

Plagioclase–melt (dis)equilibrium due to cooling dynamics: Implications for thermometry, barometry and hygrometry

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ABSTRACT

The compositional variation of plagioclase and the partitioning of major elements between plagioclase and melt have been experimentally measured as a function of the cooling rate. Crystals were grown from a basaltic melt at a pressure of 500 MPa under (i) variable cooling rates of 0.5, 2.1, 3, 9.4, and 15 °C/min from 1250 °C down to 1000 °C, (ii) quenching temperatures of 1025, 1050, 1075, 1090, and 1100 °C at the fixed cooling rate of 0.5 °C/min, and (iii) isothermal temperatures of 1000, 1025, 1050, 1075, 1090, and 1100 °C. Our results show that euhedral, faceted plagioclases form during isothermal and slower cooling experiments exhibiting idiomorphic tabular shapes. In contrast, dendritic shapes are observed from faster cooled charges. As the cooling rate is increased, concentrations of Al + Ca + Fe + Mg increase and Si + Na + K decrease in plagioclase favoring higher An and lower Ab + Or contents. Significant variations of $^{pl-liq}Kd$ are also observed by the comparison between isothermal and cooled charges; notably, $^{pl-liq}Kd_{Ab-An}$, $^{pl-liq}Kd_{Ca-Na}$ and $^{pl-liq}Kd_{Fe-Mg}$ progressively change with increasing cooling rate. Therefore, crystal–melt exchange reactions have the potential to reveal the departure from equilibrium for plagioclase-bearing cooling magmas. Finally, thermometers, barometers, and hygrometers derived through the plagioclase–liquid equilibria have been tested at these non-equilibrium experimental conditions. Since such models are based on assumption of equilibrium, any form of disequilibrium will yield errors. Results show that errors on estimates of temperature, pressure, and melt-water content increase systematically with increasing cooling rate (i.e. disequilibrium condition) depicting monotonic trends towards drastic overestimates. These trends are perfectly correlated with those of $^{pl-liq}Kd_{Ca-Na}$, $^{pl-liq}Kd_{Ab-An}$, and $^{pl-liq}Kd_{Fe-Mg}$, thus demonstrating their ability to test (dis)equilibrium conditions.

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1. Introduction

Igneous hygrometers and thermobarometers are commonly used to estimate temperatures and depths of crystallization of natural systems, and these estimates can be very useful for elucidating magma storage conditions and modes of magma transport (e.g. Klügel and Klein, 2006; Putirka and Condit, 2003). However, such models nearly always assume that natural systems approach equilibrium. But just because a mineral happens to be included in a volcanic rock is not a guarantee that equilibrium has been achieved. It is now well recognized that many volcanic rocks contain a mix of crystals that have precipitated at a range of temperatures, pressures and variable water contents, and perhaps from different liquids at different times (Cooper and Reid, 2003; Putirka, 2008; Streck, 2008). There is also now a rising recognition that crystals may grow rapidly, and in the process, may deviate from equilibrium, even if the crystal is indeed

contained within the liquid from which it grew (Hammer, 2008). A key question is: given a crystal and a host lava composition, how do we know whether or not the two are in equilibrium, so that igneous thermometers and barometers may be correctly applied? And if we mistakenly use a disequilibrium mineral–melt pair, what is the sign and magnitude of the systematic error? Mollo et al. (2010a) and Putirka (2008) show that for clinopyroxene-based models, the systematic errors resulting from accidental use of a disequilibrium pair may be very great; these studies illustrate the need for better tests of equilibrium that can be applied to natural systems, and that such information is ideally obtained by crystal growth rate studies. Here, we build on earlier work by Mollo et al. (2010a) to explore tests for equilibrium for plagioclase–liquid pairs, and plagioclase-based thermometers, barometers and hygrometers. More than perhaps any other mineral phase, plagioclase compositions are characterized by a marked sensitivity to pressure, temperature and the water content contents of magmatic systems. Therefore, thermometers, barometers and hygrometers are derived through the plagioclase–liquid exchange reaction between albite (NaAlSi₃O₈) and anorthite (CaAl₂Si₂O₈) components (Lange et al., 2009; Putirka, 2005, 2008;). Since models are

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based on assumption of equilibrium, any form of disequilibrium leading to compositional variations of the plagioclase–liquid pair will yield systematic errors on estimates of temperature, pressure and melt-water concentration for natural plagioclase-bearing rocks.

2. Experimental and analytical methods

The compositions of plagioclase–liquid pairs from cooling runs described in Del Gaudio et al. (2010) are compared to those from isothermal conditions of this study. All the experiments were carried out at the HP-HT Laboratory of Experimental Volcanology and Geophysics of the Istituto Nazionale di Geofisica e Vulcanologia in Roma (Italy). Here, we briefly summarize the experimental and analytical methods.

The chemistry of the synthetic starting material used for the experiments corresponds to a basaltic lava from Etna (Sicily, Italy) ($\text{SiO}_2 = 47.61$, $\text{TiO}_2 = 1.65$, $\text{Al}_2\text{O}_3 = 16.76$, $\text{FeO} = 10.58$, $\text{MnO} = 0.17$, $\text{MgO} = 6.51$, $\text{CaO} = 11.13$, $\text{Na}_2\text{O} = 3.31$, $\text{K}_2\text{O} = 1.82$, $\text{P}_2\text{O}_5 = 0.46$ [all wt.%]). This composition was synthesized by mixing oxide and carbonate powders in a ball mill. The resulting mixture was melted twice, to ensure the homogeneity of the glass, using a Pt-crucible loaded in 1-atm furnace at 1600 °C for 3 h. The glass was analyzed by X-ray powder diffraction and scanning electron microscopy and no crystalline phases were detected.

All the experimental charges were sealed in Pt-capsules and then run in a piston cylinder. Capsules were positioned into a 19.1 mm NaCl–crushable alumina–pyrophyllite–pyrex assembly which produces a f_{O_2} during the experiments of $\text{NNO} + 1.5$ (Freda et al., 2008; Mollo et al., 2010b and references therein).

For each experiment, pressure was held at a constant value of 500 MPa and a superliquidus temperature (1250 °C) was reached starting from room temperature (with a ramp of 100 °C/min) and it was maintained for 30 min. Notably, this temperature is about 40 °C higher relative to the liquidus temperature (1211 °C) determined by Del Gaudio et al. (2010). At this point, our experiments can be divided into two sets. For Experimental Set 1, samples were cooled using five cooling rates of 15, 9.4, 3, 2.1, and 0.5 °C/min from 1250 down to 1000 °C. For Experimental Set 2, samples were cooled at the fixed cooling rate of 0.5 °C/min applying five different quenching temperatures of 1025, 1050, 1075, 1090, and 1100 °C. In Experimental Set 3, six additional static isothermal experiments were performed by heating the starting glass from room temperature up to 1000, 1025, 1050, 1075, 1090, and 1100 °C and holding the temperature constant for 42 h.

Chemical analyses were carried out with a Jeol-JXA8200 EDS-WDS combined electron microprobe equipped with five wavelength-dispersive spectrometers, using 15 kV accelerating voltage and 10 nA beam current. Plagioclase crystals were analyzed with a beam size of 2 μm and 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) and spessartine (Mn). Sodium and potassium were analyzed first to reduce possible volatilization effects. Precision was better than 5% for all cations. Images were obtained with a Jeol FE-SEM 6500 F equipped with an energy dispersion microanalysis system.

3. Results

3.1. Textures

The overall textural characteristics of the experiments are described in Del Gaudio et al. (2010). In general, run products show different textures, varying with both cooling rate and quenching temperature. Plagioclase is a significant phase for all the experiments; it comprises up to 60 vol.% of the charges (Table 1). The main textural

Table 1

Run conditions and phases occurring in experimental products.

Run#	Plg (vol.%)	Cpx (vol.%)	Ox (vol.%)	Glass (vol.%)	Cooling rate (°C/min)	Final temperature (°C)
<i>Variable cooling rate</i>						
EB1D	46.3	31.2	3.2	19.3	0.5	1000
EB8A	44.1	29.0	3.2	23.7	2.1	1000
EB1E	36.1	35.1	3.1	25.7	3.0	1000
EB7B	39.2	32.8	1.6	26.4	9.4	1000
EB1F	8.0	11.9	0.5	79.6	15.0	1000
<i>Variable quenching temperature</i>						
EB12A	24.9	13.9	0.5	60.7	0.5	1025
EB13A	15.3	14.3	0.8	69.6	0.5	1050
EB14A	10.8	6.9	2.1	80.2	0.5	1075
EB10A	4.0	4.0	0.5	91.5	0.5	1090
EB11B	7.0	5.0	0.5	87.5	0.5	1100
<i>Variable isothermal temperature for 42 h</i>						
EB9A	46.8	32.7	3.6	16.9	0.0	1000
EB17A	29.0	18.5	3.0	49.5	0.0	1025
EB18A	20.0	16.5	2.5	61.0	0.0	1050
EB19A	16.0	10.0	2.5	71.5	0.0	1075
EB20A	11.0	9.0	2.0	78.0	0.0	1090
EB21A	10.0	8.0	2.0	80.0	0.0	1100

features can be summarized as follows: i) clinopyroxene, plagioclase and Fe–Ti-oxide occur in all the charges irrespective of the experimental conditions; ii) textural coarsening and pyroxene–plagioclase intergrowths occur at higher cooling rate and higher quenching temperature for the fixed cooling rate of 0.5 °C/min; iii) higher crystal contents are measured in run products from isothermal experiments; iv) the modal abundance of crystals increases with decreasing cooling rate; v) at the fixed cooling rate of 0.5 °C/min, higher amounts of glass are attained when the quenching temperature is increased. Cooling rate exerts the primary control on the morphologies of the plagioclase (Fig. 1). Runs cooled more rapidly than 3 °C/min contain

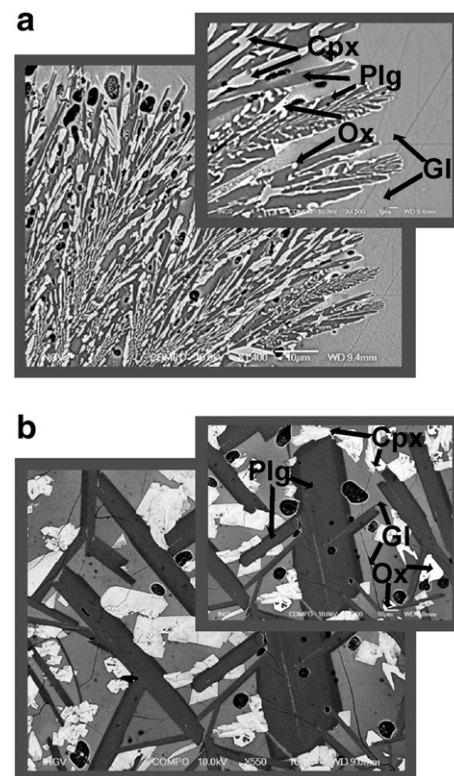


Fig. 1. Textural features of plagioclase from the anhydrous run-products at cooling rates of 15 (a) and 0.5 (b) °C/min from 1250 to 1000 °C. See also Figs. 1 and 5 in Del Gaudio et al. (2010). Plg, plagioclase; Cpx, clinopyroxene; Ox, Fe–Ti oxide; Gl, glass.

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