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Trace and Rare Earth Elements Compositional Change on Andesite Alteration in Kaligesing, Purworejo

Perubahan Komposisi Unsur Jejak dan Unsur Tanah Jarang pada Alterasi Andesit di Kaligesing, Purworejo

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Abstract- The change of chemical composition because of hydrothermal alteration process is related to the modification on mineralogy and elements mobility. Different alteration conditions could lead to dissimilar geochemical character. This study aims to discuss the alteration effect on trace and rare earth elements composition of an andesite outcrop with hydrothermal alteration in Kaligesing, Purworejo, Central Java Province. Microscopic analysis at Central for Geological Survey was applied to determine the modal mineral composition of the selected samples whilst trace and rare earth elements abundances was measured using Inductively Coupled Plasma – Mass Spectrometry. Plagioclase is the major phenocryst embedded in the fine-grained feldspar and glass groundmass of relatively fresh andesite. On the other hand, sericite, chlorite, epidote, and iron oxide are detected in the altered rock. The more Sr and Rb compositions on unaltered sample exhibit their common existence in plagioclase. The bigger Rb/Sr and the lower Ba/Sr ratios inward to the center of alteration might indicate the more degree of K-bearing mineral formation than Ca-rich mineral alteration. The Ba/Zr escalation and Zr/Y reduction from relatively fresh rock through to the vein of the studied samples are parallel to the previous investigation about andesite alteration. Chondrite-normalized rare-earth elements (REE) pattern of unaltered, altered, and vein samples depicts similar patterns: strong enrichment of Low REE, positive Eu anomaly, and relatively flat high REE. The decrease of Eu anomaly may reflect the reduction of plagioclase modal composition because of alteration and might indicate a reductive alteration state.

Abstrak- *Perubahan komposisi kimia akibat alterasi hidrotermal terkait dengan modifikasi mineralogi maupun mobilitas elemen. Perbedaan kondisi alterasi dapat menghasilkan karakter geokimia yang tidak sama. Penelitian ini bertujuan untuk menjabarkan mengenai efek alterasi terhadap komposisi unsur jejak dan unsur tanah jarang pada satu singkapan andesit di Kaligesing, Purworejo, Provinsi Jawa Tengah. Analisis mikroskopis di Pusat Survei Geologi dimanfaatkan untuk menentukan komposisi mineral pada sampel yang diteliti, sedangkan kadar unsur jejak dan unsur tanah jarang diukur menggunakan ICP-MS. Plagioklas sebagai fenokris utama, yang berada di dalam masadasar feldspar dan gelas merupakan mineral utama pada andesit segar. Pada sisi lain, serisit, klorit, epidot, dan oksida besi dapat dijumpai pada sampel teralterasi. Kelimpahan Sr dan Rb yang lebih besar pada batuan segar menunjukkan keberadaannya dalam mineral plagioklas. Kenaikan Rb/Sr dan penurunan Ba/Sr menuju pusat alterasi dapat diakibatkan oleh derajad pembentukan mineral mengandung-K ketika alterasi lebih tinggi daripada mineral kaya-Ca. Peningkatan Ba/Zr dan pengurangan Zr/Y dari sampel segar menuju pusat alterasi sesuai dengan penelitian sebelumnya mengenai alterasi andesit. Pola normalisasi unsur tanah jarang (UTJ) terhadap chondrit pada ketiga sampel menunjukkan kesamaan pola: pengayaan kuat pada UTJ-ringan, anomali Eu positif, dan UTJ-berat yang relatif datar. Penurunan derajad anomali Eu mengindikasikan pengurangan plagioklas akibat alterasi dan pertanda kondisi alterasi yang reduktif.*

Katakunci: *Alterasi, andesit, geokima, unsur jejak, dan unsur tanah jarang.*

INTRODUCTION

Alteration are mineral and geochemical compositions change that happens after rock emplacement and may because of metamorphic or magmatic activities. Rock characteristics such as permeability, texture, mineralogy, and chemical composition together with the nature of groundwater affect the alteration process (Verma et al., 2005; Pandarinath et al., 2008). The intensity of alteration was represented on the abundance of alteration mineral and escalates to the center of alteration. Sericite and chlorite are the common alteration for plagioclase and mafic minerals, respectively (i.e. Kaygusuz et al., 2013; Irzon, 2015; Kumral et al., 2016; Chukwu and Obiora, 2018). Alteration of volcanic glass might yield smectites and various types of zeolites (i.e. Christidis, 1998; Gomez et al., 2009). Instead of plagioclase, various mafic minerals including pyroxene, amphibole, and biotite amight also altered to epidote (i.e. Verma et al., 2005; Kürkcüoglu et al., 2008; Wang et al., 2018).

The formation of alteration minerals surely changes chemical composition and modify the elements mobility of altered rock in comparison to relatively fresh one. In correlation to alteration, element mobility was defined as the capacity for the element to move from any previously conditions to altered ones. The mobility of major, trace and rare-earth elements (REE) during alteration processes in different environments has been documented by numerous authors. Several elements are regarded as being mobile or immobile during specific alteration depends on the gains and losses in the system. Rare earth elements REE are relatively immobile on alteration and metamorphism of ophiolite suite in Bay Island (Suen et al., 1979), on surficial alteration of volcanic ash deposits in Yunnan and Guizhou (Zhou et al., 2000), and on Shangsu basalts alteration (Li et al., 2008). On the other hand, the group is considered mobile on komatiites metamorphism in Eastern Finland (Gruau et al., 1992) and peridotites serpentinization from Southwest Indian Ridge (Frisby et al., 2016). However, Heavy-REE (HREE) is generally more immobile than Light-REE (LREE) (Ling and Liu, 2002; Chen et al., 2016).

In compensation to the chemistry modification, alteration should also change the elemental ratios. Ba/Sr and Rb/Sr ratios are good indicators for postmagmatic alteration process and were originally propossed by Ekwere (1985). As Sr is incompatible for most common minerals except plagioclase and Ba are similarly partitioned in K-feldspar, plagioclase precipitation increase Ba/Sr ratio (i.e. Keshavarzi et al., 2014; Rezaei-Kahkhaei et al., 2014). Plagioclase feldspar is common mineral phases in the crust and thus are major reservoirs for Sr, Rb, and Eu (Ekwere, 1985; Keshavarzi et al., 2014; Frisby et al., 2016). Because rubidium generally incorporated with K-rich minerals (e.g. K-feldspar, biotite, sericite) whilst Sr coexists in Ca and Na-bearing minerals (e.g. plagioclase, carbonate, Ca-silicate) (i.e. Sharma et al., 2016; Chukwu and Obiora, 2018; Hines et al., 2019), Rb/Sr ratio is affected by the modal composition of these minerals. Relatively immobile incompatible elements such as Hf, Nb, Ta, Ti, Y, and Zr and their ratios could be used to determine mantle/crust and alteration conditions (i.e. Kaygusuz et al., 2013; Chukwu and Obiora, 2018).

Kaligesing is located in Purworejo Regency, Middle Java Province on the western part of Yogyakarta Quadrangle no 1408-2 (Rahardjo et al., 1995) at the western border of Yogyakarta Province. An outcrop of andesite alteration is identified in Kaligesing around the old Gajah Mountain's domain. The understanding of hydrothermal alteration is of value because it provides insights into the chemical attributes and origins of ore fluids and the physical conditions of ore formation. This study aims to discuss the alteration effect on trace and rare earth elements composition of the andesite outcrop in Kaligesing.

REGIONAL GEOLOGY

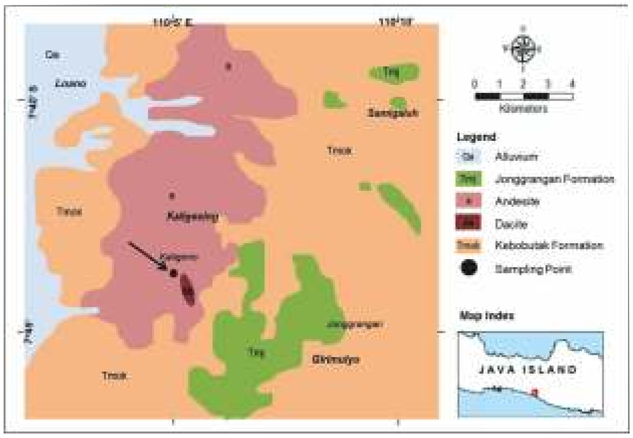
The study area comprised of two sedimentary formations, volcanic or intrusive rock, and alluvium. Kebobutak Formation is the other name of van Bemmelen's Old Andesite Formation and it is composed successively of conglomerate, sandstone, tuffaceous shale and silt, which is Late Oligocene – Lower Miocene in age. In Yogyakarta Quadrangle (Rahardjo et al., 1995), the Kebobutak Formation was sedimented over the Nanggulan Formation. The Nanggulan Formation was assigned to be deposited at Eocene to Early Oligocene using planktonic foraminifera test (Harjanto, 2011). Conglomerate, tuffaceous marl, calcareous sandstone, limestone, and coralline limestone built the Jonggrangan Formation in Lower Miocene. The Quaternary Alluvium spreads in the western part of Purworejo. Geological map with sampling locations of this study is shown in Figure 1.

Two periods of volcanic activity in Java throughout Tertiary was proposed on the previous study (Soeria-Atmadja et al., 1994). The earlier activity took place in the range of 40-18 Ma whilst the later one during 12-2 Ma. Geotectonic study of Kulonprogo stated that the basaltic andesite intrusions presumably representing the late phase magmatic activities (Syafri et al., 2013). Changes of the altered andesite to dacite rocks from mineral assemblages, rock mass, rock volume, and geochemistry views from Godean was discussed by Verdiansyah (2016). Moreover, geochemistry evidence shows that andesite and dacite are comagmatic (Irzon, 2018).

METHODOLOGY

Three samples were collected at the andesite domain on the edge of river in Kaligesing. JTQ-1 is megascopically greyish, fine to medium grain, compact, and relatively unaltered volcanic rock. JTQ-2 was taken from the alteration zone while JTQ-3 was sampled at the vein of the studied location. The altered rock is approximately 30 cm thick with a brighter color than unaltered andesite whilst the vein is relatively dark brown as shown in Figure 2. Both petrography and geochemistry analysis were done in the laboratory of the Centre for Geological Survey in Bandung. Thin sections were prepared for petrography analysis of the sample to decipher the texture and mineral composition of the rock samples.

Trace and rare earth elements contents of the selected samples were analyzed using iCAP-Q Thermo Fisher Scientific's Inductively Coupled Plasma-Mass Spectrometry. The rock samples were prepared based on Irzon (2015). The first step, all the samples were washed and dried outdoor for a day minimum. Then, whole samples were crushed with jaw crusher and were grounded using a ball mill to gain particle size of 200 mesh. Nitric acid (ultra-pure grade), formic acid (ultra-pure grade), and perchloric acid (pro analysis grade) were used in sample digestion which should be done carefully because incomplete dissolution of highly resistant minerals in geology samples may cause biased results for a number of trace and rare earth elements (Bayon et al., 2009). Full suite of rare earth elements (La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu) as well as twelve trace elements (Sc, V, Cr, Ni, Rb, Sr, Y, Ba, Zr, Nb, Th, and U) were analyzed. The CPSs (counts per second) of one blank and six levels of calibration solutions (0.1, 1, 5, 10, 25, and 50 ppb) were measured to produce the calibration curves of analyzed elements. A computer program of the ICP-MS device transformed elements' CPS of samples to concentrations using the previous calibration curves. AGV-2 and GBW 7112 were the two certified reference materials used in this study to certify the quality of measurement results.



modified from Rahardjo et al., (2012).

Figure 1. Geological map of research area and sampling points in Kaligesing, Purworejo.

Figure 2. a) The studied outcrop which is situated at the edge of a river in Kaligesing; b) bright 30 cm thick alteration zone.

RESULT AND DISCUSSION

Petrography

The relatively unaltered sample (JTQ-1) shows hypocrystalline porphyritic texture. Plagioclase (30% of the modal composition) with a lesser extent of quartz, ore mineral, and hornblende are found as phenocrysts embedded in the fine-grained groundmass consisting of microcrystalline feldspar and glass. Although minor sericite (4%) and chlorite (1%) are identified in the sample, the rock is categorized as fresh andesite. On the other hand, the sum modal composition of alteration minerals increases in JTQ-2. About 22%, 7%, 5%, and 2% of sericite, chlorite, epidote, and iron oxide are detected in the altered rock, respectively. Formation of sericite is through the alteration of feldspars mineral whilst chlorite and iron oxide are the results of mafic mineral alteration. The photomicrographs of the studied samples are shown in Figure 3.

(a) (b)

Figure 3. Photomicrographs of the studied rocks: a) JTQ-1, andesite which is mainly comprised of plagioclase with low alteration intensity; and b) JTQ-2, the altered rock shows the much higher composition of alteration minerals than JTQ-1. Cl = chlorite, Op =opaque mineral, Pl = plagioclase, Qz = quartz, and Sr = sericite.

Geochemistry

Vanadium and zirconium are the two most abundant trace elements of the vein whilst barium and vanadium in the altered sample. Sr, Ba, and Y in unaltered andesite are much more abundant than two other samples of 0.1%, 300 ppm, 22 ppm (Table 1). In comparison to the vein and unaltered andesite, the altered rock contains more Cr, Th, and U. The enrichment/depletion of the mobile large ion lithophile elements (LILE), namely Rb, Ba, K, and Sr might be correlated to the alteration (i.e. Chukwu and Obiora, 2018). The alteration process in Kaligesing decreases most of the trace elements except vanadium, zirconium, and neodymium. Barium and strontium depict sharp reductions from the system during alteration. Ba is 1,100 ppm, 426 ppm, and 65 ppm in the fresh andesite, altered andesite, and vein respectively whilst only 13 ppm is detected in the center of alteration even though unaltered andesite contains >300 ppm of Sr. Progressive decrease of Sr and Rb from relatively unaltered andesite through the vein explains the decrease of plagioclase modal composition which is parallel to petrography analysis.

The higher Ba/Sr ratio in the altered sample (6.54) than the relatively fresh one (3.74) confirms the alteration of plagioclase on microscopic analysis. The modal composition of sericite (K-rich mineral, $KAl_2(AlSi_3O_{10})(OH)_2$) is higher in JTQ-2 than JTQ-1 but with lower plagioclase (Ca-rich mineral) to explain their Rb/Sr ratio comparison. The bigger Rb/Sr and the lower Ba/Sr ratios of JTQ-3 in comparison to JTQ-2 might indicate a more degree of K-bearing mineral formation than Ca-rich mineral alteration at the vein. The Zr/Y escalation and Ba/Zr reduction from relatively fresh rock through to the vein of the studies samples (Table 1) confirm the previous study about hydrothermal alteration effects on andesitic rock in Los Azurfres (Verma et al., 2005). Although Pandarinath et al. (2008) research was focussed on rhyolitic rocks, the higher Nb/Y but lower Rb/Zr and Rb/Nb ratios of the altered andesite (JTQ-2) than the unaltered one (JTQ-1) depict the same results.

REE composition in the studied rocks ranges from 30 ppm (vein) to 72 ppm (unaltered andesite). The relatively low to medium REE content indicates that the studied outcrop is not an economic source of the suite of elements as was described by Castor and Hedrick (2006). REE in this study was divided into two groups: Light-REE (LREE, Lanthanum through europium) and Heavy-REE (HREE, Gadolinium through lutetium) as was described by Castor and Hedrick (2006). The shape of the chondrite-normalized REE (Boynton, 1984) pattern of the altered andesite is quite similar to that of the unaltered one: strong enrichments of light-REE (LREE), positive Eu anomaly and relatively flat heavy-REE (HREE) patterns (Figure 4). The total REE amount (ΣREE) tends to increase apparently in samples from the vein toward the unaltered andesite. The enrichment of LREE over HREE, expressed as the chondrite-normalized ratio $(La/Lu)_N$, ranges from 5.8 to 7.3. However, fractionation among LREE expressed as ratio $(La/Eu)_N$ shows much wider variable of 0.9 – 3.4 than HREE's of 0.4 – 1.3 $(Gd/Lu)_N$ (Table 1) to affirm the more immobile nature of HREE than LREE in several studies (i.e. Ling and Liu, 2002; Chen et al., 2016)

Accordingly, it was suggested that hydrothermal alteration removes REE, especially LREE from the fresh andesite. Sericite modal composition was increased on the altered sample based on microscopic analysis. The LREE depletion emphasizes its negative correlation with serisitification on andesitic rocks in several investigations (i.e. Khalaf, 1999; Geogieva and Hikov, 2016). Positive Eu anomaly (Eu*/Eu) tends to increase in samples from the vein outwards: 8.4, 6.9, and 4.7 in JTQ-3, JTQ-2, and JTQ-1, respectively (Table 1). The larger positive Eu anomaly (Eu/Eu*) in the relatively fresh rock reflects its higher modal plagioclase and parallel with the more Sr-Ba-Rb composition than the altered one. Moreover, a previous study stated that hydrothermal reducing solution changes the Eu^{3+} to the more mobile Eu^{2+} (i.e. Christidis, 1998; Shikazono et al., 2008). The drop of Eu anomaly inward to the vein might depict the reduction of Eu^{3+} to Eu^{2+} which was then leached away from the rock.

Table 1. Geochemical composition of the studied samples (in ppm)

Sample	JTQ-1	JTQ-2	JTQ-3	Sample	JTQ-1	JTQ-2	JTQ-3
Sc	8.14	6.07	5.53	Dy	3.16	2	1.23
V	107.4	72.2	93.77	Ho	0.61	0.39	0.3
Cr	25.58	36.72	21.39	Er	1.66	0.99	0.69
Ni	19.52	14.24	16.16	Tm	0.35	0.22	0.14
Rb	30.05	16.07	12.16	Yb	1.99	1.26	1.04
Sr	303.9	65.97	13.76	Lu	0.27	0.19	0.15
Y	22.34	9.59	4.92	Th	2.25	4.18	3.86
Zr	52.14	40.99	75.35	U	1.17	4.07	3.64
Nb	3.04	4.37	5.86	*Ba/Sr*	3.74	6.46	4.77
Ba	1138	426	65.7	*Rb/Sr*	0.10	0.24	0.88
La	15.22	13.4	8.95	*Zr/Y*	2.33	4.27	15.32
Ce	30.08	22.74	14.39	*Ba/Zr*	21.83	10.39	0.87
Pr	0.4	0.2	0.15	*Nb/Y*	0.14	0.46	1.19
Nd	16.8	11.59	5.45	*Rb/Zr*	0.58	0.39	0.16
Sm	2.35	0.49	0.1	*Rb/Nb*	9.88	3.68	2.08
Eu	4.05	1.97	0.62	*∑REE*	72.31	52.28	30.37
Gd	2.88	1.55	0.5	*Eu/Eu**	8.4	6.9	4.7
Tb	0.53	0.34	0.21				

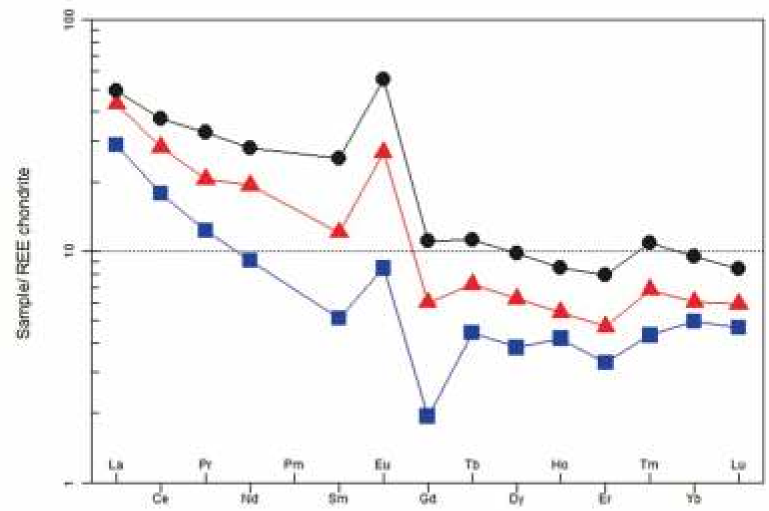


Figure 4. Chondrite (Boynton, 1984)- normalized REE patterns of studied samples.● = relatively unaltered andesite,▲= altered andesite,■= quartz vein.

CONCLUSION

Andesite alteration was indicated in Kaligesing – Purworejo, Central of Java. The relatively unaltered sample was classified as andesite which is majorly built of plagioclase. Various alteration minerals, namely sericite, chlorite, epidote, and iron ore are detected in the altered rock which modifies the rock composition. Higher Sr and Rb abundances in the unaltered sample were related to the higher plagioclase modal composition. The more degree of K-bearing mineral formation than Ca-rich mineral alteration was denoted from the bigger Rb/Sr and the lower Ba/Sr ratios inward to the center of alteration. However, the unaltered, altered, and vein samples show similar patterns: strong enrichment of LREE, positive Eu anomaly, and relatively flat HREE. The alteration might occur in reductive condition base on the decrease of Eu anomaly.

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