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Reconstruction of magmatic variables governing recent Etnean eruptions: Constraints from mineral chemistry and P–T–fO_2–H_2O modeling

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ABSTRACT

Petrological investigations of active volcanoes are often supported by mass balance, thermodynamic calculations and/or experiments performed at key conditions. Conversely, the compositions of mineral phases found in natural products are generally used as input data for predictive models calibrated to derive the intensive variables of the magmatic system. In order to evaluate the extent to which mineral chemistry records crystallization conditions, we have compared the compositions of olivine, clinopyroxene, plagioclase and titanomagnetite in 2001–2012 trachybasaltic lavas at Mt. Etna with those obtained through thermodynamic simulations and experiments conducted under anhydrous, water-undersaturated and water-saturated conditions. This systematic comparison allows us to track recent differentiation processes beneath Mt. Etna, as well as the P–T–fO_2–H_2O variables controlling the solidification path of magma. Two compositionally distinct populations of olivine and clinopyroxene phenocrysts are found in these lavas: Mg-rich and Mg-poor minerals formed at 600–1100 MPa and 1100–1250 °C, and 0.1–500 MPa and 1050–1175 °C, respectively. The oxygen fugacity varies by 1–2 log units suggesting water exsolution during magma ascent in the conduit and magma emplacement near the surface. The nucleation and growth of normally zoned plagioclases occur at $P < 100$ MPa, when the amount of H_2O dissolved in the melt abruptly decreases from about 3.0 to 0.2 wt.% due to magma decompression and degassing. This leads to the conclusion that Etnean magmas fractionate throughout the entire length of the vertically developed plumbing system where magma mixing, volatile exsolution and degassing are the most important processes driving eruptions.

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1. Introduction

Mt. Etna volcano (Sicily, Italy) is the largest volcano in Europe and one of the most active volcanoes on Earth. The onset of volcanism occurred with the eruption of tholeiitic basalts (ca. 500 ka) and, subsequently, the composition of magmas shifted towards a more alkaline affinity (ca. 220 ka) (Tanguy et al., 1997). Compared with the oldest historical eruptions, the degassing rate, the concentration of alkaline elements and the radiogenic strontium significantly increased in the last four decades (Armienti et al., 2004; Corsaro et al., 2007). A distinctive feature of this recent activity is the relatively constant degree of magmatic differentiation characterized by the eruption of trachybasaltic lavas with a uniform phenocryst assemblage of olivine, clinopyroxene, plagioclase, and titanomagnetite (Corsaro et al., 2009). The overall composition of phenocrysts is controlled by several processes that occur in (1) the deepest parts of the plumbing system during magma segregation at mantle depths (Armienti et al., 2007, 2013; Viccaro and Cristofolini, 2008), (2) at shallow crustal levels where magma differentiation, mixing and degassing are active processes (Corsaro et al., 2013; Ferlito et al., 2011; Giacomoni et al., 2014; Lanzafame et al., 2013) and (3) the emplacement conditions including the effect of cooling rate (Mollo et al., 2011a, 2013a; Scarlato et al., 2014).

Detailed petrological and geochemical studies have suggested that the plumbing system at Mt. Etna is governed by frequent inputs from mantle depths of primitive, volatile-rich magmas that mix with more evolved, degassed melts residing at shallow crustal levels (Clocchiatti et al., 2004; Corsaro et al., 2013; Ferlito et al., 2008; Métrich et al., 2004; Spilliaert et al., 2006). For the case of 2006 summit eruptive episodes, the history tracked by olivine crystals reveals that the magma pathway within the shallow plumbing system is controlled by two mafic recharge events within a few months where magma supply triggered the onset of a distinct eruptive cycle (Kahl et al., 2011).

Despite the fact that the isotope signature of some lavas reveals heterogeneities in the mantle source, both primitive and more

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differentiated magmas are systematically buffered to the composition of trachybasalt in terms of major element compositions due to the continuous supply of magma from the mantle (Ferlito and Lanzafame, 2012; Ferlito et al., 2014; Giacomoni et al., 2014). Thus whole-rock geochemistry alone is unable to comprehensively track the ascent paths of Etnean magmas. Workers primarily relying on mass balance calculations have proposed different petrogenetic mechanisms to explain the evolution of magmas, leading to number of controversies that are still matter of debate. Michaud (1995) proposed a mobility of crustal components (i.e., K, Rb and Cs) and their selective contamination of the magma as a consequence of the percolation of a carrier fluid phase through the upper parts of the magmatic feeding system. In contrast, Schiano et al. (2004) suggested that the geochemical evolution of Etnean magmas is due to a progressive transition from a predominantly intraplate-type mantle source to a different one contaminated by slab-derived fluids originated by the progressive southward migration of the slab or continuous opening of a slab window. Recently, Ferlito and Lanzafame (2010) argued that supercritical fluids coming from deeper magmas from the mantle carrying alkali Cl-complexes migrate through basic to intermediate magmas residing in the shallow feeding system. As chlorine exsolves and leaves the system, alkalis are released contributing to the observed potassium enrichment of magmas erupted after 1971.

These different petrogenetic models highlight a general paucity of information on the crystallization conditions of magma. While whole-rock data and mineral compositions are frequently documented and discussed, lesser importance has been given to their significance for the evolution of magma at both pre-eruptive and syn-eruptive conditions (Mollo et al., 2011b, 2013b). Specifically, the limited inter-crystal compositional variations recorded in the cores of larger phenocrysts hosted in lavas indicate a pristine equilibrium at nearly-static crystallization conditions. In contrast, the complex compositions of phenocryst mantles and rims, as well as those of micro-phenocrysts and microlites, mainly reflect disequilibrium crystallization under dynamic conditions due to magma mixing, rapid decompression and/or cooling during magma ascent in the conduit and lava flowage (Applegarth et al., 2013; Corsaro et al., 2013; Lanzafame et al., 2013; Mollo et al., 2013b; Viccaro et al., 2010). In order to decipher the pre-eruptive conditions of recent Etnean magmas and track their P–T–fO_2–H_2O paths, we have compared the compositions of phenocrysts found in 2001–2012 lavas with those obtained through phase equilibrium experiments and thermodynamic simulations conducted over a wide range of crystallization conditions. The crystallization conditions gained through this comparison have been integrated with the estimation of intensive magmatic variables through mineral-melt equilibrium models, thermometers, barometers, oxygen barometers, and hygrometers. Our predictions contribute to a reconciliation of most of the contradictions raised during an intense debate with regards to magma mixing, water exsolution and degassing processes operating from the mantle to the shallow feeding system to the summit crater of Mt. Etna volcano.

2. Experiments and analyses

For the purpose of this study, we have compiled three distinct datasets composed of natural, experimental and thermodynamic compositions of olivine, clinopyroxene, plagioclase, and titanomagnetite. Natural and experimental datasets from the literature have been integrated with new data from this study in order to increase the statistical significance of natural compositions and to extend the intervals of crystallization conditions reproduced in laboratory. Both experiments and analyses were conducted at the HP-HT Laboratory of Experimental Volcanology and Geophysics of the Istituto Nazionale di Geofisica e Vulcanologia (INGV) in Rome, Italy. These datasets are reported in the Excel spreadsheet provided as supplementary material together with whole-rock analyses.

The starting material used for the experiments is a trachybasalt from the 2012 eruption at Mt. Etna. In order to saturate the sample holders in iron, some aliquots of the powdered rock were loaded into the Pt crucible and Pt capsules, and run for 3 h at 1600 °C in an atmosphere chamber furnace. They were then quenched and cleaned in a hot HF solution. In this way, the iron loss from the samples during the experiments was kept to $\leq$5% of the initial amount (see Mollo et al., 2013b). The starting glass was produced by double melting in the Pt crucible 20 g of powdered rock at 1600 °C and atmospheric pressure for 30 min. The glass was analyzed by X-ray powder diffraction and scanning electron microscopy, and no crystalline phases were detected. The starting glass composition is reported in the supplementary material. Pressure experiments were performed at 250 and 400 MPa in a piston cylinder apparatus. Pt capsules loaded with the powdered glass were positioned into a 19.1-mm NaCl–crushable alumina–pyrophyllite–pyrex assembly which produced a fO_2 of NNO + 1.5 (see Mollo et al., 2013d). Water was added by microsyringe into the charge so that hydrous experiments contained 1, 1.5, and 2 wt.% H_2O. The charges were heated directly to the target temperatures of 1000, 1070, 1100, 1130, and 1150 °C that were maintained constant for 24 h. Each experiment was rapidly quenched using a quench rate of 2000 °C/min. The run product was mounted in epoxy and a polished thin section was produced from the epoxy block.

Microprobe analyses were carried out with a Jeol-JXA8200 equipped with five spectrometers. The analyses were conducted on both experimental phases and natural samples from 2001–2012 eruptions. For glasses, a slightly defocused electron beam with a size of 3 μm was used with a counting time of 5 s on background and 15 s on peak. For crystals, the beam size was 1 μm with a counting time of 20 and 10 s on peaks and background, respectively. The following standards have been adopted for the various chemical elements: jadeite (Si and Na), corundum (Al), forsterite (Mg), andradite (Fe), rutile (Ti), orthoclase (K), barite (Ba), apatite (P), spessartine (Mn) and chromite (Cr). Sodium and potassium were analyzed first to prevent alkali migration effects. The precision of the microprobe was measured through the analysis of well-characterized synthetic oxides and minerals. Data quality was ensured by analyzing these test materials as unknowns. Based on counting statistics, analytical uncertainties relative to their reported concentrations indicate that precision was better than 5% for all cations.

3. Results

3.1. Natural dataset

The natural dataset consists of phenocryst compositions from trachybasaltic lavas erupted during 2001–2012 magmatic activity (Collins et al., 2009; Corsaro et al., 2007; Giacomoni et al., 2014; Nicotra and Viccaro, 2012; Spilliaert et al., 2006; Viccaro et al., 2006, 2010), and also comprises new data from this study.

Olivine shows compositions from Fo_{67} to Fo_{88} that are generally negatively correlated with the amount of Ca (Fig. 1a). The Mg# [$Mg\# = \text{atomic } Mg/(Mg + Fe^{2+})$] of clinopyroxene changes from 54 to 85 clustering over two distinct compositional intervals of $Mg\#_{54-74}$ and $Mg\#_{75-85}$ (Fig. 1b); conversely, no clear variations are observed for Ti ranging from 0.02 to 0.07 apfu. The content of An (anorthite) in plagioclase varies widely from 56 to 89 but the amount of Fe remains almost constant with values of 0.1–0.4 apfu (Fig. 1c). Most titanomagnetite phenocrysts lie between Usp_{25} and Usp_{44} (ulvospinel) and only a few data show higher Usp_{48-55} values corresponding to low Al + Mg contents (Fig. 1d).

These phenocrysts are hosted in lavas showing almost homogeneous whole-rock compositions. The analyses indicate a limited variation for both SiO_2 and $Na_2O + K_2O$ that are in the range of 47–51 wt.% and 5–6 wt.%, respectively. This implies that magmas are buffered to the composition of a trachybasalt with $Mg\#_{47-57}$.

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