



Magmatic crystal records in time, space, and process, causatively linked with volcanic unrest

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ABSTRACT

How a volcano has behaved throughout its past is a guide to its future behaviour. Detailed knowledge of what preceded eruptions from specific volcanoes, and how this can be recognised in real-time, are pivotal questions of this field. Here, the physical history of the magma that erupted in 2010 from the flank of Eyjafjallajökull volcano, Iceland, is reconstructed in absolute time and space using only chemical records from erupted crystals. The details of this reconstruction include the number of magma bodies, their geometry, their depth, their relative inflation rate and changes to all of the aforementioned through time. Petrology and geodesy (data gathered in real-time) arrive at the same set of conclusions. As such, we report detailed agreement, which demonstrates a causative link between knowledge determined post-eruption via a physical–chemical perspective and knowledge gained syn-eruption from monitoring signals.

The composition of olivine crystal cores (~Fo74–87), and that of the chemical zonation around each core caused by disequilibrium processes, are shown to form systematic patterns at the population scale. Reverse zonation (toward Mg rich) exhibits a constant chemical offset from its crystal core (≤ 2 mol % Fo), while normal zonation (toward Fe rich) converges to a single composition (~Fo75). Conventional petrological models – for instance multiple-magma-mixing across a range of crustal depths – can explain the presence of a range of crystal core composition in the erupted rocks, but cannot explain these patterns of crystal disequilibria.

Instead, we describe how a single primitive melt produces crystals over a wide range in composition and generates systematic disequilibrium. Cooling causes crystal production from both roof and floor of a horizontal magma geometry. Crystal settling causes asymmetric thermal – and therefore compositional – stratification of the melt due to progressive insulation via development of a crystal mush at the floor, a process we term “Crystal Rain”. Crucially, each crystal’s record is both a cause and effect of the internal process of simultaneous fractional crystallisation and settling; no external processes or materials are required.

We then extract temporal information from our crystals using Fe–Mg interdiffusion modelling, and combine it with the composition and zonation data. The concept of Crystal Rain is applied, and resolves two thin (metres) sills which are staggered in time and depth, and exhibit different inflation rates. Since the approach of integrating crystal chronology within a causative physical framework may be applied to entire volcanic successions, it has potential to yield valuable insights to past, and by inference future, magmatic and volcanic behaviours by deterministic means.

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1. Introduction

Approximately 800 million people live within 100 km of one of Earth’s 1551 active volcanoes, ~80% of which are not adequately

assessed and even fewer are actively monitored (Loughlin, 2015). Volcano observatories use a range of geophysical, geochemical and petrological techniques to track magma and plumbing system behaviour (Sigmundsson et al., 2010; Tarasewicz et al., 2012; Zuccarello et al., 2013). In some cases real-time monitoring extends back decades, yet even in these the observed activity reflects only a tiny fraction of a volcanic lifetime. Since these techniques

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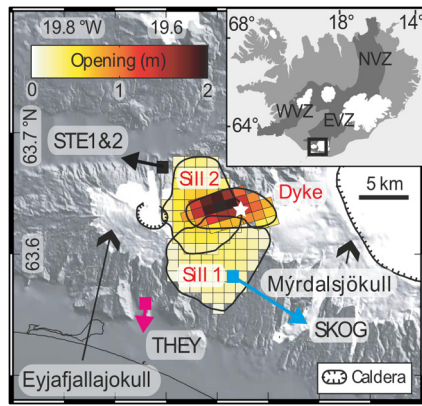


Fig. 1. Location map and summary of data gathered in real time. Fimmvörðuháls eruption site (white star) and lateral extent of dilation envelopes of the volcano plumbing system as interpreted from ground deformation and earthquake signals collected in real-time (Sigmundsson et al., 2010), presented on top of a digital elevation model. The dyke is inclined to the south from the surface to the sills (~5 km depth; scale bar at the top indicates vertical inflation). Arrows from GPS stations SKOG, THEY and STE1&2 denote the relative magnitude of displacement in the period from November 2009 to onset of eruption on 20th March 2010 in the directions reported (Sigmundsson et al., 2010); colours are the same used in later figures. Some figure elements via pers. comm. A. Hooper.

are not associated to any lasting record, the deeper past cannot be accessed to aid empirical comparisons and deterministic forecasts (Schmid et al., 2012).

Petrology, by contrast, can read the entirety of the available rock record. Recent research has demonstrated that temporal information using crystal diffusion chronometry (Bouvet de Maison-neuve et al., 2016; Hartley et al., 2016; Kahl et al., 2014; Rae et al., 2016; Ruprecht and Plank, 2013; Viccaro et al., 2016; Zellmer et al., 2005), can be compared on the same time scales as seismicity (Kahl et al., 2011; Longpré et al., 2014), ground deformation (Sigmundsson et al., 2010), gas sampling (Kahl et al., 2013), and even eruption (Charlier et al., 2012). The important limitations are that such studies can only be conducted after eruption, and crystal chronology is only possible if those rocks contain a tractable mineralogy.

The longstanding challenges have been to 1) accurately determine the dynamics of magma plumbing systems in absolute space and time from volcanic products, and 2) identify direct, *causative* links between what crystal cargoes record, and any observable signals recorded at the surface. Merging the sub-disciplines first requires us to recognise the different expressions of the same processes within independent datasets, and to link them via shared mechanisms. To achieve this using petrology, each crystal's information must be placed into a single, consistent, context. In order to link these small portions of information into such a context we must conduct investigations at the population scale, for which we require significant datasets. To provide an adequate test, it is necessary to attempt this work where high quality geophysical data are also available.

The 20th March–12th April 2010 flank eruption of Eyjafjallajökull, Iceland, situated in the Fimmvörðuháls pass (Fig. 1), presents an exceptional case study. With known eruption dates for individual samples (Keiding and Sigmarsson, 2012; Sigmarsson et al., 2011; Thordarson et al., 2011) we can correct timescales of olivine Fe–Mg inter-diffusion (Costa and Morgan, 2010; Dohmen et al., 2007; Dohmen and Chakraborty, 2007; Kahl et al., 2014) to an absolute chronology, for comparison to pre- and syn-eruption monitoring signals (Sigmundsson et al., 2010; Tarasewicz et al., 2012). Here we first seek to understand the petrogenesis of the magma(s) involved using chemical, textural, and chronological observations, and then compare our results and interpretation with that derived from the geophysical record. Our overriding question

is “can petrology provide detailed information of dynamic processes comparable to that determinable by monitoring signals?”

2. Nomenclature and methods

The term ‘macrocryst’ is used here to avoid the genetic connotations inherent in terms such as *phenocryst* (a crystal which grew free in the same magma it is observed within); *antecryst* (a ‘recycled’ crystal that grew in a previous magma of essentially the same system); and *xenocryst* (a crystal with little to do with the petrogenesis of the magma observed; its presence is due to ‘accidental’ capture; Davidson et al., 2007).

Phase rules are fundamental to petrologic interpretation and a considerable literature and range of practical tools are available to draw upon (see Spera et al., 2016 and Moyen et al., 2016 for recent introductions). The scope of this study limits us to reiterate a single point; phase diagrams are abstracted from the kinetic limitations that exist in nature, which demonstrates near ubiquitous evidence for non-equilibrium processes, as well as incomplete extent of reactions toward equilibrium (Shore and Fowler, 1996). Since dynamic scenarios are discussed throughout, for clarity we define the following.

When the term magma ‘Equilibrium’ is used, it is implied that the melt is chemically homogeneous, and all formed crystals are of the same composition (per mineral). An uncapitalised ‘equilibrium’ describes an end-point for changes that may never be realised, but for which there is driving force via equilibration mechanisms such as growth, resorption and diffusion.

Olivine composition is expressed as the molecular percentage of the forsterite (Mg_2SiO_4) endmember (Fo) in the olivine solid solutions, i.e. $(\text{Mg}^{2+}, \text{Fe}^{2+}, \text{Mn}^{2+}, \text{Ni}^{2+}, \text{Ca}^{2+})_2\text{SiO}_4$. Magmatic olivine is essentially a single solid solution between Mg and Fe endmembers; the Mn (tephroite endmember), is usually present as a minor component and Ni and Ca are common trace cations (Deer et al., 1982). An olivine's Fo content is linked to the Mg# of the surrounding melt (Roeder and Emslie, 1970) where $\text{Mg\#} = \text{Mg}/(\text{Mg} + \text{Mn} + \text{Fe})$ on an atomic basis. Olivine crystallisation and fractionation preferentially removes Mg, relative to Fe^{2+} , from a melt (Roeder and Emslie, 1970), driving Mg# down. Subsequent olivine is thus increasingly Mg-poor, forms at lower temperatures, and is described as more evolved. See also supplementary sections 4 and 5 for notes on the chemical system here, including incorporation of $f\text{O}_2$ constraints (Gunnlaugsson et al., 2013). Olivine macrocrysts (>1 mm, $n = 236$) across four samples from the flank eruption of Eyjafjallajökull (Fimm7, -5, -3, -2 in order of eruption) were mapped with high-resolution Z-contrast backscattered electron (BSE) imaging and electron backscatter diffraction (EBSD) techniques. Core-rim profiles were analysed using wavelength-dispersive electron probe microanalysis (WD-EPMA; see supplementary section 2 and also Pankhurst et al., 2017), and a subset ($n = 12$) of crystal margins were mapped for Ni and P using WD-EPMA, and Mg, Fe and Si using energy-dispersive spectroscopy (EDS).

BSE imaging produces maps of relative electron density; for olivine, brighter pixels indicate a higher concentration of Fe (darker = Mg rich). We calibrated the Fo content of an average pixel value located at a given analysis spot to within ~1 Fo unit (see supplementary section 2). Using per-image calibrations, profiles up to 100 pixels wide were extracted, which allowed us to capture major element chemical distribution with high spatial and compositional resolution, as well as textural and crystallographic context.

The textures and chemical zonation from each crystal were integrated and compared. We first establish the most likely dynamic petrological scenario that fits these data, which is used to define boundary conditions for the retrieval of diffusion timescales (from

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