



# Is gold solubility subject to pressure variations in ascending arc magmas?

Sébastien Jégo<sup>a,b,\*</sup>, Michihiko Nakamura<sup>c</sup>, Jun-Ichi Kimura<sup>d</sup>, Yoshiyuki Iizuka<sup>b</sup>,  
Qing Chang<sup>d</sup>, Georg F. Zellmer<sup>e</sup>

<sup>a</sup> Institut des Sciences de la Terre d'Orléans (ISTO), Université d'Orléans, CNRS: UMR7327, INSU, Bureau de Recherches Géologiques et Minières (BRGM), France

<sup>b</sup> Institute of Earth Sciences, Academia Sinica, 128 Academia Road, Sec. 2, Nankang, Taipei 11529, Taiwan, ROC

<sup>c</sup> Department of Earth Science, Tohoku University, 6-3 Aramaki-Aza-Aoba, Aobaku, Sendai 980-8578, Japan

<sup>d</sup> Institute for Research on Earth Evolution (IFREE), Japanese Agency for Marine-Earth Science and Technology (JAMSTEC), 15-2 Natsushima-cho, Yokosuka 237-0061, Japan

<sup>e</sup> Soil and Earth Sciences Group, Massey University, Palmerston North, New Zealand

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## Abstract

Magmas play a key role in the genesis of epithermal and porphyry ore deposits, notably by providing the bulk of ore metals to the hydrothermal fluid phase. It has been long shown that the formation of major deposits requires a multi-stage process, including the concentration of metals in silicate melts at depth and their transfer into the exsolved ore fluid in more superficial environments. Both aspects have been intensively studied for most of noble metals in subsurface conditions, whereas the effect of pressure on the concentration (i.e., solubility) of those metals in magmas ascending from the sublithospheric mantle to the shallow arc crust has been quite neglected. Here, we present new experimental data aiming to constrain the processes of gold (Au) dissolution in subduction-linked magmas along a range of depth. We have conducted hydrous melting experiments on two dacitic/adakitic magmas at 0.9 and 1.4 GPa and  $\sim 1000$  °C in an end-loaded piston cylinder apparatus, under  $fO_2$  conditions close to NNO as measured by solid Co–Pd–O sensors. Experimental charges were carried out in pure Au containers, the latter serving as the source of gold, in presence of variable amounts of H<sub>2</sub>O and, for half of the charges, with elemental sulfur (S) so as to reach sulfide saturation. Au concentrations in melt quenched to glass were determined by LA-ICPMS. When compared to previous data obtained at lower pressures and variable redox conditions, our results show that in both S-free and sulfide-saturated systems pressure has no direct, detectable effect on melt Au solubility. Nevertheless, pressure has a strong, negative effect on sulfur solubility. Since gold dissolution is closely related to the behavior of sulfur in reducing and moderately oxidizing conditions, pressure has therefore a significant but indirect effect on Au solubility. The present study confirms that Au dissolution is mainly controlled by  $fO_2$  in S-free melts and by a complex interplay of  $fO_2$  and melt S<sup>2-</sup> concentration in sulfide-saturated melts, at given temperature. In addition, we propose that the transition from sulfide (S<sup>2-</sup>) to sulfate (S<sup>6+</sup>) species in melt is shifted towards more oxidizing conditions when pressure and the degree of melt polymerization increase. If this is true, this may have important consequences during mantle melting and magma ascent. In particular, if mantle melting occurred in moderately oxidizing conditions, a small degree of partial melting would allow the primary melts to become Au-enriched but the relatively high pressure would move the sulfide-sulfate transition to more oxidizing conditions, making the primary melts saturated with sulfide phases that would sequester gold from the melt. During magma ascent, the

\* Corresponding author at: Institut des Sciences de la Terre d'Orléans (ISTO), Université d'Orléans, CNRS: UMR7327, INSU, Bureau de Recherches Géologiques et Minières (BRGM), France. Tel.: +33 2 38 25 53 99.

E-mail address: [Sebastien.Jego@univ-orleans.fr](mailto:Sebastien.Jego@univ-orleans.fr) (S. Jégo).

decreasing pressure would favor the destabilization of sulfides and the release of gold to the silicate melt. However, at shallow levels, decreasing pressure, magma evolution, and varying redox conditions would be continuously competing to concentrate or fractionate gold.

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## 1. INTRODUCTION

Au–Cu–Mo deposits represent major metal resources known to be spatially and temporally associated with intrusive, subduction-linked calc-alkaline magmatism and, according to some studies, notably with particular arc magmas whose petrogenesis may involve garnet fractionation (i.e., adakitic melts), either by slab partial melting in the upper mantle or by deep crustal melting/fractionation (e.g., Jégo et al., 2005 and references therein; Zellmer et al., 2012; see also Richards and Kerrich, 2007 for a synthesis). Although the bulk of the ore metals seems to originate from the magmas, there is as yet no consensus on processes responsible for their concentration. However, the fact that primary ore minerals are predominantly sulfides (e.g., Kesler et al., 2002) has led to the suggestion that sulfur may play an important role in metal enrichment processes at the magmatic stage (Sillitoe, 1997).

Most of previous studies focusing on the behavior of gold in silicate melts aimed to determine the partitioning of gold between silicate liquid and sulfide phases (e.g., Bezmen et al., 1994; Fleet et al., 1996, 1999; Crockett et al., 1997; Jugo et al., 1999; Simon et al., 2007, 2008; Bell et al., 2009). Even though these experimental studies quantify the solubility of gold in the melt, the reported melt gold concentrations depend on the starting composition of the metal-doped sulfides. Nevertheless, Borisov and Palme (1996) report Au solubility values from ~300 to 2000 ppb in anhydrous haplobasaltic S-free melt at 1 atm and 1300–1400 °C, and suggest that gold dissolution is dependent on the oxidation state by involving  $Au_2O$  species. Jégo et al. (2010) report much lower solubility values (30–240 ppb Au) in S-free dacitic compositions at 4 kbar and 1000 °C, but also show an increase of melt Au contents with  $fO_2$  in a way consistent with the dissolution of gold as both  $Au^{1+}$  and  $Au^{3+}$  species. In presence of sulfur, Simon et al. (2007) report gold solubility values up to 1100 ppb in Cl-bearing haplogranitic melt at ~NNO (800 °C, 1–1.5 kbar) and Jugo et al. (2005) give evidence for a solubility on the order of ~500 ppb in anhydrous basaltic melt (1300 °C, 10 kbar) in reducing conditions, while a solubility value of ~800 ppb is reported by Bezmen et al. (1994) in the Bushveld Complex melt (1300 °C, 4 kbar, ~NNO). Those three solubility values are noticeably lower than those obtained by Jégo and Pichavant (2012) in dacitic compositions (1000 °C, 4 kbar), which range from 1200 to 4250 ppb Au at ~NNO-1 and from 865 to 2400 ppb at ~NNO+1.5, depending mainly on the melt S content. Jugo et al. (1999) also report high gold solubility values, i.e., ~4 ± 2 ppm in hydrous haplogranitic melt (850 °C, 1 kbar) in

moderately oxidizing conditions (~NNO ± 0.5). Recently, Botcharnikov et al. (2010) presented the first experimental Au solubility data in S-bearing hydrous intermediate (i.e., andesitic) silicate melts around NNO. Similarly to Jégo and Pichavant (2012), the data reported in Botcharnikov et al. (2010) suggest a positive correlation between the concentrations of Au and S dissolved in the melt under reducing/moderately oxidizing conditions. However, Botcharnikov et al. (2010) show significantly lower Au solubility values (from 300 to 2500 ppb) compared to Jégo and Pichavant (2012), and Zajacz et al. (2012, 2013) also report similarly lower Au concentrations (from 220 to 1550 ppb Au and from ~60 to 3200 ppb Au, respectively) in reducing to moderately oxidizing conditions at 1000 °C and 2 kbar. Nevertheless, another study by Botcharnikov et al. (2011) reports much higher gold concentrations (from 250 to 8000 ppb Au) in basaltic to andesitic melts over an  $fO_2$  range going from NNO – 1 to ~NNO+2, which emphasize the role of  $fO_2$  in controlling the incorporation of gold in melt. Under more oxidizing conditions ( $fO_2 \geq NNO+3$ ), Jégo and Pichavant (2012) show that gold dissolution is much less effective – with solubility values around 600 ppb Au – and seems independent on the melt S content, yet very high. Thus, the respective effects of the oxidation state and the presence of sulfur on gold solubility are partly understood so far, contrarily to the influence of pressure and temperature.

Although no previous study has focused on its pressure dependence, the solubility of gold in silicate melts may vary with depth in a significant way, independently of any fractionation process. If this is true, it may lead to important implications for the gold budget of arc magmas rising up through the mantle wedge and the arc crust. A few other ore-forming metals (Mo, Pt, Te, Se, ...) as well as sulfur have been previously studied to estimate the pressure effect on their solubility in silicate melts (e.g., Mavrogenes and O'Neill, 1999; Ertel et al., 2006; Rose-Weston et al., 2009; Burkemper and Agee, 2010), but all experiments were performed at much higher pressures, some of these studies aiming to constrain early Earth evolution and core-mantle differentiation. Here, we aim to evaluate the ability of the silicate melt to incorporate gold during partial melting of a deep source (upper mantle or lower crust), saturated with sulfide phases or not, and see how the initial gold budget of a primary melt will evolve during its adiabatic ascent through the arc crust. Such information is indeed decisive to understand if the pre-concentration of Au at the magmatic stage is pressure-dependent and, therefore, to determine to which extent magmas are able to participate in the formation of ore deposits.

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