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**Eruption dynamics inferred from microlite crystallization experiments:
Application to Plinian and dome-forming eruptions of Mt Pelée
(Martinique, Lesser Antilles).**

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ABSTRACT

Decompression experiments have been conducted to simulate syn-eruptive crystallization in the volcanic conduit, in order to infer magma ascent rates and conditions during dome-forming and Plinian eruptions of silicic arc volcanoes. The experiments were carried out starting from Mt Pelée rhyolitic interstitial melt (76 wt. % SiO₂) and consisted in three consecutive steps: hydration, decompression, and annealing. Hydration (saturated and undersaturated) was performed at 850°C and 200 MPa and followed by isothermal decompression, either linearly or stepwise, to a final pressure, P_f , of 30 or 5-10 MPa. Decompression rates range from 0.003 to 25 MPa/min (decompression durations of 15 min to 40 days). Two samples were cooled by 25°C and 50°C during a 3-days step at P_f . Subsequent to decompression, the samples were held up to 15 days at P_f . The experiments generated three types of crystals: pre-, syn-, and post-decompression crystallization. The experiments basically differ from previous studies in that they are specifically designed to discriminate crystal nucleation from growth and to evaluate the influence of pre-decompression crystals on the decompression-induced crystallization.

The effects of pre-decompression crystals, decompression rate, undercooling (P_f), and terminal cooling have been determined on plagioclase nucleation, growth, morphology, and composition. The main results i) suggest a positive correlation between decompression rate and the number density of plagioclases nucleated at P_f and ii) highlight the effect of pre-decompression crystals in further decompression-induced crystallization.

The relations between the decompression conditions and the plagioclase characteristics have been used to infer Mt Pelée eruption dynamics, suggesting that i) Plinian magmas ascend from the reservoir within less than 1 hour (1-10 m/s), ii) dome and block-and-ash flows magmas ascend within more than 2-5 days, giving time for syn-decompression crystallization

around pre-existent microlites, iii) dome magmas evidence long stagnation and cooling at low pressure, and iv) surge magmas ascend without significant crystallization (within less than ~4 days) and massively nucleate plagioclase at very low pressure. The extent and violence of dome destruction may depend on the size/age of the dome, with large/old domes favouring mildly-explosive BAFs, whereas small/young protodomes may generate highly-explosive surges.

Keywords: microlite crystallization; experimental petrology, decompression rate; eruptive style; Mt Pelée;

INTRODUCTION

Arc volcanism is characterized by a wide variety of eruptive styles, from Plinian to dome-forming eruptions (e.g. block-and-ash flows and surges), with drastically different consequences in terms of risks for local population and environment. For volcanoes emitting small volumes ($< 1\text{-}3 \text{ km}^3$) of H_2O -rich magmas, such a variety of eruptive behaviours have been ascribed to different magma ascent rates (Jaupart & Allègre, 1991; Woods & Koyaguchi, 1994) rather than differences in the magma storage conditions or pre-eruptive melt volatile contents (Martel et al., 1998). Magma rise rate affects the eruption behaviour by constraining the time interval available for processes such as degassing, crystallization, and fragmentation. Thus, there is a constant competition between the time scale of magma ascent and the kinetics of the magma processes. Yet, we lack constraints on magma ascent rate and residence time in the volcanic conduit, which may be mainly ascribable to the difficulty in interpreting the seismic signals in terms of magma movements and upward migration (Kilburn, 2003; Roman & Cashman, 2006).

Rates at which magmas rise to the surface have been assessed by indirect methods such as kinetic studies of volatile exsolution, degassing-related mineral destabilisations, and xenolith transport. Based on these different studies, Rutherford & Gardner (2000) proposed average ascent rates for silicic magmas of the order of 1-15 mm/s for dome-forming eruptions and from several m/s to sonic velocities for Plinian events. Nevertheless, magma ascent rates for the 1980, May 18th, Plinian eruption of Mount St Helens, USA, have yielded quite different values, depending on the method used: ~3 m/s from mass eruption constraints (Geschwind & Rutherford, 1995), ~15 m/s from numerical models (Papale et al., 1998), and 37-64 m/s from water diffusion profiles in melt inclusions (Humphreys et al., 2008). This highlights the need

to better constrain magma ascent rates to accurately link the eruptive behaviour to the magma processes.

Magma ascent rate can be inferred from the kinetics of the syn-eruptive magmatic processes occurring in the volcanic conduit, such as degassing or crystallization. Timescales of decompression-induced bubble nucleation and growth in silicic H₂O-rich magmas are rather short (< hours) (Mourtada-Bonnefoi & Laporte, 1999, 2004; Gardner et al., 1999; Martel & Schmidt, 2003; Toramaru et al., 2008) and are de facto propitious to investigations of rapid magma ascents, such as during Plinian eruptions. However, for the longer magma ascent durations prevailing during dome-forming eruptions, ascent rate must be investigated through processes with longer timescales, such as crystallization.

Crystal nucleation and growth in H₂O-rich melts are driven by undercooling, either by isobaric cooling or isothermal decompression. In a realistic case of magma ascent in a volcanic conduit, crystallization is best driven by adiabatic decompression. Depressurizing a H₂O-saturated melt at constant temperature leads to a decrease of its dissolved H₂O content (following H₂O solubility laws) and a subsequent increase of the liquidus temperature of the crystallizing anhydrous minerals (Tuttle & Bowen, 1958). Thus, the resulting temperature difference between the liquidus and the magma, referred as to the degree of effective undercooling, ΔT_{eff} , increases with decompression. ΔT_{eff} controls rates of crystal nucleation and growth, which determine crystal morphology, volume fraction, and number density (Lofgren, 1980; Hammer & Rutherford, 2002; Faure et al., 2003). Decompression-induced crystallizations often proceed out of equilibrium, leading to high number densities of crystals with small sizes (length < 100 μm), referred as to microlites, often showing hollow or tortuous morphologies with overgrowths. Therefore, investigating nucleation and growth rates during decompression provides diagnostics on the crystallization conditions of natural microlites.

Several studies aiming at correlating textural and compositional microlite characteristics with ΔT_{eff} and ascent rates have been conducted: (i) on natural tephra (Cashman 1992; Geschwind & Rutherford, 1995; Hammer et al., 1999, 2000; Nakada & Motomura 1999; Castro & Mercer 2004; Noguchi et al., 2006, 2008; Martel & Poussineau 2007; Clarke et al., 2007), (ii) through decompression experiments (Hammer et al., 2000; Hammer & Rutherford, 2002; Couch et al., 2003; Martel & Schmidt, 2003; Suzuki et al., 2007; Brugger & Hammer, 2010; Cichy et al., 2011), and (iii) via numerical modelling of magma ascent in volcanic conduits (Papale & Dobran 1994; Melnik & Sparks 1999, Melnik et al., 2005, 2011). Most of the natural and experimental studies focused on a particular eruptive behaviour of a given volcano, but little has been devoted to the comparison and the transitions between effusive and explosive events (Castro & Gardner, 2008). Attempts have been made to interpret groundmass textures as magma geospeedometer by linking microlite number density with magma discharge rate (Blundy & Cashman, 2008). This provides useful information on magma ascent conditions, but turned out not to be precise enough for volcanoes, whereof conduit geometries are unconstrained or syn-eruptive crystallization is perturbed by pre-decompression microlites or microphenocrysts.

The present paper is meant to infer the magma ascent conditions in silicic volcanoes by investigating syn-eruptive crystallization down to the least details and evaluate the impact on the eruption regime. Such a detailed study required a target volcanic system particularly well-known in terms of eruption phenomenology, pre-eruptive conditions, and characterization of the natural products, which turn out to be fulfilled conditions at Mt Pelée. The present study is specifically worked out to consider (i) the large range of Mt Pelée eruption styles, from domes, block-and-ash flows, surges, to Plinian events, (ii) the syn-eruptive crystallization conditions through the presence or absence of pre-decompression microlites, and (iii) a systematic feedback between the experimental and natural data.

MT PELÉE: RECENT ACTIVITY, MAGMA STORAGE CONDITIONS, AND MICROLITE CHARACTERISTICS

Main eruptive events

Recent period (13 500 years BP to present) of edifice building at Mt Pelée has been characterized by dome-forming and Plinian eruptions of an andesitic magma (Westerkamp & Traineau, 1983; Smith & Roobol, 1990).

The youngest volcanic activity is dated to 1929-32 and was witnessed and well-documented by Perret (1935). The eruption consisted in dome gravitational destabilizations into block-and-ash flows (BAFs; nuées ardentes of Merapi type) and ended with the extrusion of the present dome.

The previous dome-forming events occurred in 1902-1904 and were first reported by Lacroix (1904). The eruptions similarly began with growing domes, the destabilization of which produced high-energy and high-velocity surges from 1902, May 8th to August 30th (nuées ardentes of pelean type). From Lacroix's data, Tanguy (2004) estimated the 1902-1905 dome volume and its growth rate at various stages of building. In particular, during the week preceding the 1902, May 8th catastrophe, dome growth was between 28 and 38 m³.s⁻¹, leading to a volume of 17-23x10⁶ m³. Considering that significant parts of the dome (~1/3?) were removed by the May 8th and 20th climactic surges, a high magma flux could have continued until the end of May. From November 1902 to July 1903, most of the effusive activity was concentrated in the great spine (erupted volume ~15x10⁶ m³, magma flux of 1.2 m³.s⁻¹), which eventually collapsed into minor pyroclastic flows. The eruption ended by a dome extrusion at a very slow effusion rate < 1 m³.s⁻¹ from August 1903 to October 1905 (Tanguy, 2004).

The youngest Plinian event of Mt Pelée is P1 eruption in the stratigraphy nomenclature of [Westerkamp & Traineau \(1983\)](#), dated to 650 years BP. This eruption began with one or two surge events of pelean type ([Boudon et al., 1994](#)) that were directly followed by pumice fallout.

The eruptions from the recent period of Mt Pelée activity emitted constant and rather low volumes of tephra, i.e. $\sim 0.1 \text{ km}^3$ dense rock equivalent. The behaviour changes within a single eruptive event (surge to Plinian, Plinian to dome) offer the rare opportunity to study the transition between effusive and explosive activity.

Magma storage conditions

The recent activity of Mt Pelée tapped an andesitic magma ($58 < \text{SiO}_2 < 65$ wt. %, 61-62 wt. % in average) that contains ~ 47 vol. % of phenocrysts (29-49 vol. % plagioclase, 4-9 vol. % orthopyroxene, 1-2 vol. % magnetite, minor (< 1 vol. %) clinopyroxene, ilmenite and apatite, destabilized amphibole and olivine; [Gourgaud et al., 1989](#); [Martel et al., 1998](#)) embedded in a rhyolitic matrix glass (74-77 wt. % SiO_2 ; [Martel et al., 2000](#)). The range of pre-eruptive storage conditions proposed for the upper andesitic reservoir is $875_{\pm 25}^\circ\text{C}$, $200_{\pm 50}$ MPa, an oxygen fugacity (f_{O_2}) between 0.4 and 0.8 log unit above the Nickel-Nickel Oxide buffer (NNO), and melt H_2O contents of 5.3-6.3 wt. % ([Martel et al., 1998](#)). There is evidence for magma mixing at macroscopic scale through mafic enclaves (51-59 wt. % SiO_2) and banded rocks in the 1902-1929 products ([Fichaut et al., 1989](#); [Gourgaud et al., 1989](#)). At microscopic scale, magma mixing is also evidenced by ubiquitous high-Ca plagioclase microlites interpreted as crystallizing from a mafic melt intruding the andesitic reservoir ([Martel et al., 2006](#)).

Microlite characteristics

The presence of inherited microlites in the magma chamber may play a role during magma ascent in the conduit, by providing preferential sites for further syn-eruptive crystallization from the interstitial rhyolitic melt. In subduction zones, andesitic to rhyolitic liquids are particularly rich in calcium and mainly crystallize plagioclase. The recent study on the natural microlites of Mt Pelée andesite (Martel & Poussineau, 2007) suggests that the chemical compositions of the decompression-induced plagioclase microlites poorly discriminate between the eruption styles, as they mainly range between 35 and 55 mol. % of anorthite (An_{35-55}). In contrast, decompression-induced microlites from the different eruptive styles show distinct textural characteristics, such as plagioclase area fraction (Φ), number density (N), and morphologies (see following section "Textural analysis" for definitions) (Figure 1). The whole population of 1929-1932 BAFs clasts displays very heterogeneous groundmass textures, with nearly microlite-free vesicular clasts to microlite-rich dense clasts (Martel et al., 2000; Martel & Poussineau, 2007). However, the dominant texture is characterized by low N ($< 50\,000\text{ mm}^{-2}$) and low Φ (< 0.25) of large tabular to skeletal plagioclase microlites. In surge deposits, microlites are small skeletal to dendritic plagioclases, with high N (100 000 to 250 000 mm^{-2}) and high Φ (0.2-0.3). Plinian pumices are dominantly microlite-free, but sometimes contain very small dendritic microlites characterized by low Φ (< 0.15) but high and variable N (40 000 to 200 000 mm^{-2}). Dome groundmass textures are too complex to be properly characterized, because a pervasive silica phase precludes clear microlite demarcation. Dome microlites are large and tabular, the textural rough estimations of which suggest high Φ (> 0.30 , up to 0.60) and low N ($< 50\,000\text{ mm}^{-2}$).

The aim of the present work is to reproduce the eruption-related domains of Mt Pelée groundmass textures (N , Φ , composition, size and morphology of the plagioclase microlites)

through decompression experiments simulating crystallization during magma ascent and dome emplacement.

EXPERIMENTAL AND ANALYTICAL METHODS

Starting material

The starting material is a synthetic composition corresponding to the rhyolitic microlite-free interstitial glass of the P1 Plinian pumices (Martel et al., 2000). The glass was prepared by mixing appropriate amounts of dry powders of Si, Al, Fe, Mg, Mn, and Ti oxides with Ca, Na, and K carbonates. The powder mixture was heated to 1000°C in an iron-saturated Pt-crucible at 1 atm for 30 min to decompose the carbonates. Two cycles of heating at 1600°C for 3 h (rapidly quenched in water) and glass fine grindings in an automatic mortar for 1 h were proceeded, in order to obtain a bubble- and crystal-free homogeneous dry glass powder. Electron microprobe analyses of this glass give the following average composition in wt. %: SiO₂: 75.7, Al₂O₃: 13.1, Na₂O: 3.6, FeO: 2.5, CaO: 2.4, K₂O: 1.9, MgO: 0.4, TiO₂: 0.3, and MnO: 0.1, leading to a CIPW norm of Qz₃₈Ab₃₃An₁₂Or₁₁Hy₅Ilm₁ (Quartz, Albite, Anorthite, Orthoclase, Hypersthene, ilmenite). Amounts of 20 to 50 mg of this glass powder were added to 0.5 to 3.9 mg of deionized H₂O to reach H₂O-undersaturated to oversaturated conditions at the required pressures.

The H₂O-glass mixtures were sealed into Au capsules (either 2.5- or 4.0-mm inner diameter, 20 to 30 mm long) by arc welding. The capsules were weighed before and after welding to ensure watertight sealing. The H₂O-saturated glasses at 200 MPa and 875°C contain hydration bubbles in number densities of $\sim 10^2$ mm⁻² and with diameters of ~ 10 μm. Although initial bubbles of ~ 1 μm have been recognized as providing preferential sites for

crystal nucleation in a lithium disilicate melt (Davis & Ihinger, 1998), there is no obvious spatial relation between these initial bubbles and the plagioclase crystals, maybe because their number density is 1 to 4 orders of magnitude lower than the nucleating plagioclases. Moreover, H₂O has been added within similar proportions, so that the extent of H₂O-oversaturation, and thus the amount of initial bubbles, may not significantly differ from a sample to another, and eventually not influence the experimental results.

Experimental device

Experiments requiring temperatures < 875°C were performed in horizontally-working cold-seal pressure vessels (CSPV) made of Inconel 100 (Ni-steel alloy), pressurized either with pure argon or water. The vessels were calibrated under pressure in order to determine 3-cm long hotspots free of temperature gradient. The sample capsule and a Ni-Steel alloy filler rod (80 mm) to reduce gas convection were introduced into the vessel, pressurized to target pressure and then placed at the furnace hotspot (pressure was continuously released to maintain it to target during heating). Pressure was measured using a pressure transducer calibrated against a Heise Bourdon gauge (accuracy of ± 0.2 MPa) and temperature was measured by K-type thermocouples (precision of $\pm 3^\circ\text{C}$). f_{O_2} is either close to NNO+3 or +4 where pressurized with Ar (buffered by the intrinsic vessel composition and also testified by the presence of magnetite + hematite in the isobaric runs) or close to NNO+1 where pressurized with H₂O (which provides a H₂ source for sample reduction and buffering; also testified by the presence of magnetite + ilmenite in the isobaric runs). The sample was rapidly quenched by inverting the vessel which makes the capsule and the filler rod fall into the water-cooled part of the vessel (< 50°C) within 1-2 s.

Experiments requiring temperatures $\geq 875^\circ\text{C}$ were carried out in a vertically-working internally-heating pressure vessel (IHPV) pressurized with pure argon and equipped with a

rapid-quench device consisting in dropping the sample in the cold part of the vessel ($\sim 50^\circ\text{C}$) by fusing the Pt-wire that was holding the sample. Pressure was measured using a pressure transducer calibrated against a Heise Bourdon gauge (accuracy of ± 0.2 MPa) and temperature was measured by K-type or S-type thermocouples (precision of $\pm 3^\circ\text{C}$).

Experimental strategy

Phase equilibrium runs

A set of equilibrium isobaric crystallization runs was performed between 5 and 200 MPa, in order to be used as a framework to interpret the dynamic decompression experiments. The run parameters of the phase equilibria have been chosen to cover the conditions prevailing in Mt Pelée volcanic conduit. Pressures have been varied from 200 MPa (upper magma chamber; [Martel et al., 1998](#)) to 5 MPa. Temperature ranges from 800 to 875°C . In absence of values for the redox state prevailing in the conduit, the experiments were performed at NNO+1 (close to reservoir redox state; [Martel et al., 1998](#)) and NNO+3 (as H_2O -bearing magmas show a systematic f_{O_2} increase during ascent, by as much as 2 log units; [Burgisser & Scaillet, 2007](#)). All the experiments, but one, are H_2O saturated (i.e. 5.8 wt. % H_2O at 200 MPa and 850°C , calculated after the H_2O solubility model of [Liu et al., 2005](#)). The H_2O -undersaturated run was performed at 200 MPa, 850°C , and contains 5.6 wt. % H_2O ([Liu et al., 2005](#)). Run duration ranges between 2 and 10 days, depending on pressure and temperature, in order to ensure complete hydration and crystallization ([Hammer & Rutherford, 2002](#); [Martel & Schmidt, 2003](#)).

A set of isobaric equilibrium runs was performed at temperature $> 850^\circ\text{C}$ in the pressure range of 5-200 MPa, aiming at determining the H_2O -saturated liquidus temperature of the plagioclase that is required for the estimation of ΔT_{eff} .

Decompression runs

The decompression experiments were designed to simulate magma ascent from the upper andesitic magma chamber to the fragmentation/quench level. Therefore, the starting conditions of the decompression runs must be close to the pre-eruptive conditions as defined by (Martel et al., 1998), i.e. $200_{\pm 50}$ MPa, $875_{\pm 25}$ °C, and melt H₂O content of 5.3-6.3 wt. Owing to technical limitation of the CSPV furnaces, the runs were performed at 850°C. From the isobaric runs, the plagioclase and glass compositions and proportions appeared to be comparable at NNO+1 and NNO+3 (see in section "Results"), so that we performed the decompression experiments using pure Ar as the pressure medium, since it guarantees higher success rates for the sample quench procedure than when using H₂O. The decompression experiments consist of subsequent (i) melt hydration, (ii) decompression, and (iii) annealing:

- (i) Melt hydration was performed at 200 MPa and 850°C, for 2 to 10 days for the H₂O-saturated melts, and for 3 to 6 days for the H₂O-undersaturated melts. The H₂O-undersaturated melts crystallized plagioclase. For both melts, hydration duration has not been considered as a variable, i.e. the melts are considered in the same structural state whatever hydration duration.
- (ii) Decompression was performed isothermally from 200 MPa (initial pressure, P_i), by bleeding a pressure valve either manually as timed steps of 5 MPa or continuously via an air-driven pressure valve servo-controlled by a Eurotherm. Final pressure, P_f, was either 30 MPa or 5-10 MPa (oscillating between 5 and 10 MPa), in agreement with the equilibrium H₂O pressures calculated from the H₂O contents measured in the residual glasses of the natural tephra. Namely, $1.8_{\pm 0.3}$ wt. % H₂O was measured in the residual glasses from the P1 Plinian pumices (without significant detection of other volatile species, such as CO₂; Martel et al., 2000), which converts to quench pressures of ~30 MPa following H₂O solubility laws

(Martel & Poussineau, 2007). In the dome-forming products (1902 surges and 1929 BAFs), 0.7-1.0 wt. % H₂O were measured in the melt, which convert to quench pressures of 5-10 MPa. Decompression durations were chosen to span the wide range of magma rise rates proposed for arc volcanoes (Rutherford & Gardner, 2000), i.e. from mm/s and cm/s for dome-eruptions (0.001-0.01 MPa/min, assuming a rock density of 2.5) to m/s and supersonic velocities for truly Plinian eruptions (> 10 MPa/min). The decompression rates varied from 0.003 to 25 MPa/min (i.e. durations from 15 min to 40 days). Three additional decompressions were conducted stepwise, hereafter referred as to stepping decompressions, typically consisting in five cycles of rapid decompressions (15 min) intercalated with 5 annealing steps of 3 days.

- (iii) Annealing at P_f lasted from 0 to 17 days before rapid quench. Two of these stepping decompression experiments were cooled to 825°C or 800°C during the last step of 3 days at $P_f = 5-10$ MPa to investigate the effect of a terminal cooling at low pressure.

The decompression experiments generated three types of crystallization: pre-, syn, and post-decompression crystals (Figure 2) that may coexist in the final sample. The influence of P_f (ΔT_{eff}), decompression rate, anneal duration, linear versus stepping decompression, terminal cooling, and pre-decompression crystals on the characteristics of decompression-induced crystallization was investigated.

Compositional analysis

The capsules with similar weights before and after experiment were opened for analyses. Sample fragments or half a capsule (to check for possible interaction processes at the sample-

capsule contact) were embedded into blocks of epoxy resins for compositional and textural analysis.

The chemical composition (major elements) of the plagioclases and glasses was determined using a Cameca SX 50 electron microprobe (EMP; BRGM-ISTO Orléans, France) with a 6-nA beam current, 15-kV accelerating voltage, 10-s counting time on peak, a focused beam for the crystals, and a defocused beam of 6 to 8 μm in diameter for the glasses. The analytical errors on the oxide analyses are 1 wt. % relative for SiO_2 and Al_2O_3 , 2 % for CaO , 3 % for FeO , MgO , and TiO_2 , and 5 % for MnO , Na_2O and K_2O . Following the procedure given in [Pichavant \(1987\)](#), alkali migration in glasses was corrected using hydrated standards of Mt Pelée matrix glass composition (alkali content determined by wet chemistry at the CRPG Nancy, France, and H_2O contents determined by Karl-Fischer titration, ISTO, France; [Martel & Poussineau, 2007](#)).

The H_2O content of the crystal-free glass hydrated at 200 MPa and 850°C (saturation conditions) has been measured by Karl-Fischer Titration, following the method given in [Martel & Poussineau \(2007\)](#). The average of three measurements gives $5.7_{\pm 0.2}$ wt. % H_2O . The glass H_2O content calculated after the solubility model of [Liu et al. \(2005\)](#) at 200 MPa and 850°C gives $5.8_{\pm 0.2}$ wt. %, so that we validated this calculation to determine melt H_2O contents in the investigated pressure and temperature ranges.

Textural analysis

The experimental charges were analyzed for crystal textures, following the method previously described in [Martel & Poussineau \(2007\)](#). This consists of sample imaging by scanning electron microscopy (SEM; JEOL JSM-6400, ISTO Orléans) and analysis using the SPO software ([Launeau & Robin, 1996](#); [Launeau & Cruden, 1998](#)), in order to determine crystal area fraction (Φ), number density (N), and morphology as following:

- Φ = plagioclase area / bubble-free glass area;
- N , in mm^{-2} = number of plagioclases / (bubble-free glass area + plagioclase area);
- L_{max} , plagioclase size in μm , is the arithmetic average of the fifth longest axes (long axis of an ellipse of the same area than the crystal); This method, used by [Fenn \(1977\)](#), [Hammer & Rutherford \(2002\)](#), and [Couch et al. \(2003\)](#), is preferred over the bulk-averaged quantity, because it avoids applying stereological corrections to crystal size distribution (particularly difficult for non tabular morphologies). Assuming a single-size crystal population (that has better chances to be verified in experimental than natural products), the largest crystals may directly represent the true length of the 3D crystals.
- Plagioclase morphology is defined as tabular (T; equant), skeletal (S; hollow and swallowtail shapes), or dendritic (D; chain of crystals), following the nomenclature of [Faure et al. \(2003\)](#);

Uncertainties on N , Φ , and L_{max} for a given sample result from adding the errors defined by minima and maxima crystal demarcations to the statistical error resulting from analyzing several images.

SEM imaging may represent a poor fraction of the sample total volume, leading to potential failure in detecting crystals where in very low proportion. Therefore, samples that were crystal-free when observed by SEM were systematically checked using an optical

microscope equipped with a quarter-wave ($\frac{1}{4} \lambda$) plate (after covering slightly crushed grains with a drop of density liquor of index 1.47 on a glass plate). Optical microscopy has the advantage of allowing a much better detection of the presence of few crystals despite a lower resolution ($\sim 2 \mu\text{m}$) than SEM. This checking was particularly important to better constrain the crystallization delay.

RESULTS

The conditions and the phase compositions of the isobaric experiments are reported in [Table 1](#). The conditions and the phase compositions of the decompression experiments are reported in [Table 2](#). The textural characteristics of the plagioclases from the decompression runs are reported in [Table 3](#).

Equilibrium phase relations, proportions, and compositions

With decreasing pressure, the crystallization sequence is Fe-Ti oxides, plagioclase, and orthopyroxene. Fe-Ti oxides are ubiquitous in the experimental charges, but in fractions not exceeding 0.01. They are mostly titanomagnetites, together with hematite at NNO+3 to +4 (Ar pressure medium) or with ilmenite at NNO+1 (H₂O pressure medium). Orthopyroxene appears between 100 and 125 MPa at 800°C, and between 75 and 100 MPa at 850°C. The textural and compositional data for titanomagnetite and orthopyroxene are too scarce (owing to the low number and small size of the crystals) to be further considered in this study.

[Figure 3a](#) shows that the plagioclase composition does not significantly depend on f_{O_2} . The liquidus pressure of the plagioclase (H₂O-saturation conditions) decreases with increasing temperature, i.e. from 175-200 MPa at 850°C to 150-175 MPa at 875°C. The plagioclase

liquidus composition increases from An₅₄ (54 mol. % of anorthite) at 850°C to An₅₆ at 875°C. At 850°C, the plagioclase An-content decreases with decreasing pressure (Figure 3a), following an exponential law:

$$An(mol.\%)_{850^{\circ}C} = e^{\left(\frac{\ln(P,MPa)+9.19}{3.58}\right)} \quad (1)$$

The plagioclase fraction, Φ , decreases with increasing temperature, with a particularly marked effect at $P < 100$ MPa (Figure 3b). At 850°C, the pressure-dependence of Φ is given by:

$$\Phi_{850^{\circ}C} = e^{\left(\frac{P(MPa)+27.3}{-50.7}\right)} \quad (2)$$

Figure 3c shows that the SiO₂ content of the residual glass does not seem to be significantly affected by f_{O_2} in the investigated range. The glass SiO₂ content increases from 75-76 to 79-80 wt. %, with pressure decreasing from 200 to ~100 MPa. From 100 to 50 MPa, SiO₂ remains constant at 79-80 wt. %. Below 50 MPa, the glass SiO₂ content seems to decrease by 1 to 3 wt. %. Several reasons may be proposed to explain this apparent SiO₂ depletion: (i) the crystallization of a silica polymorph, presumably cristobalite as analyzed by Martel & Schmidt (2003), between 75 and 100 MPa at 850°C and between 100 and 120 MPa at 800°C, could locally consume the melt SiO₂, (ii) the chemical analysis of the glass may be contaminated by unresolved dendritic plagioclases (depleted in SiO₂ compared to the melt), or (iii) the slow crystallization kinetics at pressures < 75 MPa could prevent achievement of the equilibrium melt composition. A combination of two or the three of them is possible.

The glass compositions have been projected in the normative Quartz-Albite-Orthoclase ternary corrected for systems bearing less than 20 wt. % of anorthite (Qz'-Ab'-Or', in wt. %; Blundy & Cashman, 2001; Table 1), in order to build a polybaric compositional trend at

equilibrium (Figure 4a). At 850°C and from 200 to 100 MPa, plagioclase crystallization forces the residual glass to migrate towards the Qz-Or join. Between 100 and 75 MPa, where plagioclase crystallizes with $\Phi \sim 0.10$, the silica phase precipitates, defining the isobaric Qz-feldspar cotectic. Further pressure decrease leads to a co-crystallization of plagioclase, orthopyroxene and silica, still driving the residual melt towards the Qz-Or join, but with a smaller increase in Qz than before crossing the cotectic.

Plagioclase liquidus

A set of isobaric runs was devoted to the determination of the plagioclase H₂O-saturated liquidus (Table 1), which is further used for the determination of ΔT_{eff} . A plot of the plagioclase-free samples in a pressure-temperature diagram shows that the plagioclase liquidus of Mt Pelée rhyolitic melt roughly parallels the liquidus in other H₂O-saturated rhyolitic systems (Figure 5). In comparison to the liquidus determined in melts with normative compositions in anorthite of 0 and 10 wt. % (melt_{An0} and melt_{An10}), Mt Pelée liquidus is shifted towards higher temperatures due to its higher anorthite content (melt_{An12}) (Tuttle & Bowen, 1958; James & Hamilton, 1969; Johannes & Holtz, 1996). Mt Pelée liquidus temperature increases from ~850°C at 200 MPa to > 1100°C at 10 MPa. For a crystallization temperature of 850°C at 10 MPa, ΔT_{eff} roughly equals 250°C. We can note that the plagioclase liquidus composition increases from An₅₄ at 200 MPa to more than An₆₂ at pressures below 50 MPa.

Pre-decompression crystallization

Plagioclases crystallized isobarically in melts that were H₂O-undersaturated at 200 MPa and 850°C. A phase equilibrium run undersaturated with 5.6 wt. % H₂O has been performed at 200 MPa and 850°C, in order to characterize the initial (pre-decompression) plagioclase

fraction (Φ_i), number density (N_i), size (the five longest crystals, L_i), and composition (An_i) (#D36; Table 1; Figure 6a). The H₂O-undersaturated samples from the dynamic experiments that did not further crystallize upon or after decompression (no syn- or post-decompression crystals; #D3, #D9, #D10; Table 3; Figure 6b) have been added to the phase equilibrium run to define the evolution of Φ_i , N_i , L_i , and An_i with melt H₂O content at 200 MPa.

Figure 7a shows that for melt H₂O content ranging from 4.6 to 5.8 wt. %, the N_i curve shows an asymmetric bell-shape, with a maximum at ~5.4 wt. % H₂O for which plagioclase L_i is minimum. Φ_i and An_i evolve linearly with melt H₂O content, showing a negative and a positive correlation, respectively (Figure 7b). The pre-decompression crystals are always tabular in shape in the range of investigated H₂O content (Figure 6a-b).

The H₂O content dependence of N_i , Φ_i , L_i , and An_i is used for deducing the characteristics of the pre-decompression crystals in H₂O-undersaturated melts that further crystallized during or after decompression, from their initial melt H₂O content (recalculated from the amount of H₂O introduced in the capsule). For instance, Figure 7 suggests that the samples hydrated at 200 MPa with 5.6 wt. % H₂O (#D4, #D6, #D15, #D38) must have contained pre-decompression tabular plagioclases with $N_i = 10^3 \text{ mm}^{-2}$, $\Phi_i = 0.03$, $L_i = 10 \text{ }\mu\text{m}$, and $An_i = 52\text{-}53 \text{ mol. \%}$, and the samples hydrated with 5.1 wt. % (#D11) must have had $N_i = 10^4 \text{ mm}^{-2}$, $\Phi_i = 0.13$, $L_i = 20 \text{ }\mu\text{m}$, and $An_i = 50 \text{ mol. \%}$ before decompression.

Syn-decompression crystallization (without pre-decompression crystals)

Textures

Syn-decompression crystallization from crystal-free melts occurs for decompression rates faster than 0.10 MPa/min, i.e. for decompression durations longer than 1 day. Plotting N as a function of decompression rate shows a slow N increase up to 5000 mm^{-2} as decompression rate decreases from 0.10 to ~0.01 MPa/min and an N increase to 12000 mm^{-2}

for decompression rates slower than ~ 0.01 MPa/min (Figure 8a). This suggests a major nucleation event at linear decompression rate of ~ 0.01 MPa/min (i.e. decompression duration of ~ 14 days to $P_f = 5-10$ MPa). De facto, the negative relationship between Φ and decompression rate (Figure 8b) primarily records the ongoing crystal nucleation.

L_{max} in the samples decompressed to $P_f = 5-10$ MPa show a bell-shape evolution with decompression rate, with a maximum value of $60 \mu\text{m}$ for 0.02 MPa/min (Figure 8c). For $P_f = 30$ MPa, L_{max} likely follows a similar bell-shape evolution with decompression rate, with a maximum value $\geq 100 \mu\text{m}$.

The crystals mainly display tabular shapes for decompression rates slower than ~ 0.01 MPa/min and are skeletal/hollow for faster decompression rates (Figures 6e-g and 8c). This morphology evolution reflects plagioclase crystallization under an increasing ΔT_{eff} (from $< 100^\circ\text{C}$ for tabular crystals to between $\sim 100-200^\circ\text{C}$ for skeletal habits) and a subsequent increase of the growth rate relative to diffusion rates of the plagioclase components as decompression rate decreases (Lofgren, 1980; Kirkpatrick, 1981; Hammer et al., 2000).

Evaluating a slightly more mafic melt composition (71 wt. % SiO_2 ; crystal-free) saturated with H_2O , Cichy et al. (2011) and Brugger & Hammer (2010) reported no clear relationships between N or Φ and decompression rate. Regarding L_{max} (or growth rate, which is the time-derivative of L_{max}), Brugger & Hammer (2010) reported a positive correlation with decompression rate, whereas Cichy et al. (2011) reported a negative trend. The present data show both trends, depending on whether nucleation occurred or not (Figure 8c). Thus, all these different results may be primarily ascribable to the progression state of the nucleation event, with a probability for the more mafic composition to reach equilibrium nucleation faster than for Mt Pelée's. Additionally, the samples of Brugger & Hammer (2010) contain

pre-decompression crystals, which may modify the decompression-induced crystallization by providing preferential sites for crystal growth, at least during early crystallization.

Compositions

The plagioclases have An-content decreases from An₅₅ (sub-liquidus composition) to An₄₈ as decompression rate increases from 0.003 to 0.04 MPa/min (Figure 8d). This trend is in apparent contradiction with the syn-decompression crystallizations in Brugger & Hammer (2010), who reported no relationship between plagioclase composition and decompression rate. The presence of initial plagioclases in the starting melts of Brugger & Hammer (2010) potentially drives decompression-induced crystallization slightly away from the crystallization conditions prevailing in the present crystal-free starting melts.

Figure 4b shows that the residual glasses are enriched in normative-Qz with increasing decompression rate, in agreement with the polybaric trend defined at equilibrium (Figure 4a). The Qz-feldspar cotectic is crossed (appearance of the silica phase) at decompression rate between 0.02 and 0.01 MPa/min (decompression duration between 6 and 14 days).

Stepwise decompressions with terminal cooling

The two samples decompressed stepwise and cooled to 825°C (#D24) and 800°C (#D21) at $P_f = 5\text{-}10$ MPa during ~5 days both show coexistence of skeletal and dendritic crystals (e.g. Figure 6h), the latter habit commonly characterizing crystal growth under $\Delta T_{\text{eff}} > 200^\circ\text{C}$ (Lofgren, 1980; Kirkpatrick, 1981; Hammer et al., 2000).

The two samples show very different N depending on terminal cooling, i.e. ~1000 and ~14000 mm⁻² in #D24 and #D21, respectively (Figure 8a). This suggests that small temperature variations at low pressure may decide on the trigger of a low-pressure nucleation

event. However, #D21 shows only a slightly higher Φ than #D24 (Figure 8b), suggesting that the nucleation event in #D21 was not followed by significant crystal growth.

Another impact of lowering temperature at low pressure is the generation of large heterogeneities in glass composition and microlite spatial distribution. Whereas #D24 shows chemically homogeneous glasses, the residual glasses of #D21 divides into clear K-rich areas where plagioclase crystallized and dark Ca-rich parts where plagioclase did not have time to significantly crystallize (Figures 4b and 6h). Both samples contain a silica phase in the matrix, so that the chemical heterogeneities may reflect the proximity of the Qz cotectic at temperatures lower than 875°C.

Post-decompression crystallization (without pre-decompression crystals)

Textures

Crystallization is delayed by 6-12 h at $P_f = 30$ MPa and 2-3 days at $P_f = 5-10$ MPa (decompression rate of 2.5 MPa/min; Figure 9a). For $P_f = 30$ MPa, N increases to reach a plateau value of $\sim 2 \times 10^4$ mm⁻² after 4 days. For $P_f = 5-10$ MPa, N drastically increases up to values $> \sim 10^5$ mm⁻² within 2-3 days following the onset of nucleation, suggesting a massive nucleation event. Figure 9a also shows that decompression rate positively correlates with N . Indeed, increasing decompression rate from 2.5 to 25 MPa/min at $P_f = 30$ MPa increases N from $\sim 13\,000$ to $43\,000$ mm⁻² (after 2 days at P_f). Similarly, increasing decompression rate from 0.5 to 2.5 MPa/min at $P_f = 5-10$ MPa increases N from $\sim 30\,000$ mm⁻² (#D32) to $\sim 107\,000$ mm⁻² (#D12).

□ Φ increases up to ~ 0.2 after 4 days at $P_f = 30$ MPa and drastically increases to at least ~ 0.3 within the 2-3 days following nucleation at $P_f = 5-10$ MPa (Figure 9b). Decompression rate is positively correlated with Φ , as shown by #D34, in which Φ is greater than in melts decompressed at 2.5 MPa/min (0.23 versus ~ 0.15 after 2 days at P_f). Similarly at $P_f = 5-10$

MPa, increasing decompression rate from 0.5 to 2.5 MPa/min increases Φ from 0.16 (#D32) to 0.27 (#D12).

L_{max} increases to $\sim 25 \mu\text{m}$ within ~ 4 days for plagioclases nucleated at $P_f = 30$ MPa and to $\sim 15 \mu\text{m}$ within less than 2 days at $P_f = 5\text{-}10$ MPa. L_{max} in #D32 and #D34 seems to follow the trend of the samples decompressed at 2.5 MPa/min (Figure 9c).

Plagioclases are mainly dendritic, sometimes mixed with skeletal habits (Figure 6i-1), as expected for $\Delta T_{\text{eff}} > 200^\circ\text{C}$ where crystallization is rate-limited by crystal growth (Lofgren, 1980; Kirkpatrick, 1981; Hammer et al., 2000).

Compositions

There seems to be a decrease from An_{51} to An_{33} as decompression rate increases from 0.5 to 2.5 MPa/min at $P_f = 5\text{-}10$ MPa and from $\text{An}_{39\text{-}41}$ to An_{33} as decompression rate increases from 2.5 to 25 MPa/min at $P_f = 30$ MPa, although the data are too scarce to ascertain the effect (Figure 9d).

The glasses enriches in normative-Qz with increasing dwelling at P_f (Figure 4c), in agreement with the polybaric trend defined at equilibrium (Figure 4a). The cotectic is crossed between 12 h and 1 day at $P_f = 30$ MPa and between 2 and 3 days at $P_f = 5\text{-}10$ MPa (Figure 4c).

Syn- and post-crystallization starting with pre-decompression crystals

Figure 6c-d shows that pre-decompression plagioclases are barely distinguishable from syn-or post-decompression ones, partly because these initial crystals are small (typically $\sim 5\text{-}10 \mu\text{m}$ in length when crystallized from melts undersaturated with 5.1 to 5.6 wt. % H_2O ; Figure 7b). Nevertheless, plagioclase-bearing melts ($N_i < 10^4 \text{ mm}^{-2}$) that further crystallize syn-decompression crystals have much higher N than crystal-free starting melts (Figure 10a).

For instance, in samples decompressed at a rate of 0.006-0.01 MPa/min, increasing N_i from 0 to 10^3 and 10^4 mm^{-2} leads to final N of 10^3 , 6×10^4 , and 10^5 mm^{-2} , respectively. These samples also show higher Φ than in initially crystal-free melts (Figure 10b). In the case of post-decompression crystallization, however, there is no clear evidence of reaching either higher N_A or Φ when starting with pre-decompression crystals (Figure 10d-e).

Pre-decompression crystals play a role on the final crystal size, in showing smaller L_{max} than from initially crystal-free melts (Figures 10c,f). Actually, L_{max} are very close to L_i (5-15 versus 5-10 μm ; Figures 10c,f), so that decompression-induced growth around the pre-decompression crystals must have been limited.

Linear (#D4) and stepwise (#D15) decompressions (integrated rate of 0.008 MPa/min) of a melt containing similar N_i (10^3 mm^{-2}) lead to comparable N (Figure 10a), Φ (Figure 10b), and L_{max} (Figure 10c).

DISCUSSION

Important parameters controlling decompression-induced plagioclase crystallization

Nucleation versus growth

With ΔT_{eff} increasing from 220 to $> 275^\circ\text{C}$ (P_f decreasing from 30 to 5-10 MPa), the nucleation lag for post-decompression crystallization increases from ~12 h to 2-3 days, and the following single nucleation event increases in intensity (Figure 9a). For $\Delta T_{\text{eff}} > \sim 275^\circ\text{C}$, the nucleation event is brief, massive, and accompanied by a drastic increase in Φ and L_{max} , which suggests that nucleation and growth are simultaneous processes. For ΔT_{eff} of 220°C , N , L_{max} , and Φ are close to reaching plateau values, suggesting that crystallization nearly achieved equilibrium within 4-6 days (Figure 9a-c).

Comparatively, syn-decompression crystallizations illustrate the complexity of the crystallization characteristics, i.e. dominance of nucleation versus growth, associated with variable ΔT_{eff} upon decompression. With decreasing decompression rate, crystal size passes through a maximum value ($\sim 60 \mu\text{m}$ for 0.02 MPa/min to $P_f = 5\text{-}10 \text{ MPa}$; [Figure 10c](#)), reflecting the dominance of crystal growth (low ΔT_{eff}) over nucleation. At lower decompression rate, however, the prevailing ΔT_{eff} favours the onset of an early (plagioclase composition close to liquidus; [Figure 10d](#)) and massive (drastic N increase; [Figure 10a](#)) nucleation event, for which the absence of plateau value suggests incompleteness within 40 days of decompression.

Effect of the decompression rate on nucleation upon dwelling at P_f

Increasing the decompression rate before crystallization at P_f (post-decompression crystallization) leads to an increase of N and Φ , and a possible decrease in the plagioclase An-content ([Figure 9](#)). The positive correlation between N and decompression rate agrees with what is observed for bubble nucleation in hydrous melts (e.g. [Mourtada-Bonnefoi & Laporte, 2004](#); [Toramaru, 2006](#); [Gondé et al., 2011](#)), potentially addressing similar nucleation triggering mechanisms on an energetic point of view. The positive correlation between N and decompression rate also agrees with previous experimental studies based on post-decompression isobaric crystallization following instantaneous decompressions ([Couch et al., 2003](#); [Martel & Schmidt, 2003](#)). Through a combined simulation including kinetics of both vesiculation and crystallization, [Toramaru et al. \(2008\)](#) and [Toramaru & Miwa \(2008\)](#) suggested that the variation in N with decompression rate can be attributed to the variation of the driving force for the diffusive bubble growth with variable decompression under a nearly constant number density of bubbles.

Effect of pre-decompression crystals

The presence of pre-decompression plagioclases (N_i up to 10^4 mm⁻², Φ up to 0.13, size ~ 10 μ m) has crucial effects on the further conditions of decompression-induced crystallization by controlling final N , Φ , and L_{max} .

The effect is particularly important during syn-decompression nucleation, with an N increase of about 1 log unit in comparison to crystal-free starting melts (Figures 10a). The initial crystals may influence decompression-induced crystallization following a Boolean choice: either they act as nucleation sites, so that decompression-induced crystallization begins as growth around these crystals, or they do not act as preferential nucleation sites and crystallization begins as a new nucleation event in a way that may resemble crystallization in crystal-free melts. From the crystal size, one can assume a limited growth of the pre-decompression crystals (L_i and L_{max} of 5-10 and 5-15 μ m, respectively; Figure 10c). From N , samples containing pre-decompression crystals show N that could be an extrapolation of the trend expected from initially crystal-free melts that crystallized upon dwelling at P_f (Figure 10d), suggesting a massive nucleation event as expected under high ΔT_{eff} . Similarly, the samples containing initial crystals reach Φ values that compare with post-decompression crystallization starting from crystal-free melts (Figure 10e), suggesting close growth conditions.

Thus, N and Φ during syn-decompression crystallizations of crystal-bearing melts may be comparable in essence to crystallization in crystal-free melts upon dwelling at constant pressure. Yet, the final crystal morphologies are different: tabular for syn-decompression crystallizations from crystal-bearing melts (Figures 6c, 10c) and skeletal to dendritic for initially crystal-free post-decompression crystallizations (Figures 6i-1, 9d), reflecting crystallization under lower ΔT_{eff} (higher pressure) than expected at $P_f \leq 30$ MPa. Therefore, it is likely that syn-decompression crystallization from crystal-bearing melts begins as growth

around the pre-existent crystals, thus lowering the driving force for crystallization (ΔT_{eff}), and goes on with nucleation at low pressure under a reduced ΔT_{eff} ($< 100^\circ\text{C}$, in order to generate tabular crystals).

Achievement of equilibrium

Comparing Φ in the dynamic and phase-equilibrium samples shows that decompression-induced crystallization is far from equilibrium. For instance, phase equilibrium at 30 MPa and 10 MPa predicts Φ of 0.3 and 0.5 using (2), respectively, whereas maximum Φ values of 0.2 and 0.3 are measured in the samples decompressed to 30 and 5-10 MPa (Figure 9b), respectively, only representing two thirds of the expected values at equilibrium. This incomplete plagioclase crystallization is emphasized by the plagioclase and residual glass compositions that did not reach the expected values at equilibrium. Indeed, the plagioclase compositions of the decompressed samples best reach An_{33} (Figure 9d), whereas An_{20-25} would be expected at 5-10 MPa using (1) and the residual glass compositions are far from the equilibrium trend at 30 or 5-10 MPa (Figure 4b-c). The difference to equilibrium Φ and phase composition is even worth in case of syn-eruptive crystallization, even after decompression duration as long as 40 days.

Although not useful for prediction of the plagioclase final proportions and compositions during dynamic experiments, phase equilibrium experiments may be used to assess crystallization conditions during decompression. Plagioclase core compositions in #D35 are An_{55} (Figure 8d). Assuming that plagioclase composition does not re-equilibrate on the timescale of the experiment (owing to the slow kinetics of chemical re-equilibration in plagioclase), this composition suggests nucleation pressure close to 200 MPa using (1). However, the Φ value of 0.16 measured in #D35 (Figure 8d) corresponds to an equilibrium pressure of ~ 66 MPa using (2), suggesting that the plagioclase nucleated close to 200 MPa

grew until reaching a pressure of ~66 MPa. In contrast, #D33 shows plagioclase compositions of $An_{48\pm 2}$ (Figure 8d), corresponding to a nucleation pressure between 91 and 123 MPa using (1) and Φ of 0.10 (Figure 8b), suggesting plagioclase growth until 89 MPa using (2). Therefore, increasing decompression rate leads to a decrease in the nucleation pressure of the plagioclase (An-poorer core composition), but growth over a smaller pressure range, leading to lower final Φ (at least while nucleation is not finished).

Post-decompression crystallizations better approach equilibrium: the closest composition to the expected An_{20-25} at 5-10 MPa is $An_{33\pm 1}$ (Figure 9d), corresponding to an equilibrium pressure of 28 MPa using (1). Additionally for post-decompression crystallizations at 5-10 MPa, cristobalite appears whilst plagioclase Φ is very low (0.13-0.16) and glass composition still stands below the Qz volume portion of the ternary (#D31 and #D32 in Figure 4c). This suggests that cristobalite may not be interpreted as resulting from silica oversaturation in a residual melt crystallizing large amount of silica-poor phases, but as evidence of a phase stability field constrained in temperature and pressure (melt H_2O content).

Magma ascent paths and eruption dynamics at Mt Pelée

The results of the decompression experiments have been used to investigate magma ascent path and rate prevailing during the recent Plinian and dome-forming eruptions of Mt Pelée (Figure 11). Mt Pelée recent eruptions are supposed to be, at least partly, triggered by a hot basaltic andesite (~1050°C; Pichavant et al., 2007) intruding the cooler andesitic upper reservoir (~875°C; Martel et al., 1998), leading to the crystallization of few, but ubiquitous, cooling-induced mafic microlites that mixed up with the rhyolitic interstitial glass of the andesite prior to magma ascent/decompression in the volcanic conduit (Martel et al., 2006).

These pre-decompression crystals have $\Phi_i < 0.1$, $N_i < 30\,000\text{ mm}^{-2}$, and length $< 100\ \mu\text{m}$ (Martel et al., 2006). The presence of pre-decompression microlites differs from that of phenocrysts in terms of further decompression-induced crystallization because their much higher number densities act more efficiently as preferential sites for further crystallization. The decompressions have been conducted isothermally (850°C), although in real magmatic systems, decompression may rather be adiabatic, with expected magma cooling (due to gas expansion and heat loss at conduit wall) and heating (from crystallization latent heat). However, the importance of these temperature variations has never been evaluated in the case of a magma ascent.

Plinian eruptions

Plinian eruptions are commonly viewed as rapidly ascending magmas that accelerate prior to fragmentation at pressures of the order of tens of MPa (Sparks et al., 1978; Wilson et al., 1980). Thus, their ascent dynamic may be reasonably simulated by rapid linear or accelerated decompressions from reservoir to fragmentation.

Most of the pumices from Mt Pelée Plinian events lack decompression-induced microlites, which has been experimentally reproduced for decompression rates $> 0.1\text{ MPa/min}$ (Figures 8a), corresponding to magma ascent from a 6-km deep reservoir to $\sim 30\text{ MPa}$ within less than 1 day. Nevertheless, some rare pumices contain very small dendritic microlites characterized by a wide range of high N from $\sim 40\,000$ to $180\,000\text{ mm}^{-2}$ and low $\Phi < 0.2$ (Figure 1). Only post-decompression crystallization generates small and numerous plagioclases. Decompressions at a rate of 2.5 MPa/min , followed by crystallization at $P_f = 30\text{ MPa}$ for more than 2 days, duplicates the required Φ (Figure 9b), but fails reaching $N > 25\,000\text{ mm}^{-2}$ (Figure 9a). A way to increase N is crystallization at P_f lower than 30 MPa . The lower P_f calculated from the statistical standard deviation on the mean H_2O content measured

in P1 residual glasses (1.8 ± 0.3 wt. %; Martel et al., 2000), i.e. 1.5 wt. %, is ~ 22 MPa (Liu et al., 2005), which gives $N \sim 75\,000$ mm⁻² (Figure 9a). In this case, however, Φ might increase up to 0.25 (Figure 9b). An alternative means of increasing N is to depressurize faster than 2.5 MPa/min, as a 10-fold increase of the decompression rate (2.5 to 25 MPa/min) doubles N (Figure 9a). In this case also, Φ might increase up to 0.22 (Figure 9b). A combination of pressures slightly below 30 MPa, decompression rates close to 25 MPa/min, and short durations at P_f (so that Φ stays below 0.2) may produce the dendritic microlite characteristics observed in some natural Plinian samples (Figure 11a).

Therefore, an eruptive scenario for the Plinian events at Mt Pelée may consider a main magma ascending from the reservoir to the fragmentation level at ~ 30 MPa at a rate > 2.5 MPa/min (1 m/s), likely close to 25 MPa/min (~ 10 m/s), without having time to crystallize microlites, and that incorporates magma batches that could have ascended to depth < 30 MPa few hours to a couple of days before the eruption started (crystallizing small dendritic microlites). The high N variability in these pumices could reflect variability in crystallization pressure before eruption.

Block-and-ash flows

BAFs (or nuees ardentes of Merapi-type) resulted from the gravitational destabilization of the dome, so that a part of the erupted products are dome fragments and the other comes from upper conduit. In the 1929-32 BAFs products, this latter category contains large (10-50 μm) tabular microlites of plagioclase, with low $N < 40\,000$ mm⁻² and low $\Phi < 0.25$ (Figure 1). According to the experimental results, large tabular crystals cannot be obtained from post-decompression crystallizations (Figure 9c), so that magmas must have crystallized while ascending, rather than during stagnation at subsurface level.

Yet, experimental syn-decompression crystallizations are characterized by lower N ($< 15\,000\text{ mm}^{-2}$; [Figure 8a](#)) than commonly observed in Mt Pelée BAFs pyroclasts. To reach higher N , several hypotheses may be proposed. The first one concerns magma cooling at low pressure, since BAFs mostly come from collapsing domes, but the tabular morphology of the natural microlites does not support this hypothesis (dendritic overgrowths were observed in the experiments cooled at 5-10 MPa). Another way to increase N regards decompression rates slower than 0.003 MPa/min, since experimental nucleation still goes for slower decompression rates (duration > 40 days). The variability in natural N and Φ could then reflect variable decompression durations from 1-2 days to more than 40 days. A third hypothesis calls on the pre-decompression crystals, which could allow increasing N over short ascent durations. In order to keep N below $40\,000\text{ mm}^{-2}$, as observed in natural products, the experiments suggest N_i below 1000 mm^{-2} if initial sizes are about 5-10 μm ([Figure 10a](#)).

The BAFs magmas could have ascended step-by-step, as already proposed for Redoubt ([Wolf & Eichelberger, 1997](#)) and Mt St Helens ([Cashman & McConnell, 2005](#)). This favours achievement of low plagioclase An-contents ($\sim\text{An}_{35}$ in the 1929-32 BAFs; [Martel & Poussineau, 2007](#)), as experimentally evidenced ($\text{An}_{46\pm 3}$ against $\text{An}_{52\pm 3}$ for stepping (#D15) and linear (#D4) decompressions, respectively; [Table 2](#)).

Therefore, BAFs magmas at Mt Pelée may have ascended in more than 2-5 days, potentially step-by-step, leading to crystallization of large tabular syn-decompression plagioclases on pre-existent inherited microlites ([Figure 11c](#)).

Surges

Mt Pelée surge matrix is texturally heterogeneous, with areas of large (up to $\sim 50\text{ }\mu\text{m}$) skeletal plagioclase microlites intermingled with areas of numerous and small ($< 20\text{ }\mu\text{m}$) dendritic microlites, both types being embedded in a heterogeneously-distributed silica-bearing

interstitial melt (Figure 1). Overall, the surge matrix shows very high and variable N (from 110 000 to 250 000 mm⁻²) and high Φ (0.2 to 0.3), with plagioclase microlite compositions ranging from ~An₅₅ to An₃₅ (Martel & Poussineau, 2007).

Experimental post-decompression crystallization at $P_f = 5-10$ MPa reproduces the small (< 20 μm) dendritic crystals (Figure 6i-j, 9c), but lack the larger skeletal microlites present in the natural samples. Both crystal populations, small dendritic and large skeletal, and heterogeneously-distributed cristobalite areas, are actually reproduced by post-decompression crystallization starting from pre-decompression crystal-bearing melts (Figure 6d).

This gives confidence in proposing an ascent scenario for surge magmas in two steps (Figure 11b): (i) a pre-decompression microlite-bearing magma (Φ_i up to 0.10 and $N_i \sim 10^3-10^4$ mm⁻²) ascending within less than ~4 days, during which no significant crystallization occurs; this would agree with the fast magma extrusion rates prior to the 1902 surge eruptions proposed by Tanguy (2004), and (ii) a post-decompression crystallization at dome-base pressure (5-10 MPa) during 4 to 15 days, by simultaneous skeletal growth around the pre-existent microlites and new nucleation of small dendritic microlites under high ΔT_{eff} (low pressure). This results in overall heterogeneously-distributed skeletal and dendritic plagioclase microlites in a cristobalite-bearing matrix glass. The large range of natural N could reflect variability in decompression rate or crystallization depth within the upper conduit.

Domes

Dome matrixes cannot be reliably characterized because a ubiquitous pervasive silica precipitation mainly precludes clear microlite demarcation. Rough estimations suggest large

tabular microlites with high $\Phi > 0.20$ and low $N < 50\,000\text{ mm}^{-2}$. Domes seem to be in the continuation of BAFs in terms of microlite characteristics (Figure 1).

According to the experimental results, relatively few and large tabular crystals characterize syn-decompression crystallization from melts containing pre-decompression crystals (Figure 10c). In that sense, it is likely that dome magmas originally ascend like BAFs magmas, i.e., slowly and/or stepwise, with crystallization dominated by growth around pre-existing crystals. Yet, dome samples have higher values of Φ and crystal length, and An-poorer microlite compositions (An_{33-35} ; Martel et al., 2006) than in BAFs pyroclasts. According to the experimental results, a way to produce enlarged and more albitic crystals is to cool the sample at P_f (Figure 8d). In the decompression experiments, the drastic Φ increase predicted by the phase equilibria when cooling a sample from 850 to 800°C at $P_f = 5\text{-}10\text{ MPa}$ is not observed on a timescale of 16 days (Figure 8b). Yet, dome magmas stagnate more than 16 days at low pressures, promoting further crystallization and higher Φ . Besides crystallizing more albitic composition, lowering temperature by 50°C produces compositional heterogeneities, with K-enriched melts where the low-temperature albitic-plagioclase crystallized and Ca-enriched melts where low-temperature crystallization did not significantly progress (Figure 6f, 4b). Additionally, the phase equilibria show that the lower the temperature, the higher the appearance pressure, and presumably the content, of cristobalite in the melt (Figure 3b). Such compositional glass heterogeneities and silica abundances have been observed in the 1902 and 1929 domes of from Mt Pelée (Martel & Poussineau, 2007), as well as in Mt St Helens dome products (Cashman, 1992). The agreement between the natural and experimental samples gives confidence in interpreting the dome products as having experienced similar ascent conditions than the BAFs magmas, followed by post-decompression crystallization owing to cooling by at least ~50°C during more than 16 days (Figure 11c).

Transitions between eruptive styles

A growing dome may partly or entirely collapse into pyroclastic flows, the explosivity of which seems to depend on the dome size or age. Namely, BAFs commonly result from a dome that is large and/or in place for a significant amount of time, whereas surges are often triggered from small or young domes. For instance, the 1902 surges at Mt Pelée were generated from small domes of magma freshly arrived to the surface (Chrétien & Brousse, 1989). At Mt St Helens in 1980, the surge came out from a newly-emplaced crypto-dome (Voight et al., 1981). Such a relation between the dome size/age and the eruption explosivity corroborates the calculations of Taisne & Jaupart (2008), suggesting that a dense dome (i.e., large or old) contributes to seal underlying fractures by pushing gases away, whereas a small dome promotes explosivity by retaining gases within the magma.

Should a dome grow via a slow magma ascent rate, the driving force for crystallization is accommodated throughout ascent within the conduit by growth around the pre-existent crystals and ΔT_{eff} at subsurface level is not high enough to trigger a crystal nucleation event. At that point, transient dome growth and associated gravitational BAFs represent the more likely hazard. But if the dome grows at a rate that prevents significant crystallization during ascent, the high ΔT_{eff} accumulated upon decompression may lead to a terminal nucleation event at dome pressure. Such low-pressure crystallization generates high and heterogeneously-distributed proportions and numbers of plagioclase microlites, which forces massive H₂O exsolution from the saturated melt. Bubble growth and crystallization of cristobalite (the stability field of which is enlarged at low P-T) are two processes that act as pressurizing magma, since both requires extra free volume that is little available in a confined, poorly compressible, upper conduit geometry. As soon as gas pressurization exceeds magma

strength, fragmentation occurs (Alidibirov & Dingwell 1996; Martel et al., 2001; Elsworth and Voight, 2001; Melnik et al., 2005) and the major hazard may be surges.

Mt Pelée P1 eruption began with surges of pelean-type followed by a Plinian event (Boudon et al., 1994). Obviously, the depressurization caused by the surges was high enough to generate a downward fragmentation wave on a whole magma column, probably by explosively relaxing the trapped gas, which causes the Plinian event. Yet, the 1902 surges, although stratigraphically, petrologically, and volumetrically comparable to the P1 surge (Boudon et al., 1994; Martel & Poussineau, 2007), were not followed by a Plinian event. Therefore, it is possible that the magma underlying the 1902 surges already lost sufficient gas to preclude any explosive gas relaxation and Plinian fragmentation. The 1902 surges ended by a dome growth, and 30 years later in 1929, this dome reactivated and destabilized into BAFs rather than surges, presumably because of its high density related to its size and age (Taisne & Jaupart, 2008).

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Figure caption

Table 1. Experimental conditions and phase compositions of the isobaric equilibrium runs.

Table 2. Experimental conditions and phase compositions of the decompression runs

Table 3. Textural analysis of the plagioclases from the decompression runs.

Figure 1. Domains of eruptive styles defined by the textural characteristics of Mt Pelée plagioclase microlites, modified after [Martel & Poussineau \(2007\)](#). The number density of plagioclase microlites (N) is plotted as a function of its fraction (Φ). The microlite morphology is specified as tabular (T), skeletal (S), or dendritic (D), with * indicating the presence of a silica phase in the residual glass. Pl for plagioclase, Opx for orthopyroxene, Mt for titanomagnetite, Gl for glass, and Si for the silica phase.

Figure 2. Type of crystallization generated by the decompression experiments: 1) pre-decompression crystallization of the H₂O-undersaturated melts during the hydration step at 200 MPa and 850°C, 2) syn-decompression crystallization during either linear or stepping decompression, and 3) post-decompression crystallization during dwelling at $P_f = 5$ -10 or 30 MPa.

Figure 3. Characteristics of the isobaric run products. **(a)** Plagioclase composition; **(b)** fraction of plagioclase, with given compositions in anorthite mol. % (An); **(c)** SiO₂ content (wt. %) of the residual glass recalculated anhydrous. The dashed horizontal lines mark the appearance of cristobalite. H₂O solubility is calculated at 850°C after [Liu et al. \(2005\)](#).

Figure 4. Compositional path evolution of the residual glasses at 850°C. The glass compositions are projected in the normative Qz-Ab-Or ternary (in wt. %; [Tables 1 and 2](#)) corrected for anorthite-bearing systems ([Blundy & Cashman, 2001](#)); **(a)** Equilibrium

isobaric crystallizations; labels give the pressure in MPa and numbers in italic give Φ ; **(b)** Syn-decompression crystallizations; **(c)** Post-decompression crystallizations. In **(B)** and **(C)**, the polybaric compositional trend (plain line) is defined from **(A)**; labels give the run number; the decompression duration or the anneal duration at P_f are reported in brackets; * indicates cristobalite-bearing glasses.

Figure 5. Plagioclase liquidus of Mt Pelée H₂O-saturated rhyolitic melt. Experimental products bearing plagioclase (plain symbols) and free of plagioclase (open symbols) are reported in the P-T space; melt_{An12} (this study), melt_{An10} (Hammer & Rutherford, 2002; HR02), and melt_{An0} (Tuttle & Bowen, 1958; TB58) give the normative compositions in anorthite wt. % of the melts, calculated from the CIPW norm; The effective undercooling, ΔT_{eff} , is shown on top axis; *An* gives the plagioclase composition (in anorthite mol. %); melt H₂O content is calculated after Liu et al. (2005); T, S, or D refers to the tabular, skeletal or dendritic morphology of the crystals, respectively.

Figure 6. Selected SEM pictures of the samples. The left column shows samples with pre-decompression crystals from **(a)** phase equilibrium undersaturated with H₂O **(b)** H₂O-undersaturated sample that did not crystallize during rapid decompression, **(c)** pre- and syn-decompression crystallization, and **(d)** pre- and post-decompression crystallization. The centre column shows syn-decompression crystallization from initially crystal-free melts, i.e. H₂O saturated with 5.8 wt. % **(e to h)**. The right column shows post-decompression crystallization from initially crystal-free melts, i.e. H₂O saturated **(i to l)**. Run conditions as reported in Table 1 (phase equilibrium) and Table 2 (decompression runs). **Gl** for glass, **Gl(d)** and **Gl(c)** for dark and clear glass, respectively; **Mt** for magnetite, **Pl** for plagioclase, **Opx** for orthopyroxene, and **C** for cristobalite. The given scale bar of 20 μm applies for all images.

Figure 7. Textural and compositional characteristics of the pre-decompression plagioclases, as a function of initial melt H₂O content. **(a)** Plagioclase number density (N_i) and area fraction (Φ_i); **(b)** Plagioclase size (L_i) and composition (An_i). Note that pre-decompression crystallization in run #D10 was followed by a 7-days decompression, during which syn-decompression crystallization may have occurred. Yet, the sample lacks a typical population of small crystals that would suggest syn-decompression nucleation. However, its large crystals ($L_{max} \sim 45 \mu\text{m}$; [Table 3](#)) probably reflect syn-decompression crystal growth, so that the provided Φ_i must be taken as a maximum value.

Figure 8. Textural and compositional characteristics of the syn-decompression plagioclases as a function of decompression rate. **(a)** Plagioclase number density, **(b)** fraction, **(c)** size, and **(d)** composition. Point label refers to the runs in [Table 2](#).

Figure 9. Textural and compositional characteristics of the post-decompression plagioclases as a function of dwelling at final pressure (no crystals detected during decompressions at rates from 0.5 to 25 MPa/min). **(a)** Plagioclase number density, **(b)** fraction, **(c)** size, and **(d)** composition. Point labels refer to the runs in [Table 2](#). Note that the large error bars mainly reflect the difficulty in individualizing plagioclase crystals (e.g. [Figure 6j](#)).

Figure 10. Effect of the pre-decompression plagioclases on further syn-decompression **(a-c)** and post-decompression **(d-f)** crystallization. **(a)** and **(d)** Plagioclase number density; **(b)** and **(e)** area fraction; **(c)** and **(f)** crystal length. Point label refers to the runs in [Table 2](#). The subscript i stands for the pre-decompression plagioclase characteristics, as defined in [Figure 7](#). The plain lines represent the pre-decompression plagioclase-free runs shown in [Figures 8-9](#). Note that the large uncertainties associated to N and Φ for runs #D5 and #D6 reflect real crystallization heterogeneities, with the lowest values corresponding to crystal-depleted and cristobalite-free areas (lower right-hand side of [Figure 6d](#)) and the highest

values characterizing the crystal-rich cristobalite-bearing areas (centre left-hand side of [Figure 6d](#)).

Figure 11. Eruptive scenarios at Mt Pelée inferred from microlite textural characteristics. **(a)** Plinian fallout, **(b)** surges, and **(c)** block-and-ash flows and domes. Dashed red areas are natural microlite characteristics from Mt Pelée (after [Martel & Poussineau, 2007](#)). Dashed arrows in **(b)** point on the shift induced by changing decompression rate.