Experimental determination of Au solubility in rhyolite melt and magnetite: Constraints on magmatic Au budgets

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ABSTRACT

The solubility of Au metal in rhyolite melt and coexisting magnetite has been determined at 800 °C, 140 MPa, and \( f_{\text{O}_2} \approx \text{NNO} \) in a Au-metal saturated, sulfur-free, vapor-brine-silicate melt system. Whole crystals of magnetite suspended within quenched rhyolite glass were analyzed by LA-ICP-MS. These data yield a solubility of Au in magnetite on the order of 2 μg/g. The solubility of Au metal in rhyolite melt hosting the magnetite crystals is on the order of 500 ng/g. These data indicate a partition coefficient for Au between magnetite and melt, \( D_{\text{Au}}^{\text{melt}} \approx 4 \). Using reasonable estimates of the mass fraction of magnetite that crystallizes in crustal magmatic systems, we modeled the proportion of Au sequestered by magnetite during fractional crystallization. We considered fractionation in two steps: the idealized derivation of a rhyolite by fractionation of basalt at depth, and the closed-system crystallization of the rhyolite in a magma chamber below the site of ore deposition. Magnetite sequesters 14–54% Au from basaltic parent melt that produces a rhyolite melt via crystal fractionation, at modal abundances of 1–5%. Less then 4% Au is sequestered from a crystallizing rhyolite melt because of low modal abundances of magnetite (<~2%). Our experimental and model results suggest that early crystallization of magnetite can play a mitigating role in the ability of a calc-alkaline magmatic system to yield a Au-rich ore fluid.

INTRODUCTION

Gold-rich, porphyry-type ore deposits are associated with both oxidized and reduced magmatic intrusions (Sillitoe 2000; Thompson and Newberry 2000; Rowins 2000). Oxidized ore deposits commonly contain primary magnetite, and form at oxygen fugacities between the nickel-nickel oxide (NNO) and hematite-magnetite (HM) oxygen fugacity buffers at the temperatures of mineralization (Sillitoe 2000). Reduced deposits contain primary Cu-Fe and Fe sulfides (e.g., chalcopyrite and pyrrhotite, respectively), and form at oxygen fugacities below the quartz-fayalite-magnetite (QFM) oxygen fugacity buffer at the temperatures of mineralization (Rowins 2000). It is generally accepted that Au in both types of porphyry deposits is genetically related, spatially and temporally, to the magmatic intrusions that are associated with the Au ore (Lindgren 1905; Emmons 1927; Burnham 1979; Tittley 1981; Sillitoe 1989; Richards et al. 1991; Hedenquist and Richards 1998). Gold mineralization forms via the separation of a magmatic volatile phase (MVP) from a magma, concomitant partitioning of Au from the melt into the MVP, egress and ascent of the MVP from the magma chamber, and precipitation of metals as physico-chemical changes in the MVP decrease the solubility of Au (Holland 1972; Burnham 1979; Candela and Holland 1984; Gammons and Williams-Jones 1997; Hedenquist and Richards 1998). The efficiency with which Au will partition into the MVP from a melt is a function of the concentration of Au-complexing ligands (e.g., Cl–, HS–) in the MVP (Candela and Piccoli 1995, 1998; Frank et al. 2002). A fundamental limitation on the mass transfer of Au from melt into an MVP is the proportion of Au remaining in the melt at the time of volatile saturation. Ultimately, the formation of Au-rich magmatic-hydrothermal ore lies in the extent to which a differentiating magmatic system retains Au in the melt phase while undergoing progressive compositional evolution. The probability of formation of an Au-rich porphyry deposit decreases if volatile exsolution occurs after a significant amount of Au has been sequestered by crystallizing phases (Candela 1989, 1992), unless these phases (e.g., magmatic sulfides) are sufficiently reactive to release the ore metals upon volatile saturation (Halter et al. 2002b). Variations in ligand concentration in the fluid also affect the efficiency of ore-metal transfer from the melt to the volatile phase. To model the behavior of Au in both oxidized and reduced magmatic environments, it is necessary to quantify the potentially mitigating effects of early crystallized magmatic phases on the Au availability in melts reaching the porphyry environment.

In S-rich, oxidized (i.e., ~NNO) magmatic systems, the crystallizing magmatic phases of interest include Fe oxides and Fe- and Cu-Fe sulfides. Experimental studies demonstrate that sulfide minerals play a significant role in Au behavior in S-rich melts (Cygan and Candela 1995; Jugo et al. 1999; Simon et al. 2000). In contrast, the ability of oxide phases to sequester Au in S-poor, oxidized (i.e., ~NNO) magmatic systems has remained largely unconstrained. Togashi and Terashima (1997) suggested that magnetite can sequester Au in oxidized mag-