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The theory of degassing and swelling of a supersaturated-by-gas solution

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HIGHLIGHTS

- New comprehensive analysis of the degassing kinetics is given.
- Extended excluded-volume theory of degassing is formulated.
- There is no now limitation associated with gas supersaturation ratio.
- Formulas for swelling degree can be applied for different systems.

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ABSTRACT

The kinetic theory of degassing and swelling of a supersaturated-by-gas liquid solution under decompression has been formulated. The theory is based on the extended excluded volume approach to kinetics of gas-bubble nucleation. A description of the nucleation stage of supercritical gas-bubble formation with nonstationary nonuniform diffusion shells around the bubbles and mean-field mixing of the dissolved gas concentration at outer parts of the shells due to their stochastic overlapping has been built. In this way, the theory embraces the cases with any degree of initial gas supersaturation and deviation from the steady-state diffusion. It has been shown that the effects of nonstationary diffusion may be very significant in the growth of bubbles and, in particular, are responsible for a significant swelling of a supersaturated-by-gas liquid solution. Distribution of supercritical bubbles in sizes and gas concentration profiles at any moment of the nucleation stage as well as duration of the nucleation stage and the swelling ratio at the end of nucleation stage have been found.

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Introduction

The degassing of a supersaturated-by-gas liquid solution under decompression is a typical first-order phase transition in two-component medium. Fast degassing of solution can be accompanied by a considerable swelling—increasing the volume of the liquid solution due to addition of the volume of the gas phase inclusions. In the case of water vapor dissolved in a magma melt, such swelling at intensive formation of water vapor bubbles inside magma may lead to explosive volcanic eruption [1]. The theory of degassing and swelling mechanisms under fast decompression is important for developing the optimal methods of molding porous materials and polymer foams, for preventing the decompressing damage in human blood and tissues [2,3].

To describe degassing and swelling in supersaturated solutions, we should consider the kinetic frameworks of homogeneous gas bubble nucleation. The traditional approach [4] to the kinetic description of one of the most important

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stages of nucleation and growth of gas bubbles, the stage of formation of size spectrum of supercritical bubbles (i.e., the bubbles larger than one in unstable equilibrium with solution) and their total number, assumes that nucleation of supercritical bubbles proceeds with synchronous and uniform throughout the volume of the solution decrease of the dissolved gas supersaturation (the mean-field approximation for gas supersaturation). Accordingly, the rate of formation of new bubbles is reduced uniformly throughout the solution volume. It is also believed in the mean-field approximation for gas supersaturation, that the diffusion of dissolved gas molecules to a growing bubble is stationary.

First of all, let us note that the assumption of a uniform (throughout the solution volume) decrease in the concentration of dissolved gas is certainly not satisfied at the beginning of the nucleation stage, when, due to the small number of nucleating bubbles, their nonstationary diffusion shells do not overlap. Furthermore, using the assumptions of stationary diffusion of dissolved gas in solution is very restrictive since the bubble growth depends on the degree of gas supersaturation of the solution and may be strongly nonstationary at high supersaturation [5]. These facts significantly affect both the growth dynamics of each bubble and the spatial distribution of the gas molecules around the bubbles.

To account for the influence of the dissolved gas heterogeneity and unsteady diffusion to nucleating bubbles, we previously formulated [6] an approach based on the idea of the excluded volume in the nucleation process. The approach of the excluded volume takes into account that, in a non-uniform diffusion shell surrounding the growing bubble, birth of new bubbles is strongly suppressed due to lowering concentration of the dissolved gas. In other words, a some spherical shell of liquid solution around each bubble can be excluded from the scope of nucleation, while in the rest of the solution, the intensity of nucleation of supercritical bubbles remains at the initial level. A similar situation exists at droplet nucleation in a supersaturated vapor, when nucleation of new supercritical droplets is suppressed in the vicinity of a growing droplet [7]. Under excluded volume approach, nucleation of supercritical bubbles in a highly supersaturated solution had previously been discussed with using the assumption that, in case of strong gas supersaturation, the excluded volumes of individual bubbles in solution are thin spherical shells [8] which do not overlap. However, the kinetic approach of the excluded volume had recently been extended [9,10] to describe isothermal and nonisothermal nucleation of supercritical droplets in a supersaturated vapor with allowing a possibility of overlapping diffusion shells around individual droplets which can occur on the final part of the nucleation stage. Note that in the case of vapor condensation on droplets, the vapor diffusion to droplets, because of the strong differences in densities of the vapor and liquid in droplet, is close to the stationary process. Besides, the large density of liquid solution and corresponding high thermal conductivity of the solution weakens the thermal effects of bubble nucleation and allows us do not consider them.

The first goal of this work is to develop the extended approach of excluded volume to describe an isothermal nucleation in a gas-supersaturated liquid solution at any degree of gas supersaturation and, correspondingly, at arbitrary deviation from the stationary regime of gas diffusion to supercritical bubbles. It should be pointed out, that unlike the condensation growth of droplets in a supersaturated vapor, the effects of nonstationary diffusion may be very significant in the growth of bubbles. This can result, in particular, in a significant swelling of a supersaturated-by-gas liquid solution. The accurate description of arbitrary swelling ratio on the nucleation stage of bubble formation is the second goal of our work.

The paper is organized as follows. First, we will recall the basic results of the theory of the nucleation stage of supercritical bubble formation in the mean-field approximation for gas supersaturation and steady-state diffusion growth of supercritical bubbles. Then we will consider the non-stationary diffusion growth of bubbles and corresponding swelling phenomena within the extended excluded-volume approach. The conclusions will be formulated in the final section.

1. Bubble formation and solution swelling on nucleation stage under mean-field approximation

The mean-field approximation assumes that the solution (liquid together with the dissolved gas and gas bubbles) is considered an effective homogeneous medium where an excess of dissolved gas over saturated solution decreases with time under the condition that the total number of gas molecules stays fixed. Together with uniform decreasing of the dissolved gas concentration, there is a corresponding uniform-over-volume decrease in intensity of formation of new viable supercritical bubbles (the bubble nucleation rate) until the complete cessation of this intensity.

The expression for the bubble nucleation rate $I(\zeta)$ as a function of gas supersaturation ζ can be written as

$$I(\zeta) = A(\zeta)\mathrm{e}^{-\Delta F(\zeta)}, \tag{1}$$

where ΔF is the work of bubble formation, the pre-exponential factor $A(\zeta)$ is a slow varying function of ζ. The gas supersaturation at time moment t is related to the dissolved gas concentration in solution as $\zeta(t) = (n(t) - n_\infty)/n_\infty$ where $n(t)$ is the mean gas concentration in solution at the same time moment t, n_∞ is the gas concentration in saturated solution.

If relative decrease

$$\varphi(t) \equiv (\zeta(0) - \zeta(t))/\zeta(0) \tag{2}$$

of the gas supersaturation from its initial value $\zeta(0)$is small, i.e., $\varphi \ll 1$, then expression (1) for the nucleation rate can be rewritten as

$$I(\zeta) = I(0)\,\mathrm{e}^{-\Gamma\varphi}. \tag{3}$$

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