52	0.18	1.50	0.08	1.03	3.77	0.69
ppm 2.12 1.00 0.46 1.59 0.45 0.34 3.05 0.36 2.03 0.86 0.23 0.56 0.21 1.67 0.14 ppm 6.06 2.86 1.45 4.12 1.44 1.12 8.52 1.09 6.11 2.67 0.73 1.46 0.55 4.08 0.36 ppm 0.91 0.43 0.25 0.65 0.24 0.17 1.24 0.19 0.43 0.12 0.55 4.08 0.36 ppm 5.76 2.73 1.65 4.06 1.20 8.36 1.44 6.33 2.86 0.88 1.39 0.63 0.36 ppm 5.76 2.73 1.65 4.06 1.20 8.36 1.44 6.33 2.86 0.88 1.39 0.63 0.63 0.63 0.63 0.63 0.63 0.63 0.63 0.63 0.63 0.63 0.63 0.63 0.63 0.63 0.63 0.63	Dy	mqq	10.53	5.03	2.15	8.42	2.15	1.67	16.05	1.68	9.77	4.00	1.13	2.74	0.94	8.10	0.54	5.38	18.35	3.53
ppm 6.06 2.86 1.45 4.12 1.14 1.12 8.52 1.09 6.11 2.67 0.73 1.46 0.55 4.08 0.36 ppm 0.91 0.43 0.25 0.65 0.24 0.17 1.24 0.19 0.43 0.12 0.22 0.13 0.63 0.68 ppm 5.76 2.73 1.65 4.06 1.20 8.36 1.44 6.33 2.86 0.88 0.76 0.73 0.63 0.63 0.68 ppm 5.76 2.73 1.65 4.06 1.20 8.36 1.44 6.33 2.86 0.88 1.39 0.76 3.79 0.63 ppm 0.82 0.40 0.26 0.29 0.12 0.12 0.16 0.76 0.76 0.79 0.76 0.79 0.76 0.79 0.78 ppm 0.82 0.40 0.26 0.26 0.26 0.76 0.76 0.76 0.76 <t< th=""><th></th><td>mqq</td><td>2.12</td><td>1.00</td><td>0.46</td><td>1.59</td><td>0.45</td><td>0.34</td><td>3.05</td><td>0.36</td><td>2.03</td><td>0.86</td><td>0.23</td><td>0.56</td><td>0.21</td><td>1.67</td><td>0.14</td><td>1.02</td><td>3.84</td><td>0.71</td></t<>		mqq	2.12	1.00	0.46	1.59	0.45	0.34	3.05	0.36	2.03	0.86	0.23	0.56	0.21	1.67	0.14	1.02	3.84	0.71
ppm 0.91 0.43 0.25 0.65 0.24 0.17 1.24 0.19 0.43 0.12 0.22 0.13 0.63 0.08 ppm 5.76 2.73 1.65 4.06 1.64 1.20 8.36 1.44 6.33 2.86 0.88 1.39 0.76 3.79 0.54 ppm 0.82 0.40 0.26 0.59 0.27 0.19 1.20 8.36 1.44 6.33 2.86 0.88 1.39 0.76 3.79 0.54 ppm 0.82 0.40 0.26 0.19 1.20 8.36 1.44 6.33 2.86 0.88 1.39 0.76 3.79 0.54 ppm 0.82 0.40 0.27 0.19 1.20 0.26 0.90 0.42 0.16 0.16 0.56 0.09 rest 266.60 440.89 69.82 294.60 59.63 140.46 106.87 54.84 57.17 275.83 9.6		mqq	6.06	2.86	1.45	4.12	1.44	1.12	8.52	1.09	6.11	2.67	0.73	1.46	0.55	4.08	0.36	2.56	10.02	1.85
ppm 5.76 2.73 1.65 4.06 1.64 1.20 8.36 1.44 6.33 2.86 0.88 1.39 0.76 3.79 0.54 ppm 0.82 0.40 0.26 0.59 0.27 0.19 1.20 0.26 0.90 0.42 0.15 0.16 0.56 0.09 REE 266.60 440.89 69.82 260.81 150.84 294.60 596.20 63.10 140.46 106.87 54.84 128.44 67.17 275.83 9.65 19		mqq	0.91	0.43	0.25	0.65	0.24	0.17	1.24	0.19	0.94	0.43	0.12	0.22	0.13	0.63	0.08	0.45	1.94	0.31
ppm 0.82 0.40 0.26 0.59 0.27 0.19 1.20 0.26 0.90 0.42 0.15 0.16 0.56 0.09 t REE 266.60 440.89 69.82 260.81 150.84 294.60 596.20 63.10 140.46 106.87 54.84 128.44 67.17 275.83 9.65		mqq	5.76	2.73	1.65	4.06	1.64	1.20	8.36	1.44	6.33	2.86	0.88	1.39	0.76	3.79	0.54	2.99	12.97	2.04
266.60 440.89 69.82 260.81 150.84 294.60 596.20 63.10 140.46 106.87 54.84 128.44 67.17 275.83 9.65	Lu	bpm	0.82	0.40	0.26	0.59	0.27	0.19	1.20	0.26	06.0	0.42	0.15	0.21	0.16	0.56	0.09	0.44	2.15	0.32
	Tot REE		266.60	440.89	69.82	260.81	150.84	294.60	596.20	63.10	140.46	106.87	54.84	128.44	67.17	275.83	9.65	190.11	814.26	129.84

REE Comparison Between Muncung Granite Samples and their Weathering Products, Lingga Regency, Riau Islands (R. Irzon *et al.*)

decrease in RGL 17 and RGL 46 are 58% and 75% compared to their granitoid bedrocks. The soil layers (A horizon) in five locations have REE below 130 ppm. The thinnest soil layer (<10 cm) without saprolite and laterite horizons indicates the lowest degree of wheatering in RGI 60 and related to the low REE content. Moreover, roots were relatively more dominant in the RGI 60's than other locations.

REE spider diagrams are used to compare these elements between soil, laterite, saprolite layers, and granitoid samples (Figure 5). The measured compositions were normalized to primitive mantle value of Sun and McDonough (1989). REE is a set of fifteen elements in lanthanide group. Almost lanthanide elements are only trivalent with the notable exception of additional valances of Ce (form 4⁺ ions) and Eu (form 2⁺ ions) in some environments. The bivalency of Eu is common in igneous rocks because of the same size and charge as Ca²⁺ which is found in plagioclase and other minerals, but the tetravalent nature of Ce is usually only recognisable as a result of sedimentary and weathering processes that involve intense oxidation (Ball et al., 2000). The spider REE diagrams of weathered granitic layers from Lingga Regency show Ce (cerium) positive anomaly and Eu (europium) negative anomaly, whilst the host rocks only show negative Eu negative anomaly. Ce anomaly (Ce/Ce*) and Eu anomaly (Eu/Eu*) were calculated using these formulas:

 $Eu/Eu^* = 2Eu_N/(Sm_N+Gd_N)$ Ce/Ce* = 2Ce_N/(La_N+Pr_N) (*i.e.* Och *et al.*, 2014)

 Eu_N , Sm_N , Gd_N , Ce_N , La_N , and Pr_N are the normalized elements content to primitive mantle value (Sun and McDonough, 1989). The calculation results are listed in Tabel 3.

Enrichment or depletion of Eu is generally attributed to its tendency to be incorporated into plagioclase preferentially over other minerals. Most of Eu is coalesced in a magma crystallizing stable plagioclase to lead a higher than expected concentration of Eu in the mineral *versus* other REE in that mineral. However, this magma is relatively depleted in Eu with a concentration of Eu lower than expected *versus* the concentrations of other REEs in that magma. As Eu is locked up in the plagioclase left in the magma chamber, Eu negative anomaly is caused of the separation of the magma from its plagioclase crystals and subsequently solidifies. Conversely, an accumulation of plagioclase crystals before solidification in the magma displays a relatively positive Eu anomaly. The Eu anomaly behaviour in natural plagioclase in function of temperature and oxygen fugacity was described by Weill and Drake (1973). Rocks that have accumulated plagioclase or have had plagioclase removed simply show positive or negative Eu anomalies (peaks or troughs in REE spider diagrams), respectively (e.g. Sawyer, 1998; el-Baghdadi et al., 2003; Singh and Vallinayagam, 2012). All granitoid samples from Lingga Regency show negative Eu that typically suggests plagioclase fractionation or indicates separation of melt from a plagioclase-rich source (Thuy et al., 2004; Kouske et al., 2012) or retention of Eu in plagioclase at the source during partial melting (Rollinson, 1993). The latest study revealed that geochemically the Muncung Granite could be divided into two facies: granitoids from Lingga Island and Selayar Island (A facies) and from Singkep Island (B facies) (Irzon, 2015). Eu anomaly numbers versus SiO, plot of selected samples show different patern (Figure 6a) and confirm the results from previous author.

Geochemically, SiO, rises along a normal magma differentiation and is mainly concentrated in quartz. The correlation of quartz to plagioclase is investigated to study the differentiation process of Muncung Granite. The negative correlation (r=0.97) of quartz to plagioclase in A facies of selected granites indicates that plagioclase was fractionated along magma differentiation to generate Eu negative anomaly (Figure 6b). Although the negative correlation was also depicted on the two minerals correlation in B facies, the lack of correlation coefficient (r=0.28) informs that the collaboration of petrography and geochemistry data is better in studying a magma differentiation process. Moreover, Eu anomaly is related to the sample's mineral assembly. Aubert et al. (2001) stated that biotite, muscovite, and apatite produced Eu negative anomaly at the value of 0.1, 0.25, and 0.39, respectively, whilst orthoclase

REE Comparison Between Muncung Granite Samples and their Weathering Products, Lingga Regency, Riau Islands (R. Irzon *et al.*)

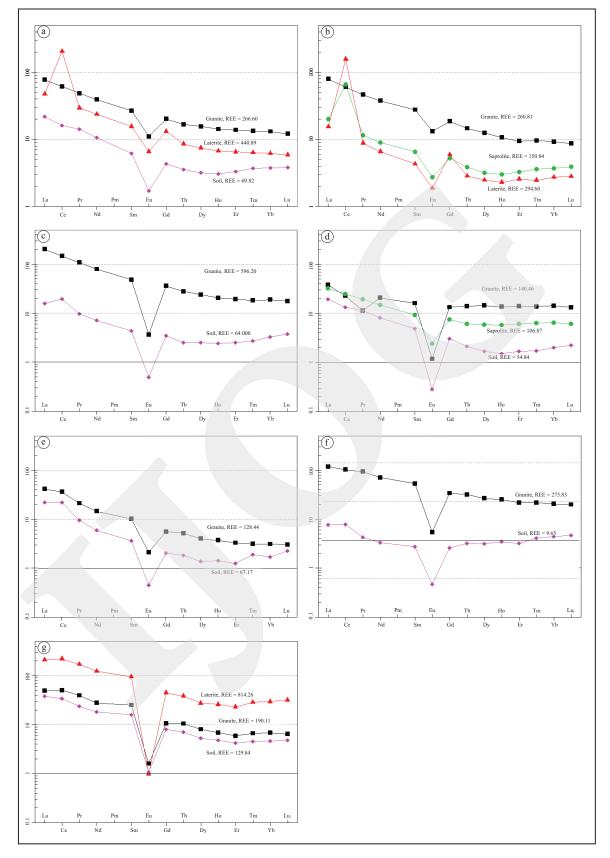


Figure 5. REE spider diagrams of studied samples: a) RGI 12; b) RGI 17; c) RGS 33; d) 46; e) 55; f) 60; g) 65. Black box = host granitoid, green circle = saprolite (C horizon); orange triangle = laterite (B horizon), pink diamond = soil (A horizon). Primitive mantle value of Sun and McDonough (1989) were used for normalization.

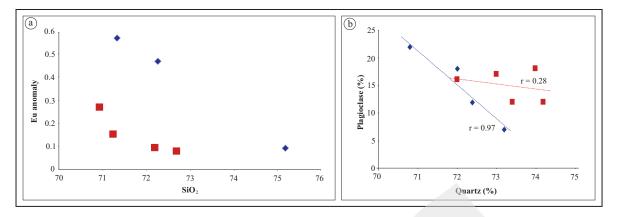


Figure 6. a) Eu anomaly *versus* SiO_2 plot of the selected samples; and b) Quartz and plagioclase correlation in the two facies of Muncung Granite. (*blue diamond* = granitoid from Lingga Island and Selayar Island; *red rectangle* = granitoid from Singkep Island).

is the mineral in granite that carries Eu positive anomaly of 3.23. Mathematically, the Eu positive anomaly from three orthoclases is equivalent to the Eu negative anomaly generated by one biotite. As discussed above, K-feldspar of Muncung granite not only consists of orthoclase but also sanidine. The abundance of biotite and muscovite (Table 1) is another clue for the Eu negative anomaly of the granites.

Cerium positive anomaly is shown in weathered granitoid layers (Figure 4) of Muncung Granite. The anomaly relates to the formation of tetravalent Ce under oxidizing condition near the surface and forms CeO, which is a stable compound in laterite/soil layers. The removal of REE, especially LREE, except Ce in an oxidizing condition near the surface develops a positive anomaly, whilst deposition of the removed REE in less oxidizing condition generates a negative anomaly (Marker and Oliveira, 1994; Sanematsu et al., 2009). The cerium enrichment of weathered Muncung Granite is detected in all saprolitelaterite layers (C-B horizons, respectively) in all location and most of the soils (A horizons). The counts of Eu and Ce anomalies are shown in Tabel 3. Chemical properties of Ce^{4+} is closer to the heavy REE (HREE) than the light REE (LREE) (Ball et al., 2000) to explain the more HREE/ LREE ratio in soil and laterite than the host rock in line with Ce positive anomaly progress (Figure 4) as other laterite studies in South China (Bao and Zhao, 2008), Shoutern Thailand (Imai et al., 2008), and Laos (Sanematsu et al., 2009).

are compared to the Ce anomaly of granite to find any corelation of this anomaly to REE's level change. $Ce_{ano-laterite}/Ce_{ano-granite}$, as the Ce anomaly number in laterite per Ce anomaly in granitoid host rock, reaches the highest in RGL 17 (13.56), whereas the REE total enrichment in laterite is only 1.13 times than that of the host granitoid. The anomaly ratio is lower in RGL 12 (5.47) with the increase of REE total enrichment (1.65)(Table 3). On the other hand, despite the lowest $Ce_{ano-laterite}/Ce_{ano-granite}$ (1.02), RGI 65 has the most REE total enrichment (4.28). This indicates that Ce is precipitated much more rapidly in horizon B than other REE and retains at the upper soil horizon during weathering (Maulana et al., 2014). The speed of settling becomes slower as weathering goes on and reaches the end where the other elements have also being completely precipitated nearby where Ce anomaly in horizon B get closer to 1. In general, no good correlation can be concluded between $Ce_{ano-soil}$ / $Ce_{ano-granite}$, or $Ce_{ano-saprolite}/Ce_{ano-granite}$, and REE change.

Ce anomalies of the three weathered layers

CONCLUSION AND RECOMMENDATION

Although the average of REE content in seven granitoid samples is 265 ppm and just below the range of the host Longnan Granite, the REE enrichment in laterite layer exceed four times than the host rock. Eu negative anomaly in granitoids reflects plagioclase fractionation dur-

Sample	es	Ce/Ce*	Eu/Eu*	$Ce_{ano-layer}/Ce_{ano-granite}$	Eu _{ano-layer} /Eu _{ano-granite}	Tot REE	REE _{layer} /REE _{host granite}
	Н	0.99	0.47	1.00	1.00	266.60	1.00
RGL 12	В	5.40	0.46	5.47	0.97	440.89	1.65
	А	0.91	0.33	0.92	0.69	69.82	0.26
	Н	0.97	0.57	1.00	1.00	260.81	1.00
RGL 17	С	4.24	0.46	4.38	0.81	150.84	0.58
	В	13.11	0.37	13.56	0.64	294.60	1.13
DCG 22	Н	0.95	0.09	1.00	1.00	596.20	1.00
RGS 33	С	1.54	0.13	1.62	1.46	63.10	0.11
	Н	0.91	0.08	1.00	1.00	140.46	1.00
RGI 46	С	0.97	0.29	1.06	3.61	106.87	0.76
	А	0.87	0.07	0.95	0.89	54.84	0.39
DCI 55	Н	1.17	0.27	1.00	1.00	128.44	1.00
RGI 55	А	1.39	0.16	1.19	0.59	67.17	0.52
RGI 60	Н	0.97	0.07	1.00	1.00	275.83	1.00
KUI UU	А	1.39	0.11	1.43	1.56	9.65	0.03
	Н	1.15	0.09	1.00	1.00	190.11	1.00
RGI 65	В	1.17	0.01	1.02	0.16	814.26	4.28
	А	1.11	0.09	0.96	1.00	129.84	0.68

Table 3. Ce and Eu Anomalies and their Relation to REE Content. H = Granitoid as Host Rock, A = Soil Layer, B = Laterite Layer, C = Saprolite Layer. All Data are normalized by Primitive Mantle Value (McDonough and Sun, 1995)

ing the partial melting. Both Eu and Ce anomaly are shown in the weathered granitoid layers. Eu negative anomaly and Ce positive anomaly in weathered horizons are the consequences of the host rock values and oxidation from the host layer, respectively. REE level in granitoid as the host rock in the studied area is higher than the A and C horizons, but lower than the B horizon. The Ce_{ano-laterite}/Ce_{ano-granite} anomaly in Muncung Granite is found to be related to REE enrichment in the laterite layer, where this number reaches the highest in the lowest REE enrichment level.

It would be better if complete host rocks and weathered layers can be found in more locations to study more about REE relation to any anomalies in different layers. This method can be applied to many granitoid provinces in Indonesia to find more prospective REE areas in Indonesia.

ACKNOWLEDGEMENTS

Thanks so much to the first author's big family for the never ending spirit. The Head of Centre for Geology Survey is thanked for the publicity permission. Mr. Baharuddin, Mr. Eko Partoyo, and Professor Hamdan Zainal Abidin again open broad scientific ideas. The very good laboratory work that was performed by Indah and Citra is highly acknowledged. Thanks a lot to Mr. Sigit Maryanto for the petrography knowledge. Geochemistry Programme of The Centre for Geological Survey assists this study financially.

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