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Garnierite mineralization from a serpentinite-derived lateritic regolith, Sulawesi Island, Indonesia: Mineralogy, geochemistry and link to hydrologic flow regime

Wei Fu[a,b,*], Yinmeng Zhang[a], Chongjin Pang[a], Xiangwei Zeng[a], Xiaorong Huang[a], Mengli Yang[a], Ya Shao[a], Henry Lin[b,c,d]

[a] Guangxi Key Laboratory of Hidden Metallic Ore Deposits Exploration, Guilin University of Technology, Guilin 541004, China
[b] Department of Ecosystem Science and Management, The Pennsylvania State University, University Park, PA 16802, USA
[c] State Key Laboratory of Loess and Quaternary Geology, Institute of Earth Environment, Chinese Academy of Sciences, Xi'an 710061, China
[d] Shanxi Key Laboratory of Accelerator Mass Spectrometry Technology and Application, Xi'an AMS Center, Xi'an 710061, China

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ABSTRACT

Garnierite represents a significant nickel ore in many lateritic Ni deposits worldwide. To gain a better understanding of its nature and origin, a well-developed garnierite-hosting transect from the Kolonodale area of East Sulawesi, Indonesia, has been investigated using field geology, mineralogy and geochemical data. Garnierite occurs mainly in veins in the lower saprolite of a serpentinite-derived regolith. Mineralogically, it can be determined as an intimate mixture of Ni-rich serpentine-like (lizardite-Népouite) and talc-like (kerolite-pimelite) phases. Results of EMP analyses indicate that Ni is preferentially enriched in the talc-like phases rather than the serpentine-like phases. A sequential precipitation of mineral phases progressively enriched in Ni and Si to form garnierite during weathering is suggested. The Ni-lizardite (2.63–8.49 wt% Ni) with elevated Fe (4.02–6.44 wt%) may have been inherited from saprolite in a first instance and enriched in Ni by cation exchange processes. Newly precipitated minerals are kerolite-pimelite (7.84–23.54 wt% Ni) and then followed by Ni-free quartz. Minor amount of Népouite (23.47–28.51 wt% Ni) occur in laths along shrinkage cracks of previously formed minerals, indicating a late stage paragenetic sequence. With emphasis on a hydrologic consideration, indicators of a preferential flow regime are identified in the garnierite-hosting regolith, including: (i) non-uniform pattern of the garnierite field occurrence, (ii) *syn*-weathering active nature of the garnierite-hosting structures, (iii) close relationship between the garnierite occurrence and vertical Fe—Mn oxides pipes as well as Fe—Mn oxides patched areas, and (iv) specific physico-chemical property of the garnierite location with higher organic matter concentrations but lower pH values compared to surroundings. It is proposed that the origin of garnierite is closely linked to a preferential flow of oversaturated solutions through accessible conduits in the regolith. Garnierite features as colloidal nature, high organic matter and low pH are key-parameters in metal transport and deposition.

1. Introduction

Lateritic Ni deposits are an important ore source, accounting for over 60% of the world's nickel resource (Berger et al., 2011; Kuck, 2013). As a consequence of developments in lateritic ore processing techniques (Mudd, 2010), the amount of Ni being extracted from lateritic ores has increased steadily in recent years, with lateritic Ni ore now contributing over 40% of annual global nickel production (Kuck, 2013). There are three general types of lateritic Ni ore, based on the dominant minerals hosting Ni: oxides, hydrous Mg silicates and clay silicates (Brand et al., 1998; Butt and Cluzel, 2013). In terms of economic significance, hydrous Mg-silicate ores represent the highest-grade type (usually beyond 2 wt% and even > 5 wt% Ni occasionally) and, historically, accounts for about 32% of total lateritic Ni resources with a mean grade of 1.44 wt%, whereas oxide ores represent the largest reserve type, accounting for about 60% of total Ni with a mean grade of about 1.0–1.6 wt% Ni (Butt and Cluzel, 2013).

Garnierite is a specific lateritic Ni ore of grass-green color, poor crystallinity and fine-grained nature, and it is solely hosted in lateritic Ni deposits of the hydrous Mg silicate type (Brand et al., 1998; Butt and

* Corresponding author at: College of Earth Sciences, Guilin University of Technology, Jiangan Road No. 12, Guilin 541004, China.
E-mail address: fuwei@glut.edu.cn (W. Fu).

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Cluzel, 2013; Golightly, 2010). Due to containing a remarkably high Ni concentration, garnierite represents a significant Ni ore type in many lateritic Ni deposits, as reported from the Goro deposit (95.1 Mt, 1.58 wt% Ni), New Caledonia (e.g., Wells et al., 2009), the Sorowako deposit (153 Mt, 1.77 wt% Ni), Indonesia (e.g., Ilyasa et al., 2016; Sufriadin et al., 2015), and the Falcondo deposit (46.5 Mt, 1.21 wt% Ni), Dominican Republic (e.g., Tauler et al., 2009; Villanova-de-Benavent et al., 2014). It is called "green gold" by some mining geologists, and is considered a reliable guide to exploring for Ni ores in laterites because of its striking color and distinctive field occurrence. Moreover, garnierite is of primary importance for determining the mining volumes that can be exploited due to its inhomogeneous occurrence (Cathelineau et al., 2016). However, as a poorly defined weathering product, to gain a definitive identification of the nature of garnierite remains a challenge. To address the nature of garnierite, extensive efforts have been directed at determining the classification and nomenclature of garnierite-forming minerals (e.g., Brindley and Hang, 1973; Brindley et al., 1977; Faust, 1966; Fritsch et al., 2016; Manceau and Calas, 1985; Springer, 1974; Soler et al., 2008; Talovina et al., 2008; Wells et al., 2009). The mineral species of garnierite has been identified as a series of Ni-bearing magnesium phyllosilicates, including serpentine, talc, sepiolite, chlorite and smectite (Manceau and Calas, 1985). In most cases, almost all garnierite samples comprise a combination of two or more of the above mineral species. However, there is still some garnierite that cannot be classified precisely due to its fine-grained nature, poor crystallinity and frequent occurrence as intimate mixtures of different mineral species (Villanova-de-Benavent et al., 2014).

Extensive efforts were made to elucidate the origin of garnierite. Most garnierite discovered in tropical belts is considered to be formed by intense weathering of ultramafic rocks, as in New Caledonia, Indonesia, Dominican Republic and other tropical countries (e.g., Cathelineau et al., 2016; Fritsch et al., 2016; Sufriadin et al., 2015; Tauler et al., 2009; Villanova-de-Benavent et al., 2014; Wells et al., 2009). Textural variations with poorly ordered materials suggest that garnierites have been precipitated from colloidal suspensions at low-temperature conditions (Brindley et al., 1977). Gali et al. (2012) gave a thermodynamic explanation for the temporal and spatial precipitation of different garnierite phases at standard temperatures and pressures.

Notably, garnierite is hosted in lateritic Ni deposits of the hydrous Mg silicate type (Brand et al., 1998; Butt and Cluzel, 2013), which indicates that it is limited to those regions where ultramafic rock exposures, tropical climatic regime and active tectonic setting coexist (Brand et al., 1998). Cluzel and Vigier (2008) and Villanova-de-Benavent et al. (2014) suggest that a brittle *syn*-tectonic environment during ultramafic-rock weathering has played an important role in the formation of garnierite. The origin of garnierite veins is also linked to the present-day topography and water table movement (Cathelineau et al., 2016). However, Talovina et al. (2008) reported that some garnierites from the Urals differ significantly from their counterparts of weathering origin. There are two paragenetic mineral associations: hydrothermal association and exogenous association, and those hydrothermal associated garnierite-forming minerals (pecoraite–chrysotile–quartz) were promoted by the contact-metasomatic impact of local thermal field and fluids of diorite massif (Varlakov, 1992), which suggest that the Uralian garnierites are of both hydrothermal and weathering origin (Talovina et al., 2008). Fritsch et al. (2016) proposed a new model for interpreting the formation of garnierite and accompanying minerals (deweylite), suggesting that some garnierite infillings in faulted peridotites of New Caledonia are probably of alteration closely linked to post-obduction tectonic activity.

Fu et al. (2014) first reported the discovery of garnierite in the Kolonodale exploration area, Sulawesi Island, Indonesia, and its contribution as an important source of high-grade lateritic Ni ore to local mining. When compared to other typical garnierite occurrences worldwide, such as those from New Caledonia (e.g., Cathelineau et al., 2016; Fritsch et al., 2016; Wells et al., 2009), Central America and South America (e.g., Gleeson et al., 2004; Soler et al., 2008; Villanova-de-Benavent et al., 2014) as well as central Africa (e.g., Prendergast, 2013), this case is marked by being associated with a tectonically active region at the circum-Pacific, a kind of serpentinite derived lateritic regolith as well as a typical tropical rain forest climate. It could provide a good example for understanding garnierite occurrences in Southeast Asia. However, the nature and formation mechanism of the garnierite from the Kolonodale area have not been studied in detail.

The aim of this study is to: (i) reveal the property of garnierite in the study site from macro- to micro-scale by detailed field examination, textural relationships, and new mineral and geochemical data; (ii) elucidate the difference between the garnierite and nearby weathered products by conducting a systematic examination along a garnierite-hosting transect; and (iii) find evidence that could bridge the garnierite occurrence and hydrologic flow regime in a profile scale, and propose a new model for garnierite formation by coupling Ni geochemical process with hydrologic process.

2. Geological background and field occurrence

The K-shaped Sulawesi Island lies at the convergence zone of three tectonic plates: Eurasian, Pacific and Indian-Australian. Four lithotectonic belts are identified in this island (Fig. 1A): the West Sulawesi Volcano-Plutonic Arc Belt, the Central Sulawesi Metamorphic Belt, the East Sulawesi Ophiolite Belt, and the Continental Fragments of Banggai-Sula, Tukang Besi, and Buton (Mubroto et al., 1994). The study site, the Kolonodale area in Central Sulawesi Province, is located within the East Sulawesi Ophiolite Belt. This ophiolite belt represents a part of the Circum Pacific Phanerozoic multiple ophiolite that were emplaced from the Cretaceous to the Miocene (Hall and Wilson, 2000). It extends 700 km from north to south and crops out over > 15,000 km^2 on Sulawesi Island (Kadarusmana et al., 2004). Ultramafic bodies are widely exposed within this belt. They mainly consist of harzburgite and lherzolite peridotite, and a large part of them has been serpentinized in varying degrees. In the study area, ultramafic rocks have been regionally metamorphosed at moderate-high grade and, petrographically, they can be determined as serpentinite (Fu et al., 2014) essentially composed of serpentine and minor orthopyroxenes with trace clinopyroxenes and spinels. There are two other lithological units in the surroundings of the ultramafic complex: alluvial and sedimentary lacustrine rocks of Quaternary and Cretaceous sedimentary rocks (Kadarusmana et al., 2004). Faults, fractures, and joints, striking mainly in NNW direction, are largely observed in the serpentinite body due to regional tectonic activities.

Eocene-Miocene and Miocene-Recent plate movements have greatly influenced the geological features of Sulawesi Island. The inferred regional uplift as a whole can be linked to underthrusting of oceanic crust, which resulted in the surface exposure of ultramafic complexes with a large area from the central to north of the Island (Golightly, 1979). It appears that these ultramafic rocks, including the serpentinite body in Kolondale area, have been exposed since the Miocene. The climate in the study area is typical of a humid tropical rainforest with high temperatures and abundant rainfall, which support lush vegetation over the entire region. Local geomorphology is characterized by a low-moderate relief with gently sloping surfaces. Under these prolonged and pervasive weathering conditions the exposed serpentinite has been subjected to strong chemical alteration, giving rise to a thick lateritic regolith. Such regolith hosts lateritic Ni deposit. Also, soils derived from ultramafic bedrock have a number of extreme chemical properties, which influences the development of a specific ecosystem renowned for high levels of endemism (e.g., plant species restricted to a limited geographic area and the occurrence of nickel hyperaccumulators) (van der Ent et al., 2013).

In the study area, the thickness of serpentinite-derived lateritic regolith is largely heterogeneous with the change of geomorphic site. It