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EFFECT OF VISCOSITY ON NUCLEATION OF BUBBLES IN DECOMPRESSED WATER-SATURATED MAGMA

S. I. Lezhnin, N. A. Pribaturin, and A. L. Sorokin

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Based on the classical nucleation theory, the effect of viscosity on bubble nucleation in water-saturated magma has been studied. A comparison with experimental data proves that viscosity has a pronounced effect on the rate of homogeneous nucleation in magma.

Key words: *homogeneous nucleation, supersaturation, bubbles, viscosity, magma.*

The volcanic-eruption pattern is largely determined by nucleation and growth of gas bubbles dissolved in magma, which are predominantly formed by H_2O and CO_2 [1]. Magma-degassing dynamics depends on the number of bubbles formed; the latter can be predicted using theoretical dependences for the nucleation rate [2, 3]. Yet, these dependences ignore the effect of magma viscosity, which is known to be quite appreciable [4].

The objective of the present work was to predict the rate of homogeneous nucleation in water-saturated magma on the basis of the classical nucleation theory [5] with allowance for viscosity. For the case of pure-liquid boiling, the rate of homogeneous nucleation was calculated with allowance for viscosity and thermal conductivity by Kagan [6]. The method proposed in [6] was used in the present study; to facilitate the comparison, we use the same notation.

Let us briefly recall the main postulates of the classical nucleation theory [5]. We assume that the nuclei of the new phase are macroscopic and their size distribution function $f(t, r)$ can be found from the following equation of the Fokker–Planck type:

$$\frac{\partial f}{\partial t} + \frac{\partial J}{\partial r} = \frac{\partial f}{\partial t} + \frac{\partial}{\partial r}\Big(Af - B\,\frac{\partial f}{\partial r}\Big) = 0. \qquad (1)$$

Here A is the growth rate of the nuclei, B is the diffusivity in the size space, and J is the nucleation rate. To calculate the growth rate of supercritical nuclei, we use the solution of an appropriate continual (heat-conduction or diffusion) problem. In constructing the steady-state solution of Eq. (1), the boundary condition at $r \to 0$ is $f(r) = f_0(r)$, where $f_0(r)$ is the size distribution function of nuclei given by the thermodynamic fluctuation theory. The boundary condition at $r \to \infty$ is $J = \text{const}$. It follows from these conditions that [5]

$$B = -A\Big(\frac{d\ln f_0}{dr}\Big), \qquad J = 2\sqrt{\frac{\sigma}{kT}}\,B(r_k)f_0(r_k),$$

where r_k is the critical-nucleus radius, σ is the surface tension at the interface between the phases, k is the Boltzmann constant, and T is the temperature in the medium.

Following the above-described approach, Toramaru [2] derived the following expression for the degassing conditions of water-saturated magma:

$$J = \frac{D_c C_0^2}{8\pi}\sqrt{\frac{kT}{\sigma}}\,\frac{2\sigma}{r_k P_0}\exp\Big(\frac{V_m\Delta P}{kT}\Big)\exp\Big(-\frac{4\pi\sigma}{3kT}\,r_k^2\Big). \qquad (2)$$

Kutateladze Institute of Thermophysics, Siberian Division, Russian Academy of Sciences, Novosibirsk 630090; lezhnin@itp.nsc.ru. Translated from Prikladnaya Mekhanika i Tekhnicheskaya Fizika, Vol. 46, No. 1, pp. 21–28, January–February, 2005. Original article submitted March 30, 2004; revision submitted May 11, 2004.

Here C_0 is the initial concentration of water in magma, D_c is the diffusivity, P_0 is the initial pressure at the concentration C_0, ΔP is the pressure difference corresponding to r_k, and V_m is the molecular volume of water in the melt. In deriving Eq. (2), the condition of mechanical equilibrium for transcritical nuclei $p = p_0 + 2\sigma/r$ was assumed to be valid; this made it possible to find A by solving the diffusion problem (p is the vapor pressure in a bubble of radius r and p_0 is the pressure in the melt).

Within the framework of the classical nucleation theory, the pre-exponential factor cannot be determined uniquely [5]. Based on the model of "molecule transition frequency", Hurwitz and Navon [3] derived the following expression for the nucleation rate:

$$J = 2\,\frac{n_0^2 V_m D_c}{a_0}\sqrt{\frac{\sigma}{kT}}\exp\Big(-\frac{4\pi}{3}\,\frac{\sigma}{kT}\Big(\frac{2\sigma}{\Delta P}\Big)^2\Big). \tag{3}$$

Here n_0 is the concentration of (undissociated) water molecules and a_0 is the distance between water molecules in the melt. A comparison of dependences (2) and (3) in [7] showed that the preexponential factors in the region of intense nucleation differ within one order of magnitude. Note that the magma viscosity does not explicitly enter dependences (2) and (3).

Let us pass now to solving the problem posed. Instead of Eq. (1), the equivalent equation

$$\frac{\partial f_n}{\partial t} + \frac{\partial J}{\partial n} = \frac{\partial f_n}{\partial t} + \frac{\partial}{\partial n}\Big(\dot n f_n - D\,\frac{\partial f_n}{\partial n}\Big) = 0$$

.

was used in [6] for the distribution function f_n of nuclei over the number n of molecules normalized by the condition $\int_1^\infty f_n\,dn = N_b$ (N_b is the number of nuclei per unit volume). Here and below, the dot denotes the derivative with respect to time. The scheme of [5] applied to constructing the solution of this equation yields the expression

$$J = \frac{C_0 kT}{4\pi\sigma}\Big(\frac{d\dot n}{dr}\Big)_k\sqrt{\frac{\sigma}{kT}}\exp\Big(-\frac{4}{3}\,\frac{\pi\sigma r_k^2}{kT}\Big). \tag{4}$$

Using the Hertz–Knudsen formula, we write the expression for the rate of change of the number of molecules in a gas bubble

$$\dot n = \pi\beta u_t r^2[p_r - p]/(kT), \tag{5}$$

where p_r is the equilibrium vapor pressure at the bubble surface, $u_t = \sqrt{8kT/(\pi m)}$ is the average thermal velocity of vapor molecules, and β is the condensation coefficient. The diffusion process in the vicinity of the bubble is governed by the solution of the steady-state equation $\Delta C = 0$ with the boundary conditions $C(r) = C_r$ and $C(\infty) = C_m$; this solution has the form

$$C(\bar r) = C_m + (C_r - C_m)r/\bar r$$

($\bar r$ is the distance from the bubble center). This solution yields

$$\dot n = 4\pi r^2\Big(\frac{\partial C}{\partial \bar r}\Big)_{\bar r = r} = 4\pi r D_c(C_m - C_r), \qquad C_r = C_m - \frac{\dot n}{4\pi r D_c}.$$

At local thermodynamic equilibrium on the bubble surface, the relation between the pressure p_r and the concentration of water molecules is described by the Henry law $p_r = AC_r^2$. We substitute these expressions into Eq.(5) to obtain

$$\dot n = \frac{\pi\beta u_t r^2}{kT}\Big[A\Big(C_m - \frac{\dot n}{4\pi r D_c}\Big)^2 - p\Big].$$

A fairly accurate approximation for transcritical bubbles is

$$\dot n = \frac{\pi\beta u_t r^2}{kT}\Big[AC_m^2 - 2AC_m\,\frac{\dot n}{4\pi r D_c} - p\Big].$$

After transformations, we obtain

$$\dot n = \pi\beta u_t r^2[AC_m^2 - p]/(kT(1+\delta)), \tag{6}$$

where $\delta = \beta u_t r A C_m/(2D_c kT)$. In calculating the derivative $(d\dot n/dr)_k$, Kagan [6] used the Rayleigh–Lamb equation

$$\rho r\ddot r + 3(\dot r)^2/2 = p - p_0 - 2\sigma/r - 4\eta\dot r/r,$$

which describes the dynamics of a spherical bubble in a viscous liquid. We express the gas pressure in the bubble from this equation, neglect the term $3(\dot r)^2/2$ ($\dot r = 0$ for $r = r_k$) and use the relation $\ddot r = \dot r\, d\dot r/dr$; then, the insertion of the resultant expression into (6) yields

$$\dot n = \frac{\pi\beta u_t r^2}{kT(1+\delta)}\Big[AC_m^2 - p_0 - \frac{2\sigma}{r} - 4\eta\,\frac{\dot r}{r} - \rho r\dot r\,\frac{d\dot r}{dr}\Big].$$

We calculate the derivative $d\dot n/dr$ by neglecting the derivative of the first multiplier and all terms that contain $(\dot r)^2$. After cumbersome yet simple transformations, we obtain the following expression for $r = r_k$:

$$\Big(\frac{d\dot n}{dr}\Big)_k = \frac{\pi\beta u_t r_k^2}{kT(1+\delta_k)}\Big\{\frac{2\sigma}{r^2} - \frac{4\eta}{r}\,\frac{d\dot r}{dr} - \rho r\Big(\frac{d\dot r}{dr}\Big)^2\Big\}_k. \tag{7}$$

We neglect imperfection of vapor in the bubble and write

$$\dot n = \frac{d}{dt}\Big(\frac{4}{3}\,\frac{\pi r^3 p}{kT}\Big) = \frac{d}{dt}\Big(\frac{4}{3}\,\frac{\pi r^3}{kT}\Big[\rho r\ddot r + p_0 + \frac{2\sigma}{r} + 4\eta\,\frac{\dot r}{r}\Big]\Big).$$

We differentiate this expression as previously and calculate the derivative at $r = r_k$ to obtain

$$\Big(\frac{d\dot n}{dr}\Big)_k = \frac{4\pi r_k^2}{kT}\Big(\frac{d\dot r}{dr}\Big)_k\Big[p_0 + \frac{4\sigma}{3r_k} + \frac{\gamma\eta}{3}\Big(\frac{d\dot r}{dr}\Big)_k + \frac{1}{3}\,\rho r^2\Big(\frac{d\dot r}{dr}\Big)_k^2\Big]. \tag{8}$$

Equating the right sides of Eqs.(7) and (8) yields the following cubic equation for $d\dot r/dr$:

$$\frac{\rho r_k^2}{3}\Big(\frac{d\dot r}{dr}\Big)_k^3 + \Big(\frac{4\eta}{3} + \frac{\beta u_t\rho r_k}{4(1+\delta_k)}\Big)\Big(\frac{d\dot r}{dr}\Big)_k^2 + \Big(p_0 + \frac{4\sigma}{3r_k} + \frac{\eta\beta u_t}{(1+\delta_k)r_k}\Big)\Big(\frac{d\dot r}{dr}\Big)_k - \frac{\sigma\beta u_t}{2(1+\delta_k)r_k^2} = 0.$$

By introducing the dimensionless variables

$$z = \frac{(1+\delta_k)r_k}{\beta u_t}\Big(\frac{d\dot r}{dr}\Big)_k, \quad \omega = \frac{3}{2}\,\frac{\beta u_t\eta}{\sigma(1+\delta_k)}, \quad \omega' = \frac{1}{3}\,\frac{(\beta u_t)^2\rho r_k}{\sigma(1+\delta_k)^2}, \quad b = \frac{2\sigma}{p_k r_k},$$

we nondimensionalize the equation

$$\omega' z^3 + \frac{8}{9}\,\omega\Big(1 + \frac{27}{32}\,\frac{\omega'}{\omega}\Big)z^2 + \frac{2}{3}\Big(\frac{3-b}{b} + \omega\Big)z - \frac{1}{2} = 0.$$

We assume that $\omega'/\omega^2 \ll 1$ (which is equivalent to neglecting the inertial terms) and obtain the equation

$$z^2 + \frac{3}{4}\,\frac{1}{\omega}\Big(\frac{3-b}{b} + \omega\Big)z - \frac{9}{16}\,\frac{1}{\omega} = 0.$$

We substitute the positive solution of this equation into (7) and then insert the result obtained into Eq. (4); this yields the following expression for the nucleation rate:

$$J = 3C_0\,\frac{\beta u_t}{1+\delta_k}\sqrt{\frac{\sigma}{kT}}\,\frac{1}{b}\Big(\omega + \frac{3+b}{b} + \sqrt{\Big(\omega + \frac{3-b}{b}\Big)^2 + 4\omega}\Big)^{-1}\exp\Big(-\frac{4\sigma r_k^2}{3kT}\Big). \tag{9}$$

Apart from viscosity, diffusivity, and the constant in the Henry law, the preexponent in (9) contains the condensation coefficient to be predicted by methods of the kinetic theory of gases. The resultant dependence calculated for different values of β is compared with dependence (2) in Fig. 1. The calculations were performed for the following values of physical parameters: $C_0 = 3.45 \cdot 10^{27}$ m^{-3}, $P_0 = 200$ MPa, $T = 1173$ K, $D_c = 2.3 \cdot 10^{-11}$ m^2/sec (these values were chosen to be in agreement with the experimental conditions of [8]), $\sigma = 0.08$ N/m, and $\eta = 10^{-3}$ Pa $\cdot$ sec. Variation of β from 1 to 10^{-5} weakly affects the value of the nucleation rate, and dependences (2) and (9) calculated with $\beta = 10^{-6}$ are almost coincident.

Generally, the condensation coefficient depends on the surface curvature, degree of supersaturation, etc. All the quantities in (9) are calculated for the critical nucleus. Assuming that dependences (2) and (9) differ insignificantly at low viscosity and that the condition of mechanical equilibrium is approximately fulfilled for transcritical bubbles (as in [2]), we determine the value of β from the condition of identical diffusion and kinetic fluxes of molecules at the surface of the transcritical bubble:

$$\dot n = \pi\beta u_t r^2(p_r - p)/(kT) = 4\pi r D_c(C_m - C_r)$$

and

$$\beta = \frac{4D_c kT}{u_t r}\,\frac{C_m - C_r}{p_r - p}.$$

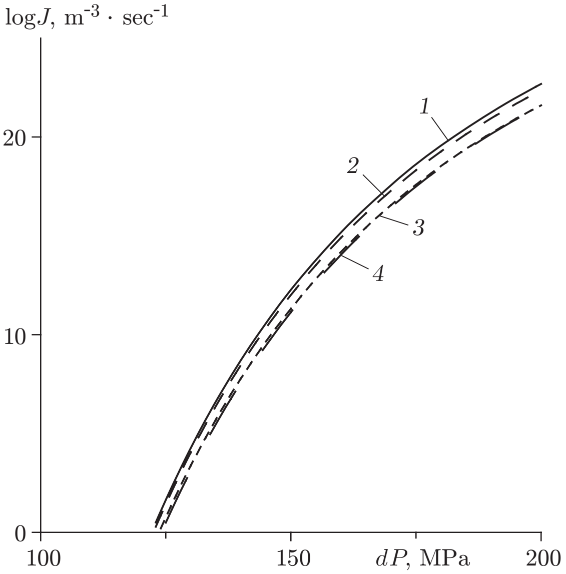

Fig. 1. Nucleation rate versus supersaturation pressure for different values of β: curves 1–3 refer to the calculation by formula (9) for $\beta = 1$ (1), 10^{-5} (2), and 10^{-6} (3); curve 4 refers to the calculation by formula (2).

As $r \to r_k$, we have $C_r \to C_m$ and $p \to p_r(C_m)$. With due allowance for this, we obtain the following expression for the condensation coefficient:

$$\beta = \frac{4D_c kT}{u_t r_k}\Big(\frac{dP}{dC}\Big)^{-1}_{C_m}.$$

With this choice of β, the formulas for the dimensionless parameters and the dependence obtained acquire the form

$$\omega = \frac{3}{2}\,\frac{\eta D_c kT}{\sigma r_k A C_m}, \qquad \omega' = \frac{1}{3}\Big(\frac{D_c kT}{AC_m}\Big)^2 \frac{\rho}{r_k\sigma}, \qquad b = \frac{2\sigma}{p_k r_k},$$
$$J = \frac{3C_0^2 D_c kT}{r_k p_0}\sqrt{\frac{\sigma}{kT}}\,\frac{1}{b}\Big(\omega + \frac{3+b}{b} + \sqrt{\Big(\omega + \frac{3-b}{b}\Big)^2 + 4\omega}\Big)^{-1} \exp\Big(-\frac{4\sigma r_k^2}{3kT}\Big), \tag{10}$$

and the condition $\omega'/\omega^2 \ll 1$ reduces to the condition $\omega'/\omega^2 = 4\rho\sigma r_k/(27\eta^2) \ll 1$, which is satisfied with high accuracy for typical values of physical parameters of magma. Thus, in calculating the preexponential factor, we take into account the contribution of pressure and viscous forces into the bubble dynamics, whereas the condition of mechanical equilibrium for the bubble is used only in estimating the condensation coefficient.

Dependences (10) and (2) calculated for different values of viscosity are compared in Fig. 2. The calculations were performed for the following values of physical parameters: $C_0 = 3.45 \cdot 10^{27}$ m^{-3}, $P_0 = 200$ MPa, $T = 1173$ K, $D_c = 2.3 \cdot 10^{-11}$ m^2/sec, and $\sigma = 0.08$ N/m. The dependences almost coincide for low viscosity, whereas the nucleation rate at $\eta = 10^4$ Pa · sec decreases by one or two orders, depending on the actual degree of supersaturation. The effect of viscosity is especially pronounced for $\eta = 10^6$ Pa · sec.

The mechanism of homogeneous nucleation of gas bubbles in magma was experimentally studied in [8, 9] at high pressure and temperatures typical of conditions in volcano channels. The experiments in [9] were conducted at an initial pressure of 200 MPa and initial temperature of 1073 K. In those experiments, however, the homogeneous nucleation regime was observed only in samples that contained (in addition to H_2O) a substantial amount of CO_2. The homogeneous nucleation regime in water-saturated samples was observed for the first time in [8], in experiments performed at a higher temperature (1173 K).

The values calculated by formula (10) for different values of surface tension are compared with the experimental data of [8] in Fig. 3, which also shows the experimental data corrected by the dependence for the nucleation duration reported in [10]. The nucleation rate was determined in [8] from the total decompression time. The time interval with intense decompression, however, is short as compared to the total decompression time. The nucleation

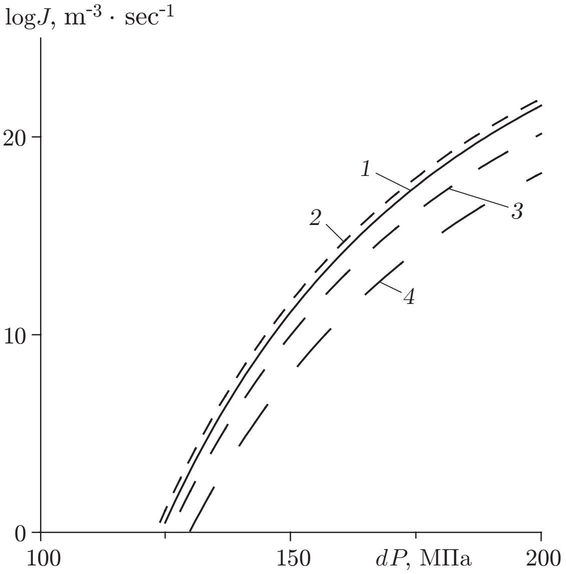

Fig. 2. Nucleation rate versus supersaturation pressure: curve 1 refers to the calculation by formula (2) and curves 2–4 refer to the calculation by formula (10) for $\eta = 10^2$ (2), 10^4 (3), and 10^6 Pa·sec (4).

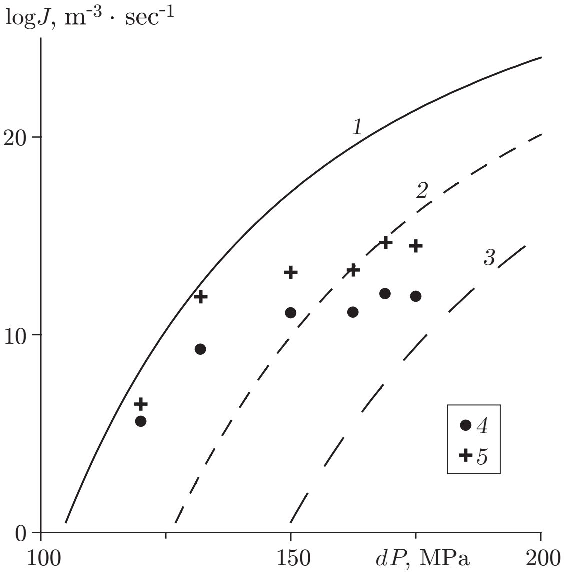

Fig. 3. Nucleation rate versus supersaturation pressure for different values of surface tension: curves 1–3 refer to the calculation by formula (10) for $\sigma = 0.07$ (1), 0.08 (2), and 0.09 N/m (3); points 4 and 5 refer to the experimental data of [9] and corrected data of [9], respectively.

rate measured in [8] was corrected by dividing the number concentration of bubbles by the nucleation duration calculated by the dependence derived in [10]. As it follows from Fig. 3, the corrected nucleation rate turns out to be two to three orders of magnitude higher. In performing the comparison, it should be taken into account that the experimental data obtained at moderate decompression rates (0.025 MPa/sec) could be affected by heterogeneous nucleation on the capsule walls. This circumstance was emphasized in [8], where the experimental data were processed with allowance for the residual water content in the samples. The results obtained for a decompression rate of 8.5 MPa/sec involve no such inaccuracies. With these remarks taken into account, it can be concluded that

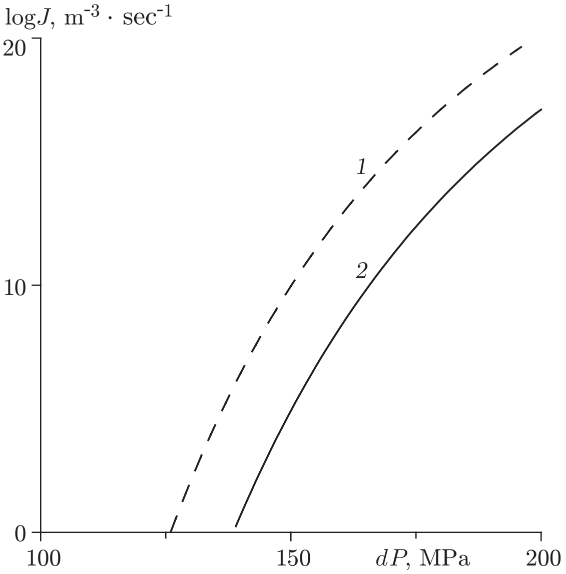

Fig. 4. Nucleation rate versus supersaturation pressure calculated by formula (10) for $\sigma = 0.08$ N/m: curve 1 refers to $T = 1173$ K and $C = 5.2\%$ [8] and curve 2 refers to $T = 1073$ K and $C = 4.6\%$ [9].

TABLE 1

Sample	dP, MPa	J_1, $\mathrm{m}^3/\mathrm{sec}$	J_2, $\mathrm{m}^3/\mathrm{sec}$
VGD6	160	$6.5 \cdot 10^{11}$	$2.18 \cdot 10^{8}$
VGD10	178	$1.2 \cdot 10^{16}$	$1.12 \cdot 10^{13}$
VGD13	190	$1.9 \cdot 10^{18}$	$2.81 \cdot 10^{15}$

$\sigma = 0.08$ N/m. This value is close to $\sigma = 0.076$ N/m for which good agreement between numerical results and the experimental data of [3] was obtained in [11]. The difference of $\sigma = 0.08$ N/m from the value $\sigma = 0.10$–0.11 N/m obtained in [8] with the use of Eq.(3) results from taking account of viscosity in relation (10).

The difference between the experimental results in [8] and [9] can be considered as an indication pointing to the effect of viscosity on the nucleation rate. The temperatures in those experiments differed by 100 K. This difference seem to be the reason for the fact that no homogeneous nucleation (in samples without CO_2) was observed in [9] for close initial values of water content and pressure. A 1-% decrease in mass concentration and a 100-K decrease in temperature in this range of parameters increase viscosity by an order of magnitude [4]. The nucleation rates calculated for $\sigma = 0.08$ N/m by formula (10) with due allowance for the difference in pressure and water content differ from each other by five to six orders of magnitude (Fig. 4). The nucleation rates J_1 and J_2 calculated by formulas (3) and (10), respectively, for typical experimental conditions of [9] are summarized in Table 1 (here, dP is the decrease in pressure experimentally observed in the indicated sample). The value of J_1 was calculated for $\sigma = 0.105$ N/m [this value of σ was used in [9] to perform estimates by Eq. (3)], whereas the value of J_2 was calculated for $\sigma = 0.08$ N/m. Under the indicated supersaturation, the value of J_2 is three orders of magnitude lower than J_1 even for much lower surface tension; this difference results from the effect of viscosity. Nonetheless, the estimated nucleation rates are much higher than the sensitivity threshold $J_0 = 10^4$ $\mathrm{m}^3/\mathrm{sec}$ reported in [9] and provide no unambiguous explanation to the absence of homogeneous nucleation in those experiments.

The formula derived in the present study takes into account the effect of viscosity on the rate of homogeneous nucleation in decompressed water-saturated magma. With an appropriate correction, this formula can be used to predict the effect of viscosity on the rate of heterogeneous nucleation. A comparison with experimental data confirms that viscosity has a pronounced effect on the rate of homogeneous nucleation in magma.

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