



Mobility and fractionation of REEs during deep weathering of geochemically contrasting granites in a tropical setting, Malaysia

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ARTICLE INFO

Article history:

Received 26 September 2012

Received in revised form 24 March 2013

Accepted 18 April 2013

Available online 27 April 2013

Editor: J. Fein

Keywords:

REE

Lanthanides

Granite

Saprolite

Tropical weathering

REE fractionation

ABSTRACT

In this study we describe the mobility and fractionation of REEs in two deep (up to 30 m) tropical weathering profiles developed on two granites from the Kuala Lumpur pluton, Malaysia, sampled at Cheras and Rawang. On the basis of Na₂O and K₂O both are S-type granites, but Rawang has higher CaO, MgO and FeO than Cheras and lower SiO₂. With respect to Al-saturation Rawang is I-type and Cheras is S-type. We compared the two profiles in terms of total REEs, magnitude and changes in Ce and Eu anomalies, REE mobility and LREE/HREE fractionation. Rawang profiles have higher REE contents, display lower mobility for most except the heaviest REEs and show higher LREE/HREE fractionation than those from Cheras. These differences can be linked to differences in primary mineralogy and degree of weathering, the latter controlling the type and volume of secondary minerals. Specifically, bowl-shaped parent-rock-normalised patterns in the Cheras saprolites appear to be a result of apatite dissolution. Moreover, moderate weathering evident in lower Mineralogical Indices of Alteration (MIA) at Cheras has conserved parent rock REE patterns and fractionation factors in the saprolites. By contrast, more intense weathering observed in Rawang profiles has produced abundant kaolinite group minerals that have preferentially retained LREE, which consequently display high LREE/HREE fractionation. This study provides important insights into the factors controlling REE mobility during tropical weathering, and its potential as an indicator of weathering intensity.

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1. Introduction

The lanthanides or rare earth elements (REE), together with Yttrium (Y), form a group of (mostly) trivalent elements that behave coherently in nature owing to systematic variations in their ionic charge to radius ratio (Henderson, 1984). Nevertheless, a variety of processes can disrupt this coherency, fractionating the elements when concentrations are normalised to chondrite or some other accepted standard. Consequently, a fractionation pattern observed on any sample provides a useful tool for elucidating natural processes leading to the formation of the sample, and its provenance (Takahashi et al., 2004). Examples of such processes include, but are not limited to (i) magmatic crystallisation (Bau, 1996; Irber, 1999); (ii) water-rock interaction and hydrothermal alteration (Takahashi et al., 2002), groundwater mixing and evolution (Smedley, 1991; Johannesson et al., 1997, 1999; Dia et al., 2000; Nelson et al., 2003; Tweed et al., 2006) and weathering (Nesbitt, 1979; Alderton et al., 1980; Banfield and Eggleton, 1989; Öhlander et al., 1996; Taunton et al., 2000; Aubert et al., 2001; Bao and Zhao, 2008; Beyala et al., 2009; Harlavan et al., 2009; Ndjigui et al., 2009; Gong et al., 2011; Ma et al., 2011); (iii) sediment dynamics and evolution, including sediment provenance

(McLennan, 1989; McLennan et al., 1993) and monitoring downstream processes (Stille et al., 2006; Steinmann and Stille, 2008); (iv) pollution source tracing (Nozaki et al., 2000; Kulkarni et al., 2007); (v) biological activity (Stille et al., 2006, 2009) (vi) interaction with particulate matter (Gruau et al., 2004) and (vii) solution complexation (Ohta and Kawabe, 2001; Johannesson et al., 2004).

Within the context of catchment-scale weathering, a common approach to understanding REE behaviour is to examine the distribution of REEs with depth in weathering (soil/saprolite) profiles, either in the porewaters (Stille et al., 2009; Ma et al., 2011) or in the mineral fraction (Aubert et al., 2001). The resulting profiles are indicative of the major processes/mechanisms that may be responsible for the mobilisation or otherwise of the REE, leading to fractionation. The main mechanisms often cited include (i) preferential dissolution of REE-containing primary minerals, (ii) solution complexation and the differential mobility of the respective solution complexes, (iii) the role of colloids and secondary minerals and (iv) biological activity. The potential for these mechanisms to interact means that despite a large number of studies over the last three decades, there is still no consistent model for predicting REE mobility and fractionation in granites and other rock types.

Most of the REEs in granites and other igneous rocks tend to be concentrated in accessory mineral phases such as apatite, allanite and monazite (Alderton et al., 1980; Sawka and Chappell, 1988; Bao and Zhao, 2008). The preferential dissolution of these primary

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minerals is therefore likely to play a key role in the mobility and fractionation of the REEs depending on the inherent differences in the mineral patterns (Möller and Giese, 1997; Möller, 2002). As might be expected, accessory minerals that dissolve easily will yield depth profiles showing bulk depletion in total REE in the top horizons of the weathering profile, although the degree of fractionation will likely depend on the REE proportions in the accessory mineral (Bao and Zhao, 2008) and possibly the degree of chemical weathering (Laveuf and Cornu, 2009; Ma et al., 2011). Some accessory phases can occur as inclusions in plagioclase and alkali feldspar, the main rock-forming minerals, so that release of REE will, to a degree, be coupled to major mineral degradation.

Differential stability of the complexes formed in solution can also lead to fractionation of the REE during weathering. For example, the heavy REE (HREE, Gd to Lu) and Y form stronger complexes with carbonate ions (Wood, 1990), reducing their ability to sorb to particles (Tang and Johannesson, 2010). Consequently, natural waters in equilibrium with CO₂ under neutral to alkaline pH display HREE-enriched fractionation patterns whilst the particles inherit LREE-enriched patterns (Sholkovitz et al., 1994). A similar particle scavenging mechanism has been invoked to explain the preferential enrichment of HREE in groundwater (Nelson et al., 2003).

Since weathering results in formation of secondary minerals, some of which are in the colloidal size range, the chemical and structural composition of these secondary minerals can affect the distribution of REEs in weathering profiles. For example, clays are known to adsorb, and act as carriers for REEs in weathering profiles (Öhlander et al., 1996; Galán et al., 2007). The extent of adsorption varies with clay type, with chlorite reportedly showing highest contents (Laveuf and Cornu, 2009). Fe and Mn oxides are important in the distribution and contents of the REEs, although the REE contents do not always correlate with Fe and Mn contents (Yan et al., 1999). These oxides scavenge REEs by several processes, including co-precipitation, adsorption and ion exchange but not by isomorphous substitution. Apparently, the REEs tend to be higher in amorphous than in crystalline forms of these oxides, due to expulsion during ageing and crystallisation (Compton et al., 2003). A characteristic feature of REE association with Mn and Fe oxides is the presence of positive Ce anomalies, particularly when these oxides form concretions in the weathering profile (e.g. Tripathi and Rajamani, 2007). Lastly, REEs are also enriched in secondary phosphates, in particular rhabdophane, florencite and xenotime (Banfield and Eggleton, 1989; Aubert et al., 2001; Köhler et al., 2005). However, as weathering becomes more intense, secondary phosphates re-dissolve, leading to remobilisation of the REEs (Taunton et al., 2000).

Biological activity plays an important role in the cycling of REEs in weathering and soil profiles. Plants recycle REEs within the root zone, although uptake levels are generally low, and in general plants do not fractionate REEs (Tyler, 2004). However, Stille et al. (2006) have shown that vegetation tends to preferentially recycle light REEs, leading to river waters that are depleted in light REEs in catchments with dense vegetation cover. Bacteria and fungal hyphae can also mobilise REEs through organic complexation and dissolution of REE-containing minerals (Feng et al., 2011), and hence control the vertical distribution of REEs in weathering profiles, as demonstrated by Taunton et al. (2000).

Our efforts are concerned with understanding the mechanisms of weathering of granites under tropical conditions in Malaysia, based on two sites, at Cheras and Rawang, in the Kuala Lumpur area (Fig. 1). The sites were chosen because they provided recently engineered cut surfaces and core material that gave a continuous section from the surface soil, through 10–30 m thick saprolite, across a weathering boundary and into unweathered bedrock. We have used changes in mineralogy, major and trace element concentrations as tools to assess the degree of weathering and how this affects the engineering properties of the resulting saprolites. The present paper focusses on the distribution of REEs in the weathering profiles with a view to (i) assessing the mobility

of the REEs during deep weathering of granites; (ii) assessing the degree of REE fractionation during weathering and what mechanisms might be responsible for both mobility and fractionation; and (iii) establishing whether the observed mobility and fractionation can be correlated with degree of weathering based on other alteration indices. We approach the problem by using granites, which although come from the same intrusion display slight differences in bulk rock chemistry, and hence allow us to explore the effect of parent rock chemistry on REE behaviour during weathering.

2. Study areas, sampling and analysis

2.1. Study areas

The study was conducted at two locations, Cheras and Rawang, near Kuala Lumpur, Malaysia (Fig. 1). Both localities are in the large Kuala Lumpur granite pluton, a member of the West Malaya Main Range granite province. Petrographic and chemical details of Malaysian granites can be found in a number of previous studies (Cobbing et al., 1986, 1992; Khoo and Lee, 1994; Azman, 2000; Azman et al., 2008). High precision zircon U–Pb ages of 210 ± 7 and 215 ± 7 Ma for the Kuala Lumpur granite have been presented recently by Searle et al. (2012). The West Malaya granites are predominantly S-type, as defined by Chappell and White (1974), and on the important criterion of Na₂O and K₂O contents (Table 1), Cheras and Rawang are both S-types. However, Rawang has higher CaO, MgO, Fe₂O₃ and P₂O₅ than Cheras, and is less aluminous, indicating a more I-type character. Details are given later.

2.2. Profile characteristics and sampling strategy

Fig. 2 shows the actual profiles that were sampled for this study. The Cheras profile is a cut rock–saprolite–soil slope surface, produced by recent engineering work, comprising 3 berms, each 3 to 5 m in height. The total height of the slope is 13 m. The slope is positioned close to the edge of a granite quarry. The quarry is an open excavation to a depth of about 33 m. Fresh granite samples were obtained from the quarry base. The Rawang profile is a cut soil–saprolite slope about 15 m high. The foot of the slope rests on land levelled by quarrying. Exploration holes were drilled in this levelled surface to obtain samples of saprolite and rock from below the cut slope surface, giving a total section of 33 m.

Two types of sample were used for this study. The first type of samples was taken from the surface of the cut slopes. Soils, saprolites and rock samples were taken at appropriate intervals measured vertically down the cut slope. The soft materials were sampled by driving plastic pipes with 50 mm inner diameter and 3 mm wall thickness into the saprolite. The cores were then placed in airtight plastic bags. The sampler was driven horizontally to a depth of about 15 to 20 cm to avoid the roots of grasses growing on the slope surface. The second sampling type was taken from the base of the exposed cut slope down to known depth in percussion drill holes. Saprolite samples were taken out with a split-spoon sampler. When rock was encountered, coring with a diamond bit head was used and continuous rock core samples were obtained.

The profiles are sub-divided into different zones based on a combination of structural, physical and mineralogical parameters. Each profile contains a *soil* zone at the top, defined as materials in which the original rock structure and material fabric has been destroyed. The material crumbles under normal pressure of the hand, and contains decaying organic matter when observed in hand specimens with an optical microscope. The minerals are imbedded in a clay matrix and contain small to large fractures. Feldspar is absent. The *saprolite* zone is similar to the soil zone except that there is no observable organic material in hand specimen. Feldspar may be present, in small amounts, increasing downwards as the weathering zone is approached. The *weathering* zone is defined as consisting of material where the original rock structure remains intact under normal

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