



Platinum-alloy and sulfur saturation in an arc-related basalt to rhyolite suite: Evidence from the Pual Ridge lavas, the Eastern Manus Basin

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Abstract

We have measured the platinum group element (PGE) and Re concentrations of arc-type lavas from the Pual Ridge and the surrounding area in the Eastern Manus Basin. These magmas followed an Fe-enrichment trend to produce a wide range of compositions with MgO varying between 8 and 0.1 wt.%. We found distinct differences in the PGE geochemistry of the high (>3 wt.% MgO) and the low-Mg lavas (<3 wt.% MgO). During the early stages of fractionation, the concentrations of Pd, Cu, Au and Re increase whereas Ir, Ru, Rh and Pt decrease. Co-variations of Ir, Ru and Rh with Pt in the high-Mg lavas suggest that the depletion of these elements is due to Pt-rich alloy saturation. This is consistent with the high Pt contents in the high-Mg lavas, which is close to the solubility of Pt in the basaltic melt at similar conditions. In contrast, the concentrations of all PGE and Re drop rapidly in the low-Mg lavas (except for Ru and Ir), with the PGE concentrations falling at a rate that is appreciably faster than Cu, which we attribute to sulfide saturation. As a consequence, there is a marked decline in Pd/Cu in the low-Mg lavas and we suggest that this ratio is the best indicator of sulfide saturation in an evolving magmatic system.

A feature of the data is that duplicate analyses of the same sample often do not agree within error. We attribute this scatter to the nugget effect, with nuggets of a Pt-rich alloy in the high-Mg lavas and sulfide blebs in the low-Mg lavas. The PGE concentrations of phenocryst-bearing high-Mg lavas are higher than in the associated glassy lavas, and scatter on MgO variation diagrams is significantly reduced if only glassy lavas are considered, which indicates that the micron scale Pt-rich alloy grains are intimately associated with the phenocrysts. Our results provide strong evidence that Pt-rich alloys can crystallize from a basaltic-andesitic magma, along with the silicate minerals, and fractionate Pt from Pd during magmatic differentiation. As a consequence, Pd/Pt increases during Pt alloy fractionation and this ratio can be used to identify Pt-rich metal saturation. The Pual Ridge alloys are Pt-rich because the primary magmas are Pt-enriched and Ir-depleted, which is typical of arc-related magmas.

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1. INTRODUCTION

There is an abundance of platinum group element (PGE) data for mafic and ultramafic rocks because (1) important Ni–Cu(-PGE) deposits are hosted by mafic–ultramafic rocks so there is considerable economic interests in their PGE geochemistry and (2) PGE data of these rocks provide

important constraints on mantle petrogenesis, and fractional crystallization and differentiation of mantle-derived magmas. In contrast, the PGE geochemistry of felsic-intermediate rocks is virtually unknown due to scarcity of data. This is principally because the concentrations of PGE of these rocks are so low that analysis is challenging. However, many of arc-related intermediate to felsic magmatic systems are associated with hydrothermal Cu, Au and Cu–Au deposits and considered to be a primary source for the ore-forming metals (Robb, 2004). The behavior of chalcophile elements including Cu and Au will be sensitively

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controlled by sulfide or volatile saturation because these elements are highly compatible with both phases. Identifying sulfide saturation in an evolving felsic-intermediate system is therefore critical to understanding the geochemistry of chalcophile elements and its potential for Cu–Au mineralization. This could be accomplished by direct analyses of these elements. However, one advantage of studying the PGE in addition to Cu and Au is that they have much higher partition coefficients with regard to immiscible sulfides than Cu and Au (Francis, 1990; Peach et al., 1990; Bezmen et al., 1994; Crocket et al., 1997; Ripley et al., 2002; Fonseca et al., 2009) and are therefore more sensitive indicators of sulfide saturation. Furthermore, the PGE are less mobile in low temperature fluids than Cu and Au (e.g. Crocket, 2000). Mobility of Cu and Au in the altered rocks associated with Cu, Au and Cu–Au mineralization often leads to erratic data that are difficult to interpret.

With these overall aims in mind, and as a first step towards understanding the geochemistry of the PGE in felsic-intermediate systems, we analyzed a suite of arc-type lavas from the Pual Ridge and vicinity that underwent extreme fractionation from basalt to rhyolite. This suite was chosen because its major and trace element geochemistry had previously been well documented by Kamenetsky et al. (2001), Moss et al. (2001), Sun et al. (2007), Jenner et al. (2010) and Park et al. (2010) who showed that it followed an Fe-enrichment trend to produce a range of compositions with MgO varying between 12 wt.% to 0.1 wt.%, and SiO₂ between 51 wt.% and 75 wt.%. Jenner et al. (2010) showed that the evolving suite becomes magnetite-saturated at 4 wt.% MgO, which lowered Fe³⁺/Fe²⁺ of the melt, consequently the *f*O₂, leading to sulfide saturation at 3 wt.% MgO. We will show that the most primitive samples analyzed in this study were saturated with a Pt-rich alloy and that fractionation of this alloy depleted all of the PGE except Pd whose concentration increased with fractionation. We will also show that the rate of depletion of all PGE, including Pd, accelerated markedly once the magma reached sulfide saturation at 3 wt.% MgO.

2. GEOLOGICAL BACKGROUND AND SAMPLES

The Pual Ridge is a recent volcanic complex located in the western part of the South East Rift, Eastern Manus Basin, southwest Pacific (Fig. 1; Taylor et al., 1994; Martinez and Taylor, 1996). The Pual Ridge lavas show typical features of island arc lavas such as enrichment of Pb and LILE relative to HFSE and REE, and depletion of Nb and Ti relative to adjacent elements on a chondrite-normalized trace element diagram (Sinton et al., 2003; Park et al., 2010). These features originated from the mantle below the Pual Ridge region, which is interpreted to contain an ancient subduction component (Sinton et al., 2003; Park et al., 2010). The major and trace element data as well as Sr, Nd and Pd isotopic compositions suggest that the wide range of compositions displayed by the lavas from the Pual Ridge area is mainly caused by low pressure (~1 kbar) fractional crystallization and that the magma is derived from a homogeneous mantle source (Sinton et al., 2003; Sun et al., 2007; Jenner et al., 2010; Park et al., 2010).

The Pual Ridge lavas can be subdivided into two groups based on the onset of sulfide crystallization; sulfide-undersaturated high-Mg lavas with >3 wt.% MgO and sulfide-saturated low-Mg lavas with <3 wt.% MgO (Fig. 2). The early crystallizing mineral assemblage in the high-Mg lavas consists mainly of olivine, plagioclase and clinopyroxene, with minor Cr spinel that occurs as inclusions in olivine phenocrysts (Kamenetsky et al., 2001; Sinton et al., 2003). Fractional crystallization of plagioclase, clinopyroxene and magnetite controls the compositional variation in the low-Mg lavas (Kamenetsky et al., 2001; Sinton et al., 2003). The Pual Ridge lavas investigated here are the same samples that were analyzed by Moss et al. (2001), Sun et al. (2003, 2004, 2007) and Jenner et al. (2010), and the MgO, Cu, Au and Yb data were taken from their works.

3. ANALYTICAL TECHNIQUES

Glassy parts of the Pual Ridge lavas were hand-picked for analyses using a stereoscopic microscope to exclude altered portions. Two samples (MD3-p and MD101A-p) with phenocrysts (~10% modal phenocrysts) were analyzed in order to investigate the effect of phenocrysts on PGE and Re abundances. The selected lava fragments were cleaned ultrasonically in dilute HCl and Milli-Q water, dried, and powdered in an alumina mill. The alumina mill is known to contribute the lowest PGE contamination among conventional mills (e.g. Evans et al., 2003). Whole rock PGE and Re concentrations of the Pual Ridge lavas were measured using a Ni–sulfide fire assay – isotope dilution method, which is described in detail by Park et al. (2012c). Briefly, 0.45–3.1 g of sample powder was mixed with Ni, S and sodium borax powder in the ratio of sample:Ni:S:Na-borax = 10:1:0.5:10. A mixed enriched PGE (⁹⁹Ru, ¹⁰⁵Pd, ¹⁸⁵Re, ¹⁹¹Ir, and ¹⁹⁵Pt) spike solution was added to the powder. The mixture was dried for 60 min at 100 °C and fused in a preheated furnace at 1100 °C for 30 min. In order to provide reducing conditions during fusion, a second outside crucible, containing ~0.1 g of flour was used and N₂ gas was introduced into the open furnace at a flow rate of ~0.03 m³/min. After quenching, Ni–sulfide beads were collected and dissolved in 6M HCl. The solution was then filtered through a filter paper (0.45 μm cellulose membrane, Millipore), prior to digestion of the filter paper in aqua regia. After complete digestion, the solutions were dried down to approximately ~100 μl and diluted with 2% HNO₃.

An Agilent 7500 quadrupole ICP-MS at the Australian National University (ANU) was used to measure the isotopes of PGE and Re. All samples were analyzed in duplicate to assess heterogeneity of a sample powder aliquant. The sensitivities in each analysis using the Agilent 7500 were 6.7–9.0 × 10⁴ cps/ppb for mass 89, 4.1–5.6 × 10⁴ cps/ppb for mass 140 and 2.5–3.9 × 10⁴ cps/ppb for mass 205. The argide production rate measured from ⁶¹Ni⁴⁰Ar/⁶¹Ni was 0.9–1.0 × 10⁻⁴. The duplicate (second sample) was re-analyzed by an Agilent 7700x quadrupole ICP-MS, which provided higher sensitivity of 1.5–2.3 × 10⁵ cps/ppb for mass 89, 140 and 205 and a lower argide production rate of 0.4 × 10⁻⁴ than the Agilent 7500 ICP-MS.

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