

Fluid-melt partitioning of sulfur in differentiated arc magmas and the sulfur yield of explosive volcanic eruptions

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Abstract

The fluid-melt partitioning of sulfur ($D_S^{\text{fluid/melt}}$) in differentiated arc magmas has been experimentally investigated under oxidizing conditions (Re–ReO₂ buffer) from 800 to 950 °C at 200 MPa. The starting glasses ranged in composition from trachyte to rhyolite and were synthesized targeting the composition of the residual melt formed after 10–60% crystallization of originally trachy-andesitic, dacitic and rhyodacitic magmas (Masotta and Keppler, 2015). Fluid compositions were determined both by mass balance and by Raman spectroscopy of fluid inclusions. $D_S^{\text{fluid/melt}}$ increases exponentially with increasing melt differentiation, ranging from 2 to 15 in the trachytic melt, from 20 to 100 in the dacitic and rhyodacitic melts and from 100 to 120 in the rhyolitic melt. The variation of the $D_S^{\text{fluid/melt}}$ is entirely controlled by the compositional variation of the silicate melt, with temperature having at most a minor effect within the range investigated. Experiments from this study were used together with data from the literature to calibrate the following model that allows predicting $D_S^{\text{fluid/melt}}$ for oxidized arc magmas:

$$\ln\left(D_S^{\text{fluid/melt}}\right) = 9.2 - 31.4 \cdot \frac{nbo}{t} - 1.8 \cdot \text{ASI} - 29.5 \cdot \text{Al\#} + 4.2 \cdot \text{Ca\#}$$

where $\frac{nbo}{t}$ is the non-bridging oxygen atoms per tetrahedron, ASI is the alumina saturation index, Al# and Ca# are two empirical compositional parameters calculated in molar units $\left(\text{Al\#} = \frac{X_{\text{Al}_2\text{O}_3}}{X_{\text{SiO}_2} + X_{\text{TiO}_2} + X_{\text{Al}_2\text{O}_3}} \text{ and } \text{Ca\#} = \frac{X_{\text{CaO}}}{X_{\text{Na}_2\text{O}} + X_{\text{K}_2\text{O}}}\right)$.

The interplay between fluid-melt partitioning and anhydrite solubility determines the sulfur distribution among anhydrite, melt and fluid. At increasing melt polymerization, the exponential increase of the partition coefficient and the decrease of anhydrite solubility favor the accumulation of sulfur either in the fluid phase or as anhydrite. On the other hand, the higher anhydrite solubility and lower partition coefficient for less polymerized melts favor the retention of sulfur in the melt. At equilibrium conditions, these effects yield a maximum of the sulfur fraction in the fluid phase for slightly depolymerized melts ($\frac{nbo}{t} = 0.05\text{--}0.15$). Our data allow quantitative predictions of the sulfur yield of explosive volcanic eruptions over a wide range of magma compositions.

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1. INTRODUCTION

Sulfur compounds (H₂S and SO₂) constitute a major fraction of volcanic gas emissions, with abundance next

only to H₂O and CO₂ (Symonds et al., 1994). The average annual sulfur degassing from volcanoes may add up to 35% of the total atmospheric SO₂ and sulfate burden, including anthropogenic and ocean sources (Graf et al., 1997). Explosive eruptions can inject large amounts of sulfur into the stratosphere in a single event, inducing atmospheric perturbations that may eventually result in changes of the Earth's

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average temperature. Once injected into the stratosphere, SO₂ is photochemically oxidized to sulfate aerosols that increase the Earth's albedo by backscattering the incoming solar radiation, while at the same time, they warm the stratosphere by absorbing the upwelling infrared radiation (McCormick et al., 1995). Depending on which of these effects prevails, which in turn depend on the size of the sulfate aerosol particles, the consequences can be either the cooling or warming of the Earth's surface (Lacis et al., 1992). The 1991 eruption of Mt. Pinatubo produced one of the largest climate perturbations of the 20th century, by injecting around 17–20 Tg of SO₂ into the stratosphere that were responsible for a global cooling of 0.5 °C (McCormick et al., 1995; Robock, 2002). The amount of sulfur erupted by Mt. Pinatubo has been estimated to be ~23 larger than the amount that could have been dissolved in the volume of erupted melt at the pre-eruptive T, P, and *f*O₂ (Gerlach et al., 1996). As for Mt. Pinatubo, many other recent eruptions emitted larger amounts of sulfur than the amount expected from the degassing of the erupted silicate melt (Wallace, 2001 and references therein). The mismatch between the amount of sulfur predicted for an eruption (based on a petrologic estimate of the sulfur content in the erupted melt) and the actual mass discharged is usually called the “sulfur excess” (Andres et al., 1991) and it has been explained by the accumulation of a sulfur-rich fluid phase in the magma chamber before the eruptions (Keppler, 1999).

In addition to the fluid phase, sulfur can be stored in a magma either as a dissolved species in the silicate melt or in accessory minerals, such as pyrrhotite (Fe_(1-x)S) and anhydrite (CaSO₄). The sulfur contained in these minerals is likely not released into the fluid during explosive eruptions, because their decomposition rate is slow relative to the eruptive timescale (Hanic et al., 1985). However, the solubility of minerals such as anhydrite controls the concentration of sulfur in the melt and, indirectly, the amount of sulfur that partitions in the fluid. An accurate determination of the fluid-melt partition coefficient of sulfur is essential to determine the sulfur distribution among the magmatic phases (melt, fluid and minerals) and, ultimately, the sulfur budget of volcanic eruptions.

There is a broad consensus among available experimental studies on the striking effect of oxygen fugacity on the fluid-melt partition coefficient, which decreases by about an order of magnitude with increasing *f*O₂ (e.g., Keppler, 1999; Webster and Botcharnikov, 2011 and references therein; Zajacz et al., 2012; Jago and Dasgupta, 2014). Sulfur partition coefficients for most of arc magmas at oxidizing conditions are however limited to a few compositions and large uncertainties sometimes exist because of various experimental difficulties. Some recent experimental studies provided insightful data for the partitioning of sulfur between fluid and melt in basaltic (Lesne et al., 2011), andesitic (Zajacz et al., 2012; Fiege et al., 2014), haplogranitic (Keppler, 2010; Huang and Keppler, 2015) and other simplified aluminosilicate melts (Zajacz, 2015). These studies investigated the behavior of sulfur either alone, or in presence of other volatiles, at either isobaric conditions or dur-

ing decompression, and altogether they suggest that the composition of the silicate melt has a key role on sulfur partitioning between fluid and melt. The full range of melt composition and temperature relevant to differentiated arc magmas remains yet uncovered. Since these magmas are the main source of volcanogenic sulfur in the stratosphere (e.g., Oppenheimer et al., 2011), reliable data on fluid-melt sulfur partitioning are required in order to predict the environmental impact of explosive eruptions.

In this study, we experimentally determined the fluid-melt partition coefficients of sulfur in differentiated arc magma compositions at oxidizing conditions and for a range of relevant temperatures. By combining our data with experimental data from the literature fulfilling certain criteria (i.e., fluid/melt ratio ~1, multiple experiments with same melt composition that obey Henry's law), we calibrated a general model to predict the sulfur partitioning between fluid and melt. This model, taken together with the anhydrite solubility model presented in Masotta and Keppler (2015), allows calculating the budget of sulfur for most of differentiated arc magmas and, ultimately, the atmospheric sulfur yield of volcanic eruptions.

2. EXPERIMENTAL METHODS

2.1. High pressure experiments

Seven glassy starting materials were synthesized using as target the composition of the interstitial melt of anhydrite solubility experiments selected from those reported in Masotta and Keppler (2015). Each glass was prepared from a stoichiometric mixture (about 10 g) of analytical grade SiO₂, TiO₂, Al(OH)₃, Fe₂O₃, Mg(OH)₂, CaCO₃, Na₂CO₃, and K₂CO₃. Each mixture was first dehydrated and decarbonated by slowly heating to 1100 °C in 12 h and holding at the same temperature for a further 12 h, then melted for 2 h at 1600 °C in a iron-saturated platinum crucible, and finally quenched in distilled water. The resulting glasses were free of crystals and very similar in composition to the experimental glasses selected as target (Table 1).

Each glass was crushed into powder and loaded into 20 mm long gold capsules (2.5 mm outer diameter, 2.1 mm inner diameter) with an equal amount (~10 mg) of H₂SO₄ solution at concentration ranging from 0.05 to 4 M (a total of 8 capsules for each starting composition; Table 2). Capsules were then sealed by arc welding and checked for weight loss after heating in a drying furnace for few hours at 130 °C. The high fluid/melt ratio (1:1 by weight) was chosen in order to allow a more accurate determination of the amount of sulfur in the fluid (calculated by mass balance) and, by consequence, to reduce the error in determining the fluid/melt partition coefficients (see Zajacz et al., 2012). In each experiment, two capsules were loaded into a 30 mm long outer capsule (5 mm outer diameter, 4.6 mm inner diameter) together with 40 mg of water and 400 mg of Re–ReO₂ buffer (1:1 mixture). The high permeability of gold to hydrogen at high temperatures allows the equilibration with the external buffer within few hours or days (Chou, 1986; Keppler, 2010). At the same time,

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