



The effect of oxygen fugacity on the rheological evolution of crystallizing basaltic melts

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ABSTRACT

Storage and transport of silicate melts in the Earth's crust and their emplacement on the planet's surface occur almost exclusively at sub-liquidus temperatures. At these conditions, the melts undergo crystallization under a wide range of cooling-rates, deformation-rates, and oxygen fugacities (f_{O_2}). Oxygen fugacity is known to influence the thermodynamics and kinetics of crystallization in magmas and lavas. Yet, its influence on sub-liquidus rheology remains largely uncharted.

We present the first rheological characterization of crystallizing lavas along natural cooling paths and deformation-rates and at varying f_{O_2} . Specifically, we report on apparent viscosity measurements for two crystallizing magmatic suspensions 1) at $\log f_{O_2}$ of -9.15 (quartz–fayalite–magnetite buffer, QFM, -2.1) and 2) in air. These fugacities span a range of reduced to oxidized conditions pertinent to magma migration and lava emplacement. We find that: 1) crystallization at constant cooling-rates results in a quasi-exponential increase in the apparent viscosity of the magmatic suspensions until they achieve their rheological cut off temperature (T_{cutoff}), where the melt effectively solidifies 2) the rheological departure and T_{cutoff} increase with increasing f_{O_2} and 3) increasing f_{O_2} results in decreased crystallization-rates. Based on the experimental results and by comparison with previous rheological isothermal studies we propose a generalisation of the effect of f_{O_2} on the dynamic rheological evolution of natural magmatic and volcanic suspensions. We further discuss the implications for magmatic transport in plumbing and storage systems (e.g. conduits, dikes and magma chambers) and during lava flow emplacement.

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1. Introduction

1.1. Motivation and scope of this study

Basaltic compositions represent the most abundant volcanism on Earth and other terrestrial bodies such as the Moon and Mars (Chevrel et al., 2013a; Sigurdsson et al., 2015). Their eruptions can drastically alter global climate and lead to regional disruption of agriculture and aviation (Sigurdsson et al., 2015). Understanding their flow behaviour and emplacement mechanisms informs the prediction of volcanic hazards and risks. It also informs the deduction of lava compositions and associated environmental parameters of planetary volcanism.

Magma is forced towards the surface by buoyancy forces resulting from the density contrast with the host rock (Sigurdsson et al., 2015). Its migration in the crust and emplacement on the

surface are governed by its effective viscosity (Chevrel et al., 2015; Kolzenburg et al., 2017; La Spina et al., 2016; Nicolas and Ildefonse, 1996; Pinkerton and Sparks, 1978; Sato, 2005; Vona and Romano, 2013), the flow/effusion-rate (Harris and Rowland, 2009; Nicolas and Ildefonse, 1996), the plumbing system geometry (La Spina et al., 2015) and the underlying topography (Cashman et al., 2013; Kolzenburg et al., 2016a). Forecasting the flow behaviour of magmas and lavas, to date, is hindered most severely by an incomplete understanding of how lava-rheology evolves during emplacement and when flow ceases (Chevrel et al., 2013b; Giordano et al., 2007; Kolzenburg et al., 2017).

Predicting the flow dynamics of magmas and lavas relies thus on a detailed understanding of their rheology, which evolves during migration, eruption and emplacement. This evolution in rheology is caused by changes in phase state, melt composition, texture and temperature of the magma resulting from gas loss, cooling and crystallization. This creates strongly heterogeneous flow conditions, textures and morphologies (Arzilli and Carroll, 2013; La Spina et al., 2016; Nicolas and Ildefonse, 1996; Shaw et al., 1968; Sigurdsson et al., 2015; Vetere et al., 2017) that evolve in space

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and time. Understanding the rheological evolution of crystallizing magmas/lavas requires direct measurement of the flow properties at emplacement conditions. Few direct measurements of lava rheology under natural conditions are reported (Pinkerton and Sparks, 1978; Shaw et al., 1968). These measurements are crucial for benchmarking of experimental data, but insufficient to develop a systematic understanding of the evolution of lava-flow properties in response to varying external and internal parameters (composition, cooling- and shear-rate, oxygen fugacity etc.), as they represent snapshots of the system at one specific condition.

Magmas are generated in the Earth's interior at low oxygen fugacities (fO_2) and then transported to and erupted on the Earth's surface where they encounter increasingly oxidizing conditions. The effect of oxygen fugacity on the transport properties of natural silicate melts at super-liquidus temperatures has been investigated for a range of compositions (Chevrel et al., 2013a; Dingwell, 1989; Dingwell and Virgo, 1987; Mysen et al., 1984; Sato, 2005).

However, migration and transport of magma generally occurs in settings where the melts crystallize. Oxygen fugacity affects the stability of Fe-bearing phases, the crystallization- and degassing-onset, -path and -kinetics and the glass transition (T_g) of magmas under both static (i.e. constant T and P) and dynamic (decreasing P and T) conditions (Arzilli and Carroll, 2013; Bouhifd et al., 2004; Hamilton et al., 1964; La Spina et al., 2016; Markl et al., 2010; Sato, 1978; Toplis and Carroll, 1995).

Crystallization, in turn, increases the effective magma viscosity and therewith influences its migration and the eruptive style of volcanoes. This sparked a series of sub-liquidus rheology experiments on a variety of compositions, dominantly basaltic and andesitic. This is because they provide favourable crystallization kinetics, resulting in a manageable time frame of experimentation (Bouhifd et al., 2004; Chevrel et al., 2015; Sato, 2005; Vetere et al., 2017; Vona and Romano, 2013; Vona et al., 2011; Wilke, 2005). Most existing rheological data stem from measurements at constant temperature and atmospheric conditions (i.e. equilibrated in air) that may not necessarily be representative of the dynamic emplacement situations in nature. Evaluating the influence of the evolving flow properties of lavas on their emplacement dynamics requires characterization of the rheological properties of lavas at non-isothermal and non-equilibrium conditions. In such conditions, the lava undergoes transient increases in viscosity, reaching increasing degrees of undercooling until a "rheological cut off temperature" (T_{cutoff}) (Giordano et al., 2007; Kolzenburg et al., 2016b, 2017) is reached, where the effective viscosity rises steeply, terminating its capacity to flow. This transient rheological gradient governs the lavas' emplacement. Studies on the effect of oxygen fugacity on the sub-liquidus rheological evolution of magmas/lavas are scarce (Bouhifd et al., 2004; Sato, 2005) and few measurements of the cooling-rate dependent, disequilibrium rheology of natural melts exist to date (Giordano et al., 2007; Kolzenburg et al., 2016b, 2017). In fact, to our knowledge no study on the effect of oxygen fugacity on the disequilibrium rheology of natural melts under either static or dynamic thermal conditions has been presented.

We present the first dataset on the sub-liquidus rheological evolution of Etna and Holuhraun melts during crystallization at reduced conditions and in air. Reduced experiments are performed at low $\log fO_2$ with respect to magmatic conditions to assess the maximum possible effect that changes in fO_2 may have on the systems' crystallization dynamics and rheological evolution.

1.2. The role of iron in melt-structure and eruptive dynamics

The role of iron in silicate melt structure and viscosity has been extensively investigated via experiments (Di Genova et al.,

2017b; Dingwell and Virgo, 1987; Giordano et al., 2015; Mysen et al., 1984; Poe et al., 2012; Toplis and Carroll, 1995). Viscosity models, such as (Giordano et al., 2008b; Russell et al., 2003), however, generally do not include this effect. This is largely because most studies do not report iron speciation for the investigated melts and the understanding of the structural role of iron in melts, especially during migration and eruption remains incomplete (Cicconi et al., 2015; Giordano et al., 2015; Poe et al., 2012; Wilke, 2005). Based on literature results, it is generally accepted that the iron coexists in both reduced (FeO) and oxidized (Fe_2O_3) states with coordination environments ranging from tetrahedral to pentahedral and octahedral depending on the melt composition, temperature and oxygen fugacity (Mysen et al., 1984; Wilke, 2005). Because of this unique behaviour, several studies show that increasing Fe_2O_3 content, during magma ascent and oxidation, results in increasing melt viscosity (Bouhifd et al., 2004; Di Genova et al., 2017b). However, the structural role (i.e. network former vs network modifier) of iron is still highly debated (Alderman et al., 2017; Poe et al., 2012) and *in-situ* studies such as Cicconi et al. (2015) are required to better constrain the iron environment in melts, especially in dynamic temperature and fO_2 conditions.

Iron oxidation state also shifts phase equilibria (Hamilton et al., 1964; Markl et al., 2010), influencing the magmas' crystallization path (Toplis and Carroll, 1995) and rheological evolution (Bouhifd et al., 2004; Sato, 2005). Moreover, it has been demonstrated that changing the dissolved iron content in melts is responsible for iron nanolite formation which, in turn, affects the viscosity and may facilitate the exsolution of volatiles (Di Genova et al., 2017a).

2. Physical constraints on the ascent and emplacement conditions of basaltic magma

Common oxygen fugacities for basaltic lava flows and shallow magmatic systems at eruptive temperatures lie in the range of $\log fO_2 = -8$ to -11 (nickel-nickel oxide buffer (NNO) to QFM -1) (Hamilton et al., 1964; Markl et al., 2010; Mollo et al., 2015; Sato, 1978).

Once flowing towards the surface, lavas are exposed to increasingly oxidizing conditions and fO_2 starts to equilibrate towards atmospheric values. Redox equilibration of silicate melts in experiments occurs on the timescale of hours to days (Dingwell and Virgo, 1987; Mysen et al., 1985). In most lavas, especially ones that crystallize rapidly, redox equilibrium with the atmosphere is unlikely to be achieved. Experiments performed under equilibrated atmospheric conditions, therefore, represent an end-member case at the most oxidized conditions to be expected in nature.

Coherent basaltic (low viscosity) magmas rising in a conduit or in narrow and shallow dikes commonly undergo cooling-rates from few degrees per hour to $\sim 10^\circ C min^{-1}$ (Giordano et al., 2007; La Spina et al., 2015, 2016), due to thermal exchange with the conduit wall and gas expansion and escape. These cooling-rates represent steady flow conditions after an initial rapid chilling event of the first batch of magma of up to $50^\circ C min^{-1}$ (Giordano et al., 2007). However, in extreme events, such as basaltic plinian eruptions (Schauroth et al., 2016), high flux-rates may result in heating of the conduit walls. The data presented here are, therefore only to be applied to emplacement conditions where the thermal history can be constrained. Cooling-rates of basalts, measured in active lava channels range from 0.01 to $15^\circ C min^{-1}$, where high cooling-rates represent the exterior and lower values the insulated interior of the lava (Cashman et al., 2013; Kolzenburg et al., 2017; Witter and Harris, 2007).

Shear-rates during viscous transport in dike, conduit, or storage systems range from $\sim 70 s^{-1}$ in plinian eruptions (Papale, 1999) to as low as $10^{-9} s^{-1}$ in convecting magma chambers (Nicolas and Ildefonse, 1996). Effusion-rates for basaltic eruptions

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