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Vapor transport of silver and gold in basaltic lava flows

Peishu Li and Alan E. Boudreau*
Division of Earth and Ocean Sciences, Nicholas School of the Environment, Duke University, Durham, North Carolina 27708, USA

ABSTRACT

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INTRODUCTION

In recent years, volcanic gas emission data, vapor inclusion compositions, and experimental studies have independently suggested that gold (Au) and silver (Ag) can be more enriched in surface volcanic vapor than in silicate liquid (Lowenstern et al., 1991; Allard et al., 2000; Migdisov and Williams-Jones, 2013). Few studies, however, examined evidence of vapor-assisted metal transport and deposition in basaltic systems. Kindle (1970) reported native copper (Cu) deposits in Mesoproterozoic Coppermine River flood basalt. Sisson (2003) noted a single native Au bleb in fresh basanite glass in an early submarine lava flow at Kīlauea volcano (Hawaii, USA). Zhang et al. (2006) reported native Au and Cu inclusions in the olivine phenocrysts of picritic lava at Emeishan large igneous province in southwest China. Other authors, while finding no native metals directly, noted localized metal enrichments in various mafic to mafic-intermediate rocks (Keays and Scott, 1976; Plail et al., 2014).

Here, we report native Cu, Ag, and Au in a pāhoehoe flow from Kīlauea volcano, an a'ā flow from Mauna Loa volcano (Hawaii), and a mid-oceanic-ridge basalt (MORB) from the Chile Ridge in the southeastern Pacific Ocean (Fig. 1). To our knowledge, this is the first sighting of native Ag in basalt, and the only other finding of native Au in Hawaiian lava flows after Sisson (2003). We attribute the relatively large native Ag and Au grains to igneous vapor transport during lava solidification.

*E-mail: boudreau@duke.edu

METHODS AND RESULTS

Detailed methods are described in the GSA Data Repository[1]. In short, polished and carbon-coated basalt thin sections were scanned with a Cameca CAMEBAX electron microprobe at Duke University (North Carolina, USA). Native metals with large atomic weights like Ag and Au were recognized by their brightness in backscattered electron (BSE) imagery. Energy dispersive spectrometry (EDS) analysis was conducted on these grains to obtain specific mineral composition, and X-ray composition maps were constructed to record the specific mineral and textural environment proximal to the native metal.

Native Cu and Ag were present in all samples except ML-2 (Mauna Loa), and native Au grains were observed in samples KL-2 (Kīlauea), ML-1, and MORB (Table DR1 in the Data Repository). All grains were found along grain boundaries, groundmass cracks, or embedded in groundmass or glasses, but not as mineral inclusions. Ag and Au typically occurred as nearly pure metals. Ag-Au alloys were comparatively rarer, though one electrum (a gold-silver alloy, with trace amounts of copper) grain was found in KL-2 (Fig. DR3). Aside from the lone occurrence of a Cu-Au alloy, no native Au was discovered in KL-1, which was collected near KL-2 from the same lava flow. In hand samples, the vesicle diameters of KL-1 (0.5–0.9 cm) were considerably larger than those of KL-2 (0.1–0.3 cm), consistent with the former being closer to the more rapidly cooled upper flow margin (Sahagian et al., 2002).

Cu occurred in native phase, as Cu-O ± Fe oxide or a poorly defined Cu-Fe-S ± O sulfide phase (Table DR1). Cu-Fe sulfides (5–10 μm in diameter), and sulfides in general, were rare among Hawaiian basalts. When present, they were spheroidal and typically heavily oxidized. The sulfides were found in groundmass in Kīlauea samples, and exclusively enclosed in olivine phenocrysts in Mauna Loa samples. Sulfides (mainly pyrrhotite) were more common in the MORB, but Cu-Fe sulfides were still rare (Table DR1).

In Hawaiian samples, spheroidal native Cu ranged between 3 and 12 μm in diameter (Fig. DR1). Two native Cu grains from Kīlauea had a modest sulfur signature. The MORB sample contained the largest native Cu grain (~20 μm diameter). Minor Ag and Au signatures were noted in several Cu-rich grains in MORB and Hawaiian samples (both <10 wt%), and two Cu-rich grains in KL-1 and ML-1 were alloyed with a strong Au signature (>20 wt%).

Native Au ranged from <3 to 50 μm in diameter (Fig. DR2). Smaller grains were spheroidal and present in KL-2, ML-1, and MORB. Larger grains were found in KL-2 only. They were irregularly shaped and mostly found in cracks/voids in the groundmass matrix. Most native Au did not alloy with Cu or Ag, except the one electrum (Fig. DR3) and the two Cu-Au alloys reported above.

Spheroidal native Ag ranged from 3 to 40 μm in diameter in Hawaiian basalts. The largest native Ag grain was found in MORB. A low, but consistent, Cl signature (typically <1 wt%; Cl = 0.46 wt% in the Ag grain in Figure 2) and varying sulfur signatures (ranging from 0 to 20 wt%) were found in native Ag from all samples. No other regions were associated with Cl aside from native Ag grains. One native Ag grain in KL-2 was situated in the center of a roughly circular area ~120 μm across composed of dendritic magnetite

[1]GSA Data Repository item 2019317, sample descriptions, methods, supplemental figures, and a table with whole-rock analysis of samples, is available online at http://www.geosociety.org/datarepository/2019/, or on request from editing@geosociety.org.

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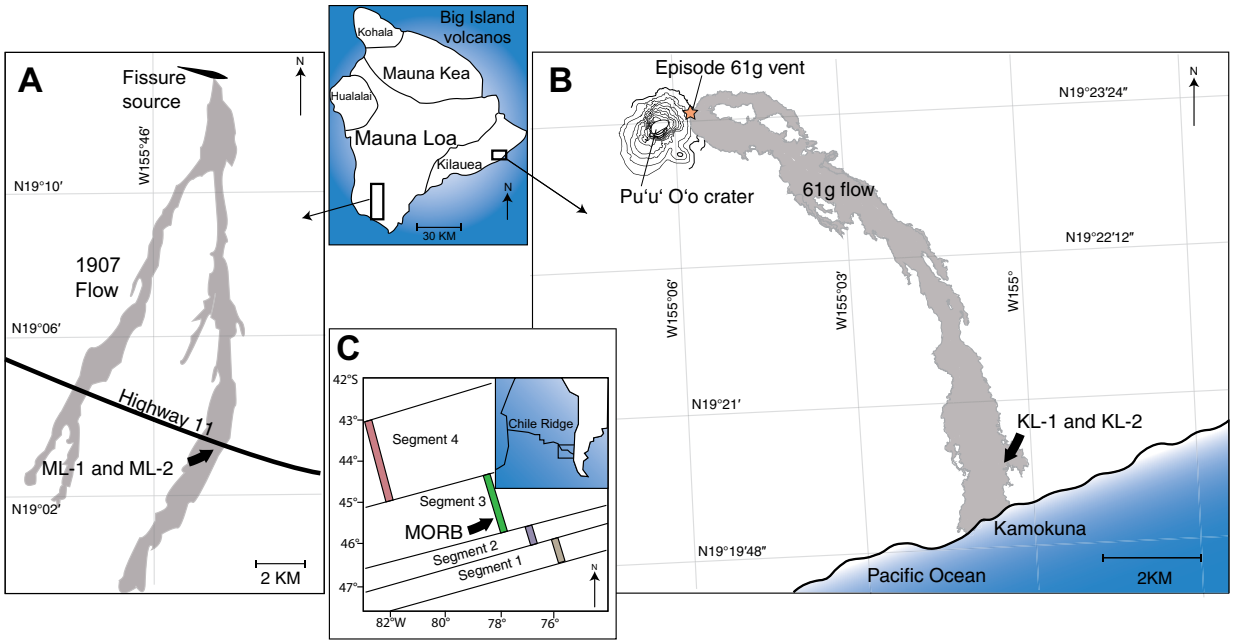

Figure 1. Sample locations. (A) C.E. 1907 lava flow at the Southwest Rift Zone of Mauna Loa (Hawaii, USA), redrawn from Zimbelman et al. (2008). (B) Location of current 61g flow (the 7th flow[g] in the sequence of events that compose the 61st episode of the ongoing East Rift Zone eruption) at Kīlauea, Hawaii, redrawn from the Hawaiian Volcano Observatory (2017). Kamokuna—Kamokuna ocean entry location. (C) Location of the mid-oceanic ridge basalt (MORB) sample from the Chile Ridge (southeastern Pacific Ocean), redrawn from Klein and Karsten (1995).

intergrown with plagioclase. Surrounding this patch, there was magnesium-rich pyroxene (Fig. 2). Proximal to the native Ag was a Mg-poor, Fe-rich center zone, transitioning to a Mg-rich, Fe-poor zone farther away (Figs. 2C and 2D). The magnetite in the circular patch was compositionally similar to other magnetites on the same thin section. Similar crystallization morphologies were observed elsewhere in KL-2 (Fig. DR3) and in the MORB (Fig. 3).

DISCUSSION

The total number of native metals reported here is by no means an exhaustive survey of all metals in our samples. Nonetheless, it sufficiently demonstrates that native precious metal abundance in basaltic lava flows may have been previously underestimated. The morphology and precipitation environment of the native metals observed here differ from previous reports (Sisson, 2003; Zhang et al., 2006). Unlike the Cu and Au grains in olivine reported by Zhang et al. (2006), large olivine phenocrysts were only present in the Mauna Loa a‘ā flows and were metal-free. A typical Hawaiian basalt has bulk concentrations of Cu = 133 ppm, Ag = 0.09 ppm, and Au = 0.0027 ppm, which are significantly lower than saturation levels of 600 ppm, 6 ppm, and 0.49 ppm, respectively (Crocket, 2000; Ripley et al., 2002; Bell et al., 2011; Zajacz et al., 2013; Greaney et al., 2017; see the Data Repository). A *post-hoc* whole-rock analysis also failed to detect Ag and Au in our samples, whereas Cu concentration was consistent with values reported by Crocket (2000) and Klein and Karsten (1995) (see also the Data Repository). These observations suggest that the native metals found here were unlikely to remain xenocrysts during transport, given the metal-undersaturated composition of the magma. Sisson (2003) inferred that the native Au in the Kīlauea submarine basanite precipitated out of a resorbed Au-rich immiscible sulfide liquid. Given the high bulk sulfur content (1000–3000 ppm; Sisson, 2003) and submarine eruption setting, the basanite was unlikely to have undergone major sulfur loss, while the Hawaiian samples here experienced extensive shallow sulfur degassing based on the relative paucity of sulfides.

Three hypotheses may account for the native metals observed here:

(1) The metals were concentrated by an immiscible sulfide liquid (given their chalcophile nature) and formed later through sulfur loss similar to that suggested by Sisson (2003).

(2) The metals were directly precipitated from the interstitial silicate liquid.

(3) The metals were transported and precipitated from a vapor phase.

Although Cu may be partially partitioned into Fe-Ti oxides, its affinity for sulfide is at least 2–3 orders of magnitude higher in basaltic systems (e.g., Ripley et al., 2002; Liu et al., 2015). Therefore, sulfur loss from a magmatic Cu-sulfide phase is the preferred explanation for the observed Cu-Fe-S ± O, Cu-O ± Fe, and native Cu grains. These phases are expected when sulfur loss and variable oxidation of the residual assemblage (Stone and Fleet, 1991; Li and Boudreau, 2017) occur through a general redox reaction:

$$\underset{\text{Chalcopyrite}}{3CuFeS_2} + \underset{\text{gas}}{9O_2} \rightarrow \underset{\text{metal}}{Cu^0} + \underset{\text{oxide}}{Fe_3O_4} + \underset{\text{oxide}}{2CuO} + \underset{\text{gas}}{6SO_2}. \quad (1)$$

Given the chalcophile nature of Cu, Ag, and Au in the absence of an Fe metal alloy, if these metals were pre-concentrated in an immiscible sulfide liquid and later formed alloys and native metals as a result of sulfur loss, one would expect to see either Cu being the predominant metal in the alloy or, perhaps, mixed clusters of Au-Ag grains with Cu grains. The few native Cu grains with minor amounts of Ag and Au (both below 10 wt%) likely formed through such a pathway. However, precious metal grains, in general, were rarely associated with Cu-rich phases, suggesting that simple desulfurization of sulfide cannot account for the isolated occurrences of native Au and Ag.

The data reported here imply that a typical Hawaiian basalt is undersaturated in Au and Ag by ~2 orders of magnitude. Using these bulk rock concentrations, direct precipitation of metals from a silicate liquid would require 10.2 cm^3 and 0.17 cm^3 of magma in order to form a 5 µm native Au or Ag grain, respectively, assuming 100% removal efficiency. A simple partitioning model under Rayleigh fractionation and without sulfide saturation (Wernette et al., 2019; see also the Data Repository) suggests that Au and Ag can become increasingly concentrated in a separating vapor over the course of solidification, forming hydrated complexes (Lowenstern et al., 1991). This suggests that a vapor can be more efficient in transport and precipitation than a silicate liquid alone.

We propose a two-stage enrichment model to explain the unusual size and separate precipitation of Au and Ag. During lava-flow emplacement and solidification, extensive sulfur loss suppressed sulfide saturation and resorption of any earlier precipitated sulfide, if present. Ag and Au were initially enriched in the residual interstitial liquid and then further concentrated by preferential partitioning into a separating vapor phase. Under surface conditions, in Hawaiian basalts, Au was likely transported as an

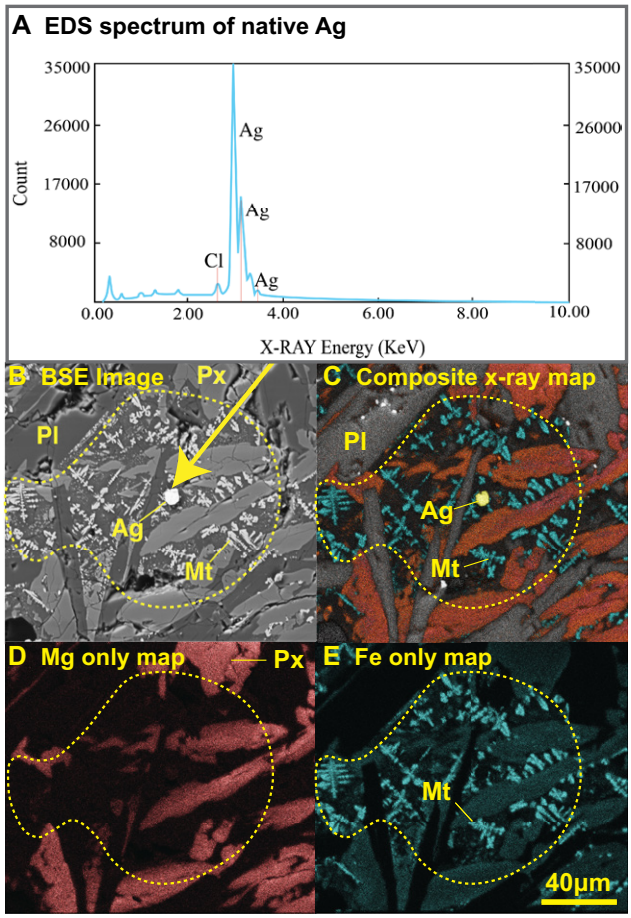

Figure 2. Native silver (Ag) in sample KL-2 from Kīlauea, Hawaii. (A) Energy dispersive spectrometry (EDS) spectrum of the silver grain shown at arrow in panel B. Note the modest but distinct Cl peak (0.46 wt%). (B) Backscattered electron (BSE) image. Ag grain occurs in center of a roughly circular patch ~120 μm in diameter (circled dashed line area) composed of abundant dendritic magnetite (Mt) intergrown with plagioclase (Pl), surrounded by magnesium-rich pyroxene (Px). (C) X-ray composite composition map. (D) Mg X-ray map. (E) Fe X-ray map. Deficiency of magnesium minerals around the patch and an abundance of magnetite inside the patch can be seen in Mg and Fe X-ray maps, respectively.

AuHS·H_2O complex (Gibert et al., 1998), while Ag was likely transported as AgCl·H_2O (Migdisov and Williams-Jones, 2013). Both HS^- and Cl^- are present as vapor phases at Kīlauea (Andres et al., 1989; Mather et al., 2012). The Cl signature in the native Ag in the deep-ocean MORB sample suggests that metal was transported as a hydrated chloride complex in a hydrothermal fluid during late volatile saturation. As the metal-rich vapor rose from the hot lava flow interior to cooler upper margins in both the MORB and Hawaiian cases, the metals precipitated as a result of solubility decreases during cooling and/or changes in the oxidation state of the ligands (e.g., S^{2-} in H_2S to S^{4+} in SO_2; Christie et al., 1986).

Separate transport and precipitation mechanisms with different carriers (sulfides versus vapor, Cl^- versus HS^- complexes) led to the decoupled occurrences of the three native metals and the paucity of alloys. The morphology and bulk composition of the native Ag and Au in this study resemble native metal condensations in volcanic fumarole deposits (Yudovskaya et al., 2008; Chaplygin et al., 2015). No evidence of hydrothermal alteration was observed around the native metals, which is commonly associated with fumarole deposits (Chaplygin et al., 2015). However, volcanic fumaroles have far higher discharge fluxes over longer time scales than basaltic lava flows, and will have more-pronounced alteration. Indeed, no alteration was seen in our study, even around the vesicle walls in our samples (e.g., Fig. DR6).

The metal-bearing patches of aphyric or Fe-rich mineral assemblages in the MORB and Kīlauea samples are inferred to be pipe vesicles infilled with residual volatile-rich liquids. During degassing, upward vapor migration raises local porosity, attracts more evolved silicate liquid, and lowers liquidus temperature, which further promotes vapor migration (Fowler et al., 2015). This mechanism allows vapor to scavenge metals from a much larger volume of liquid more effectively than either local sulfide saturation or random bubble growth. A basalt flow with a depth <3 m is stratified into an upper and lower vesicular zone and a relatively vesicle-free middle zone (Sahagian et al., 2002; see Fig. DR5). At the upper and lower margins, rapid cooling will preserve exsolved bubbles and pipe vesicles and produce a vesicular texture. The bubble-free core represents fully degassed lava, with vesicles either collapsed or infilled. The vesicular Kīlauea samples come from the transition depth (1 m below surface) between the upper vesicular and middle infilled zone, with evidence of vesicle infilling by evolved silicate liquid when examined under microprobe. It is inferred that after

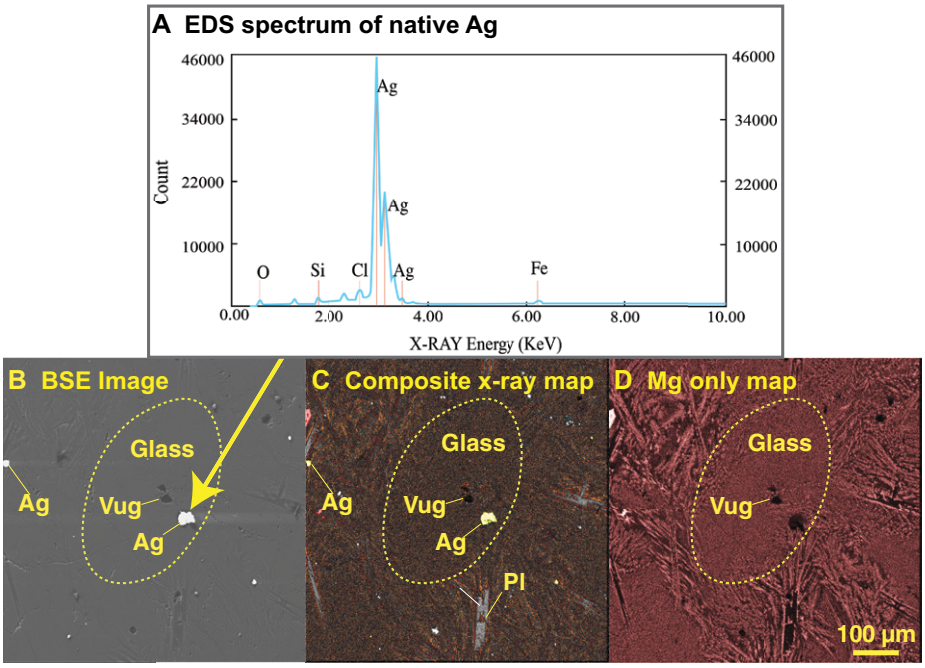

Figure 3. Native silver (Ag) in mid-oceanic-ridge basalt (MORB) from the Chile Ridge (southeastern Pacific Ocean). (A) Energy dispersive spectrometry (EDS) spectrum of the silver grain shown at arrow in panel B. Note the distinct Cl peak. (B) Backscattered electron (BSE) image. The native silver is embedded in a roughly circular area of ~200 μm in diameter (yellow dashed line area) of aphanitic glass with a vug (cavity in the rock), similar to the patch structure identified in Figure 2. (C) X-ray composite composition map. (D) Mg X-ray composition map. X-ray maps show the patch surrounded by a much more crystallized region defined by numerous needle-like intergrown magnesium-enriched minerals and plagioclase (Pl).

metal precipitation in the vesicles, they are infilled at the transition zone (Fig. DR5). Notably, open vesicle rims in the Kīlauea samples are covered with dendritic magnetite similar to that seen in infilled vesicles, presumably at the onset of vesicle infill (Fig. DR6). The compositional similarity of magnetites inside and outside the infilled vesicles supports the hypothesis that the minerals inside the infilled vesicles formed from a late-stage liquid.

In Hawaiian samples, the native metals are more commonly found in the Kīlauea pāhoehoe samples than in the Mauna Loa a'ā samples. The pāhoehoe flow formed from a flow lobe that stopped flowing and underwent inflation with concurrent degassing. Such flows typically exhibit frozen pipe vesicles at the lower and upper margins, but these features are absent in the central flow. In contrast, a pāhoehoe flow can transition to an a'ā flow morphology under sustained movement and prolonged cooling, crystallizing, and degassing. Loss of metals to degassing or resorption of earlier-precipitated Au and Ag grains as they are reincorporated into the moving a'ā flow can cause the relative native metal paucity in the Mauna Loa samples.

Given the otherwise incompatible nature of Cu, Au, and Ag, their behavior is strongly controlled by either the presence of sulfide or a vapor phase. Vapor transport of chalcophile metal in mafic systems may be most effective in sulfur-poor lavas or when vapor saturation occurs before sulfide saturation, which may have occurred in the possibly CO_2-rich MORB sample. The high-temperature metallogenic mechanisms discussed here also support vapor transport of ore elements in plutonic mafic rocks, especially layered intrusions formed during basaltic magma crystallization (e.g., the Au horizon of the Skaergaard intrusion in East Greenland). Ongoing work by our laboratory has noted native Ag throughout the Skaergaard intrusion (Wernette et al., 2019), which is abnormally sulfur-poor relative to other intrusions, and may represent another example of Ag remobilization via vapor transport. Finally, Au in basaltic rocks/greenstones is commonly considered to be easily mobilized by hydrothermal fluids, to produce gold deposits. Our results suggest that Au may be pre-concentrated during lava solidification as relatively large grains before subsequent hydrothermal remobilization.

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