



Thermometry of the magma ocean: Controls on the metal–silicate partitioning of gold

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Abstract

We have performed experiments to investigate the solubility and metal–silicate partitioning of gold as a function of metal sulphur content (X_S), silicate melt polymerization (NBO/T) and pressure (P). These experiments show that Au becomes less siderophile both with increasing pressure and as the metal phase becomes more sulphur-rich. For the studied range of compositions, melt polymerization has no effect on the solubility of Au. The reduction in the siderophile tendency of gold with increasing metal sulphur content is greater than expected on the basis of activity–composition relationships in the metal phase. This suggests a significant role for complexing between Au and S in the silicate melt. Our new experimental results are combined with literature data to yield a parameterisation for the exchange coefficient of Au ($Kd_{Au}^{Met/Sil}$) as a function of P , T and X_S :

$$\log Kd_{Au}^{Met/Sil} = -1.10(0.04) + 1.11(0.01) \frac{10^4}{T(\text{K})} - 67.8(10.9) \frac{P(\text{GPa})}{T(\text{K})} + 5.81(0.24) \log(1 - X_S)$$

Using this relationship, alongside similar parameterisations for Ni and Co selected from the literature, we performed continuous accretion models to delineate regions of parameter space in which Au and moderately siderophile element (MSE) abundances in the primitive upper mantle (PUM) could be reproduced. These models suggest that for metal–silicate equilibrium at very high pressures, Au will be overabundant in the PUM if equilibrium also occurs at extreme temperatures. Instead, most successful models are found when equilibrium occurs at high pressure but sub-liquidus temperatures. This result is in keeping with the physical conditions expected for a scenario where core-forming metal ponds and equilibrates at the rheological base of a magma ocean (e.g. Wade and Wood, 2005).

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1. INTRODUCTION

Elements with an extreme affinity for iron metal are termed the highly siderophile elements (HSE) and comprise

Au, Ir, Os, Pd, Pt, Re, Rh and Ru. During planetary differentiation, liquid iron alloy segregates from molten silicate to form a metallic core composed primarily of iron and nickel. As core segregation proceeds, experiments predict that HSEs will partition into the metal phase, leaving an HSE impoverished mantle with fractionated inter-element ratios (e.g. Borisov and Palme, 1996, 1997; Brenan and McDonough, 2009). Estimates of the primitive upper mantle (PUM) composition however, reveal only a modest depletion relative to

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chondrite (~ 2 orders of magnitude) and broadly chondritic element ratios. This discrepancy is termed ‘the highly siderophile element problem’ (Capobianco et al., 1993) and is often accounted for by the addition of chondritic material to the PUM subsequent to core-formation. This ‘late-veener’ of chondritic material does not undergo equilibration with the core, thus raising HSE abundances in the PUM and endowing it with chondritic element ratios.

Gold occupies a unique position among the HSEs by exhibiting several orders of magnitude greater solubility in silicate melt than is typical for the group (Brenan and McDonough, 2009; Bennett and Brenan, 2013). The partitioning of Au into the metal phase during planetary core-formation is therefore expected to be correspondingly weaker than for other HSEs, and chondrite-normalised concentrations of Au in the PUM following core-formation should be elevated relative to elements such as Ir, Os and Re. The distribution of trace elements between liquid metal and silicate can be described by the partition coefficient ($D_i^{Met/Sil}$):

$$D_i^{Met/Sil} = \frac{C_i^{Met}}{C_i^{Sil}} \quad (1)$$

where C_i is the concentration of element i in the super-script phase. Values of $D_{Au}^{Met/Sil}$ typically decrease with increasing T and are essentially independent of fO_2 at reducing conditions (Brenan and McDonough, 2009; Bennett and Brenan, 2013; Righter et al., 2015). This suggests that the Au content of PUM may provide a constraint on the thermal history of core–mantle equilibrium that is not complicated by the effects of progressive oxidation or reduction during accretion. This principle has been used previously to estimate a maximum temperature for metal–silicate equilibration during core segregation of ~ 3200 K, based on experiments performed at 2 GPa (Bennett and Brenan, 2013). To more confidently use Au partitioning as a thermometer for Earth’s primary differentiation, the effect of other parameters, such as silicate melt composition, metallic melt composition and pressure, must be better understood.

In this study, we have performed series of experiments to systematically determine changes in $D_{Au}^{Met/Sil}$ as a function of pressure, melt composition and the sulphur content of the metal phase. These new results are combined with literature data to parameterise $D_{Au}^{Met/Sil}$ as a function of P , T and metal sulphur content. Continuous core-formation models were performed using this new parameterisation for Au, alongside similar relationships for Ni and Co selected from the literature (Siebert et al., 2012). The results of these models suggest that Au partitioning is a useful constraint on core–mantle equilibration T . Experiments to investigate sulphur however, suggest further work is required to understand and parameterise the coupled effects of pressure and sulphur content of the system on the metal–silicate partitioning of Au.

2. THEORETICAL BACKGROUND

2.1. Calculation of metal–silicate partition coefficients

Metal–silicate partitioning was determined using two types of experiments: Those that contain Au only as a trace

constituent and those which are saturated in Au–metal. Two different approaches are therefore required in order to calculate values of $D_{Au}^{Met/Sil}$. Experiments to determine Au partitioning as a function of sulphur contents of the metal phase contain only minor amounts (<1 wt%) of Au and partition coefficients for a given composition may be calculated simply by means of Eq. (1). Experiments that explore the effect of silicate melt composition or pressure on Au solubility in silicate melt require a different approach. These experiments are saturated in an Au-rich metallic melt and partition coefficients are calculated from concentrations measured in the silicate phase at the solubility limit (C_{Au}^{SilSat}), by extrapolating to infinite dilution using the formulation of Borisov et al. (1994):

$$D_i^{Met/Sil} = \frac{1}{A \times C_i^{SilSat} \times \gamma_i^{Fe\infty}} \quad (2)$$

where A is a mole to weight conversion factor, and $\gamma_{Au}^{Fe\infty}$ is the activity coefficient of Au at infinite dilution in liquid iron metal. For a Fe–Au binary composition, A tends toward a value of 2.84×10^{-7} . A value of 12 was chosen for $\gamma_{Au}^{Fe\infty}$, following Brenan and McDonough (2009) who determined this parameter from solubility and metal–silicate partitioning experiments done at 2588 K. This is similar to the value of 6 calculated by Liu et al. (2009) for the Fe–Au system at 1623 K.

3. MATERIALS AND METHODS

3.1. Au partitioning in sulphur-bearing systems

The effect of adding sulphur to the metal–silicate system was investigated at 0.1 MPa, 1573–1753 K and $\sim IW - 1.4$ to -1.1 (where $IW \pm R$ is the deviation in log units from the fO_2 defined by the iron–wüstite buffer). The sulphur content of liquid Fe–S alloy co-existing with solid Fe decreases with increasing temperature (e.g. Waldner and Pelton, 2005). Experiments were therefore performed at different temperatures in order to generate a range of Fe–S alloy compositions. Temperatures were chosen which correspond to positions on the iron–sulphur binary liquidus where the system remains saturated in Fe–metal. Target compositions for Fe–S alloy were typically between $X_S = 0.1$ – 0.3 , as the shallow slope of the liquidus in this compositional range helps to minimize the T difference between experiments. Starting materials for sulphur-bearing experiments consisted of synthetic basalt, pure Fe and a Fe–S mixture, which were placed as powders into chromite crucibles in a gravitationally stable arrangement. Initially, Au was added to experiments as a pure metal bead, which comprised no more than ~ 1 wt% of the starting materials in order to ensure Henrian behaviour. In later experiments (AuFeS 9; AuFeNiS 1a, 1b), Au powder was added to the Fe powder portion of experiments to form a 100:1 mixture. The synthetic basalt was prepared by grinding pure oxide and carbonate powders under ethanol in an agate mortar and pestle. The ground mixture was dried at room temperature and calcined overnight at 1373 K before being re-ground then equilibrated at 1673 K in a controlled

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