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# Ultrasonic Dispersion of Agglomerated Particles in Metal Melt

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**Abstract.** This work considers the deagglomeration and wettability of particles by metal melt and proposes a mechanism of particle agglomerate dispersion by ultrasonic cavitation. The main dependences connecting the processing time and intensity with the physical and chemical properties of particles and the melt as well as acoustic parameters are obtained. For the first time found that melt during ultrasonic treatment, inclusive the particles agglomerates proportional to melt viscosity and the size of the agglomerates. It has been established that time ultrasonic treatment melt containing the particles agglomerates is proportional to melt viscosity and the size of the agglomerates. The required time for successful melt infiltration in the agglomerates, wettability and their introduction into the melt takes ten minutes. The suggested equation allows estimating the intensity of ultrasonic radiation, required to destroy the agglomerates of particles in the melt. It was found that intensity of the ultrasound must be inversely proportional to the radius of the agglomerates. The theoretical results are confirmed by comparing with experimental dates.

## INTRODUCTION

There are specific problems associated with the introduction of nanopowders into the molten metal when creating composite alloys [1]. This happens because nanoparticles are not wetted by the melt and, therefore, it is next to impossible to introduce them directly into the melt. Ultrasonic processing is experimentally proved to facilitate the introduction of fine particles into the melt and their homogeneous distribution in the liquid [2-4]. For processing of metallic melt and introduction of particles into the melt in practice use ultrasonic field [2, 3, 10]. It is shown that ultrasonic cavitation promotes deagglomeration and better distribution of nonmetallic particles in the liquid. One of hypotheses explaining the increased apparent wettability connects it to the improved access of the melt to a surface of nonmetallic particles and penetration of the melt into defects of a surface (capillaries) with the ultrasonic cavitation [3].

Scientists connect positive effects of ultrasonic processing of metal melt containing nanoparticles with ultrasonic cavitation. In this work, we describe theoretically the process of particle deagglomeration and improvement of effective wettability through the sono-capillary effect [5] caused by ultrasonic cavitation. A model based on the analysis of capillary effects and cavitation allows explaining known effects for accelerating the introduction of particles in the melt by ultrasound. The analysis of such a model makes it possible to estimate the important parameters of the ultrasonic melt treatment.

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It is fair to assume that the acoustic field facilitates the impregnation of capillary channels and pores of agglomerated particles, even though their surface is poorly wetted by the liquid. Cavitation bubbles, collapsing near agglomerates, create overpressure, which is higher than the negative capillary pressure. As a result, liquid metal penetrates the pores and cracks of the agglomerate, which facilitates its dispersion under the acoustic wave.

The purpose of this work is to consider a possible mechanism of deagglomeration under ultrasonic melt processing.

## MATERIALS AND METHODS

### Mathematical Model

The nanoparticles exist in the form of micron and submicron agglomerates (particle diameter about 0.1 ÷ 10 μm), since due to their agglomerated advanced for directly their surface area creation after synthesis [2]. Such agglomerates contain micro- and nano-pores. The specific surface area of these nanopowders is ~ 350 $m^2/g$, and they are containing the mass of gas comparable to the mass of the particles themselves. If the particles are not wetted by the liquid melt, they will float on the liquid surface due to adhering of air bubbles, which are difficult or impossible to remove by simple stirring.

So, nanopowder which containing micron size agglomerates can be deagglomerated when its introduced in to the metal melt [4, 11]. Agglomerates contain pores, channels, and cracks. It is known that ultrasound increases efficiency of filtration and impregnation of filters and preforms [4, 5, 11]. The intensification of these processes is related to the excessive pressure formed close to the capillary entry by pulsating and imploding cavitation bubbles, so-called sono-capillary or ultrasonic capillary effect (UCE), that leads to the sharp increase of depth and speed at which liquid penetrates into micro-capillary channels (e.g., filtration channels, cracks, pores). Ultrasonic cavitation is the leading phenomenon in this effect.

Ultrasonic processing makes it possible to create the mode of the developed cavitation with many cavitation bubbles pulsing and collapsing in liquid metal at any given moment. The ratio of the volume of bubbles to that of the cavitation zone approaches unity in this regime [6], which means that each agglomerate is surrounded by collapsing bubbles sized 50÷300 μm (at a frequency of 17.5 kHz) [7]. So at any point of time, at least one cavitation bubble will implode near a capillary (pore, or crack). It will create a pressure impulse $p_{ex}$ proportional to the intensity of ultrasound [6].

If all particles have a spherical form, the same size and pores (capillaries) of identical radius and depth, the excessive pressure $p_{ex}$ will cause the movement of the meniscus in the capillary with the following velocity:

$$v = \frac{r^2 p}{8\eta\, l} \tag{1}$$

where $r$ is the capillary radius, η is the viscosity of the melt, $l$ is capillary length filled with the melt, and $p$ is the total pressure

$$p = p_{ex} + \frac{2\sigma\cos\theta}{r} = p_{ex} + \Delta p \tag{2}$$

Here $\Delta p$ is the Laplace capillary pressure, (greater negative quantity in case of poor wettability and low $r$), θ is the wetting angle, and σ is the surface tension.

Under the conditions of $p_{ex} > \Delta p$, the capillary will be filled with liquid metal after some time. This time can be obtained by integrating Eq. 1:

$$t = \frac{4\eta l^2}{r^2 p} \tag{3}$$

The ratio of the length of the pores to their radius is proportional to the specific surface of powder $S$, diameter of agglomerates $D$, and density of particles $\rho$. We can determine this value using a geometrical model. For example, if



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the agglomerate is evenly filled with identical cone-shaped pores, the relation holds: $\frac{l}{r} = \frac{8}{3} SD\rho$. Then the time of agglomerate impregnation is as follows:

$$t \approx A \frac{\eta SD\rho}{p} \tag{4}$$

where coefficient $A$ depends on the structure and shape of the pores; in the simple geometrical model considered above, $A$=28.4.

Suppose the agglomerate is at the front of the acoustic wave. Then, following Ref. [6], we analyze the forces acting on it and obtain an expression for the threshold intensity for breakup:

$$I = 2c\rho_l \left( \frac{\sigma_{st}}{D\omega\rho_l} \right)^2 \tag{5}$$

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 [6], and the tensile strength of particle agglomerates is orders of magnitude lower than for a monolithic particle.

## Experimental Procedure

Aluminum alloy alumina nanoparticles containing produced by chill casting method. An aluminum alloy A356 was melted in a graphite crucible with a total melt volume of 700 g. The ultrasonic processing was performed using a 5-kW water-cooled magnetostrictive transducer (PMS 15A-18) with a working frequency of 17.1 kHz and with a Nb sonotrode. First, ultrasonic degassing was performed at a melt temperature of 760°C for 1 minute. Then, the preformed mixture Al-10 wt% $Al_2O_3$ was introduced into the ultrasonic cavitation zone in the melt, directly under the sonotrode. After the introduction of the preformed mixture, the melt was treated with ultrasound for another 2 minutes at 720-740 °C. Then the obtained mixture was poured at 710 °C into a metallic mold with the cavity size of dia. 30 × 200 mm. The nominal nanoparticles content in the alloy was 0.2 and 1 wt%.

## RESULTS AND DISCUSSION

### Saturation Time of Agglomerated Particles with the Melt

The possibility of impregnating particle agglomerates with liquid metal and saturation time primarily depends on the value of total pressure $p_{ex}$+ $\Delta p$. The authors in [8] gaged the values of excess pressure $p_{ex}$ created in capillaries when cavitation bubbles implode as a result of ultrasonic cavitation in the aluminum melt. Such pressure may reach very high values (depending on the proximity of the imploding bubble to the entry of a capillary) but the average is 40÷50 MPa.

In the case of poor wettability of the particle surface with liquid, the capillary pressure $\Delta p$ takes negative values can reach magnitudes of the same order as $p_{ex}$: according to formula (1), 50-400 MPa for r=5÷30 nm in the system of liquid aluminum–aluminum oxide (wetting angle $\theta$=150°, surface tension $\sigma$=0.9 N/m).

The impregnation time for an agglomerate in accordance with (4) depends on the specific surface, diameter and density of the particles and viscosity of the melt and inversely proportional to the pressure $p=p_{ex}+\Delta p$.

In experiments with fine particles introduced into metal melt, the duration of ultrasonic processing is measured in minutes. This corresponds to a situation when excess pressure and capillary pressure have unlike signs and the same order of magnitude: $p_{ex} \sim -\Delta p$. If both of these values are about tens of megapascals, the resultant pressure will reach $p$~10÷50 Pa.



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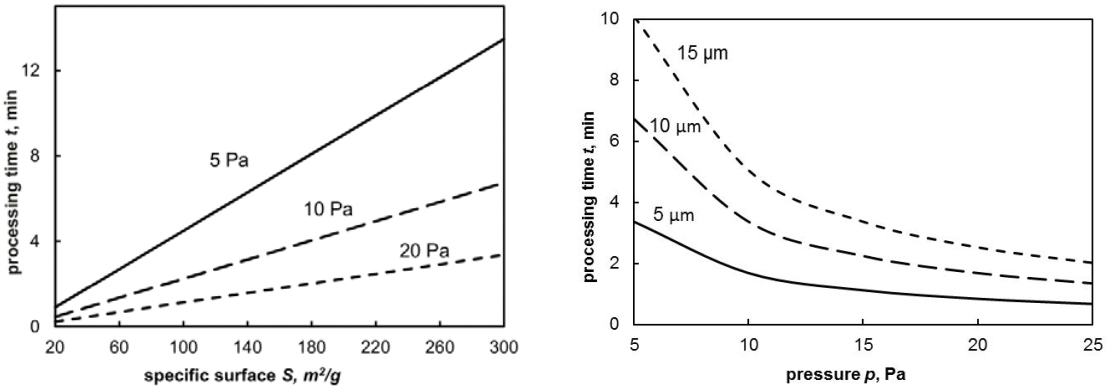
Figure 1a shows the dependence of the penetration time of melt into the pores of the particles on the specific surface of particles for various values of pressure $p$. Figure 1b shows the dependence of the penetration time on the pressure $p$ for various sizes of agglomerates ($S$=150 m$^2$/g) .



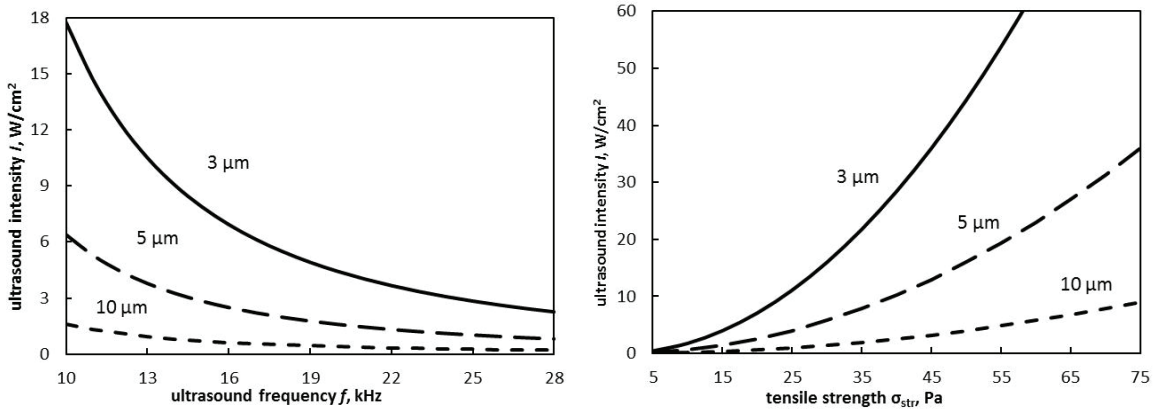

a) b)

**FIGURE 1.** Dependence of the penetration time of melt into the pores of the particles on the pressure p, specific surface S and diameter of the particles D: a) *t(S)* for different *p* (*D*=10 μm), b) *t(p)* for different *D* (*S*=150 m$^2$/g).

An increase in excess pressure (in proportion to the growing ultrasonic intensity) first leads to a sharp fall of penetration time, but a further increase in pressure does not give a noticeable time gain (Fig. 1b).

## Threshold Intensity of Deagglomeration

Figure 2a shows the calculated (formula 5) dependence of the threshold ultrasonic intensity for deagglomeration of aluminum oxide particles in aluminum melt at ultrasound frequencies for three sizes of agglomerated particles.




a) b)

**FIGURE 2.** Dependence of the threshold intensity of deagglomeration on ultrasonic frequency (a) and agglomerate strength (b).

With small agglomerates, with a radius of about 3 μm, high ultrasonic intensity is required for their dispersion. With the reduction of the agglomerate size, the threshold intensity sharply increases, reaching technically unachievable values over 1 kW/cm$^2$ (Fig. 2b). Here, the lower the sound frequency, the higher the intensity required for the deagglomeration.



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Figure 2b illustrates the dependence of the threshold ultrasonic intensity on the strength of agglomerates. The calculation is performed for the ultrasonic frequency $f$=20 kHz. Large, loose agglomerates with low strength are much easier to break than strong small ones.

The obtained theoretical results are indirectly confirmed by experimental data on ultrasonic processing of aluminum melts containing particles of oxides, borides, and carbides [2]. The main parameters of exposure are the processing time and the amplitude of vibration. The wettability of particles and their distribution in the liquid volume are improved with an increase in processing time.

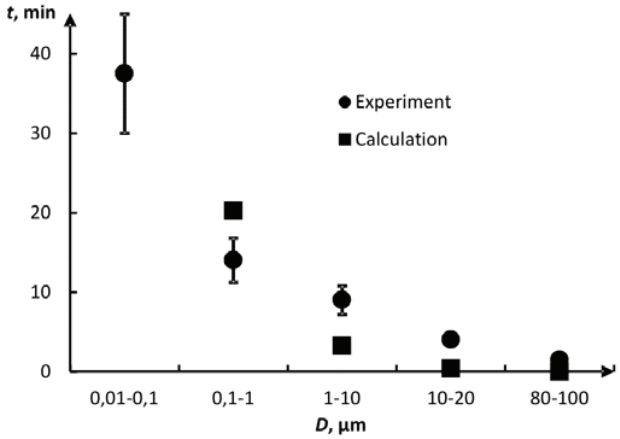


**FIGURE 3.** Time of ultrasonic cavitation processing of aluminum melt for deagglomeration of $\gamma Al_2O_3$ particles: experiment [9] and calculation by formula (4).

The obtained experimental dependence shows that the required time of processing increases with the decrease in particle diameter [9]. We did the calculation by formula (4) for $p$=50 Pa. Figure 3 shows the calculated and experimental results. Though the estimated time of impregnation of particles grows quicker with decrease of their sizes, than it is observed in an experiment, we can claim that obtained results of mathematical modelling correspond to the experimental ones.

## CONCLUDING REMARK

The parametrical study of the proposed model simulating the ultrasonic dispersion of particle agglomerations in metal allows us to draw the following conclusions:

– Developed ultrasonic cavitation facilitates impregnation of agglomerates by liquid metal, effective wettability and introduction of particles into the melt.

– The time required for the impregnation is proportional to the viscosity of the melt, specific surface of the powder and size of agglomerates; it is inversely proportional to the ultrasonic intensity and amounts to minutes or tens of minutes.

– The deagglomeration of the particles requires the ultrasonic intensity inversely proportional to the squared radius of agglomerates.

A comparison of calculated values of impregnation time of aluminum oxide particles by liquid aluminum with experimental data validates the feasibility of the model. The proposed calculation formulas will make it possible to estimate the intensity, frequency, and time of the ultrasonic processing that are necessary for successful introduction of particles into the metal melt.

The proposed model is an association of consideration of physical and chemical effects related to capillary phenomena, and known models of the ultrasonic cavitation in liquids. The combination of these models allows first expression propose important parameters for the evaluation of ultrasonic treatment of the metal melt, containing particle agglomerates: time and intensity treatment. Knowledge of these parameters is important for engineering practice and can be used in metallurgy, at receiving of new composite materials.



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## REFERENCES

1. O. Kudryashova and S. Vorozhtsov, J. Miner. Met. Mater. Soc. **68 (5)** 1307–1311 (2016).
2. G. I. Eskin and D. G. Eskin, Chem. Rubber Comp. Press, 2014.
3. S. A. Vorozhtsov, D. G. Eskin, J. Tamayo, A. B. Vorozhtsov, V. V Promakhov., A. A. Averin and A.P. Khrustalyov, Metallurgical and Mater. Trans A., **46**, 7, 2870–2875 (2015).
4. P. P. Prokhorenko, N.V. Dezhkunov and G.E. Konovalov, *Ultrasonic Capillary Effect* (Minsk, Nauka i Tekhnika (in Russian), 1981).
5. T. Matsunaga, K. Ogata, T. Hatayama, K. Shinozaki and M. Yoshida, Compos. A, **38**, 771–778 (2007).
6. L. Rozenberg High-intensity ultrasonic fields, NY, Plenum Press, 1971.
7. I. Tzanakis, W. W. Xu, G. S. B. Lebon, D.G. Eskin and K. Pericleous, P.D. Lee, Physics Procedia. **70,** 841–845 (2015).
8. I. Tzanakis, W. W. Xu, D.G Eskin, P.D. Lee and N. Kotsovinos, Ultrasonic Sonochemistry, **27,** 72–80 (2015).
9. G. I. Eskin, Technol. Legk. Splavov, **2,** 17–24 (2000).
10. Y. Yang and X. Li, J. Manuf. Sci. Eng. **129**, 498–505 (2007).
11. E. G. Konovalov and I. K. Germanovich, Reports of Belorussia Academy of Sciences (Dokladi Akademii Nauk Belorusskoy. *SSR* – in Russian), **6**,8, 492–498 (1962).



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