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Mechanisms of Acoustic Processing of a Metal Melt Containing Nanoparticles

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Abstract. Wave processing with the frequencies from subsound (vibration) to ultrasound is used to produce nanopowder-modified composite alloys. This work considers mechanisms of such processing of metal melts, which lead to deagglomeration and wettability of particles of a metal melt and to the destruction of growing crystals during solidification. The main dependences for the threshold of the turbulence and cavitation were obtained. Resonance phenomena that contribute to positive changes in the melt are discussed. Possible mechanisms of the destruction of growing crystals and agglomerates of particles at the high-frequency processing of the melt are considered, including the destruction of agglomerates in the front of an acoustic wave and the destruction of crystals by oscillating solid particles.

INTRODUCTION

A perspective direction in metallurgy is related to the modification of metals and alloys with solid refractory nanoparticles. Such modification allows producing metals with new properties, including for use in the extreme external environment, for the aerospace industry, etc.

The introduction of nanoparticles to a molten metal is accompanied by great difficulties [1]. Particles are badly wetted with fluid; because of their large surface energy, nanoparticles coagulate and form agglomerates. The molten metal containing such agglomerates can be considered as a three-level hierarchical structure (metal, agglomerates, particles). Oscillating effects (from subsound and vibration to ultrasound) are used for the introduction of nanoparticles to the melt and improvement of the quality of casting [2–4].

The purpose of this work is to consider possible mechanisms of destruction of agglomerates and reduction of a crystal grain, to offer a mathematical description of the effects accompanying acoustic processing of a melt with particles, and to define optimum modes of such processing.

DESTRUCTION OF CRYSTALS AT LOW-FREQUENCY PROCESSING OF A MELT

In the low-frequency processing of melts composed of alloys, it is necessary to provide a turbulent mixing of the whole mass of the melt. At the low-frequency (long-wave) processing in comparison with ultrasonic ones, the processed volume can apparently be increased in proportion to the wavelength.

Microflows in the liquid metal during solidification hinder the dendrite growth and lead to the grain reduction. On the other hand, cavitation, which may also be realized under certain conditions in the liquid metal, causes the same effect. Microflows also contribute to the uniform distribution of particles in the metal volume, and the cavitation contributes to the destruction of agglomerates and wetting of particles [3].

Thus, under the low-frequency vibration processing of the melt two phenomena contribute to the destruction of the growing crystals: turbulent mixing and cavitation.

Ignatyev et al. solve the problem of motion of liquid particles in the field of elastic vibrations [5]. The cavitation mode is realized in the melt volume when the critical frequency of vibration is reached.
where $f$ is the frequency and $A$ is the amplitude of vibrations, $g$ is the gravitational acceleration, $\Delta p$ is the pressure difference, and $\rho_l$ is the liquid density. The turbulence in the melt will occur under vibrations, for which the acoustic Reynolds number exceeds a certain critical value

$$Re = \frac{\rho_c V}{\omega b} \approx \frac{\rho_c A}{b} > Re_{cr},$$

(2)

where $V = A \omega$ is the characteristic scale of the vibrational velocity, $\omega = 2\pi f$ is the circular frequency, $c$ is the sonic speed, $b = a + v$ is the dissipation parameter proportional to the sum of thermal diffusivities $a$ and kinematic viscosity $v$ of the melt.

The amplitude required for the turbulence mode, regardless of the impact frequency (2), but the amplitude required for the cavitation mode, decreases with increasing frequency (1). A sufficiently high frequency, at which the amplitudes of turbulence and cavitation match, will be optimum ($f_{opt}$)—two useful modes are realized at that frequency.

The intensity of the wave field $I$ is related to the frequency $f$ and oscillation amplitude $A$ as

$$A = \frac{1}{\pi^2} \sqrt{\frac{I}{2c\rho_l}}.$$

(3)

From expressions (1)–(3) follows minimum intensity (and amplitude) and optimum frequency of exposure, which are sufficient for creating cavitation regimes and turbulence in the melt

$$I_{min} = \frac{\pi^2}{16} \Delta p c, \quad A_{min} = \frac{1}{\pi^2} \sqrt{\frac{I_{min}}{2c\rho_l}}, \quad f_{opt} = \frac{\Delta p}{32\rho_l (\nu + a) Re_{cr}}.$$

(4)

At $f_{opt}$ both useful mechanisms—turbulence and cavitation—are realized, which yields favorable conditions for grain refining. This explains the well-known experimental fact [6, 7] that with increasing frequency of vibration processing (up to a certain limit) the microstructure of the casting is improved. With a frequency increase, maintaining of the given amplitude of the processing requires too larger intensity, according to equation (3), which is not always accessible to the used devices. Therefore, the amplitude becomes insufficient for the realization of the useful modes, and the effect decreases.

As appears from expressions (4), both the required amplitude and frequency of vibroprocessing significantly depend on the metal properties, namely, speeds of sound, density, viscosity, and heat diffusivity. Therefore, for different metals and alloys these parameters will be different. The calculated values of optimum frequency and minimum amplitude of the processing are given in Table 1 for various metals.

### SATURATION OF PARTICLES BY MOLTEN METAL AT ACOUSTIC PROCESSING

Modifying nanoparticles introduced into molten metal are micron-sized agglomerates that can be deagglomerated. Agglomerates contain pores, channels, and cracks.

Acoustic processing makes it possible to create the mode of the developed cavitation with many cavitation bubbles pulsing and collapsing in the liquid metal at any given moment. The cavitation bubble collapse creates a pressure impulse $p_{ex}$ proportional to the intensity of ultrasound. If all particles have a spherical form, the same size, and pores (capillaries) of the identical radius and depth, the excessive pressure $p_{ex}$ will cause the movement of the meniscus in a capillary. Kudryashova and Vorozhtsov obtained the expression for time of saturation of particles by the molten metal [3]

### TABLE 1. Optimum frequency, intensity and minimum amplitude of the vibration processing

<table>
<thead>
<tr>
<th>Metal</th>
<th>Ni</th>
<th>Pb</th>
<th>Bi</th>
<th>Al</th>
<th>Sn</th>
<th>Li</th>
<th>Cu</th>
</tr>
</thead>
<tbody>
<tr>
<td>$f_{opt}$, Hz</td>
<td>85.05</td>
<td>56.42</td>
<td>41.66</td>
<td>73.90</td>
<td>42.82</td>
<td>114.80</td>
<td>13.18</td>
</tr>
<tr>
<td>$I$, MW/m²</td>
<td>294.55</td>
<td>110.30</td>
<td>101.68</td>
<td>289.63</td>
<td>141.73</td>
<td>299.49</td>
<td>213.21</td>
</tr>
<tr>
<td>$A$, mm</td>
<td>3.95</td>
<td>5.45</td>
<td>7.76</td>
<td>8.79</td>
<td>8.91</td>
<td>12.70</td>
<td>25.90</td>
</tr>
</tbody>
</table>
Table 2. Time of ultrasonic cavitation processing of the aluminum melt for deagglomeration of γAl₂O₃ particles: experiment [8] and calculation by formula (5)

<table>
<thead>
<tr>
<th>D, μm</th>
<th>t, min (experiment [8])</th>
<th>t, min (calculation)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.01–0.10</td>
<td>37.5</td>
<td>810</td>
</tr>
<tr>
<td>0.1–1.0</td>
<td>14.0</td>
<td>20.25</td>
</tr>
<tr>
<td>1–10</td>
<td>9.1</td>
<td>3.24</td>
</tr>
<tr>
<td>10–20</td>
<td>4.1</td>
<td>0.26</td>
</tr>
<tr>
<td>80–100</td>
<td>1.5</td>
<td>0.01</td>
</tr>
</tbody>
</table>

\[ t = B \frac{nSDp}{\rho}, \]  
\[ p = p_{ex} + \frac{2\sigma \cos \theta}{r} = p_{ex} + \Delta p_L. \]

where the coefficient \( B \) depends on the structure and shape of pores, \( S \) is the specific surface area of powder, \( D \) is the particle diameter, \( \rho \) is the particle density, and \( p \) is the summary pressure. \( \Delta p_L \) is the Laplace capillary pressure (negative quantity in case of poor wettability and low \( D \)), \( \theta \) is the wetting angle, and \( \sigma \) is the surface tension. In the case of the uniform filling of the agglomerate volume with identical conical pores, \( B = 28.4 \).

The experimental dependence of the time of ultrasonic processing on the particle size for γAl₂O₃ in the 99.99% aluminum melt at the temperature from 700 to 720°C at an acoustic power of 500 W was obtained [8]. It shows that the required time of processing increases with a decrease in the particle diameter. We used formula (5) for \( p = 50 \) Pa. Table 3 shows the calculated and experimental results.

**DESTRUCTION OF AGGLOMERATES IN THE ACOUSTIC WAVE**

Suppose an agglomerate is at the front of the acoustic wave. Then, according to Rozenberg [9] we analyze forces acting on it and obtain an expression for the threshold intensity for breakup

\[ I = 2c\rho_l \left( \frac{\sigma_{st}}{D\omega D_0} \right)^2, \]  

where \( \sigma_{st} \) is the tensile strength of the agglomerate, \( \omega \) is the ultrasonic frequency, \( c \) is sound velocity in liquid metal, and \( \rho_l \) is liquid density. Here, wave impedance \( c\rho_l \) in the mode of developed cavitation is approximately three times lower than without any exposure [9], and the tensile strength of particle agglomerates is orders of magnitude lower than for a monolithic particle.

**RESONANCE PHENOMENA**

Disturbance with sound speed \( c \) extends from an acoustic source through the melt volume. If the wavelength \( \lambda = 2L \), where \( L \) is the height of the melt bath, then a resonance will be established. It corresponds to the frequency \( f = c/\lambda = c/2L \). There can also be resonances connected with higher harmonics of oscillations, multiple influencing frequency if it is equal to the relation \( c/2L \). Resonance oscillations of the liquid and particles in it reach the maximum amplitude. The resonant frequency grows with a decrease of the melt thickness. So, for an aluminum melt 0.1 m thick the resonant frequency is \( f = 23.5 \) kHz, and for that 0.5 m thick it is \( f = 4.7 \) kHz.

**DESTRUCTION OF CRYSTALS BY OSCILLATING SOLID PARTICLES**

A particle weighed in the liquid in the acoustic field is involved into the oscillating motion. Depending on the properties of the environment, sizes and density of the particle, it can be dragged by the liquid with different degree that is defined by the drag coefficient \( k_{drag} \) (the powder particle speed amplitude related to the environment particle speed amplitude).
TABLE 3. Drag coefficient of diamond particles 10 μm in diameter under ultrasonic processing

<table>
<thead>
<tr>
<th>$f$, kHz</th>
<th>12</th>
<th>13</th>
<th>14</th>
<th>15</th>
<th>16</th>
<th>17</th>
<th>18</th>
<th>19</th>
<th>20</th>
<th>21</th>
</tr>
</thead>
<tbody>
<tr>
<td>$k_{drg}$</td>
<td>0.90</td>
<td>0.88</td>
<td>0.87</td>
<td>0.85</td>
<td>0.84</td>
<td>0.82</td>
<td>0.81</td>
<td>0.79</td>
<td>0.78</td>
<td>0.76</td>
</tr>
</tbody>
</table>

For the Reynolds number $Re = DV/\nu > 1$, where $V$ is the speed of motion of an environment relating to the particle, $\nu$ is the kinematic viscosity, Oseen obtained the expression for the drag coefficient \[10\]

$$k_{drg} = \frac{1}{\sqrt{1 + (\omega \tau / (1 + 3/8 Re))}}$$

where $\tau = \rho D^2/18\eta$ is the relaxation time of the particle.

The drag coefficient is equal to 1 in the subsound frequency range, and powder particles are completely dragged by the melt and move with the speed and amplitude equal to the speed and amplitude of the liquid particles. On the contrary, under ultrasonic frequencies of the processing the drag coefficient can significantly differ from 1. For a diamond particle 10 μm in diameter, the drag coefficient is given in Table 3.

Powder particles in their movement are behind the movement of liquid particles. It contributes to the destruction of arising metal crystallites. It can be considered as one of the mechanisms of grain reduction during ultrasonic processing of melts containing solid particles of micron-sized powders.

CONCLUSION

Mechanisms of the refinement of crystal grains and destruction of agglomerates of particles in metal melts modified by nanoparticles under acoustic processing are considered. It is shown that at the low-frequency processing of the metal melts such mechanisms could be turbulence and cavitation. Formulae for the optimum frequency and amplitude for the processing were obtained.

Under the ultrasonic processing of the melt, because of the cavitation mode, agglomerates of modifying particles are saturated by the liquid metal and break up in the front of the acoustic wave. The phenomenon of an acoustic resonance in the molten metal is considered. Formula for the time of processing is obtained. A comparison of the calculated values of the saturation time of aluminum oxide particles by liquid aluminum with the experimental data indicates the adequacy of the proposed model. It was shown that the mechanism of destruction of arising crystals by solid particles is realized at high frequencies of processing when a lag of particle motion in a wave is considerable. The proposed formulae will allow estimating parameters of acoustic processing of the molten metal modified by particles to improve the structure of casting.

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