



# On the second nucleation of bubbles in magmas under sudden decompression



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## ABSTRACT

I conducted numerical experiments to evaluate the condition for the second nucleation of bubbles when a sudden change in the decompression rate occurs in magmas with preexisting bubbles. I took into account homogeneous nucleation and diffusive bubble growth. Results from numerical experiments with variable number densities and sizes of preexisting bubbles were able to provide the limiting condition for the decompression rate, above which the second nucleation effectively takes place. The limiting condition as a function of number density and size of preexisting bubbles can be divided into two regimes that are controlled by the mass ratio of dissolved volatiles to exsolved volatiles as preexisting bubbles at the initial state. In the case with a realistically high mass ratio, namely, with a relatively small preexisting bubble volume, the detectable second nucleation requires decompression rates that are higher by a factor of about 10 than those that are hypothetically required to form the preexisting bubbles. By consulting a simple scaling argument, we derive the expression for the limiting decompression rate as a function of the number density and size of preexisting bubbles. The graphical display of the expression allows us to evaluate whether the second nucleation will effectively take place for practical cases with a given number density and size of preexisting bubbles. In natural pumice and scoria, the preexisting bubbles may be observed as larger pheno-bubbles that provide buoyancy at the storage regions, which is needed to trigger the eruption. Such pheno-bubbles are isolated in smaller matrix-bubbles that are formed by the rapid decompression that takes place during the ascent of magma to the surface.

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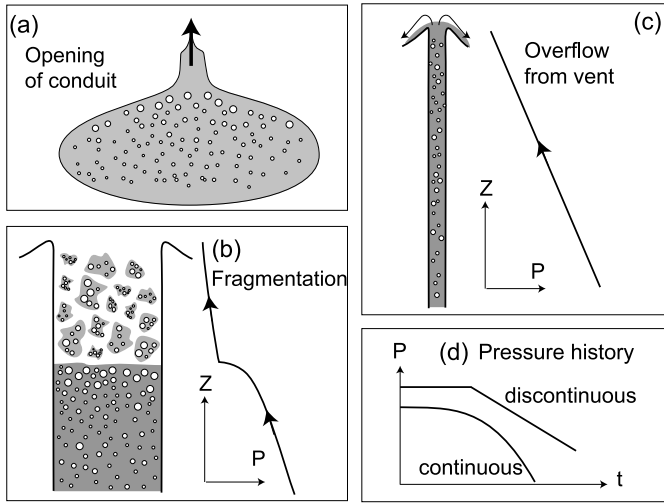
## 1. Introduction

The second nucleation of bubbles, in which bubbles newly nucleate in magmas with preexisting bubbles, can be expected to occur when magmas are further decompressed at fast rates. A variety of situations can lead to rapid decompression and these are depicted in Fig. 1. The first scenario involves the onset of magma ascent from the supersaturated reservoir. During this scenario, preexisting bubbles are formed by heterogeneous nucleation on the wall and the overpressurization of the magma reservoir eventually opens the conduit, which leads to the beginning of magma ascent along with sudden decompression. The second scenario involves magma fragmentation in the conduit flow during which the pressure changes discontinuously according to the fluid mechanics. The third scenario involves the moment at which the eruption is triggered either by fracturing of the conduit cap during Vulcanian eruptions or by the overflow of less viscous magma from a vent like in Hawaiian eruptions. In addition, in Plinian eruptions the bubbly magmas that fill the conduit just before the main

phase of the activity undergo sudden decompression (Toramaru, 2006). Thus, whether the second nucleation occurs may provide key signatures for the ascent processes of magmas, especially if the textural characteristics of pyroclasts can be analyzed in an appropriate manner for the second nucleation. To do this, we first have to understand the condition of the second nucleation of bubbles in regards to the relationship between the physical variables and textural properties.

As a natural consequence, the second nucleation may produce vesicle textures that are more complex, i.e., with multiple peaks in the vesicle size distribution (VSD), than those with unimodal VSDs that often occur from the single event of nucleation. Actually, such complex VSDs have been reported in many cases (Giachetti et al., 2010; Shea et al., 2010; Formenti and Drit, 2003; Klug and Cashman, 1994; Klug et al., 2002; Parcheta et al., 2013; Polacci et al., 2008; Sparks and Brazier, 1982), and the second nucleation has been advocated as the reason for the origin of the multiple peaks in VSDs; otherwise, bubble coalescence is thought to be the mechanism for the formation of minor populations of larger sized bubbles. However, these interpretations for the multiple peaks in VSDs lack conclusive physical evidence and are

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**Fig. 1.** Schematic figures showing possible situations that encounter the sudden change in decompression rates for the second nucleation of bubbles. (a) The onset of magma ascent from supersaturated magma reservoir. Preexisting bubbles separate from the wall and concentrate near the roof by buoyancy and convection. (b) The magma fragmentation in the conduit flow. (c) The overflow from vent. (d) The expected changes of decompression rates; discontinuous as in situations (a) and (c), and continuous as in situation (b).

impaired by the limited understanding of the second nucleation and coalescence processes.

Only a few decompression experiments with silicate melts have been conducted that focus on the condition of the second nucleation. Gardner (2009) demonstrated that high decompression rates maintain the degree of supersaturation at high levels through disequilibrium degassing in decompressed magmas with preexisting bubbles. In numerical simulations of conduit flows, Massol and Koyaguchi (2005) showed that the second bubble nucleation occurs at low pressures where the decompression is accelerated because of magma fragmentation.

Some arguments and suggestions have been proposed based on experimental and theoretical works. Mourtada-Bonnefoi and Laporte (2004) and Cluzel et al. (2008) postulated that the second bubble nucleation at the fragmentation level could explain the discrepancy between common values of bubble number densities in natural samples and the prediction from their experimental results with decompression rates of  $10\text{--}10^6$  Pa/s. Hamada et al. (2010) succeeded in the experimental production of a high number density by applying the high decompression rates that were expected at the fragmentation level. Navon et al. (1998) argued, on the basis of their analysis of bubble growth, that tiny bubbles in the melt space between large bubbles are formed by the second nucleation because of the inhibited growth of large bubbles in highly viscous magmas.

Under a single constant decompression rate, it has been understood that the nucleation occurs as a single event with an acute peak in the nucleation rate; this understanding was derived from numerical and theoretical studies (Toramaru, 1989, 1995) and from experimental studies of systems without preexisting bubbles (Mourtada-Bonnefoi and Laporte, 2004). However we have no fundamental understanding on the system with preexisting bubbles, which undergoes further decompression with different rates. It can be qualitatively inferred that magmas with low number densities of preexisting bubbles will experience further supersaturation with sudden increases in the decompression rate, which is due to the small total surface area and depletion rate of gas in melt (Navon and Lyakhovskiy, 1998; Toramaru and Miwa, 2008), and this will lead to the second nucleation.

In the present study, in order to quantitatively evaluate the condition for the second nucleation of bubbles, a numerical experiment was conducted whereby we systematically varied parameters including the number density, size of preexisting bubbles, effective interfacial tension, and decompression rate using an existing model (Toramaru, 1989, 1995). As a result, we obtain the condition for the second nucleation of bubbles in terms of number density and size of preexisting bubbles. First, we briefly explain the numerical model and show the results. Second, we perform a simple analysis in order to better understand the physical processes behind the numerical results. Third, we rearrange the numerical results into a useful expression and show graphs that may be used for practical purposes in future experimental studies and for interpretations of VSDs in natural pyroclasts. Finally, we provide some examples of the application of the results to natural samples and discuss issues remaining in the present model.

## 2. Methods

I first assumed a unique size for the preexisting bubbles. Then, giving the number density and size of preexisting bubbles, we calculated the vesiculation process of nucleation, growth, and expansion of bubbles with a constant decompression rate using an existing model (Toramaru, 1995). A more detailed description of the calculation scheme is given in Appendix A. The symbols used along with their definitions, units and the scales employed for dimensionless variables are listed also in Table 1 of Appendix A.

The pressure decreases linearly with time as shown in Fig. 2 and according to

$$P = P_0 - \left| \frac{dP}{dt} \right| t \quad (1)$$

where  $P_0$  is the initial storage (saturation) pressure and  $|dP/dt|$  is the linear decompression rate, which is related to the decompression time scale  $t_{\text{dec}}$  defined as  $t_{\text{dec}} = P_0/|dP/dt|$ . In numerical calculations, the actual initial melt pressure  $P'_0$  is slightly different from  $P_0$ , given the solubility of the initial concentration  $C_0$  due to the thermodynamic conditions for the mechanical and chemical equilibrium states with preexisting bubbles, as shown later.

The model has four dimensionless controlling parameters that represent the nucleation barrier ( $\alpha_1$  or surface tension), solubility curve ( $\alpha_2$ ), effective decompression rate ( $1/\alpha_3$ ), and effective viscosity ( $\alpha_4$ ); the definitions for these parameters are given in Toramaru (1995) and Table 1 in Appendix A. Among them, we changed two parameters; the nucleation barrier ( $\alpha_1$ ) and the decompression rate ( $1/\alpha_3$ ). The other two parameters were held constant at  $\alpha_2 = 0.3$  and  $\alpha_4 = 10^5$ , which correspond to the diffusion-limited regime. The nucleation barrier related to the interfacial tension  $\gamma$  is defined by

$$\alpha_1 = \frac{4\pi\gamma R_0^2}{3kT} \quad (2)$$

where  $k$  is the Boltzmann constant,  $T$  is temperature, and  $R_0 = 2\gamma/P_0$  is the scale of the nucleus. The effective decompression rate, which is the ratio of the decompression time scale  $t_{\text{dec}}$  to the time scale of diffusional bubble growth  $t_g$ , is defined by

$$\alpha_3^{-1} = \frac{t_g}{t_{\text{dec}}} = \frac{R_0^2}{\phi_{G0} D t_{\text{dec}}} = \frac{R_0^2}{\phi_{G0} D P_0} \cdot \left| \frac{dP}{dt} \right| \quad (3)$$

where  $D$  is the diffusivity of water in melts,  $\phi_{G0} = kTC_0/P_0$  is the total gas-equivalent volume of dissolved water at the initial saturation pressure  $P_0$ , and  $C_0$  is the corresponding solubility (water concentration) defined in terms of the number of molecules in the unit melt volume.

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