



# Discriminating secondary from magmatic water in rhyolitic matrix-glass of volcanic pyroclasts using thermogravimetric analysis

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## Abstract

Pyroclasts from explosive eruptions, such as the 1060 CE explosive Glass Mountain eruption of Medicine Lake volcano, California, contain large amounts of water. This may be the consequence of diffusive rehydration of the volcanic glass by meteoric (secondary) water after the eruption. Discriminating between magmatic and secondary water in the matrix glass of pyroclasts is important, because the degassing of magmatic water affects the intensity of volcanic eruptions. Such discrimination has remained a challenging problem, especially because some aspects of water diffusion in silicate glasses at low temperatures and atmospheric pressure remain poorly constrained. We used thermogravimetry to analyze the loss of water from natural volcanic glasses and glasses that were hydrated in the laboratory at magmatic temperatures and pressures. Numerical modeling of diffusive water loss during thermogravimetric analyses accounted for the interconversion of molecular water ( $\text{H}_2\text{O}_m$ ) and hydroxyls groups (OH), and indicates that Glass Mountain pumices contain 0.2–0.5 wt% primary water, but gained 1–2 wt% of meteoric water by diffusive rehydration during the past 950 years. These results confirm that the majority of magmatic water is lost from the magma during explosive eruptions. Furthermore, the integration of thermogravimetric analysis and numerical modeling facilitates discrimination between the magmatic and secondary water content of volcanic glasses.

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## 1. INTRODUCTION

The style and intensity of volcanic eruptions are largely controlled by the degassing of magmatic volatiles during magma ascent to the surface (e.g., Sparks, 1978, 2003; Eichelberger, 1995; Gonnermann and Manga, 2007). Among the different volatiles produced during volcanic

eruptions, water is in many cases the most abundant. It is also the most important volatile species for the eruption of intermediate to felsic magmas, because of its influence on magma rheology and, thereby, on the dynamics of eruptive magma ascent (e.g., Shaw, 1972; Dingwell et al., 1996; Richet et al., 1996; Giordano and Dingwell, 2003).

Typically, the amount of water dissolved in an aliquot of melt, some of which may be preserved as dissolved water within the volcanic glass upon eruption and quenching, continuously decreases as the eruption proceeds. During ascent, the ambient pressure exerted on the magma

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decreases and, because of pressure-dependent water solubility, the melt becomes supersaturated in dissolved water, resulting in the nucleation and growth of bubbles of a supercritical water-bearing fluid phase (e.g., Hurwitz and Navon, 1994; Thomas et al., 1994; Lyakhovsky et al., 1996; Proussevitch et al., 1998; Gardner et al., 1999, 2000; Cashman, 2004; Gonnermann and Manga, 2007). Consequently, during eruptive magma ascent the concentration of dissolved water decreases and in principle the erupting magma may be almost ‘dry’ when it reaches the Earth’s surface.

Recent work by Gonnermann and Houghton (2012) suggested that this may not necessarily be the case. Detailed modeling of magma degassing during Episode III of the 1912 Plinian eruption of Novarupta volcano, Alaska, indicated the possibility that the magma could have retained a significant fraction of its pre-eruptive water as dissolved water within the melt, due to disequilibrium degassing. Instead, the magma may have lost a large fraction of its dissolved water by open-system degassing shortly before or after fragmentation (e.g., Namiki and Manga, 2008), or the melt may have retained several weight percent of dissolved water upon quenching. Total water contents in the matrix glass of pyroclasts from the 1912 Novarupta eruption are approximately 2–3 wt% (Giachetti and Gonnermann, 2013), perhaps consistent with incomplete degassing of the erupting magma, but contrary to the magma losing most of its water upon ascent to the surface (e.g., Jaupart and Allègre, 1991; Gardner et al., 1996).

Alternate explanations for the surprisingly high water content of pyroclasts from the 1912 Novarupta eruption, as well as in pyroclasts from other explosive eruptions of silica-rich magmas (Giachetti and Gonnermann, 2013), could be bubble resorption (e.g., Westrich and Eichelberger, 1994; Yoshimura and Nakamura, 2008, 2010; Watkins et al., 2012; McIntosh et al., 2014), interaction of the ascending magma with hydrothermal fluids or ice (e.g., Burnham, 1979; Villemant and Boudon, 1999; Tuffen et al., 2010), or the gradual diffusion of meteoric water into the volcanic glass after eruption and deposition (e.g., Ross and Smith, 1955; Friedman et al., 1966; Friedman and Long, 1976; Newman et al., 1986; Denton et al., 2009, 2012; Giachetti and Gonnermann, 2013). The latter is referred to as ‘rehydration’. Upon rehydration, the fraction of dissolved water that does not represent pre-eruptive magmatic (‘primary’) water is often referred to as ‘secondary’ water. Obviously, the ability to discriminate between magmatic and secondary water is of critical importance to understanding the dynamics that govern explosive volcanic eruptions. For example, quantifying the relative proportion of magmatic and secondary water would help to further constrain the relative degrees of pre- vs. post-fragmentation magma open-system degassing and, hence, the conditions at magma fragmentation.

The discrimination between magmatic and secondary water in the matrix glass of pyroclasts is the objective of our study. Although several different methods to distinguish primary from magmatic water exist (e.g., Tuffen et al., 2010), each has significant limitations. For

example, the D/H ratio of meteoric water is distinct from magmatic water (e.g., DeGroot-Nelson et al., 2001; Harford et al., 2003; Tuffen et al., 2010). However, Nolan and Bindeman (2013) showed that the D/H ratio can be rapidly changed by minor diagenesis, even at relatively low temperatures of 20 °C (and despite no significant water gain after 2 years at 70 °C was observed), making its interpretation complicated. Among the different methods available to distinguish magmatic from secondary water, oxygen isotopes may be the most reliable technique (Goff and McMurtry, 2000; Tuffen et al., 2010; Nolan and Bindeman, 2013). Here we focus on alternate method: Thermogravimetric analysis (TGA). During TGA a sample is heated (in this case at atmospheric pressure) to a specified temperature and at a specified rate, while its change in mass is continuously recorded (e.g., Eichelberger and Westrich, 1981; Roulia et al., 2006; Anovitz et al., 2008; Denton et al., 2009, 2012; Stevenson et al., 2009; Applegarth et al., 2013). Because the solubility of water in silicate glasses at atmospheric pressure decreases during heating, water diffuses out of the sample during TGA. The discrimination between magmatic and secondary water is based on the hypothesis that secondary water is more ‘weakly bound’ within the silica network than magmatic water, and that it is therefore lost from the sample at lower temperatures (Newman et al., 1986; Westrich, 1987; Roulia et al., 2006; Denton et al., 2009, 2012). We performed a series of TGA experiments of rhyolitic glasses, which were hydrated at known and controlled conditions in the laboratory (magmatic pressures and temperatures) and, thus, of known magmatic water content. Subsequent modeling of water loss during TGA allowed us to establish a framework for quantifying magmatic water content and its application to pumices from explosive volcanic eruptions.

## 2. WATER IN RHYOLITIC MELT AND GLASS

In this section we summarize the key points on speciation, solubility and diffusivity of water in rhyolitic melts and glasses. We refer to the reviews by Zhang et al. (2007) and Zhang and Ni (2010) for further information.

### 2.1. Speciation

Water dissolves in silicate melts as molecular water ( $\text{H}_2\text{O}_m$ ) and hydroxyls (OH). The total amount of dissolved water is denoted as  $\text{H}_2\text{O}_t$  and herein sometimes referred to as water. Both  $\text{H}_2\text{O}_m$  and OH can be quantified using infrared and Raman spectroscopy (e.g., Stolper, 1982b; Ihinger et al., 1994; Chermiak et al., 2010; Ni et al., 2013).  $\text{H}_2\text{O}_m$  and OH can interconvert through the reaction (e.g., Stolper, 1982a,b)



where O is an anhydrous oxygen ion and the ionic charge is not shown. The equilibrium constant of Eq. (1) can be written as

$$K = \frac{[\text{OH}]^2}{[\text{H}_2\text{O}_m][\text{O}]}, \quad (2)$$

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