An experimental study of dissolution and precipitation of forsterite in a thermal gradient: implications for cellular growth of olivine phenocrysts in basalt and melt inclusion formation

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Abstract

The morphology of crystals in magmas strongly depends on the temperature regime of the system, in particular the degree of undercooling and the cooling rate. To simulate low degrees of undercooling, we developed a new experimental setup based on thermal migration, in which large cylinders of forsterite (single crystals) immersed in haplobasaltic melt were subjected to a temperature gradient. As forsterite solubility is sensitive to temperature, the forsterite on the high-temperature side undergoes dissolution and the dissolved components are transported toward the low-temperature side where a layer of newly grown forsterite forms (up to 340 μm thick after 101 h). A striking feature is that the precipitation process does not produce a planar front of forsterite advancing at the expense of liquid: the growth front shows a fingered outline in planar section, with solid lobes separated by glass tubes that are perpendicular to the growth front. We ascribe this texture to cellular growth, a type of growth that had not been experimentally produced so far in silicate systems. We find that the development of cellular growth requires low degrees of undercooling (a few $^{\circ}\text{C}$) and large crystal–liquid interfaces (~ 1 mm across or more), and that it occurs at a growth rate of the order of 10^{-9} m/s. We found natural occurrences of cellular growth on the rims of olivines from basanites, but otherwise cellular textures are poorly documented in natural volcanic rocks. Melt inclusions were produced in our experiments, showing that they can form in olivine at relatively slow rates of growth (10^{-9} m/s or lower).

Keywords Crystal growth and dissolution · Cellular growth · Temperature gradient · Melt inclusion formation · Thermal migration

Introduction

Crystals and melt inclusions are precious tools to decipher the processes from magma production to final emplacement, and they have been the topic of an impressive amount of studies in the past decades. Melt inclusions are parcels of liquid trapped in crystals, which hold precious information on the chemical composition (major and trace elements, volatiles) of primitive magmas (Roedder and Bodnar 1980; Schiano 2003). Zoning patterns, shape and texture of crystals are a record of the composition of the liquid they grew from and the conditions during crystallization (Welsch et al. 2013). To decipher igneous processes, we need to know the growth mechanisms of minerals. The rate at which crystals grow is of particular interest to investigate the timescale of magmatic processes, such as magma ascent rate or crystal residence time in a reservoir prior to eruption (Armienti et al. 1994; Brewer 2000; Murphy et al. 2000; Blundy et al. 2006; Schiano et al. 2006; Humphreys et al. 2006; Morgan

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and Blake 2006; Castro and Dingwell 2009; Costa et al. 2010; Viccaro et al. 2012; Zellmer et al. 2012; Druitt et al. 2012; Eppich et al. 2012).

Crystal growth is controlled by a dominant rate controlling process that defines the morphology of the crystal. Such processes are reaction at the crystal–melt interface, chemical diffusion in the melt or latent heat flow (Kirkpatrick 1975, 1981; Sunagawa 1981). The reactions at the crystal–melt interface are attachment mechanisms such as continuous growth, surface nucleation or screw dislocation. In this case, there are not significant compositional gradients in the melt along the interface. Diffusion controls the crystal growth when it is slower than attachment mechanisms. Growth controlled by chemical diffusion may produce spatial compositional gradients and usually results in morphologies like skeletal or dendritic crystals. The mechanisms that define the crystal shape and size depend on the variation of extrinsic (temperature, pressure...) or intrinsic (chemical composition, volatile content) parameters. A determinant parameter affecting the mechanism of crystal growth is temperature. In most cases, the increase of the crystal growth rate is attributed to the cooling regime of the magmatic system (O'hara et al. 1968; Takei and Kobayashi 1974; Faure and Schiano 2005). For instance, the olivine growth shape switches from polyhedral, to skeletal and to dendritic with increasing degree of undercooling and cooling rate (Donaldson 1976; Faure et al. 2003a). This crystallization shape sequence does not include cellular growth that produces an intermediate morphology (between polyhedral and skeletal/dendritic) at relatively low growth rates. Cellular growth is well known in metallurgy but poorly documented in silicate minerals (Tiller et al. 1953; Keith and Padden 1963; Shechtman et al. 1984).

Most experimental studies of olivine crystal morphologies were performed at relatively large undercooling degrees and cooling rates (Donaldson 1976; Jambon et al. 1992; Faure et al. 2003a, 2007; Faure and Schiano 2005; Goldstein and Luth 2006), which are specific to scenarios like the emplacement of a small volume of magma at or near the Earth surface. However, the large abundance of dominantly polyhedral olivine phenocrysts in igneous rocks implies low degrees of undercooling and cooling rates in numerous natural magmatic environments. At present, the study of the effect of low undercooling degrees ($\leq 10^\circ\text{C}$) on crystal growth is nearly absent from the literature due to experimental limitations. To fill this gap, we conducted thermal migration experiments, in which large forsterite crystals immersed into a basaltic liquid were subjected to a low-temperature gradient (typically, $5\text{--}10^\circ\text{C}/\text{mm}$). In the presence of a thermal gradient, solid–liquid systems are known to undergo dissolution–precipitation processes that contribute to mass transport and can strongly affect the development of cumulates in layered intrusions or the compaction

of partially molten rocks (Leshner and Walker 1988; Walker et al. 1988; Schiano et al. 2006; Sonzogni et al. 2011). At a smaller scale, a thermal gradient can cause the migration of a melt inclusion in its host crystal (Leshner and Walker 1988; Walker et al. 1988; Schiano et al. 2006; Sonzogni et al. 2011).

Figure 1 illustrates the concept of thermal migration (Buchwald et al. 1985; Leshner and Walker 1988; Walker and Agee 1988) and how it was adapted in our experimental study. Thermal migration is a process of mass transport and phase redistribution in a mixture of crystals and liquid subjected to a temperature gradient. It comprises two components: chemical diffusion resulting from variations of mineral solubility with temperature, and thermodiffusion (Soret effect) resulting from differences in temperature in the liquid. We consider a simple binary system A–B, with crystals of phase A in suspension in an interconnected liquid phase. If this system is placed in a temperature gradient, then local equilibrium between phase A and liquid will impose a more A-rich liquid composition on the high-temperature side (T^+ in Fig. 1), and a more B-rich composition on the low-temperature side (T^- in Fig. 1). As the liquid is interconnected, this gradient in liquid composition will give rise to chemical diffusion of component A from regions of high temperature to regions of low temperature, and of component B in the opposite direction. This process, which is dictated by local mineral saturation, is called saturation gradient chemical diffusion (Leshner and Walker 1988; Walker and Agee 1988). To maintain local equilibrium between crystals

Fig. 1 Illustration of saturation gradient chemical diffusion in a binary system A–B. In contrast to the conventional usage, temperature is increasing downwards to be consistent with the experimental conditions. The sketch on the right shows one of the configurations used to study forsterite growth and dissolution; a thin layer of liquid is placed between two forsterite monocrystals in a temperature gradient. The small arrows indicate the displacement of the upper solid–liquid interface (growth front at temperature T^-) and of the lower interface (dissolution front at T^+). The large arrow shows the diffusion of dissolved forsterite components in the liquid

and liquid, phase A must continuously dissolve on the high-temperature side and precipitate on the low-temperature side. In parallel to saturation gradient chemical diffusion, the liquid phase will also undergo thermodiffusion. In the case of basic silicate liquids, thermodiffusion leads to the transport of dissolved olivine components (MgO, FeO) toward the low-temperature side of the thermal gradient (Leshner and Walker 1986). Accordingly, in our study, Soret effect will assist saturation gradient chemical diffusion and lead to additional concentration of forsterite on the cold end of experimental charges.

In comparison to former thermal migration experiments (Leshner and Walker 1988; Walker and Agee 1988; Walker et al. 1988), the main difference in our study is that we replaced the small olivine crystals by large forsterite cylinders (single crystals). The most simple configuration is sketched in Fig. 1: it is composed of a thin layer of liquid between two forsterite monocrystals. Because of the difference of temperature, the lower forsterite–liquid interface (at T^+) will continuously dissolve, and the lower forsterite–liquid interface (at T^-) will continuously grow during the experiment. We show that studying the behavior of large monocrystals in a thermal gradient allows to bridge the gap between Ostwald ripening experiments, in which degrees of undercooling are $\leq 1^\circ\text{C}$ (Cabane et al. 2005), and cooling experiments, in which crystal growth data can only be obtained at degrees of undercooling of $\approx 10^\circ\text{C}$ or more (Donaldson 1976; Kirkpatrick 1981). Another advantage

of our experimental configuration is to provide at the same time an insight into the dissolution and crystallization processes at low degrees of superheating and undercooling, respectively.

Experimental procedure

Starting materials and experimental protocol

The experiments were run in the simplified system $\text{Na}_2\text{O}-\text{CaO}-\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2$ (Na-CMAS). The haplobasaltic glass was produced by Schott AG Company (see chemical compositions in Table 1) and crushed down into fine powder. Cylinders of forsterite 2–2.5 mm diameter were cored in a large synthetic monocrystal of pure Mg_2SiO_4 (Ito et al. 2003a, b) (Table 2). The thickness of the forsterite cylinders ranged from 0.6 to 2.3 mm. The faces of the cylinders were polished down to the micron. The starting materials were loaded in platinum capsules with an outer diameter of 3.0 mm and an inner diameter of 2.6 mm. The capsules were inserted into an assembly for a 3/4-inch piston–cylinder apparatus with Pyrex glass and NaCl sleeves used as pressure medium and thermal insulator. All experiments were conducted at 10 kbar (1 GPa) and 1400 or 1425 $^\circ\text{C}$ (Table 3). The temperature was generated by a graphite cylinder and read by a C-type (tungsten–rhenium alloy) thermocouple placed about 0.8 mm below the capsule. Prior to experiment,

Table 1 Electron probe microanalyses of the glass (2 σ)

Exp.#	Location (number of analyses)	SiO ₂	Al ₂ O ₃	MgO	CaO	Na ₂ O	Total	An
Synthesis ^a	Starting glass (54)	49.25	19.41	16.4	12.83	2.11	100	0.77
		0.46	0.25	0.25	0.25	0.12	0.68	0.01
A4	Bath at the top (12)	49.52	17.63	19.18	11.80	1.87	100.34	0.78
		0.29	0.56	0.36	0.33	0.18	0.76	0.02
	Between pillars (32)	49.53	17.58	19.16	11.83	1.90	100.36	0.78
		0.22	0.56	0.48	0.31	0.15	0.91	0.01
B6	Bottom (17)	49.30	17.88	18.91	11.96	1.95	100.14	0.77
		1.96	1.65	1.95	0.54	0.20	1.89	0.01
	Middle (78)	49.34	18.84	17.32	12.42	2.07	99.73	0.77
		0.38	0.41	0.82	0.52	0.17	1.23	0.02
	Top cyl. (23)	49.11	19.48	16.81	12.41	2.18	99.60	0.76
		0.56	0.47	0.35	0.32	0.12	0.83	0.01
Melt inclusions								
A2	Euhedral Fo, Fig. 6c (5)	50.32	22.96	8.53	13.51	2.89	98.20	0.72
		0.56	0.48	0.20	0.74	0.36	0.84	0.02
A4	Overgrowth, Fig. 7c, d (5)	50.96	21.32	10.33	14.25	2.35	99.20	0.77
		0.28	0.32	0.12	0.26	0.20	0.20	0.02
B1	Overgrowth, Fig. 5b (5)	49.17	19.99	13.84	14.24	2.06	99.31	0.79
		0.84	0.67	1.18	0.42	0.26	0.58	0.01

^aFrom M. Hardiagon (unpublished data)
^bAnorthite content An is the Ca/(Ca + Na) molar ratio

Table 2 Electron probe microanalyses of the different types of forsterite (2 σ): the cores of the monocrystalline cylinders; the overgrowth layers on the forsterite cylinders; the pillars on the dissolution side of the cylinders; the large euhedral forsterites that grew during the experiments; the small forsterite grains in suspension ("Fo bath")

Location	Exp.# (number of analyses)	SiO ₂	Al ₂ O ₃	MgO	CaO	Na ₂ O	Total	Mg/Si (Fo)
Cylinder core	Starting (8)	42.83	0.02	57.15	0.00	0.00	100.76	1.99
		0.45	0.03	1.04	-0.01	0.01	1.22	0.04
	A0 (6)	42.46	0.01	57.52	0.01	0.01	100.77	2.02
		0.55	0.02	0.47	0.01	0.02	0.84	0.03
	A4 (46)	42.82	0.02	57.13	0.01	0.01	101.18	1.99
		0.66	0.03	0.66	0.03	0.02	1.22	0.01
	B6 (48)	42.62	0.01	57.34	0.01	0.00	99.10	2.01
		0.63	0.02	0.62	0.04	0.01	1.06	0.05
Cylinder overgrowth	A4 (8)	43.21	0.13	56.41	0.24	0.01	101.80	1.95
		0.40	0.08	0.66	0.13	0.03	0.72	0.03
	B6 (14)	42.63	0.13	56.96	0.28	0.01	99.02	1.99
		0.32	0.08	0.34	0.10	0.02	0.69	0.03
Cylinder pillars ^a	A4 (1)	42.87	0.05	56.96	0.12	0.01	101.30	1.98
	B6 (1)	42.49	0.03	57.35	0.13	0.00	99.38	2.00
Euhedral Fo	B6 (16)	42.56	0.14	57.00	0.28	0.01	99.23	2.00
		0.68	0.04	0.69	0.04	0.03	0.61	0.05
Fo bath	A4 (9)	42.73	0.14	56.84	0.28	0.02	100.22	1.98
		0.32	0.04	0.36	0.03	0.04	0.61	0.03

^aRepresentative compositions of the inner part of the pillars (analyzed at 15–20 μ m from the liquid)

Table 3 Summary of experimental conditions

Exp ^a	m Fo	Fo axis ^b	m bath	m Fo/m bath	Fo/gl	Height (mm) Fo1; Fo2	T °C ^c	Duration (h)	Phases
A0	12.8	<i>a</i> -axis	25.8	0.33	0.55	1.02	1425	0.1	gl, ol
A2 ^d	31.2	<i>a</i> -axis	27.0	0.54	0.69	2.19	1400	176.0	gl, ol, cpx, sp
A4	29.8	<i>a</i> -axis	36.4	0.45	0.63	2.10	1425	48.1	gl, ol
B1 ^d	11.7	<i>a</i> -axis	34.5	0.25	0.25	0.68; 0.58	1425	45.2	gl, ol, sp
B6	32.0	<i>a</i> -axis	16.0	0.67	0.78	0.86; 1.41	1400	101.0	gl, ol
B7	28.6	<i>b</i> -axis	14.8	0.66	0.77	1.05; 0.94	1400	50.4	gl, ol
B8	47.1	<i>c</i> -axis	15.4	0.75	0.83	1.75; 1.53	1400	47.6	gl, ol
B9	33.9	<i>a</i> -axis	15.5	0.69	0.79	1.24; 1.11	1400	22.9	gl, ol

^aIn type A experiments, a single forsterite cylinder was immersed into a mixture of glass (67 wt%) plus small forsterite grains (33 wt%). In type B experiments, a glass layer was placed between two forsterite cylinders (Fo1 is the upper cylinder; Fo2 the lower cylinder)

^bMost cylinders were drilled along the *a*-axis of the forsterite lattice, except those in run B7 (parallel to *b*-axis) and in run B8 (parallel to the *c*-axis); see Fig. S2

^cThermocouple temperatures: the temperatures at the hot post are about 20 °C higher than these temperatures (see Fig. S1)

^dNo Au–Pd sputtering

m Fo mass of forsterite in mg, *m bath* mass of the bath (glass or glass + small forsterite) in mg, *height* height of the forsterite cylinder(s), *phases* phases present in the run products (gl, glass; ol, olivine; cpx, clinopyroxene; spl, spinel)

the assemblage containing the capsule was stored at 150 °C for 12 h minimum. The experiments started with the pressurization of the assembly to 3–4 kbar to ensure good electrical contacts between the graphite furnace and the power supplier without fracturing the solid pieces of the assembly. Then the temperature was raised to 650 °C (close to the

Pyrex glass transition) with a rate of 60 °C/min and kept constant for the end of the pressurization of the assembly. Once close to the target pressure (10 kbar), the temperature was increased to the desired set point (1400 or 1425 °C) and maintained constant during the experiment by a Eurotherm controller. Run duration ranged from 0 to 176 h. At the end

of the experiment, the sample was quenched by switching off the power, resulting in a cooling rate of ~ 80 °C/s, and then slowly decompressed to 1 bar. Details of the experimental conditions are presented in Table 3.

Setup geometry

In our experiments, a system composed of forsterite monocrystal(s) in contact with a haplobasaltic melt is subjected to a temperature gradient. In a first series of experiments, we used the experimental configuration A, in which a single forsterite crystal (Fig. 2a) about 1–2 mm thick is immersed in haplobasalt. In the final series, we used the configuration B, in which a liquid layer of the order of mm is sandwiched between two crystals (Figs. 1, 2b). Based on temperature calibration experiments, we adjusted the position of the capsule into the piston–cylinder assembly so that its bottom was less than 1 mm below the hot spot in every experiment (Figs. 2 & S1). Consequently, most of the capsule (70–80%) was above the hot spot and was subjected to a temperature decreasing upwards.

The principle of our thermal migration experiments is best explained by considering configuration B (Figs. 1, 2b). In type B experiments, the opposite faces of the cylinders are at temperatures T^+ (upper face of lower cylinder) and T^- (lower face of the upper cylinder), with $T^+ > T^-$. Let us assume that the liquid is homogeneous at the beginning, with a composition in equilibrium with forsterite at the mean temperature $(T^+ + T^-)/2$. As forsterite solubility depends on temperature, the liquid is slightly oversaturated in forsterite on the colder side of the liquid layer, and undersaturated on the hotter side. This situation triggers a process of forsterite dissolution, transport, and precipitation. The driving force is

proportional to the temperature difference $\Delta T = T^+ - T^-$, and we assume that the degree of undercooling along the upper face of the monocrystal and the degree of superheating on the lower face are equal to $\approx \Delta T/2$. Thus, for a thermal gradient of 6 °C/mm (typical of type B experiment; see below) and a melt thickness of 1 mm, the degree of undercooling driving forsterite growth will be 3 °C. Type B configuration provides a straightforward path for solute transport from the dissolution site to the precipitation site (Fig. 1).

In theory, the same reasoning applies to experimental configuration A (Fig. 2a), with a difference of temperature between the lower face of the forsterite monocrystal and its upper face equal to the thermal gradient times the thickness of the monocrystal. In this case, we expected the lower face of the forsterite to dissolve, the dissolved components to be transported upwards (in the thin melt layer between the forsterite and the platinum wall), and the upper face of the forsterite to crystallize. In practice (see “Results”), we observed that at the end of type A experiments, the platinum capsule was pressed against the forsterite monocrystal and that there was almost no melt layer left to transport the dissolved forsterite components upwards. Hence, the upper and lower parts of the sample behaved as two independent subsystems in all type A experiments.

Sample preparation

In type A geometry (runs A0, A2 and A4; Fig. 2a; Table 3), the sample was composed of a forsterite cylinder embedded in a mixture of haplobasaltic glass and small grains of forsterite (33 wt% forsterite/67 wt% glass). The purpose of adding micron-sized grains of forsterite in type A experiments was to ensure a fast establishment of equilibrium between

Fig. 2 Sketch of the experimental geometries used for experiments of types A (a) and B (b), with a typical qualitative temperature (T) distribution along the capsule axis; the hot spot is located in the lower third of the capsule, 0.5–0.7 mm above the bottom. The height of the samples ranges from 3.1 to 5.1 mm. c Experiment A0 at time zero ($t=0$ h; Table 3); SEM picture of the whole sample and enlarge-

ments of the upper and lower faces of the forsterite cylinder; the matrix is composed of small grains of forsterite and Na-CMAS basaltic glass. The bright small balls are metallic droplets produced by the destabilization of a thin layer of $\text{Au}_{80}\text{Pd}_{20}$ sputtered on the faces of the forsterite cylinder

forsterite and liquid at the beginning of the experiments. In type B geometry (runs B1, B6–9; Fig. 2b), a layer of pure haplobasaltic glass was sandwiched in between two forsterite cylinders.

In all type A experiments and in experiments B1, B6 and B9, the cylinders were drilled along the *a*-axis of the forsterite lattice (i.e., perpendicular to (b, c) plane; see Fig. S2). Some cylinders were prepared differently to explore the effect of crystal orientation on growth mechanisms: cylinders of run B7 were prepared parallel to *b*-axis, and those in B8 were drilled along the *c*-axis of the starting forsterite single crystal (Fig. S2). The others axes of the cylinders were not constrained due to free rotation during their insertion in the capsule.

The faces of forsterite cylinders can advance or recede several hundred microns during an experiment due to precipitation or dissolution, respectively. To measure precisely these displacements, it is necessary to be able to locate the initial position of the forsterite faces. We used a Denton Vacuum Coater to sputter the planar surfaces of each forsterite cylinder with a layer of Au₈₀Pd₂₀ (except in two experiments, B1 and A2, to compare the dissolution and growth textures with and without gold–palladium sputtering). At run temperature, the thin layer (a few tens of nanometers) decomposes into an array of coplanar droplets, which mark the original surfaces without disturbing the dissolution and precipitation processes (Fig. 2c). The decomposition of the continuous Au–Pd layer into droplets is due to the fact that the experiments are run well above the melting temperature of the alloy and that when the alloy layer melts, it breaks down into droplets to minimize interfacial energy.

Thermal profile

Our experiments were conducted at constant temperature, the main driving force for forsterite growth and dissolution being the temperature difference along the sample height. In a piston–cylinder assembly, the temperature difference between the top and the bottom of a sample depends on its vertical position relative to the hot spot of the furnace (Fig. 2a). The thermal profile along the axis of our piston–cylinder assemblies has been calibrated previously using either multiple temperature readings (up to five) at different heights or the reaction $\text{MgO} + \text{Al}_2\text{O}_3 \rightarrow \text{spinel}$ (Watson et al. 2002) (Fig. S1). These calibrations show that the position of the hot spot is rather well constrained: it is at 17 ± 0.5 mm from the basis of the assembly, with values closer to 16.5 mm at high temperatures (1200 °C and above). This is about the mid-height of the furnace (34.3 mm high, including a 1.3-mm-thick graphite disc at the top). In our experimental configuration, the thermocouple tip is located at a height of 14.5 mm, that is about 2 mm below the hot spot, and the temperature difference between the thermocouple

and the hot spot is 20 °C (note that the temperatures in Table 3 are the thermocouple temperatures; Fig. S1). There is a thin alumina disc separating the thermocouple tip and the platinum capsule, and the bottom of the silicate sample is located 0.5–0.7 mm below the hot stop (depending on the thickness of the platinum weld at the bottom of the capsule).

To estimate the temperature at the top of the capsules (the low-temperature side, on the opposite side to the thermocouple tip), we relied on two independent techniques: (1) the temperature calibration curve in Fig. S1, and (2) the appearance of secondary mineral phases (spinel and clinopyroxene) using the phase diagram of our CMAS–Na haplobasaltic composition as reference (M. Hardiagon, personal communication). At 10 kbar, the liquidus temperature lies between 1450 and 1425 °C, and the liquidus phase is forsterite (Fo). The second solid phase that crystallizes at 10 kbar is spinel (Sp), which appears between 1400 and 1375 °C, and the third one is clinopyroxene (Cpx), which appears between 1350 and 1300 °C. At 1300 °C, Cpx and plagioclase (Pl) are stable, in addition to Fo and melt, while spinel is absent. To compute the temperature difference between the bottom and the top of the samples, we considered that the Spl- and Cpx-in temperatures are equal to $1385 (\pm 10 \text{ °C})$ and $1330 (\pm 20 \text{ °C})$, respectively.

The thermal data are summarized in Table 4. The temperature difference between the warmest and the coldest regions of a capsule ranges from 20 to 80 °C depending on the length of the capsule. The 80 °C difference corresponds to experiment A2, which was the one with the longest capsule (5.1 mm); it is the only one in which Cpx was found at the top of the capsule (see below). The temperature difference between the two faces of a forsterite cylinder in type A experiments ranges from 8 to 25 °C, depending on their thickness and position in the capsule. The melt layer in type B experiments is placed about 1 mm above the hot spot; thus, the temperature difference across the glass layer between the two forsterite cylinders is less than ~10 °C and the thermal gradient is typically 5–6 °C/mm (Fig. S1), except in B1 (10 °C/mm) because the capsule was longer.

Analytical techniques

At the end of an experiment, the whole platinum capsule was mounted in epoxy, sectioned lengthwise, polished and carbon coated. Textures of the experimental products were observed with a JEOL JSM-5910 LV scanning electron microscope (SEM). Crystal fractions and sizes were determined from SEM photographs of 2D sections (automatic threshold based on the gray level) using SPO2003 software (Launeau 2004). Crystal orientation and face recognition were done from interfacial angle measurements following Welsch et al. (2013). Chemical analyses of the run products were made by electron probe microanalysis (EPMA)

Table 4 Main characteristics of experimental products

Exp#	Thickness ^a cylinder 1		Thickness ^a cylinder 2		Temperature		Cylinder 1				Euhedral Fo growth ^e					
	Before	After	Before	After	Ratio Fo1	Ratio Fo2	Bulk ratio	Sample height (mm)	Dif-ference ^b (sample)	Average gradient ^b (°C/mm)	Dissolution ^c		Growth ^d			
											Thick. (mm)	Rate (m/s) × 10 ¹⁰	Thick. (mm)	Rate (m/s) × 10 ¹⁰		
A0	1.01	1.00				0.99	0.99		80	15.8	0.33	5.3	0.31	5.0	0.13	2.1
A2	2.14	2.12			0.99		0.99	5.07	45	10.1	0.19	10.7	0.20	11.3	0.05	2.9
A4	2.10	2.11			1.00		1.00	4.45	45	10.0	0.12	7.4	0.20	12.3	0.10	6.2
B1	0.68	0.76	0.58	0.36	1.12	0.62	0.89	4.48	20	5.8	0.33	9.1	0.34	9.4	0.19	5.2
B6	0.86	0.83	1.41	1.18	0.97	0.84	0.89	3.43	20	6.4	nd	nd	0.11	6.3	0.06	53.3
B7	0.99	nd	1.08	nd	nd	nd		3.11	20	4.7	nd	nd	0.17	10.1	0.07	4.1
B8	1.57	1.52	1.71	1.51	0.97	0.88	0.92	4.27	20	6.0	nd	nd	nd	nd	0.06	6.7
B9	1.15	1.11	1.28	1.18	0.97	0.92	0.94	3.33	20		nd	nd	nd			

^aThicknesses before and after the experiments were measured with calipers and from SEM image, respectively. In type A experiments, there is only one cylinder (no. 1); in type B experiments, cylinder 1 is the upper forsterite cylinder and cylinder 2 the lower one

^bTemperature differences (from bottom to top) and temperature gradients in absolute values; temperatures are decreasing upwards

^cThe column “Dissolution” gives the thickness of forsterite dissolved and the rate of dissolution. The thickness of forsterite dissolved is equal to the initial cylinder thickness plus the thickness of the growth layer minus the final cylinder thickness

^dThe column “Growth” gives the thickness of the growth layer (equal to the distance from the alignment of Au–Pd droplets to the growth front) and the rate of growth

^e“Size” corresponds to the radius of the largest euohedral crystals

using a Cameca SX 100. Analytical conditions were focused beam ($< 1 \mu\text{m}$)—15 nA and 22 kV for crystals—and unfocused beam ($< 15 \mu\text{m}$)—8 nA and 15 kV for glasses—during 10 + 20 + 10 s (background, sample, background). The probe was calibrated using wollastonite, corundum, albite and forsterite standards.

Results

The textural results are illustrated in Figs. 3, 4, 5, 6, 7, and supplementary Figures S3 and S4. In all figures, the polarity of the sample during the experiment is respected (that is, the lower part of the microphotograph corresponds to the bottom of the sample in the hot spot). The location of electron probe microanalyses is shown using the same shape and color

codes in Figs. 3, 4, 5, 6, and 7. If we put aside experiment A0 that was quenched shortly after heating, major changes in the thickness of the forsterite cylinders and in the shape of their surfaces occurred during all experiments due to dissolution and precipitation processes.

Dissolution and precipitation textures: type B experiments

Experiments B6–B9 (Figs. 3, 4, S3–S4) show the evolution of type B geometry with increasing duration (at $T = 1400^\circ\text{C}$). This geometry is designed to produce a dissolution/precipitation process in three steps (Fig. 1): (1) dissolution of the upper face of the lower cylinder, which is at a higher temperature; (2) solute transport upwards through the melt layer; (3) precipitation along the lower face of the

B6 - 1400°C - 101h

Fig. 3 SEM photographs of experiment B6 (1400 °C, 101 h). **a** Overview of the sample at low magnification. Black regions are due to decompression cracks and crystal snatching, white areas are the platinum capsule, and bright and dark gray are glass (gl) and forsterite (Fo), respectively. **b** Enlargement of the upper part of the sample showing dissolution of the upper face of forsterite cylinder 1 and crystallization of large euhedral forsterite hosting melt inclusions (MI). The dissolution of the upper part of the cylinder is indicated by the Au–Pd droplets deposited on its surface and by the gap between the glass–cylinder interface and the capsule shrinkage (dashed orange arrow). The *a*-axis direction is indicated by an arrow. **c** Enlargement

of the lower face of the upper forsterite cylinder experiencing crystallization. The alignment of bright metallic droplets (Au–Pd) marks the initial forsterite cylinder surface, allowing the determination of the cylinder overgrowth (yellow arrow). Symbols and segments are EPMA spots and chemical profiles, respectively (see Figs. 8, 9). In this figure and the following ones, the polarity of the sample during the experiment is respected; the upper part of the microphotograph corresponds to the top of the sample. Crystal orientation in (C) was determined from interfacial angles indexation following (Welsch et al. 2013)

B8 - 1400°C - 48h

Fig. 4 SEM photographs of experiment B8 (1400 °C, 48 h). **a** Overview of the sample at low magnification. **b** Enlargement of the lower face of the upper forsterite cylinder, showing the fingered outline of the forsterite–liquid interface; the yellow arrow gives the thickness of

the precipitated layer. **c** Enlargement of the upper face of the lower forsterite cylinder: the receding surface is almost perfectly flat. There are some Au–Pd droplets suspended in the glass or fallen on the forsterite surface

upper cylinder (at a lower temperature). The most obvious textures are those associated with step (3), especially after 101 h and 48 h (Figs. 3c, 4b). In these figures, an alignment of Au–Pd droplets is visible deep inside the forsterite cylinder: it corresponds to the Au–Pd layer that was sputtered on the original surface. The thickness of the precipitated layer (measured from the alignment of Au–Pd droplets to the forsterite front) measures 170 µm after 48 h and 340 µm after 101 h. As presented below, the alignment of Au–Pd droplets corresponds also to a compositional limit (for minor elements: Al, Ca) between the starting synthetic forsterite and the newly grown forsterite.

A striking feature is that the precipitation process does not produce a planar front of forsterite advancing at the expense of liquid. There is a kind of self-organization that develops, in which domains of faster growth give rise to forsterite lobes protruding into the liquid. As a result, the growth front shows a fingered outline in two dimensions (2-D), with solid lobes separated by tubes of glass that are roughly perpendicular to the growth front (Figs. 3c, 4b). The average lobe spacing is of the order of 0.1 mm (that is, 10 lobes per mm of interface). The longest tubes can be up to ~150 µm deep, but are still shorter than the growth layer (that is, they do not reach the alignment of Au–Pd droplets). The lobes are elongated parallel to the orientation of the forsterite cylinder: parallel to *a*-axis in run B6 and parallel to *c*-axis in run B8. Facets controlled by the crystallographic orientation of the cylinder develop at the growth front (Figs. 3c, 4b, S3C and S4B); in Fig. 3c, these crystallographic facets join to form pyramids with sharp edges. In some cases, neighboring lobes merge at their ends and thus entrap melt in the observed section. We checked

by optical microscopy that some of these regions of trapped melt in 2-D are real glass inclusions in 3-D (i.e., inclusions totally closed).

As expected, the upper face of the lower cylinder receded due to dissolution. This is not obvious at first glance because it remained almost flat (Figs. 3a, 4c, S3A, S4C). In experiment B9 (23 h; Fig. S4C), the slightly curved alignment of Au–Pd droplets visible in the glass few tens of µm above the forsterite cylinder demonstrates that the upper part of the lower cylinder has been dissolved. The Au–Pd alignment is no longer visible in the longer duration experiments, but there are still a few larger Au–Pd droplets visible either suspended in the glass or fallen on the receding forsterite surface (Fig. 4c). The dissolution of the lower cylinder is also demonstrated by the downward displacement of the upper surface compared to its initial position marked by the shrinkage of the capsule (displayed by an orange arrow on Fig. 3a). This shrinkage is due to a higher degree of compaction of glass powder compared to forsterite cylinder during the compression at the beginning of the experiment. Finally, the forsterite right below the receding surface has levels of CaO and Al₂O₃ recalling those in the starting synthetic forsterite and lower than those expected at equilibrium with the haplobasaltic melt (see below).

In our first type B experiment (B1), we used a slightly different initial configuration, including forsterite cylinders with a smaller outer diameter (2 mm instead of 2.5 mm). Figure 5a shows that this configuration was not stable and that the upper forsterite cylinder fell into the lower part of the liquid reservoir. We observe, however, the same textures as above: dissolution along the top of the lower cylinder produced a flat surface and rounded cylinder edges, and

B1 - 1425°C - 45h

Fig. 5 SEM photographs of experiment B1 (1425 °C, 45 h). **a** Overview of the sample at low magnification, showing the two cylinders at the bottom and a layer of large euhedral forsterite (with a few small spinel crystals) at the top. **b** Enlargement showing precipitation microtextures on the lower face of the upper forsterite cylinder, and

the flat dissolution surface on the top of the lower cylinder. Note the planar facets at the ends of the forsterite lobes and the melt inclusions (MI) and tubes. The orange dashed line corresponds to the position of the cross section shown in c. **c** Cross section through the growth front on the lower side of the upper forsterite cylinder

precipitation at the base of the upper cylinder yielded a fingered interface, with glass tubes separating solid lobes terminated by crystallographic facets (110) (Fig. 5b). The melt inclusions are especially well developed in this experiment, reaching up to 80 μm and showing a range of shapes from rounded, to ovoid, to negative crystals. A section perpendicular to the growth front is illustrated in Fig. 5c. On a technical point of view, it is interesting to note that experiment B1, in which the forsterite cylinders were not sputtered with Au–Pd, shows the same microtextures as in experiments with Au–Pd-sputtered forsterite cylinders. Accordingly, the precipitation and dissolution processes are not affected by the presence of droplets of gold–palladium.

Unlike the sketch in Fig. 2b, some of the liquid was able to sneak above the upper cylinder during type B experiments (and to a lesser extent under the lower forsterite cylinder).

As a result, a second dissolution–precipitation cell operated at the top of the capsule, as a parallel system. The effect is best visible in experiment B6 (Fig. 3), with the recession of the upper face of the top forsterite cylinder and the growth of large euhedral crystals of forsterite hanging from the roof of the capsule. The dissolution of the upper face left a few vertical pillars of forsterite standing several hundreds of microns above a slightly wavy floor (on the right of Fig. 3b). These special features are best visible in type A experiments and will be discussed below.

Dissolution and precipitation textures: type A experiments

A major complication arose in runs A2 (1400 °C, 176 h; Fig. 6) and A4 (1425 °C, 48 h; Fig. 7) due to the absence of

A2 - 1400°C - 176h

Fig. 6 SEM photographs of experiment A2 (1400 °C, 176 h). **a** Overview of the sample at low magnification showing its mineralogical stratification. The orange arrow shows the thickness of forsterite dissolved on the upper side of the cylinder; the kinks in the platinum wall mark the initial position of the upper surface of the forsterite cylinders before dissolution. The yellow arrow indicates the thickness of forsterite precipitated on the lower side. The orange dashed line corresponds to the position of the cross section shown in (E). **b** Close-up view of the upper part of the forsterite cylinder: pillars of residual for-

sterite stand several hundreds of microns above the receding surface; note the small grains of forsterite suspended in glass above the dissolution zone. The forsterite dendrites (white arrows) presumably grew during quench. **c** SEM micrograph (back-scattered electrons) of large euhedral forsterite in the upper part of the capsule. **d** Calcium map of the same area (with concentration scale in wt% CaO on the right). **e** Cross section through the layer of forsterite pillars and glass in **b**. *Sp* spinel, *Cpx* clinopyroxene, *Fo* forsterite, *gl* glass, *MI* melt inclusion

Fig. 7 SEM photographs of experiment A4 (1425 °C, 48 h). **a** Overview of the sample at low magnification. **b** Close-up view of the dissolution texture at the top of the forsterite cylinder, with pillars of residual forsterite almost 200 µm high. Small grains of forsterite are suspended in the glass above the dissolution zone. **c** Close-up view of the precipitation layer at the bottom of the forsterite cylinder; the layer of Au–Pd droplets is well visible. White arrows: quench overgrowths. Symbols and segments are EPMA spots and chemical profiles, respectively (see Figs. 8, 9). **d** Chemical map of magnesium in part of image (c). **e** Chemical map of magnesium in part of image (b)

connection or the poor connection between the lower face of the forsterite cylinder and its upper face. As a result, the system did not work as sketched in Fig. 2A because two independent subsystems developed in the lower half and upper half of capsules. In the lower half, the lower face of the forsterite cylinder grew at the expense of the small forsterite grains at the bottom of the capsules, giving rise to crystal-free layers right below the forsterite cylinder (Figs. 6a, 7c). Forsterite overgrowth is best visible in run A4 owing to the presence of a distinct alignment of Au–Pd droplets in the cylinder, 200 µm behind the front. In the upper half of the capsules, the upper face of the forsterite cylinder underwent dissolution (see below) and large, euhedral forsterite crystals grew at the top of the capsules (Figs. 6a, 7a). These crystals are ≈ 100–800 µm in size. They have euhedral outlines, with planar faces and sharp edges; some of them show large embayments and contain glass inclusions (Fig. 6c). Large euhedral crystals are actually present in all experiments of A and B types (Figs. 3, 4, 5, 6, 7, S3 and S4). In all cases, they are located at the top of the capsule, which is the coldest

part of the system. In experiment A2, they are not right at the top, but just below a layer of clinopyroxene + spinel + glass, which occupies the upper part of the capsule (Fig. 6a).

Type A experiments show a very special kind of dissolution texture: in experiments A2 and A4, the upper face of the forsterite cylinder displays vertical pillars standing several hundreds of microns above a slightly wavy floor (Figs. 6b, 7b). There is no doubt that these pillars represent a dissolution texture because we found some Au–Pd droplets kept at the top of the pillars or hanging on their walls (Fig. 7e; they are absent in run A2, in which the forsterite cylinder had not been sputtered with Au–Pd), and because the forsterite making the pillars has low levels of CaO and Al₂O₃, which approach the levels of the starting synthetic forsterite and which are lower than those measured in forsterite grown from the haplobasaltic melt (see below). In addition, all overgrowth layers show a very distinct alignment of entrapped Au–Pd droplets underlining the original forsterite surface (for example, sample A4 in Fig. 7c); as we do not see any Au–Pd droplets entrapped at the basis of the pillars in

Fig. 7b, the forsterite pillars are not growth textures. There is also a geometrical argument because the initial position of the upper face of the cylinder is marked by a kink in the platinum wall (Figs. 6a, 7a); the vertical pillars are well inside the original outline of the forsterite cylinders. Finally, we observe that small forsterite crystals (from the starting material) are present in the glass above the dissolution zone, but not in the dissolution zone between the pillars (e.g., the “gulfs” in Fig. 6b).

We prepared a cross section through the layer of forsterite pillars and glass of sample A2 (Fig. 6e). In the center of the sample, the pillars form numerous, sub-rounded sections isolated in glass with $\sim 35 \mu\text{m}$ diameter and a density of 254 pillars per mm^2 (Fig. 6e). Toward the rim, the forsterite increases in proportion relative to glass and it forms polyhedral sections or rods. It is important to note that the forsterite sections from all sides of the capsule display the same crystallographic orientation, which demonstrates that all these crystal units correspond to the same initial forsterite crystal before dissolution. In addition to the forsterite pillars in experiments A2 and A4, a few pillars are also observed in experiment B6 (Fig. 3a, b). Given the crystallographic orientations of the forsterite cylinders in these experiments, the pillars are all elongated preferentially along the a -axis of forsterite.

Mineralogical stratification and temperature gradient

The presence of Sp or Sp + Cpx (in addition to Fo + melt) primarily depends on the nominal temperature of the experiment (1400 or 1425 °C) and on the length of the capsule (3.1–5.1 mm; Table 4). Clinopyroxene was only found

at the top of experiment A2 (Fig. 6), which was made at 1400 °C with a capsule length $> 5 \text{ mm}$. Thus, this combination resulted in a temperature lower than 1330 °C at the very top of the capsule. A few small Sp crystals, but no Cpx, were observed in the layer of large euhedral forsterites at the top of experiments B1 and A4, indicating a temperature lower than 1385 °C but higher than 1330 °C. Again, the presence of an additional phase is related to the length of the samples (4.5 mm). In all other experiments (B6–B9), the capsule length was $< 4.3 \text{ mm}$, and the only solid phase was forsterite even when the nominal temperature was 1400 °C. If we take into account the temperature difference between the thermocouple reading and the maximum temperature at the hot spot (Watson et al. 2002), the temperature interval in these experiments was $< 35 \text{ °C}$ (Table 4).

Chemical results

Chemical compositions of forsterite are presented in Fig. 8a and Table 2. Three main populations can be identified: compositions similar to the starting forsterite, compositions in equilibrium with the haplobasaltic melt at experimental conditions and intermediate ones. The starting forsterite is characterized by a molar ratio Mg/Si of about 1.99, and negligible levels of CaO and Al_2O_3 ($< 0.02 \text{ wt\%}$). The population similar to the composition of starting forsterite has CaO and Al_2O_3 concentrations lower than 0.05 wt% and corresponds to the inner parts of forsterite cylinders, from the interface on the dissolution side to the initial forsterite–melt interface revealed by the alignment of Au–Pd droplets (Figs. 3, 7, 8a). The equilibrium forsterite composition is best represented by the small forsterite grains in experiments such as A4, which have an average Mg/Si ratio of 1.98 and concentrations of

Fig. 8 **a** Diagram CaO vs. Al_2O_3 showing the composition of forsterite in experiments A4 and B6. “Equil. (new cryst.)” corresponds to CaO and Al_2O_3 concentrations in equilibrium with the haplobasalt used here, according to partition coefficients in the literature (Libourel 1999; Pack and Palme 2003). **b** Diagram MgO vs. Al_2O_3

showing glass compositions in experiments A4 and B6. The location of analyses is shown in Figs. 3 and 7. Quench crystallization of forsterite at the end of the experiments led to glass compositions locally strongly depleted in MgO and enriched in Al_2O_3 (see also Fig. 9)

CaO and Al_2O_3 equal to ~ 0.30 and 0.15 wt%, respectively (Table 2). These relatively large concentrations are typical of forsterite in equilibrium with basaltic melts at high temperatures (Libourel 1999). For Al_2O_3 , the data in sample A4 yield a forsterite/liquid partition coefficient of ~ 0.01 , consistent with values of 0.003 – 0.02 reported by (Pack and Palme 2003) (Fig. 8a). The layers of forsterite overgrown on the initial cylinders and the large, euhedral forsterite crystals are also in equilibrium with the haplobasaltic melt.

The intermediate population of forsterite compositions is associated with the pillars in the dissolution zones, with CaO concentrations approaching the equilibrium defined above and low aluminum contents (< 0.10 wt%; Fig. 8a). In fact, there is a gradient from the inner parts of the pillars, which have very low levels of CaO and Al_2O_3 (down to 0.12 wt% and 0.03 wt%, respectively; Table 2) to the rims in contact with haplobasalt, which contain significant concentrations of these two oxides, especially CaO (Fig. S5). The low levels of CaO and Al_2O_3 in the inner parts of the pillars are a strong argument for an origin of this special texture by dissolution. The enrichment in CaO and Al_2O_3 in the outer parts is presumably due to a process of reequilibration with the surrounding melt, with CaO diffusing faster than Al_2O_3 in the forsterite crystalline lattice. The effect of solid-state diffusion can also be observed along profiles performed across the Au–Pd droplet alignments (Fig. 9a); the CaO concentration profiles measured in B6 and A4 experiments show different slopes that are consistent with the run durations. The dissolution side of forsterite cylinders is also likely to be affected by calcium diffusion up to ~ 80 μm from the melt.

Glass compositions analyzed in experiments A4 and B6 are shown in Fig. 8b. The interpretation of glass compositions is complicated by the late crystallization of forsterite during the quench, which results in a strong depletion of MgO (down to 14 wt%) and enrichment in CaO and Al_2O_3 (Fig. 8b). In fact, MgO depletion is observed over a thickness of several tens of micrometer wherever the glass is in contact with forsterite and is particularly visible in tubes on the precipitation side and in gulfs close to pillars on the dissolution side of forsterite cylinders (e.g., Fig. 7d–e). Putting aside the compositions analyzed close to forsterite, the glass contains in average ~ 19.1 wt% MgO and 17.8 wt% Al_2O_3 in sample A4, and ~ 16.8 wt% MgO and 19.5 wt% Al_2O_3 in sample B6 (Table 1). Glass B6 is close to the starting glass (16.4 wt% MgO and 19.4 wt% Al_2O_3) whereas glass A4 is strongly enriched in MgO and depleted in Al_2O_3 in comparison to the starting glass, indicating bulk forsterite dissolution during this experiment. The difference in glass compositions between A4 and B6 is due to the difference in temperature: 1400 $^\circ\text{C}$ in run B6 vs. 1425 $^\circ\text{C}$ in run A4. Accordingly, the average MgO content in melt increases by 2 – 2.5 wt% for a temperature increase of 25 $^\circ\text{C}$, that is, a variation of 0.08 – 0.1 wt% MgO/ $^\circ\text{C}$.

Fig. 9 a Chemical profiles (CaO) of forsterite in A4 and B6 vs. distance from the face of the cylinder at the beginning of the experiment (marked by Au–Pd sputtering). b Chemical profiles (MgO) in the glass in experiments A4 and B6 vs. distance. The concentration of the starting glass is indicated. See Figs. 3 and 7 for the location of the chemical profiles

The melt inclusions of experiments A2, A4 and B1 are texturally homogeneous with no crystals discerned but chemical mapping reveals a heterogeneous distribution of calcium or magnesium (Figs. 6d, 7d). Chemical gradients in glass inclusions are due to crystallization of the host forsterite during the quench. Even the cores of the glass inclusions yield compositions depleted in MgO (8.5 – 14 wt%; Table 1) in comparison to the compositions measured far from any forsterite. Accordingly, they do not reflect equilibrium conditions. Based on the compositions of forsterite, equilibrium glass composition, and glass in the melt inclusion of experiment A4 (Fig. 7a), we estimate that about 24 wt% of the melt inclusion crystallized during the quench, forming a layer of forsterite a couple of microns thick along the walls.

Rates of dissolution and growth

The growth rate (v_{Growth} , m/s) of the cylinders was determined from the maximum thickness of the growth layer (beyond the Au–Pd sputtering line) divided by run duration (Fig. 10). Similarly, the growth rate of euhedral crystals was calculated by dividing the mean size of the largest crystals

Fig. 10 Growth and dissolution rates of the forsterite cylinder plotted as a function of temperature gradient. The growth rate of large, euhedral forsterite crystals is also shown

by time. To estimate the dissolution rate, we computed the thickness of the dissolution layer as the initial cylinder thickness plus the thickness of the growth layer minus the final cylinder thickness. The thickness of dissolved forsterite can always be closely confirmed by measuring the distance between the final position of the cylinder surface and its initial position marked by the kinks of the capsule wall due to shrinkage, assuming no displacement of the cylinder. In type B experiments, only the upper cylinder was characterized so, because it gives access to both dissolution and growth. The growth rates and dissolution rates range from 5.0×10^{-10} to 1.2×10^{-9} m/s (Table 4; Fig. 10). The growth rate of large, euhedral forsterite ranges from 2×10^{-10} to 7×10^{-10} m/s.

Discussion

Thermal migration as a technique to study growth and dissolution of crystals in silicate liquids

We were able to produce regular growth or dissolution layers reaching thicknesses up to 400 μm by placing monocrystalline cylinders of forsterite into a haplobasaltic melt in a temperature gradient. Of the two experimental configurations tested (Fig. 2a, b), type B involving a layer of melt sandwiched between two forsterite cylinders is by far the more suitable for a quantitative study (Fig. 1). The main advantage is that the transport step of the dissolution/precipitation process is straightforward in comparison to type A experiments, in which the dissolved forsterite components have to find their way between the cylinder wall and the capsule. A potential improvement for the future would be to use forsterite cylinders whose faces correspond to natural faces of olivine phenocrysts. Our experiments show, however, that

similar growth and dissolution textures develop on starting surfaces with different crystallographic orientations. Sputtering a thin layer of Au–Pd over the cylinder faces is of great help to locate the initial forsterite–melt interfaces and to quantify the thickness of the growth layers. Since the same textures are observed in experiments with and without sputtering, the thin layer of gold–palladium does not seem to disturb the dissolution/precipitation processes as long as experiments are run at temperature above the melting point of Au–Pd.

As discussed in “Introduction”, thermal migration encompasses two diffusion potentials that operate simultaneously: saturation gradient chemical diffusion and Soret diffusion. To evaluate the relative importance of these two components, we computed separately the variations of melt composition arising from Soret effect and saturation gradient. Using the Soret coefficients for SiO_2 , Al_2O_3 , MgO , CaO and Na_2O determined experimentally for a MORB composition (Leshner and Walker 1988; Walker and Agee 1988), the composition of melt in experiment B6 (16.81 wt% MgO ; Table 1), and considering two points separated by 1 $^\circ\text{C}$, we obtain a variation of MgO equal to 0.45×10^{-3} in molar fraction (or a variation of 0.03 wt% MgO). This is almost three times smaller than the variation of MgO due to saturation: 1.19×10^{-3} in mole fraction per $^\circ\text{C}$ (based on the difference in liquid compositions in the experiments at 1400 and 1425 $^\circ\text{C}$, B6 and A4, respectively). Accordingly, the dominant process in our thermal migration experiments is saturation gradient chemical diffusion, but Soret effect is not negligible.

The total flux, J (in moles per unit area per unit time), associated with thermal migration is (Leshner and Walker 1988; Walker and Agee 1988)

$$J = -D \frac{dc}{dz} - D' c \frac{dT}{dz}, \quad (1)$$

where the first term on the right-hand side is the flux arising from saturation gradient chemical diffusion, and the second term is the flux arising from the temperature gradient alone. Parameters are as follows: D (cm^2/s), chemical diffusivity; D' ($\text{cm}^2/\text{s}/^\circ\text{C}$), thermal diffusion coefficient; c (mol/cm^3), solute concentration; z (cm), position along the diffusion axis; T ($^\circ\text{C}$), temperature. Introducing the Soret coefficient $\sigma = D'/D$ ($^\circ\text{C}^{-1}$), Eq. (1) can be rewritten:

$$J = -D \frac{dc}{dz} + \sigma c D \frac{dT}{dz}, \quad (2)$$

where $[dc/dT]_{\text{Liq}}$ is the variation of concentration with temperature along the liquidus. Equation (2) shows that the flux J is directly proportional to the temperature gradient and is constant as long as the saturation relations and temperature remain unchanged (Leshner and Walker 1988; Walker and Agee 1988).

In our experimental configuration (Fig. 1), there is a direct relationship between the upward flux of MgO by thermal migration, J_{MgO} , and the growth rate of the upper forsterite cylinder:

$$v_{\text{Growth}} = \frac{1}{2} J_{\text{MgO}} V_{\text{Fo}}, \quad (3)$$

where v_{Growth} is the growth rate (cm/s) and V_{Fo} is the molar volume of forsterite ($\approx 46 \text{ cm}^3/\text{mol}$ in our experimental conditions; (Li et al. 2007)). Thus, if as expected, the flux is time independent, then the growth rate of forsterite remains constant during a thermal migration experiment. Using the relation $c = mp/M$, where m is the mass fraction of an oxide, M its molar mass, and ρ is the liquid density (assumed constant: 2.7 g/cm^3), we can recombine (1) and (3) into

$$\frac{2v_{\text{Growth}}}{V_{\text{Fo}}} = -\frac{D_{\text{MgO}} \rho}{M_{\text{MgO}}} \left(\frac{dm_{\text{MgO}}}{dz} + \sigma_{\text{MgO}} m_{\text{MgO}} \frac{dT}{dz} \right). \quad (4)$$

For experiment B6, we have $v_{\text{Growth}} = 9.4 \times 10^{-8} \text{ cm/s}$, $m_{\text{MgO}} = 0.1681$, $dm_{\text{MgO}}/dz = -0.05 \text{ cm}^{-1}$ (Fig. 9b), $dT/dz = -58 \text{ }^\circ\text{C/cm}$ (Table 4); the Soret coefficient σ_{MgO} is equal to 0.00239 (Leshner and Walker 1988; Walker and Agee 1988). Thus, we obtain a chemical diffusivity D_{MgO} equal to $7.8 \times 10^{-7} \text{ cm}^2/\text{s}$, which is in good agreement with the chemical diffusivity estimated from previous thermal migration experiments run at conditions close to ours: $D \approx 10^{-6} \text{ cm}^2/\text{s}$ (Walker et al. 1988).

Dissolution and growth of forsterite in a temperature gradient

The eight experiments of this study yielded qualitatively similar dissolution and growth textures: a planar or slightly wavy forsterite–melt interface, residual pillars (mostly in type A experiments), and rounded cylinder edges in dissolution areas; fingered forsterite–melt interfaces with melt tubes and melt inclusions in the growth areas. We, therefore, conclude that these textures are independent of temperature over the small range investigated (1400–1425 $^\circ\text{C}$; Table 3), forsterite over glass ratio (in the range 0.25–0.83; Table 3), and run duration (no qualitative difference between experiments A2 and A4 conducted for 176 and 48 h, respectively).

Concerning the dissolution textures, the smooth melt–crystal interfaces and rounded cylinder edges produced here are similar to textures described in previous experimental work (Zhang et al. 1989; Donaldson 1990; Soulié et al. 2017). The pillars represent, however, an original texture undocumented in the literature to our knowledge, and do not follow the rule that olivine dissolution produces almost perfect planar interfaces (Zhang et al. 1989; Chen and Zhang 2008). We suggest the following scenario. At the beginning of the experiment, the upper face of the cylinder

was at the level of the tops of the pillars. Because of its higher temperature relative to the upper part of the capsule, it began to dissolve and recede. With time, the original face retreated several hundred micrometers except in areas that were somehow protected from dissolution and that gave rise to the vertical pillars. As the most characteristic pillars are found in runs A2 and A4, the presence of suspended crystals above the forsterite cylinder is presumably the reason why some areas have remained immune to dissolution. As a matter of fact, a number of pillars are capped by a small forsterite grain (at the center of Fig. 6b, or on the right side of Fig. 7b), and thus resemble the so-called fairy chimneys. In contrast, type B experiments, in which there were no crystals in suspension above the forsterite cylinder, show dissolution surfaces almost perfectly planar with only rare pillars or no pillars at all.

Most pillars are robust structures: their length increases with time (by dissolution of the neighboring forsterite) since they are best developed in the longest duration experiment (A2). Some pillars may, however, become unstable with time. For instance, there are a few short pillars in Fig. 6b that may be in the process of resorption. At this stage, our experimental database is not sufficient to establish any systematic relationship between the width or the spacing of the pillars and the experimental parameters (time, temperature, crystal orientation).

Melt composition on the dissolution and precipitation sides of forsterite cylinders

During the experiments, the melt in contact with forsterite is anticipated to be slightly enriched in MgO and depleted in CaO and Al_2O_3 (in comparison to the average melt composition) on the dissolution side of forsterite cylinders, and slightly depleted in MgO and enriched in CaO and Al_2O_3 on the precipitation side. We do not have access to the equilibrium glass composition at the contact with forsterite due to the effect of quench crystallization. To circumvent this difficulty, we acquired three EPMA profiles in experiment B6 (FF', GG', and HH'; Fig. 3a and Online supplementary Table). Profile GG' is a nearly horizontal profile in the melt layer between the two forsterite cylinders, and is rather flat (Fig. 9b). Profiles FF' and HH' are vertical and extend from a zone of forsterite growth to a zone of forsterite dissolution. If we ignore the effect of quench crystallization (well visible for instance close to F), we observe that the MgO content in the glass increases very slightly with a gradient of about 0.5–0.6 wt% MgO per mm from the growth side to the dissolution side (Fig. 9b). Due to the effect of quench crystallization, it is not possible to know the exact MgO levels at the contact with the two forsterite cylinders, but the difference in MgO between the dissolution and growth sides is of the order of 0.35 wt%. Considering a variation of 0.08

to 0.1 wt% MgO/ °C for melt in equilibrium with forsterite (see above), this difference of 0.35 wt% MgO corresponds to a temperature difference $\Delta T \approx 4$ °C, in good agreement with the temperature gradient in experiment B6 (5.8 °C/mm; Table 4) and the thickness of the melt layer (640 μ m). Accordingly, the well-developed growth layer on the lower face of the upper forsterite cylinder and the thick dissolution layer on the upper face of the lower cylinder formed at degrees of undercooling and superheating, respectively, of ≈ 2 °C ($\Delta T/2$).

Cellular growth of olivine in basaltic melts

During the solidification of a melt, solid–melt interfaces become unstable with increasing growth rate, evolving from planar, to cellular, to dendritic. Cellular growth, which occurs at low to intermediate growth rates, is well known in metallurgy (Tiller et al. 1953; Shechtman et al. 1984), but poorly documented in silicate systems and natural magmas. Vermiform rims on spinel in natural basalts were interpreted as the result of a cellular growth mechanism, when diffusion in the melt is no longer able to support the growth of a smooth interface, creating protuberances (Roeder et al. 2001). Takei and Kobayashi suggested that cellular growth was responsible for the shape and distribution of minute gas bubbles in the core of large forsterite monocrystals grown under a cooling rate of 300 °C/h (Takei and Kobayashi 1974). With the exception of this reference, cellular morphology is absent from the catalog of olivine shapes and from the classical sequence from polyhedral, tabular, skeletal to dendritic shape with increasing degree of undercooling and cooling rate (Donaldson 1976; Faure et al. 2003a).

Our experiments show that cellular growth may be a major growth mechanism for olivine at low to moderate degrees of undercooling (of the order of 2 °C in experiment B6, for instance) and growth rates of $\approx 10^{-9}$ m/s. The textures produced support the breakdown of a planar forsterite–melt surface into a series of lobes, but we could not relate the number, frequency or shape ratio of lobes to any experimental parameter. We note, however, that cellular growth occurs on surfaces with a lateral extension greater than 1 mm (1.1 mm in experiment A4, and about 2 mm in most cases). In comparison, the large euhedral forsterites whose faces have a lateral extension of at most 300 μ m, and more commonly of the order of 100 μ m, display a planar morphology (Fig. 3b). This contrast is consistent with the average spacing of lobes (≈ 100 μ m). Therefore, cellular growth seems to require a minimum lateral extension of the olivine–melt interface of the order of 1 mm. The formation of a boundary layer enriched in the incompatible elements rejected by the growing crystal and not able to diffuse away fast enough could locally impede or slow down the crystallization and, beyond a certain value of the growth rate, cause

the switch from a planar growth front to a cellular morphology (Jackson 2004).

The rates of dissolution and crystallization of forsterite cylinders and euhedral forsterites are limited to a narrow range (4×10^{-10} – 1.4×10^{-9} m/s; Table 4). Those values are very close to the growth rate observed for olivine with an undercooling of 15 °C (2×10^{-9} m/s) and to the one assessed for lower undercooling values [6.3×10^{-10} m/s; (Jambon et al. 1992)]. They fit into the range of natural growth rates determined for olivines from Etna basalt [2×10^{-11} m/s; (Armienti et al. 1994)] and from Lanzarote [$\sim 3 \times 10^{-7}$ m/s; (Armienti et al. 1991)] without and with a 30 °C undercooling, respectively, and for olivine from an oceanic basalt quickly chilled against sediments [3.2×10^{-9} m/s; (Donaldson 1975)], although these values were determined by postmortem observation of the crystal size distribution. They are slower by half an order of magnitude than the growth rates measured *in situ* by (Ni et al. 2014) on olivine. In contrast, the growth rates in our study are similar to the ones measured during transcrystalline migration of melt inclusions within olivine at very small degrees of undercooling (< 1 °C): 3×10^{-10} to 3.2×10^{-9} m/s (Schiano et al. 2006). Both our experiments (Fig. 5b) and transcrystalline melt migration experiments produced (001) faces that are characteristic of a polyhedral olivine habit at very low degrees of undercooling (Deer et al. 1962). Morphologies with (001) faces are also observed on olivine dendrites re-equilibrated under near equilibrium conditions (Faure et al. 2003b). The slow growth rate, the absence of marked chemical gradients at crystal–liquid interfaces and the presence of (001) faces are evidence of very low degrees of undercooling during experiments.

Faure et al. retained four main morphologies of olivine among the ten described by Donaldson (1976): polyhedral, tabular, hopper and dendritic (Faure et al. 2003a). Those morphologies depend on the type of growth mechanism, steady-state vs. rapid growth mechanisms, which depends, in turn, on the cooling rate and the degree of undercooling (Faure et al. 2003a). Here, we show a new type of growth mechanism, cellular growth, and new growth morphologies that have not been described in previous studies, in which olivine growth was investigated using cooling experiments (Donaldson 1975, 1976; Coish and Taylor 1979; Kirkpatrick 1981; Lofgren 1983; Jambon et al. 1992; Faure et al. 2003a; Faure and Schiano 2005; Welsch et al. 2012; Ni et al. 2014). Compared to classical cooling experiments, our thermal migration experiments allow to combine two parameters that are required for the occurrence of cellular growth and for the development of cellular morphologies: low degrees of undercooling (estimated at 2 °C in experiment B6) and large forsterite–melt interfaces (typically 2 mm in diameter). Based on our experiments, the classical degree of undercooling vs.

Fig. 11 SEM images showing microtextures suggestive of cellular overgrowth in natural olivine phenocrysts from the basanitic tephra of **a** Montcineyre volcano (Chaîne des Puys, France), **b** Souilhol volcano (Ardèche, France), and **c, d** Thueyts volcano (Ardèche). The core of the crystal in **b** is a xenocryst of MgO-rich composition, which is completely surrounded by lobes of olivine in equilibrium with the basanitic matrix. The crystallographic orientation is indicated in pink font

cooling rate vs. olivine morphologies scheme (Faure et al. 2007) needs to be completed to include cellular growth of olivine. Integrating the new data with previous ones, we are able to link growth texture in olivine to growth rate: polyhedral, tabular, and cellular morphologies occur at crystallization rates of $\approx 10^{-9}$ m/s and below, while hopper and dendritic morphologies are generated for higher growth rates ($\approx 10^{-8}$ m/s and $> 10^{-8}$ m/s, respectively) (Faure et al. 2007; Ni et al. 2014).

Natural occurrences of cellular growth in magmas

The main goal of our thermal migration experiments was to access the low degrees of undercooling characteristic of most magmatic systems. Under conditions pertaining to natural systems (low degrees of undercooling and large phenocryst faces), our major finding is the development of cellular morphologies. To date, cellular growth has been poorly documented in the literature, with few natural examples on pyroxene, plagioclase and spinel (Singer and Pearce 1993; Andersson and Eklund 1994; Hibbard and Sjöberg 1994; Roeder et al. 2001). However, we believe that cellular morphologies are much more frequent and may not have been detected in the past. For instance, we found microtextures that are suggestive of cellular growth in olivines from the basanitic tephra of Montcineyre (Chaîne des Puys, France; Fig. 11a) and Souilhol and Thueyts volcanoes (Ardèche, France; Fig. 11b–d). The olivine phenocrysts are a few mm long at Montcineyre, and between 0.5 and 1 mm at Souilhol and Thueyts. Most olivine phenocrysts in these basanites are characterized by a discontinuous outer part showing lobes or blocks of growth separated by depressed areas or embayments: the result is a crenulated outline. Olivine xenocrysts, which are frequent in these basanites, show the same morphology at their periphery; the MgO-rich olivine core in Fig. 11b is completely surrounded by lobes of olivine in equilibrium with the basanitic matrix. Small melt inclusions are found in the rim of some olivines from Thueyts (Fig. 11d), yielding microtextures reminiscent of a process of cellular growth in which neighboring lobes merged at their ends and entrapped melt droplets at their roots. In the examples of Fig. 11, the final stage of olivine growth was frozen by the eruption, allowing the preservation of what we interpret as textures of cellular growth.

Constraints on melt inclusion formation

A possible consequence of crystal growth is the entrapment of melt inclusions. With the experimental configuration used here, we could test for the necessity of a temperature decrease to form melt inclusions, such as suggested by Goldstein and Luth (2006) who produced inclusions at cooling rates higher than 1 °C/h only. Here, we generated melt inclusions resembling natural ones in the absence of temperature variations, but with temperature gradients of a few degrees per mm which drove olivine growth under low degrees of undercooling. Therefore, melt inclusions can be formed in steady P–T systems at low degrees of undercooling; their formation does not require a specific cooling regime or rapid crystallization (skeletal of dendritic growth) as previously proposed (Albarede and Bottinga 1972; Roedder 1979; Watson 1996; Schiano 2003; Kohut and Nielsen 2004; Faure and Schiano 2005; Goldstein and Luth 2006; Faure et al. 2007). Cellular growth could favor the formation of melt inclusions by giving rise to melt tubes that will subsequently get closed at their ends (Higuchi et al. 1995; Nakamura and Shimakita 1998; Colin et al. 2012). This process is well illustrated by the merging of neighboring lobes terminated by crystallographic facets in experiment B6 (Fig. 3c), and closely resembles the mechanism described by Colin et al. in skeletal olivine crystals found in a MORB (Colin et al. 2012). However, we do not claim that our experiments represent the unique or the most likely way of making inclusions, and there is natural and experimental evidence that melt inclusions form under varying pressure and/or temperature (Kohut and Nielsen 2004; Faure and Schiano 2005; Faure et al. 2007; Manzini et al. 2017). Cellular growth is another mechanism to generate melt inclusions hosted in polyhedral forsterite, in addition to the interaction between spiral growth dislocations (Faure and Schiano 2005), partial dissolution of host mineral followed by recrystallization (Anderson 1991; Nakamura and Shimakita 1998; Welsch et al. 2014; Manzini et al. 2017), dissolution of preexisting crystals or impurities (Davey and Mullin 1974; Michael et al. 2002), or fluctuation of growth rate (Roedder 1979).

Conclusions

We used an original experimental setup based on thermal migration to monitor the growth of large forsterite cylinders (single crystals) at low degrees of undercooling (≈ 2 °C). As forsterite solubility is sensitive to temperature, the temperature gradient within the capsules generated a dissolution–precipitation process involving (1) dissolution of forsterite on the high-temperature side (giving rise to rounded crystal–melt interfaces and pillars of residual forsterite); (2) transport of dissolved components in the melt;

(3) precipitation on the low-temperature side (giving rise to thick layers of newly grown forsterite with a fingered interface and melt inclusions, and to the growth of large, euhedral crystals of forsterite). The starting forsterite cylinders were sputtered with Au–Pd allowing us to mark the initial melt–forsterite interface and to quantify crystal growth.

The fingered crystal–melt interfaces observed on the crystallization side of forsterite cylinders result from cellular growth, a type of growth that had not been produced experimentally so far in silicate systems. Our experiments show that cellular growth requires a low degree of undercooling (a few °C) and a large substrate (~ 1 mm across or more), and that it occurs at growth rates of the order of 10^{-9} m/s. Cellular growth can lead to the formation of melt inclusions at relatively slow rates of growth. Although we found microtextures suggestive of cellular growth in olivines from mafic rocks of the French Massif Central, cellular textures are poorly documented in natural products, probably because they were not recognized as such.

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