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Melt cavitation at its volumetric crystallization

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ABSTRACT

It is shown that at volumetric crystallization of the undercooled melt the high negative pressures are generated in this melt; they are caused by matter shrinkage at solidification, what leads to intensive cavitation of uncrystallized melt. The mechanisms are studied and the kinetic model of this process is presented here. Considerable dependence of cavitation intensity on the cooling rate is shown. Numerical solutions to the problem are found at the example of cavitation of crystallizing metal melts. The sizes of crystalline grains and formed cavitation inclusions in the solidified material are determined.

1. Introduction

There are many reasons for intense liquid cavitation, and all of them somehow relate to generation of tensile stresses (negative pressures) in this liquid. For instance, cavitation takes place at static expansion of liquid, in decompression wave, propagating in liquid (gas–liquid) medium, in the accelerating liquid flow, etc. This phenomenon is also observed at melt crystallization.

One of possible mechanisms of cavitation of gas-saturated melts relates to the fact that since gas solubility in the solid phase is significantly less than in the liquid phase, gas dissolved in the melt will be displaced by the moving fronts of crystallization (the so-called gas segregation in the melt). When its concentration near crystallization surface exceeds the critical one, corresponding to equilibrium concentration at the given temperature and pressure, nucleation and growth of gas bubbles start; then these bubbles are entrained by the solid phase, and, finally, the solidified material has the porous structure. Many works deal with the study of this problem (e.g., see [1–5]). However, most of them consider the case of equilibrium successive crystallization (the flat front of solidification) with very low rates of crystal growth.

At higher cooling rates melt crystallization occurs in the result of fluctuation nucleation and growth of new phase sites in the melt volume [6]. This situation can be achieved, for instance, at ultra rapid quenching of microvolumes, in particular, at spraying of melt microdroplets on a cold surface [7,8]. It was noted in some studies dealt with experimental investigation of this process that micropores and cracks in the solidified material can be caused by matter shrinkage during solidification [9–13]. Not sufficient attention is paid to theoretical modeling of this phenomenon in literature.

The current study is aimed at investigation of the process of melt cavitation at its volumetric crystallization. The problem considered in the paper is interesting both from the point of science and practice. The phenomenon observed in experiment is angled theoretically towards fundamental science; this phenomenon has not had the complete explanation until now. From the point of practice the ability of correct calculation of the microstructure of solidified material is required for some technologies based on the method of ultra-rapid quenching from the liquid state and aimed at production of new promising materials and coatings with given properties, such as nano- and microcrystalline, bioactive and other ones; since namely the microstructure of materials determines mainly their mechanical, physical–chemical, electrical and other properties.

2. Model of the process

As it was already mentioned in Section 1, the phenomenon of liquid cavitation relates to its volumetric expansion, which causes nucleation and growth of vapor-gas bubbles. At this, intensity of cavitation processes depends directly on the value of tensile stress (negative pressure) preceding nucleation of cavitation sites, which in turn is proportional to relative expansion of liquid:

$$\Delta p = -\frac{1}{\beta}\frac{\Delta V_l}{V_l}, \tag{1}$$

where ΔV_l is a change in initial volume of liquid at its expansion; V_l is liquid volume; β is coefficient of volumetric expansion.

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Nomenclature

a	thermal diffusivity ($m^2\, s^{-1}$)
B	kinetic coefficient (s^{-1})
C	heat capacity ($J\, kg^{-1}\, K^{-1}$)
f	function of size distribution of cavitation bubbles
J	nucleation frequency ($m^{-3}\, s^{-1}$)
K	kinetic coefficient ($ms^{-1}\, K^{-1}$)
Ku	Kutateladze number, $L/(c\Delta T)$
K	shrinkage coefficient
k_g	coefficient
k_B	Boltzmann constant ($J\, K^{-1}$)
L	specific heat of melting ($J\, kg^{-1}$)
N	number of sites of a new phase per a volumetric unit (m^{-3})
N_m	number of molecules per a unit of melt volume (m^{-3})
N_h	number of molecules on the surface of heterogeneous particle (m^{-3})
q	rate of melt cooling ($K\, s^{-1}$)
Δp	tensile stress (Pa)
R	nucleus radius (m)
$\bar{R}$	average nucleus radius (m)
r_T	thickness of temperature boundary layer around a crystal (m)
T	temperature (K)
T_{mel}	melting temperature (K)
ΔT	melt undercooling, $T_{mel} - T$(K)
t	time (s)
V	volume (m^3)
V_T	volume of "forbidden" region around a crystal (m^3)
v_0	typical rate of crystal growth (ms^{-1})
W^*	work of nucleation of a critical nucleus during homogeneous process (J)
X	part of crystalline mass
X_T	volume of "forbidden" region per a unit of melt volume

Greek symbols

β	coefficient of volumetric expansion (Pa^{-1})
θ	equilibrium wetting angle
λ	heat conductivity ($W\, m^{-1}\, K^{-1}$)
ν	kinematic viscosity ($m^2\, s^{-1}$)
ρ	density ($kg\, m^{-3}$)
σ	surface tension ($J\, m^{-2}$)
ϕ	volumetric concentration of cavitation inclusions
ψ	function of θ

Subscripts

cav	meets cavitation process
cr	meets crystallization process
g	gas phase
l	liquid phase
s	solid phase

Supersripts

i	initial state
f	final state

It is shown in [6] that limit expansion Δp^*, when liquid starts cavitating intensively, can be estimated with accuracy acceptable for practice with the help of the theory of homogeneous nucleation:

$$\Delta p^* = -\left(\frac{16\pi\sigma_{lg}^3}{3k_BT\ln[N_mB_{cav}/J_{cav}]}\right)^{1/2}, \quad (2)$$

where J_{cav} is frequency of cavitation site nucleation, described below. However, in some experiments limit expansions of liquid are lower than these values. This relates to the fact that real liquids always include micrononuniformity in the form of microbubbles of free gas and solid microparticles, whose volumetric concentration is estimated by different authors from 10^8 to $10^{12}\, m^{-3}$[14]. Naturally, all these microparticles (which are in fact the heterogeneous sites) play the role of cavitation nuclei. If in solid body mechanics the destruction process starts from defect generation, in liquid the structural defects in the form of cavitation sites exist from the very beginning. This means that cavitation of real liquids has always the heterogeneous character. However, only heterogeneous nucleation is observed in the case, when liquid is expanded relatively slow. With a rise of expansion rate (what brings liquid to the range of deeper metastable states) the probability of homogeneous nuclei generation in the volume increases significantly, and finally, the contribution of a homogeneous component to the total process of nucleation becomes predominant [15]. At this, heterogeneous nucleation takes place as before, but its intensity becomes significantly less than that of the homogeneous process. This is caused by the fact that despite the work of critical nucleus formation in the homogeneous process is higher than that in the heterogeneous process, the number of potential nucleation sites in the first process is considerably higher than in the second.

As it was already mentioned in the Introduction, the phenomenon of cavitation is observed at volumetric (spontaneous) crystallization of undercooled melts, what relates to their shrinkage at solidification. The latter leads to generation of high negative pressures in uncrystallized melt and, as a sequence, to nucleation and growth of cavitation bubbles (naturally, this phenomenon is observed only for substances whose solid phase density is higher than that of their liquid phase). It is shown schematically in Fig. 1. The goal of our research is investigation of this cavitation mechanism.

Let's consider the melt volume, which was initially cooled fast relative to the equilibrium temperature of melting. At this, we con-

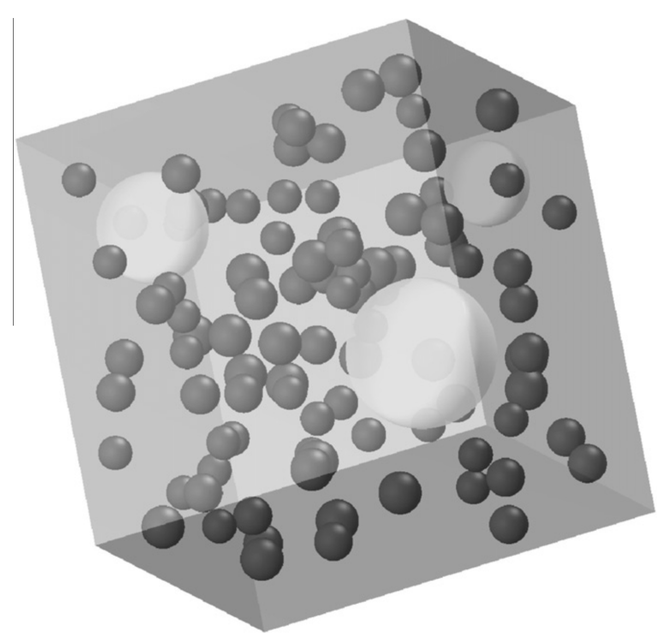

Fig. 1. Schematic drawing of melt cavitation process at its volumetric crystallization: black balls, crystals; white balls, bubbles.