



## Relaxation of concentration variance: A new tool to measure chemical element mobility during mixing of magmas

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### ABSTRACT

The ability of chemical elements to diffuse in silicate melts is central to igneous processes. It controls the rates of phase transitions such as crystal growth and dissolution kinetics, the rate of homogenization of compositional gradients generated by fractional crystallization and assimilation of country rocks as well as one of the most intriguing processes of all, magma mixing.

A very useful measure, commonly used to quantify the rate of homogenization of chemical elements in silicate melts, is the diffusion coefficient. It is widely approximated to be of a constant value (at a fixed pressure and temperature) for a melt with a given composition and rheology. When dealing with magma mixing processes, melts with different initial compositions and rheological properties (e.g. basalt and rhyolite) coexist in the same system. Under such circumstances, the compositional and rheological dependence of diffusion coefficients must be considered, leading to a considerable increase of complexity in the modeling of magma mixing. Yet, an additional and even more dramatic increase in complexity is due to the fact that the mixing of magmas is undoubtedly a very dynamic process. Scale-invariant distributions of filaments of different melts are generated by stretching and folding dynamics. This has a dual effect on the mobility of chemical elements. On the one hand their mobility increases because of an exponential increase of contact area. On the other hand, mobility can be buffered by the different diffusivities in the melts (larger in the low-viscosity than in the high-viscosity melt). Uphill diffusion of chemical elements is likely to develop at the interface between interacting magmas, further increasing the complexity of the process.

Here we aim to understand chemical element mobility during melt homogenization in a magma mixing event under dynamic conditions. We have performed experiments by mixing phonolitic and alkali-basaltic melts. The mixing process was induced using a high-temperature centrifuge apparatus. The rotating speed and acceleration during all experiments were 1850 rpm and 1000 g, respectively. Experiments were performed for 5, 20 and 120 min. Samples were arranged in a buoyantly unstable geometry, with the denser material placed at the inner side of the rotating circle, resulting in injection of the mafic melt into the felsic melt during rotation. The temperature during experimental runs was 1200 ( $\pm 1$ )°C.

From the resultant glasses, vortex-like structures generated by repeated stretching and folding dynamics were observed at the interface between the two melts. The concentrations of major and trace elements were then measured along interfaces by electron microprobe and LA-ICP-MS. The mobility of each element was next quantified by calculating the decrease (or relaxation) of concentration variance with time. The first notable result is that for major and trace elements, concentrations variance decays exponentially. The exponent of the exponential function is then chosen as a measure of element mobility. Our results show clearly that different chemical elements homogenize in the melt at differing rates. Amongst the major elements Na is the fastest element followed by Al, Mg, Ca, K and Si. The trace elements, Ba, Rb, Sr, Nb and Zr exhibit similar mobilities. The REE display the lowest mobility and they show a systematic decrease from light to heavy.

The results from this study indicate that the decay of concentration variance is a robust tool for obtaining new insights into chemical exchanges during the mixing of silicate melts. Concentration variance includes in a single measure an expression of the influence of all possible factors (e.g. viscosity, composition, fluid-dynamic regime) controlling the mobility of chemical elements during the mixing of two liquids. A new parameter, the Relaxation of Concentration Variance (RCV), is proposed as an effective tool for quantifying the homogenization of chemical elements during the mixing of silicate melts.

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## 1. Introduction

Magma mixing is a process of undoubted importance in the petrogenesis of igneous rocks (e.g. Russell, 1990; Didier and Barbarin, 1991; Bateman, 1995; Wiebe, 1996; Jellinek and Kerr, 1999; Perugini et al., 2003b; Kratzmann et al., 2009). At any point during the life span of a magmatic system, compositional gradients may develop and lead to subsequent processes of magma hybridization, or “magma mixing”. This does not necessarily imply an interaction between melts generated from different sources (e.g. in the extreme, for example, crustal- and mantle-derived melts). For example, compositional gradients can also occur during anatexis where temperature gradients may yield different degrees of partial melting and, therefore, melts with different trace element contents. Another example is fractional crystallization along wall of conduits or magma chambers that would deplete the magmatic mass in those elements incorporated into the crystallizing minerals, generating compositional gradients from the core of the magma body to its peripheral portions. Yet another process in which compositional gradients are generated is assimilation of country rocks. Here the differential assimilation of country rocks, in different regions of a magma body, for instance, due to the presence of thermal gradients in the magma chamber, can produce melts with different compositions coexisting in the same system. As a consequence, mixing between different silicate melts can emerge.

Thus understanding the development of magma mixing, in space and time, is crucial for understanding these igneous processes.

Recent geochemical, numerical and experimental studies revealed what a high degree of spatiotemporal complexity is possible during the mixing process (e.g. Perugini et al., 2006; De Campos et al., 2008; Perugini et al., 2008; De Campos et al., 2011). It has been suggested that magma mixing is governed by chaotic dynamics (e.g. Perugini et al., 2003a; De Campos et al., 2011), whose temporal evolution, via stretching and folding of interacting melts, generates a scale-invariant (i.e. fractal) distribution of filaments of melts and, consequently, a fractal distribution of compositional gradients. These phenomena have been recognized previously in natural samples (e.g. Perugini et al., 2002; Poli and Perugini, 2002; Perugini et al., 2003b) as well as in experiments performed at high temperatures using synthetic and natural melts of high viscosities (e.g. De Campos et al., 2004, 2008; Perugini et al., 2008; De Campos et al., 2011).

One of the most remarkable outcomes from those studies was the recognition of a diffusive fractionation of chemical elements caused by the evolution in time of chaotic mixing. Diffusive fractionation can generate volumes of magmas with extreme geochemical heterogeneities at short length scale [down to the order of a few microns; (Perugini et al., 2006, 2008)]. It was also shown experimentally (e.g. Chakraborty et al., 1995a; Perugini et al., 2008) that the classic conceptual model that magma mixing should generate linear trends in binary plots (e.g. Langmuir et al., 1978; Fourcade and Allegre, 1981) is not necessarily true. During homogenization, the occurrence of complex trends is more likely to be the rule than the exception. Further, differing mobilities for different chemical elements have first order consequences. The larger the difference in mobility between pairs of chemical elements, the more complex the trend in interelemental plots will be (e.g. Perugini et al., 2006).

A measure that is commonly used to quantify the rate of homogenization (i.e. the mobility) of chemical elements during mixing is the diffusion coefficient. It is commonly assumed to be a constant (at a given pressure and temperature) for a melt with a given composition and rheology (e.g. basalt). However, when dealing with magma mixing processes, melts with different initial compositions and rheological properties (e.g. basalt and rhyolite) coexist in the same system. Therefore, compositional and rheological dependence of diffusion coefficients should also be considered (e.g. Chakraborty et al., 1995b; Zhang, 2008). This leads to a remarkable increase of complexity in the modeling of these processes. A further and more dramatic increase of

complexity is that the mixing of magmas is a very dynamic process governed by chaotic dynamics. In fact, as discussed above, stretching and folding dynamics generate a scale-invariant distribution of filaments of melts. This has a dual effect on the mobility of elements. On the one hand their mobility is expected to increase because of an exponential increase of contact area between melts. On the other, mobility can be buffered by the different diffusivities in the two melts (larger in the low-viscosity than in the high-viscosity melt). Additionally, uphill diffusion of chemical elements (i.e. diffusion against concentrations gradients) is also likely to develop at the interface between interacting magmas, still increasing the complexity of the process (e.g. Watson and Jurewicz, 1984; Bindeman and Davis, 1999; De Campos et al., 2011). Therefore, although our understanding of chemical element mobility has benefitted greatly from studies of diffusion coefficients, they may not provide a complete enough picture when dealing with magma mixing processes.

To understand the complexity of magma mixing, and its impact on the petrogenesis of igneous rocks, is clearly a formidable but important task. Here we present new experiments performed with natural phonolitic and alkali-basaltic melts in a high-temperature centrifuge apparatus. In particular, we trigger the injection of the alkali-basaltic melt into the phonolitic one and we follow the evolution of the process in time by evaluating changes in composition due to the mixing process. We introduce the concept of the Relaxation of Concentration Variance (RCV). Our results are discussed in the light of different RCV for different elements in the mixing system. Petrological implications of the results presented in this work are discussed.

## 2. End-members and experimental setup

### 2.1. End-members

Experiments were performed using natural samples from the Campi Flegrei (Italy) volcanic area (Arienzo et al., 2009). In particular, the end-member melts derive from Agnano Monte-Spina B1-B2 phonolitic tuff (de Vita et al., 1999) and the alkali-basaltic glass matrix from the Minopoli eruption (Di Vito et al., 1999). These two compositions are thought to be the most suitable counterparts for replicating extreme interacting compositions in the Campi Flegrei system (Arienzo et al., 2009). Measured viscosities and calculated densities of the basaltic and the phonolitic melt at 1200 °C (i.e. the temperature used during the experiments, see below) are 200 and 20,000 Pa s, and 2.82 and 2.50 g/cm<sup>3</sup>, respectively. In the rest of this work the phonolitic and alkali-basaltic melt will be referred to as, felsic and mafic melt, respectively.

Starting materials have been obtained from melting these natural volcanic products. Each end-member was completely homogenized at super-liquidus temperature (1400 °C) in a high-temperature MoSi<sub>2</sub> electrical resistance furnace (NaberthermTM) for 24 hours. Then samples were drilled out and tested for homogeneity and the absence of crystals. Cylinders from the glasses with 0.5 cm height and 0.5 cm wide were then double-sided polished and mounted inside a bottom sealed Pt-capsule. After mounting the cylinders, one on top of the other (Fig. 1a), the Pt-capsule was sealed on the top, leaving a small (<1.0 mm) aperture for eventual gas release during experimental runs. Mass ratios of the felsic and mafic melts for the experiment presented in this work are 50:50. Major and trace element analyses of end-member melts are reported in Table 1.

### 2.2. The experimental apparatus

The experimental apparatus consists of a standard laboratory Cryofuge 8500i centrifuge (Heraeus InstrumentsTM) that has been modified to hold a high-temperature (1200 °C) furnace. Temperature was controlled by three independent Pt–Pt<sub>90</sub>Rh<sub>10</sub> (type S) thermocouples at the top, bottom and the middle of the sample container

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