Cavitation Disintegration of Microparticles and Nanoparticles in Dense Liquid Dispersions

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ABSTRACT

With the current rapid development of nanotechnology, top-down preparation of nanoparticles by various disintegration techniques became an important industrial sector. In addition to the presently common high speed bead mills working with particles in liquid dispersions, jet mills are also frequently used. They operate in the dry disintegration mode of extreme aerosol flow with cyclonic separation of the fine fraction. The common characteristic of these both mill types is the submicron size of disintegrated output particles. Very promising alternative to these techniques is the use of the disintegrating effect of cavitation implosion. In this work, we present experimental results and technical experience with a new experimental device for cavitation disintegration of microparticles and nanoparticles in dense liquid dispersions. In first test experiments, the mean size of the primary silicon particles of 140 nm was reduced to 40 nm by the cavitation disintegration lasting 200 minutes.

Keywords: nanoparticles, silicon, disintegration, cavitation

1 INTRODUCTION

With the current rapid development of nanotechnology, preparation of nanoparticles becomes a very important technological factor. Bottom-up processes of nanoparticle production are generally based on chemical methods [1-2]. Their basic problems are the effective management of reaction kinetics in nucleation, growth restrictions [3-4] and possible modifications [5-7]. Another technological approach is top-down production of nanoparticles using various disintegration techniques. In addition to the presently common high speed bead mills which operate in the environment of liquid particle dispersions, jet mills become also frequently used [8]. They operate in the dry disintegration mode of extreme aerosol flow with cyclonic separation of fine fractions. The common characteristic of these both mill types is the submicron size of disintegrated output particles.
Very promising alternative to these techniques is the application of the disintegrating effect of cavitation implosion on the particles dispersed in a liquid. Intense cavitation can be generated by high-energy water jet, like in the disintegrator Water Jet Mill [9]. Our experiments confirmed that it is an effective tool for nanoparticle preparation [10-11]. In this paper we present preliminary experience and experimental results of the cavitation disintegration of micro and nano particles in a dense fluid dispersion under intense ultrasonic field.

2 THEORY OF CAVITATION DISINTEGRATION OF PARTICLES IN DENSE LIQUID DISPERSIONS

The analysis [9] of the mechanism of disintegration based on the implosion of cavitation bubbles proved that the impact pressures can reach tens of GPa, sufficient to disintegrate even very hard materials. For this application, we constructed an ultrasonic disintegrator schematically illustrated in Figure 1.

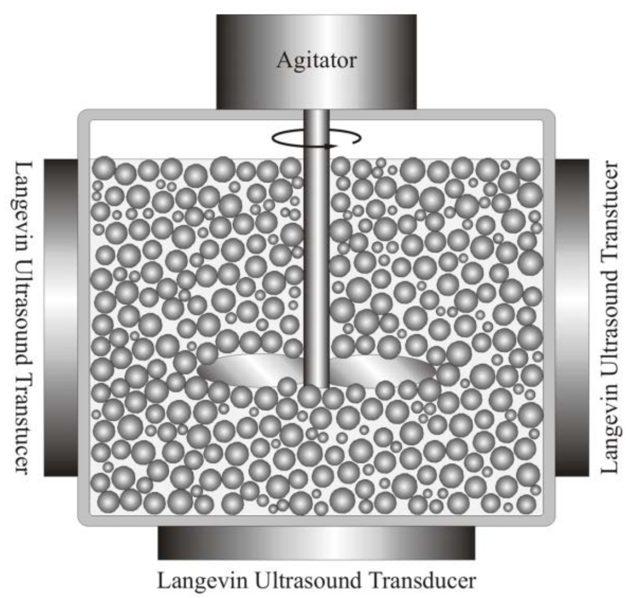

Figure 1: Sketch of the prototype cavitation ultrasonic disintegrator with the stirring of dense particle dispersion (sizes of disintegrated micro particles are significantly magnified to illustrate the required high dispersion density).

Inside the disintegration chamber, the dense liquid dispersion of microparticles is subjected to ultrasonic radiation of Langevin transducers through its metal walls. Simultaneously, the relative positions of microparticles are rapidly changing due to external mixing of dispersion. In these conditions, mutual impacts and subsequent separations of dispersed particles are very frequent. Heterogeneous nucleation of cavitation bubbles on the surface of the particles substantially prevails over homogeneous nucleation. Therefore, cavitation bubbles form mainly between two particles separated by mixing.

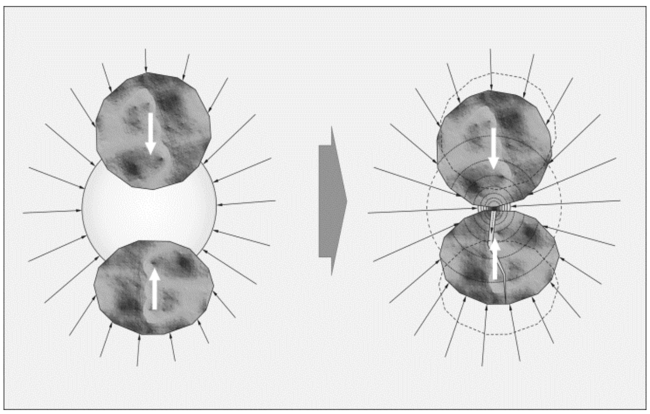

Figure 2: Illustration of the mutual impact disintegration due to the collapse of a cavitation bubble between two particles

This situation is illustrated in Figure 2. On the left, the cavitation bubble reached its maximum size and it started imploding. The two particles form the opposite walls of the collapsing bubble and they are pushed by surrounding liquid into the place of mutual collision, as displayed on the right. Reflecting the analysis of the cavitation disintegration in [9], the following experiments tested the disintegration of semiconductor-grade silicon submicron particles dispersed in degassed water.

3 EXPERIMENTAL VERIFICATION OF ULTRASOUND CAVITATION DISINTEGRATION TECHNOLOGY

The chamber of the ultrasonic cavitation disintegrator was filled with the dense water dispersion (slurry) of semiconductor-grade Si microparticles with the size distribution depicted in Figure 3, with the mean size of 140 nm.

For optimal efficiency of the disintegration it was necessary to provide the volume fraction of dispersed particles in the aqueous medium at least 50%. Such an arrangement in the microscale induces very high frequency of mutual collisions during mixing of the dispersion. Simultaneously, it also ensures plentiful formation of cavitation bubbles between particles after their separation and therefore the high intensity of cavitation disintegration.
In the arrangement according to Figure 1, the dense dispersion of the aforementioned primary microparticles was subjected to ultrasonic cavitation disintegration for 200 minutes at the power density of ultrasonic field 50 W/l and the agitator speed of 100 rpm.

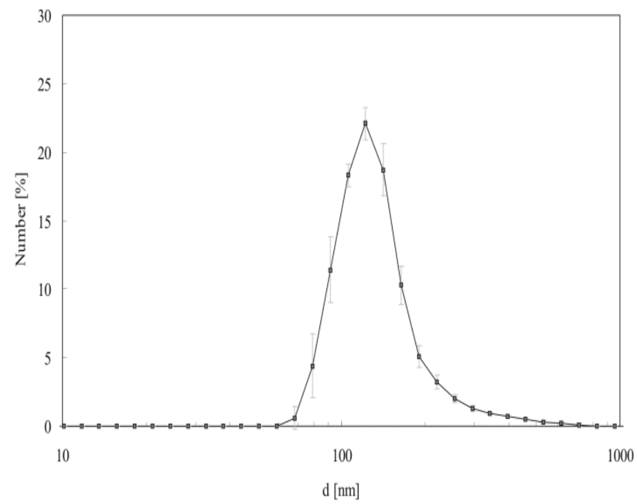

Figure 3: The statistical distribution of diameters *d* of primary silicon microparticles with the mean size of 140 nm, obtained using DLS Malvern Nano ZS considering the correction due to the hydrodynamic coefficient of 0.8. These primary microparticles were produced by milling in a planetary mill and separated by sieve analysis.

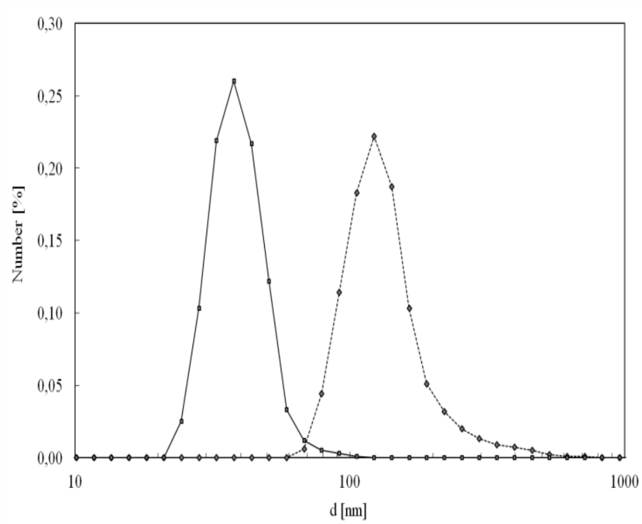

Figure 4: The effect of 200 minutes of ultrasonic cavitation disintegration. The original size distribution of microparticles is depicted in a dashed line, the distribution after the processing in a solid line. The mean diameter is 40 nm. The analysis was obtained using DLS Malvern Nano ZS considering the correction due to the hydrodynamic coefficient of 0.8.

Figure 4 shows the transition in the size distribution of silicon particles due to the disintegration. Primary microparticles (dashed line) transform to resulting nanoparticles (solid line). Following the statistical analysis of [7], the mean size of the particles is reduced to 27%. The resultant nanoparticle liquid dispersion was finally vacuum freeze dried to obtain dry nanopowder for further practical use. This was then analyzed on transmission electron microscope TEM Jeol JEM 1230, 80 kV.

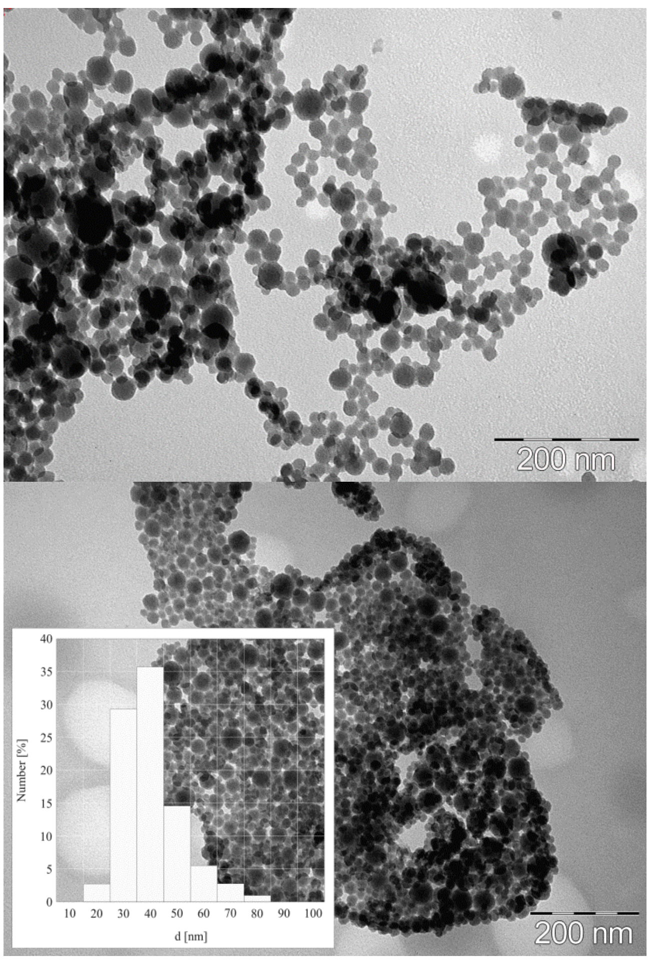

Figure 5: TEM micrograph of a final aggregate of Si nanoparticles after disintegration and vacuum freeze drying (TEM Jeol JEM 1230, 80 kV).

Image analysis of the final silicon nanoparticles gave a statistical ensemble of 100 particles with the size distribution shown in Figure 5. Its mean value of 39 nm is in good agreement with the value of 40 nm obtained by dynamic light scattering method using Malvern Nano ZS.

The disadvantage of our first experimental disintegrator is a low density of acoustic power transferred to the processed dispersion. This naturally reduces the efficiency of the disintegration, increases the time required to achieve the desired grain refinement and restrains disintegration (grinding) limits.

The achieved results prove the described technology of disintegration basically effective, so we can approach the construction of an efficient prototype of the cavitation ultrasonic disintegrator with a density of the transferred acoustic power above 100 W/l.

In the previous works [10-11], the theoretical analysis of maximum attained impact pressures has shown that they are significantly influenced by the external hydraulic pressure of the surrounding liquid. For these reasons, a new prototype of the cavitation ultrasonic disintegrator will operate with higher pressures, up to 15 MPa.

4 CONCLUSIONS AND FUTURE DIRECTIONS

In the first experiments, we tested the response to ultrasonic cavitation disintegration on sparse dispersions, always with a negligible effect. In the light of this outcome, our later results confirm the validity of fundamental theoretical ideas about the mechanism of impact disintegration due to implosion of cavitation bubbles in dense liquid dispersions of microparticles.

With regard to the theoretical calculations of the maximum impact pressures in the previous work [10-11], we estimated the dynamics of a mutual collision of two particles. It follows that the submicron particles collide with the characteristic speed of hundreds of meters per second. Such conditions lead to the destruction of even very hard materials.

Currently, we are finalizing the prototype of a new, more efficient device that will operate with higher hydraulic pressures and higher acoustic power densities. The improved parameters of the new apparatus should increase the intensity of heterogeneous nucleation of cavitation bubbles on solid particles and also the impact velocities in collapse.d

ACKNOWLEDGMENTS

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