RARE EARTH ELEMENT IN GREENSCHIST FACIES ROCK FROM BANTIMALA COMPLEX SOUTH SULAWESI INDONESIA

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ABSTRACT

The occurrences of rare earth elements (REE) have been determined using Laser Ablation Inductive Couple Plasma-Mass Spectrometry (LA-ICP-MS) method from greenschist facies rock in Bantimala Complex, South Sulawesi. Greenschist facies rocks occur as product of low grade metamorphism along with HP metamorphic rock, including eclogite and bluschist facies rocks. Major element result show that the greenschist facies rock were derived from various affinities, ranged from mid oceanic ridge basalt to upper continental crust which has intermediate to basic composition. They were also enriched in rock with intermediate composition, particularly quartzepidote chlorite schist whereas other rock which have more basic affinity show relatively low content of REE. All the greenschist facies rocks were characterized by moderate irregular REE pattern with some variation showing LREE enrichment except those from albite actinolite schist. Generally, the $\Sigma REEs+Y$ ranges from 9 – 786 ppm with average of 173 ppm. The LREE was much concentrated than the HREE except for albite actinolite schist by a factor of 2.36 - 6.27, which suggested that the LREE content in greenschist facies rocks were derived from intermediate rock composition rather than basaltic composition. It is shown from the study that REE were relatively immobile during low grade metamorphism.

Keywords: Rare Earth Elements, Greenschist facies rock, Bantimala Complex, Sulawesi.

INTRODUCTION

Bantimala Complex has been subject of study since last decades. This complex consists of lithology ranges from various various metamorphic rocks to meta-sedimentary rocks (Sukamto, 1982; Maulana, 2009). It is located in South Sulawesi and has been considered as one of the Central Indonesian Collison Complex (Wakita, 2000). While numerous studies has been conducted in this complex,

report on the occurrences of rare earth element from greenschist rocks are still limited despite their significant contribution in unraveling the petrogenetic study of the area especially the mobility of rare earth elements in metamorphic process as well as the economic values of these elements. The rare earth elements (REE) are the 15 lanthanide element with atomic number 57 to 71 (Henderson, 1984). Yttrium (Y) and Scandium (Sc) are generally included with the

REE as they occur with them in mineral and have similar chemical properties (Henderson, 1996). REE play an important role in geochemical study since their distribution in earth crust and mantle contribute to elucidate evolutionary process of geological cycles.

Nowadays, they have been widely used in modern high technology devices such as automobile, permanent magnet, optic and laser research as well as military missile (Minowa, 2008). REE are classified into two groups of: light REE or cerium group (Lanthanum to Europium) and the heavy REE comprising Gadonilium through lutetium, as well as Yttrium and Scandium (Henderson, 1996). Naturally, the light REE are more abundant than the heavy REE. It is often assumed that REE were immobile during low grade metamorphism although some reports reveal the mobility of these elements. In this study, we report the rare earth elements occurrences in greenschist facies rocks from Bantimala Complex and discuss the mobility of these elements during metamorphism. In addition, constrain on the petrogenetic of the rocks is also considered.

REGIONAL GEOLOGY

The Bantimala area is situated approximately 40 km north-east of Makassar, South Sulawesi. The detailed geology of this area was described by Sukamto (1986); Wakita et al (1996) and Maulana et al (2009) as shown in Fig.1. The geology of the Bantimala area is dominated by a Triassic – Jurassic "basement complex", the Bantimala Complex (Sukamto, 1986), which consists of high-pressure metabasites and low pressure metamorphosed clastic sequence rocks of Cretaceous age that includes sandstone, shale, conglomerate, chert, siliceous shale, basalt, ultramafic rock and "schist breccia" (Wakita et al., 1996). This complex is surrounded by Tertiary and Quaternary sedimentary and volcanic rocks,

and is unconformably overlain by Late Cretaceous volcanic products and Paleocene sediments. It is bounded by faults which were active before the Paleocene and partly reactivated in Cenozoic time (Miyazaki et al., 1996). Palaeogene diorites and andesites intruded the basement complex. The high pressure metamorphic rocks in the Bantimala Complex consist of glaucophane schist, albite-actinolite-chlorite schist, chlorite-mica garnet-glaucophane-quartz schist. schist. garnet-chloritoid-glaucophane-quartz schist, serpentinite, garnet-glaucophane rocks and eclogite (Miyazaki et al., 1996). The Early Cretaceous K-Ar phengite ages of these rocks (Wakita et al., 1994, 1996) are as follows: 132 ± 7 , 113 ± 6 Ma for garnet-glaucophane rocks; 124 ± 6 Ma for mica-rich units intercalated with garnet-glaucophane rock; and 114 ± 6 , mica-quartz 115± 6 Ma for schists intercalated with hematite-bearing glaucophane schists. The metamorphosed sedimentary comprised melange. rocks turbidite and shallow-marine clastic sediment. The melange occurs as a tectonic block, and contains rock types such as sandstone, shale, siliceous shale, chert, basalt, schist, and felsic igneous rocks within a sheared matrix (Miyazaki et al, 1996). The mid Cretaceous (late Albian - early Cenomanian) chert unconformably overlies the high-pressure metamorphic rocks (Wakita et al., 1996). The ultramafic rock is dominated by serpentinised peridotite, which contains chromite lenses in some areas.

METHODS

9 samples were crushed and milled to obtain the whole rock and trace element compositions. Whole-rock major elements were analysed by X-ray fluorescence analyses (XRF), and whole-rock and individual mineral trace element analyses by laser ablation inductively-coupled plasma mass spectrometry (LA-ICP-MS). Major elements Na, Mg, Al, Si, P, S, K, Ca, Ti, Mn, Fe, F and Cl were assessed by XRF with a Phillips (now Panalytical) PW2400 wavelength-dispersive X-ray fluorescence spectrometer. Lithium borate discs were prepared by fusion of 0.27g of dried sample powder and 1.72g of "12-22" eutectic lithium metaborate-lithium tetraborate at 1010 °C for 10 minutes in a rocker furnace. The major elements were calibrated against a set of 28 international standard rock powders.

Trace elements including rare earth elements analyses were obtained by LA-ICP-MS at the Research School of Earth Sciences, ANU. Trace elements concentration were determined on glasses made from rock powders fused with lithium borate flux (1: 3 mass ratio). The LA ICP-MS employs an ArF⁺ (193 nm) excimer laser and a Hewlett Packard Agilent 7500 ICP-MS. Laser sampling was performed in an Ar-He atmosphere using a spot size between 80 and 100 µm. The counting time was 20 seconds for the background and 60 seconds for sample analyses. The external standard for calibration was NIST 612 glass, using the standard reference values of Pearce et al. (1979). Si was employed as the internal standard, employing the SiO₂ concentration previously measured by XRF.

Loss-on-ignition (LOI) values were calculated from the mass differences in approximately 2 grams of powdered sample after heating to 1010 °C in the furnace for one hour.

RESULTS

The greenschist facies metabasites of the Bantimala Block display a wide range of composition (Table 1). SiO_2 is 43.1 - 55.8 wt% and total alkalis 2.9 - 8.3 wt%. If these components are assumed to have their original igneous concentrations, the rocks can be classified as picrobasalt (CP03B), basalt

(BM06, BM07 and CP 04A), basalt andesitic (CP04B and CP04C), dacite (CP03A) and trachyandesitic (BML03A) (Fig.2). However, some of them show extremely low Al_2O_3 contents and high MgO (CP04B and CP04C) inconsistent with the silica and alkali content for volcanic rocks, implying that they may have been derived from an ultramafic protolith by the addition of Si and alkalis. In contrast, BML03A has much higher Al₂O₃ (up to 21 wt%) as well as very high alkalis $(4.97\% \text{ K}_2\text{O} + 3.25\% \text{ N}a_2\text{O})$, which again suggests metasomatism or hydrothermal alteration if the protolith was igneous (but see below). In the AFM diagram, the greenschists mainly lie on a tholeiitic trend, although two quartz-epidote schists are much higher in alkali content, corresponding to much more felsic compositions if primary (Fig 3), as is possible for CP03A but not so for BML03A.

Primitive mantle-normalised trace elements from these rocks show five distinctive group patterns. The first group includes garnetactinolite schist (CP03B), actinolite-epidote schist (CP04A and BM07), and quartzschist (CP03A). epidote They were characterised by enriched LILE and depleted HFSE, particularly Nb and Ta (Fig.4). However, Sr shows enrichment for CP04A and BM07 but depletion in CP03B and CP03A. These have strongly enriched LREE $(La_N/Yb_N = 1.9 - 6)$ in their chondrite normalised REE patterns, with a slight negative Eu anomaly (0.7 -0.9) in all except CP03B (Fig. 5). The positive Eu anomaly in CP03B is explained by the abundance of plagioclase in this sample, whereas the depleted HREE suggest fractionation of garnet (Wilson, 1989). An arc or back-arc basin affinity is indicated by the significant depletion of Nb and Ta but enrichment in LILE, Sr and Ca, particularly in CP04A and BM07 (Danyushevsky et al., 1993).

The second group includes albite-actinolite schists (CP04B and CP04C). Primitive mantle-normalised trace elements from this group show enrichment of LILE relative to NMORB (particularly Rb and Ba) but depletion in Th and other HFSE (Fig.6). Chondrite-normalised REE show depletion in LREE $(La_N/Yb_N = 0.1 - 0.4)$ with a small positive Eu anomaly (Eu/Eu* = 1.1) in CP04C and negative anomaly in CP04B (Eu* = 0.64) and almost flat pattern of HREE, all of which are well below N-MORB concentrations (Fig.7). From these patterns, coupled with the low content of Al₂O₃ and the extremely high MgO, Cr and Ni, these rocks are deduced to be ultramafic cumulates that have been secondarily silicified.

The third group consists only of the actinolite schist with relict augite (BM06). This rock shows enrichment in LILE and HFSE (especially Nb) with negative trend in Sm – Yb (Fig. 8), suggesting an oceanic island basalt (Holm, 1985). Chondrite-normalised REE show slight LREE enrichment (La_N/Yb_N = 13.3) without a noticeable Eu anomaly (Fig.9). Apart from depletion in Rb and Ba, this is a similar pattern with the OIB from Sun and McDonough (1989).

The fourth greenschist type is BML03A, a Krich, Al-rich rock which is quite distinct from all others. PM-normalised trace elements show extreme enrichment in LILE and HFSE (except for Sr in LILE and Ta in HFSE) with a slight depletion of Ti, typical of upper continental crust pattern (Rudnick et al., 2004) (Fig.10). This rock also has a high Th/U = 2.4, relatively close to the values proposed by Taylor & McLennan (1985) for the upper continental composition. Chondritenormalised REE show enrichment in LREE with $La_N/Yb_N = 3.7$ and a slight negative Eu anomaly (Eu/Eu* = 0.18) with flat HREE. Both trace element and REE pattern are comparable to the upper continental crust pattern of Taylor and McLennan (1985) and Rudnick and Gao (2003) (Fig.11). Hence, this rock appears to be continental in origin. The composition of the rock reflects either terrigenous sediment or granodiorite caught up in the trench and accreted. The Eu anomaly, suggests plagioclase fractionation during evolution of an igneous precursor.

The last group type is intermediate to basic igneous rock which has been metamorphosed into metabasalt (CP02D). The metabasalt (CP02D) is so called because of retention of its phenocrystic texture. It is characterised by relatively high SiO₂ and Na₂O content (51.4 wt% and 7.7 wt%, respectively) (Table 1), the latter causing it to plot in the basaltic trachyandesite field in the TAS diagram (Fig. 2) and in the calcalkaline field in an AFM diagram (Fig.3). The high Na almost certainly represents metasomatism by seawater. Primitive mantle-normalised trace element patterns show depletion in mobile elements (Rb and Ba) but enrichment in Th, U and Nb (Fig. 12). Chondrite-normalised REE show LREE enrichment in LREE $(La_N/Yb_N = 15)$ without Eu anomaly (Eu/Eu* = 1.02) and with strongly depleted HREE (Fig.13). This pattern resembles those from the oceanic island basalts (Holm, 1985).

DISCUSSION AND CONCLUSION

All sample exhibit variable abundance of REE (Table 1). Generally, light REE (LREE) were enriched in most of samples compare with heavy REE (HREE) by a factor of 2.36 - 6.27 except those from albite actinolite schist. The \sum REEs+Y ranges from 9 – 786 ppm with average of 173 ppm. BML 03 A which chiefly composed of quartz + epidote + chlorite with significant amount of garnet and zircon as accessory minerals has the highest content of total REE (605 ppm) with total Y+REE reach up to 786 ppm. Based on petrographic analyses (not included in this paper) the enrichment of REE in this sample is due to significant number of zircon and epidote

which incorporated the REE. In addition, HREE enrichment also confirm with the occurrences of garnet in this sample as a host of HREE (Wilson, 1989). The LREE enrichment further suggest the LREE content in greenschist facies rocks were derived from intermediate rock composition rather than basaltic composition.

All the samples have been metamorphosed into greenschist facies metamorphism which indicated by the presence of typical greenschist facies mineral such as epidote, albite and chlorite. Therefore, it is important to evaluate the effect of metamorphism on the rare earth element geochemistry of the samples. Rollinson (1993) suggested that element mobility in metamorphic rock is controlled by solid-state diffusion, melt generation and interaction with fluid. He further suggested that at the scale of several centimeters or more, the effect of solid-state diffusion and melt generation is insignificant, and the main concern is fluid-controlled element mobility. It is generally considered that many element including K, Rb, Cs, Th and U are mobile during metamorphism. In contrast, REE are assumed to be immobile and are not affected greatly by contact or regional metamorphism up to amphibolites facies (Girty et al, 1994). In addition. Tribuzio et al (1996) reported that the blueschist and eclogite facies rocks did not introduce significant release of REE to the upper mantle. Thus, mobility of REE is a powerful tool to study the petrogenetic model.

Based on chondrite-normalised REE pattern, the first groups of the samples are assumed to be comparable to island arc basalt composition proposed by Wilson (1989). The which incorporated second group the cumulate ultramafic rocks which has been silisified also shows a relatively equivalent value with those from ultramafic rocks with some enrichment in certain elements. The third group which associated with OIB also reveals the relatively similar value of their sources. The fourth group which resembles the upper continental pattern show enrichment (5 times higher) compared to the original value of the sources though they showed a relatively similar trend with enrichment of LREE. The last groups which have metabasalt affinity shows relatively similar pattern with oceanic island basalt (OIB).

Geochemical composition coupled with similarity of chondrite-normalised REE patterns of the samples indicates the lack of large-scale remobilization of rare earth element in the samples. Therefore, although some elements such as large ion lithophile elements (LILE) likely to be mobile and with some exception in sample BML 03A, there is no significant remobilization of REE from most of the samples. Thus, it is concluded that REE mobility during greenschist facies metamorphism was not prevailed and further confirm that the REE were relatively immobile during low grade metamorphism.

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S ample	C P 03B	CP04 B	CP 04 C	BML 3A	BM 07	C P 04A	B M06	CP 02D	CP03A
	Garnet	Albite actinolite schist		Quartz	Actinolite epidote schist		Augite	Meta-	Quartz
R oc k-ty pe	actinolite			epidote	t a		bearing	bas alt	epidote
	s c his t			chlorite			actinolite		s c his t
			1	s c his t			schist		
Whole rock (wt%)									
SiO ₂	43.16	51.75	53.24	55.88	46.50	48.76	47.37	51.41	68.91
TiO ₂	1.20	0.08	0.16	0.22	1.67	0.27	2.52	2.91	0.78
AI20 ₃	17.07	5.00	5.77	21.51	11.14	16.10	9.65	14.30	12.00
FeOT	17.18	7.48	8.63	5.71	10.60	6.78	10.96	10.32	6.36
MnO	1.28	0.15	0.16	0.13	0.19	0.14	0.17	0.41	0.13
MgO	12.24	18.18	18.08	0.93	11.75	10.02	11.66	4.40	1.71
CaO	5.45	8.99	7.95	3.76	11.93	11.95	12.78	4.19	1.53
Na ₂ O	0.51	1.95	2.59	3.25	1.74	1.99	1.72	7.77	3.43
K ₂ O	0.37	0.35	0.19	4.97	0.32	0.57	0.09	0.21	1.61
P ₂ O5	0.21	0.01	0.01	0.02	0.07	0.07	0.43	0.54	0.08
SO ₃	0.00	0.00	0.00	0.90	0.00	0.00	0.00	0.26	0.00
LOI	2.35	5.51	2.57	2.65	3.86	3.35	2.93	4.56	2.46
Total	101.03	99.44	99.35	99.92	99.75	99.99	100.28	101.25	99.01
Trace elements (ppm)									
Cr	66.94	14.80	26.60	13.33	43.40	74.81	31.90	55.56	63.05
Ni	15.20	49.60	15.80	8.70	27.40	13.40	15.50	24.70	42.67
Rb	6.12	5.10	1.04	89.01	6.05	14.44	0.85	2.49	36.89
Ba	48.22	41.48	7.93	403.27	29.12	30.10	13.91	57.30	130.91
Th	0.22	0.01	0.05	12.29	0.62	0.09	3.12	4.02	3.10
U	0.70	0.03	0.06	5.01	0.55	0.08	0.90	0.98	0.74
Nb	2.11	0.46	1.86	140.86	8.93	0.96	36.53	44.47	9.28
Та	0.16	0.03	0.07	2.89	0.25	0.05	0.98	1.05	0.33
Sr	107.71	28.77	22.46	595.54	843.83	334.31	581.37	172.45	81.99
Nd	17.26	0.24	0.91	104.34	13.91	4.70	41.74	36.43	13.83
Zr	21.58	6.38	27.43	1837.20	114.02	18.46	198.16	261.16	103.75
Hf	1.40	0.85	1.07	34.38	3.06	0.84	5.14	6.57	3.61
La	12.65	0.07	0.37	108.80	8.91	2.81	37.68	36.04	14.43
Ce	26.89	0.22	0.88	228.41	20.14	6.64	69.37	71.68	34.23
Pr	3.69	0.05	0.15	25.99	2.85	1.04	8.67	8.31	3.35
Nd	17.26	0.24	0.91	104.34	13.91	4.70	41.74	36.43	13.83
S m	4.16	0.19	0.37	22.62	3.89	1.41	9.62	7.70	2.78
Eu	4.44	0.06	0.18	1.43	1.26	0.45	3.15	2.39	0.68
Gd	4.53	0.48	0.67	24.60	4.65	1.36	10.36	6.72	2.82
Tb	0.66	0.12	0.10	4.43	0.76	0.26	1.37	0.97	0.40
Dy	4.85	0.81	0.80	30.89	5.62	1.83	7.65	5.71	2.60
Но	0.99	0.20	0.20	6.41	1.07	0.32	1.33	0.90	0.58
Er	3.92	0.59	0.61	19.95	3.22	1.08	3.07	2.17	1.62
Tm	0.55	0.10	0.09	3.00	0.45	0.15	0.36	0.29	0.26
Yb	3.77	0.68	0.71	20.92	3.41	0.92	2.04	1.72	1.75
Lu	0.63	0.10	0.10	3.19	0.59	0.08	0.27	0.24	0.24
ΣLREE	69.10	0.84	2.86	491.59	50.97	17.05	170.24	162.56	69.31
ΣHREE	19.91	3.08	3.28	113.41	19.79	6.01	26.44	18.72	10.27
ΣREE	89.00	3.92	6.13	605.00	70.75	23.06	196.68	181.28	79.57
ΣREE + Y	119.39	9.16	11.91	786.39	101.50	32.13	229.51	205.35	94.08
LREE/HREE	3.47	0.27	0.87	4.33	2.58	2.84	6.44	8.68	6.75
HREE/REE	0.22	0.79	0.53	0.19	0.28	0.26	0.13	0.10	0.13
Ce*	3.94	3.85	3.72	4.30	4.00	3.88	3.84	4.14	4.92
Ce/Ce*	6.83	0.06	0.24	53.18	5.04	1.71	18.08	17.31	6.96
E u/E u*	1.42	0.10	0.16	7.72	1.39	0.45	3.28	2.35	0.91

TABLE 1. Bulk rock, trace element and rare earth elements composition of the greenschist rocks

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Sukamto, 1982; Wakita et al., 1996; Maulana,2009).

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FIGURE. 3. The greenschist facies rocks from the Bantimala Block plotted in the FeO-Alkali-MgO ternary diagram of Irvine and Baragar (1971). Symbols see Fig.2.

MgC

Na₂O+K₂O



FIGURE.4. PM-normalised (Sun and McDonough, 1989) trace element patterns of quartz-epidote schist (CP03A), actinolite schist (CP04A and BM07) and garnet-actinolite schist (CP03B) from the Bantimala Block. Note that the patterns show enrichment in LILE and depletion in Nb and Ta compared to MORB, characteristic of arc environments.



FIGURE.5. Chondrite-normalised (Sun and McDonough, 1989) REE patterns of the greenschist-facies rocks from the Bantimala Block.

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FIGURE.6. PM-normalised (Sun and McDonough, 1989) trace element patterns of albite-actinolite schist (CP04B and CP04C) from the Bantimala Block. N-MORB pattern from Sun & McDonough (1989) is shown as reference.



FIGURE.8. PM-normalised (Sun and McDonough, 1989) trace elements patterns of the augite-bearing actinolite schist (BM06). Note the relative similar pattern with the oceanic island basalt affinities from Sun and McDonough (1989) except the Rb, and Ba



McDonough, 1989) REE patterns of albite- actinolite schist (CP04B and CP04C). The samples show depletion of REE relative to N-MORB (Sun & McDonough, 1989).



FIGURE.9. Chondrite-normalised REE of BM06. Note the similar pattern with the OIB (Sun and McDonough. 1989).



FIGURE.10. PM-normalised (Sun and McDonough, 1989) trace elements for quartz-epidote-chlorite schist BML03A. Note the similarity with the upper continental crust pattern from Rudnick & Gao (2003).



FIGURE.12. PM-normalised trace element patterns of metabasalt (CP02D). Note similarity to OIB pattern (Sun and McDonough, 1989).



FIGURE.11. Chondrite- normalised (Sun and McDonough, 1989) REE for BML03. Again, these are similar to those for the upper continental crust (Rudnick & Gao, 2003).



FIGURE.13. Chondrite-normalised REE pattern of metabasalt CP02D. OIB data of Sun and McDonough (1989) is shown as reference.