# Effect of Ultrasound Waves on Emulsification and Separation of Two-Phase Liquids

Mamoru Kuwabara*, Masamichi Sano*, Sergey V. Komarov*,
Kazumichi Yoshida** and Noriyasu Kawakita**

Department of Materials Processing Engineering
Graduate School of Engineering, Nagoya University
Furo-cho, Chikusa-ku, Nagoya 464-8603, Japan

(*: Faculty members, **: Graduate students)

**Abstract**    Cold model experiments have been carried out to investigate the effect of ultrasound on interfacial phenomena between two-phase liquids simulating slag and molten metal bath. Rate processes of emulsification of two phase liquids and subsequent separation of emulsified fine droplets have been optically detected by measuring decay of transverse laser beam through the bath in acrylic resin vessel. These processes can be successfully controlled by imposing ultrasound with appropriate sets of frequency and liquid depth which allow acoustically resonant conditions in the liquid bath as well as with appropriate powers having more or less than the cavitation threshold.

## 1. Introduction

Reactive or extractive distribution of chemical species between metal and slag is an essential task of refining in ferrous or non-ferrous metallurgical processes. Therefore, increasing attention has been paid not only to an appropriate selection of slag constituents for each process but also to realizing both sufficient interfacial area and the greatest intensity of mixing of metal bath for an enhanced rate process. A disperse system may be a choice to ensure an effective interfacial transport process between metal and slag. However, a system with relatively large slag droplets in the metal phase are often unstable due to both the gravitational floatation and the aggregation or coalescence of dispersed phase, which consequently decrease free energy associated with the interfacial area to form separated two layers. Otherwise, a disperse system with fine slag droplets often leads to a permanent persistence of fine non-metallic inclusions in the metal, which are difficult to be removed in the subsequent processes in gravitational field.

An alternative and innovative processing of material in its molten state could be realized by applying electromagnetic and/or ultrasonic external forces to the dispersed system of interest. The present authors[1]-[3] have been proposing new methods of ultrasonic processing of material, which are based on such phenomena as the acoustic radiation pressure acting on the dispersed phase as well as the acoustic streaming and the cavitation in liquid associated with the use of highly intensified ultrasound. Taking account of physical natures which ultrasound and sound waves commonly possess, we call such acoustic-wave-assisted processing as "*sonoprocessing*". The application of this new technology to the processes involving pyro-metallurgical systems would become easier in the near future, since intensive investigations are currently undertaken for developing both new piezoelectric ceramics with high Curie points and a wide-bandwidth ultrasound speaker for high temperature use[4], [5] as well as for generating an electromagnetically induced ultrasound[6]-[8].

The ultimate goal of this research is to establish fundamentals of the sonoprocessing of two-phase liquids for an ultrasonically controlled dispersed system. Cold model experiment has been carried out to investigate the effect of ultrasound on the interfacial phenomena between

two-phase liquids simulating slag and molten metal bath. Rate processes of emulsification of two-phase liquids and subsequent separation of the dispersed phase have been optically detected by measuring decay of transverse laser beam through the bath. The effects of process parameters such as frequency and power of ultrasound, liquid depth, and atmospheric pressure on the interfacial phenomena of the dispersed system will be described below on the basis of the experimental results.

## 2. Emulsification of Two-Phase Liquids

High-intensity ultrasound can produce cavitation bubbles in a liquid due to stresses induced by a propagating sound wave consisting of compression and decompression cycle. High temperature and high pressure, produced during the cavitation and collapse of the bubbles, cause irregular motions of both the bubbles and the liquid, which are effective for homogenizing the liquid resulting in a dispersed system including emulsion. This section is devoted to describing the results of cold model experiments conducted for the ultrasonic emulsification of two-phase liquids simulating metal and slag.

### 2.1 Experimental

The experimental setup is shown in Fig. 1. A sinusoidal signal which was generated by a function generator and was amplified by a power amplifier was imposed to a bolted Langevin-type transducer (resonant frequency: 46 kHz, nominal maximum input power: 55 W). The transducer was fixed to a stainless steel plate of 3 mm thickness at the bottom of a vessel. The vessel made of a cylindrical acrylic resin of 5 mm thickness and 80 mm inner diameter, was filled with distilled or undistilled city water to a depth of 75 mm which is nearly a resonant depth possible to cause violent cavitation. The tetrahydronaphthalene (tetralin) solution that is basically immiscible with water and has a lower density than water, 970 kg/m$^3$, was quietly fed over the water bath to a depth of 10mm. The atmosphere in the vessel was evacuated to a given pressure using a vacuum pump. Experiments under irradiation of ultrasound were then carried out at the room temperature. A front view of the vessel was photographed sequentially during cavitation-induced emulsification of two-phase liquids and it was related with the relative decay of the He-Ne laser beam passing through the vessel using a photo-sensor with an electric recording device. The laser beam incident upon the vessel was running at 15mm under the initial interface level.

### 2.2 Results and Discussion

Figure 2 shows a typical photographic sequence of ultrasonic emulsification of the two-phase liquids under the conditions where ultrasonic power (U.S.P) was 18W, frequency 43.2kHz and atmospheric pressure 0.1MPa. Just after irradiation of ultrasound, emulsification due to cavitation proceeds from around the center part of the interface, and an almost homogeneous emulsification is already attained after 180 s as is found from the whitened zone.

The extent of emulsification can be evaluated as the turbidity factor $\square$ through a relationship between the relative decay of the laser beam $I/I_0$ and its transmitted distance D, which has the form of

$$log\ I/I_0 = \square\ \square D \qquad (1)$$

The $I/I_0$ term is nearly equal to the relative increase in the electric resistance of the photo-sensor $R_0/R$ that can be measured directly.

Figure 3 shows the effect the kind of water used on the transitional change in turbidity during emulsification. Undistilled water as a lower liquid leads to more intensified emulsification than distilled water because the former liquid involves more dissolved gas(air) that can cause intensified cavitation. When distilled water was employed, turbidity is nearly saturated after 180 s at a level of about 50(1/m) where the incident laser beam was almost scattered out due to the intensified emulsification. This fact clearly corresponds to a situation

photographically observed in Fig. 2.

Figure 4 shows the effect of atmospheric pressure on transitional change in turbidity during emulsification. It is noted that the emulsification at 0.1 MPa is more extensive than that at 0.05 MPa though an intensive cavitation can be expected at a lower pressure. Figure 5 represents photographs showing motion of cavitation bubbles in water with irradiation of 42.3 kHz ultrasound at a reduced pressure of 0.06MPa. There exist great amount of small cavitation bubbles at the initial stage (a), but the bubbles are only steadily evacuated upwards without intensified mixing. Instead, at the later stage (b), relatively large cavitation bubbles cause intensified mixing accompanied by a violent motion of the free surface. This may be the reason why the reduced pressure in Fig.4 induces less effect on emulsification with two kinds of transitional stages.

## 3. Separation of Dispersed Phase

Ultrasound irradiated to a liquid with a dispersed phase can propagate through the system with less attenuation than in a gas phase. Difference of the acoustic impedance $\Box c(\Box$: density, c: sound velocity) between the bulk liquid and the dispersed phase renders the radiation pressure acting on the interface, which can move the dispersed phase towards nodes or loops of a standing wave established in the liquid. Fundamental aspect of the ultrasonic separation of the dispersed phase from the bulk liquid is discussed below on the basis of the results from cold model experiments. Too fine dispersed phase obtained in the preceding experiment demanded an excess time and an excess ultrasonic power for its ultrasonic separation. Thus, relatively large size of polystyrene particles is arbitrarily employed here as a dispersed phase instead of fine droplets.

### 3.1 Experimental

Two kinds of experiments were carried out. In one experiment in which ultrasound was irradiated vertically, experimental setup is basically the same as that seen in Fig.1. The vessel here, made of a cylindrical acrylic resin of 3 mm thickness and 73 mm inner diameter, was filled with an aqueous sugar solution to a resonance depth of 60 mm. In another experiment in which ultrasound was irradiated horizontally, a rectangular apparatus shown in Fig. 6 was filled with an aqueous sugar solution to a resonance depth of 100 mm with or without an upper oil layer of 5mm thickness. The solution was predegassed under reduced pressure and with appropriate stirring in order to avoid possible cavitation. Polystyrene particles of 163 μm diameter were initially dispersed uniformly in the solution. Change in the concentration of the aqueous sugar solution allows us to determine the density difference between the liquid and the particles. Under the condition of about 13 wt%, the density difference becomes zero at $\rho_l = \rho_p = 1050$ kg/m$^3$ in which case the buoyancy force acting on any particle can be ignored. Photographs were taken sequentially in front of the vessel during transitional motion of the polystyrene particles. Effective value of sound pressure in the vessel was measured using an ultrasonic meter to determine the energy density of ultrasound as well as sound pressure distribution.

### 3.2 Results and Discussion

According to the theory proposed by Yosioka and Kawasima,[9] the acoustic radiation force, $f_{rp}$, on a spherical particle in the standing-wave field is given as:

$$f_{rp} = -3V_p k E_{av} \cdot F(\Lambda,\sigma) \cdot \sin(2kx), \tag{4}$$

where,

$$F(\Lambda,\sigma) = [\Lambda + 2(\Lambda-1)/3]/(2\Lambda+1) - 1/(3\Lambda\sigma^2), \tag{5}$$

where, $k$ is the wave number (= $2\Box f/c_l = 2\Box/\Box$), $c_l$ is the speed of sound, $f$ is the frequency, $\Box$ is the wavelength, $E_{av}$ is the time-averaged energy density of ultrasound, $\Box$ is the density ratio of the particle to the liquid and $\Box$ is the sound velocity ratio of the particle to the liquid.

Polystyrene particles may move toward the pressure nodes in the standing-wave field because the function $F$ for the polystyrene in water at room temperature is positive. It should be noted that inclusions in molten metal may move toward the pressure antinodes depending on the negative sign of factor $F$.

Figure 7 shows a transitional aggregation of dispersed polystyrene particles under the conditions of $\rho_l = \rho_p = 1050$ kg/m$^3$ and vertical irradiation of ultrasound with $f$=48.5 kHz and $E_{av}$ =5 J/m$^3$ which was calibrated from the effective sound pressure in the vessel. The figures show photographs at transient time intervals of 0, 10, 30 s after ultrasound irradiation. The distribution of sound pressure in the vertical direction is also shown in this figure. The aggregation of the particles was initiated as soon as ultrasound irradiation was begun and nearly finished around the nodes of the sound pressure after 15 s. A detailed discussion on the rate process of ultrasonic separation is available elsewhere[10].

Figure 8 shows a photograph of an aggregation experiment in the rectangular acrylic resin vessel in which the polystyrene particles of diameter 0.163 mm are being separated from the distilled water with incidence of a lateral ultrasonic standing wave. Such lateral irradiation of ultrasound can effectively assist natural floatation or sedimentation of the dispersed phase since large particles aggregated in the nodal vertical planes are favorable to move against the drag force. However, it should be noted that the free surface of the liquid bath is acoustically equivalent to a sound pressure node of the standing-wave field. Therefore, it may be difficult in principle to separate particles near the top surface where they are loosely constrained. As is demonstrated in Figure 9, an alternative way to avoid this situation is to settle (b) a rigid plate or (c) a liquid layer like slag with an appropriate thickness on the free surface, which can stipulate a fixed boundary condition needed for reflecting ultrasound at the top satisfactorily.

## 4. Conclusions

Cold model experiments have been carried out to investigate the effect of ultrasound on interfacial phenomena between two-phase liquids simulating slag and molten metal bath. Rate processes of emulsification of the two-phase liquids and subsequent separation of dispersed phase have been optically detected through the bath in acrylic resin vessel. These processes can be successfully controlled by imposing ultrasound with appropriate sets of frequency and liquid depth which allow acoustically resonant conditions in the liquid, as well as with appropriate powers having more or less than the cavitation threshold and with the direction in which ultrasound is imposed.

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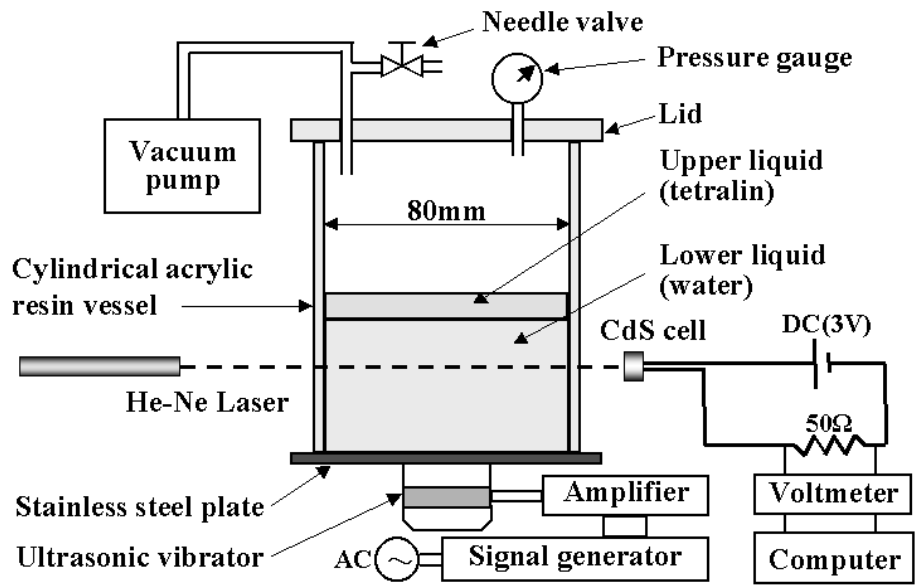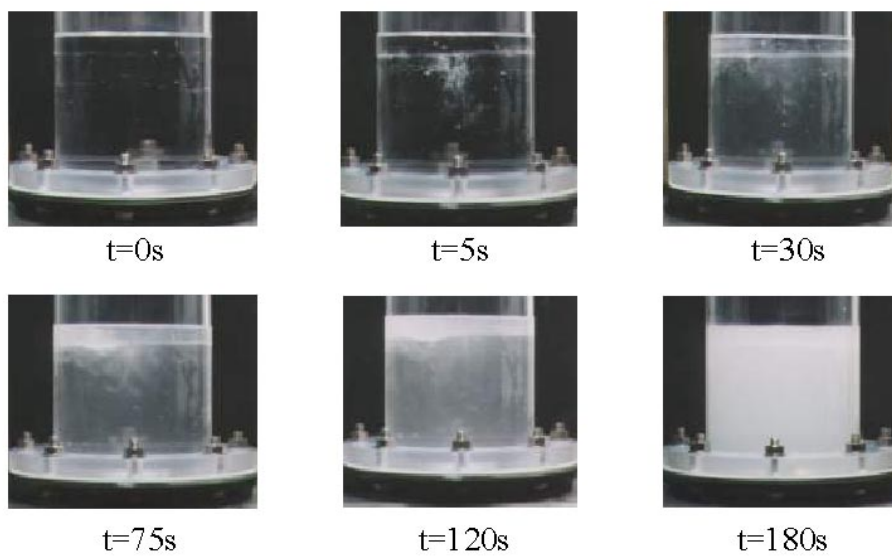
Fig. 1. Experimental apparatus for ultrasonic emulsification.

| t=0s | t=5s | t=30s |

| t=75s | t=120s | t=180s |

Ultrasonic power(U.S.P) : 18W,    Frequency : 43.2kHz,
Upper liquid (tetralin) : 10mm,    Lower liquid (distilled water) : 75mm,
Atmospheric pressure : 0.1MPa,    Temperature : 294K

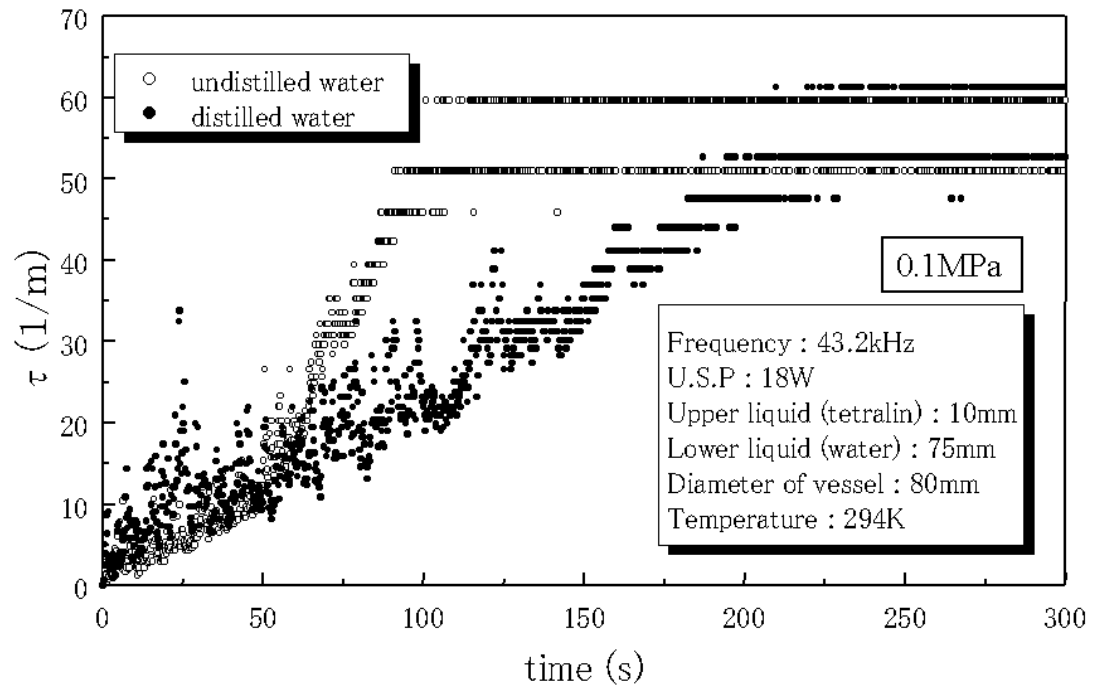Fig. 2.   Photographic sequences of ultrasonic emulsification of
two-phase liquids.

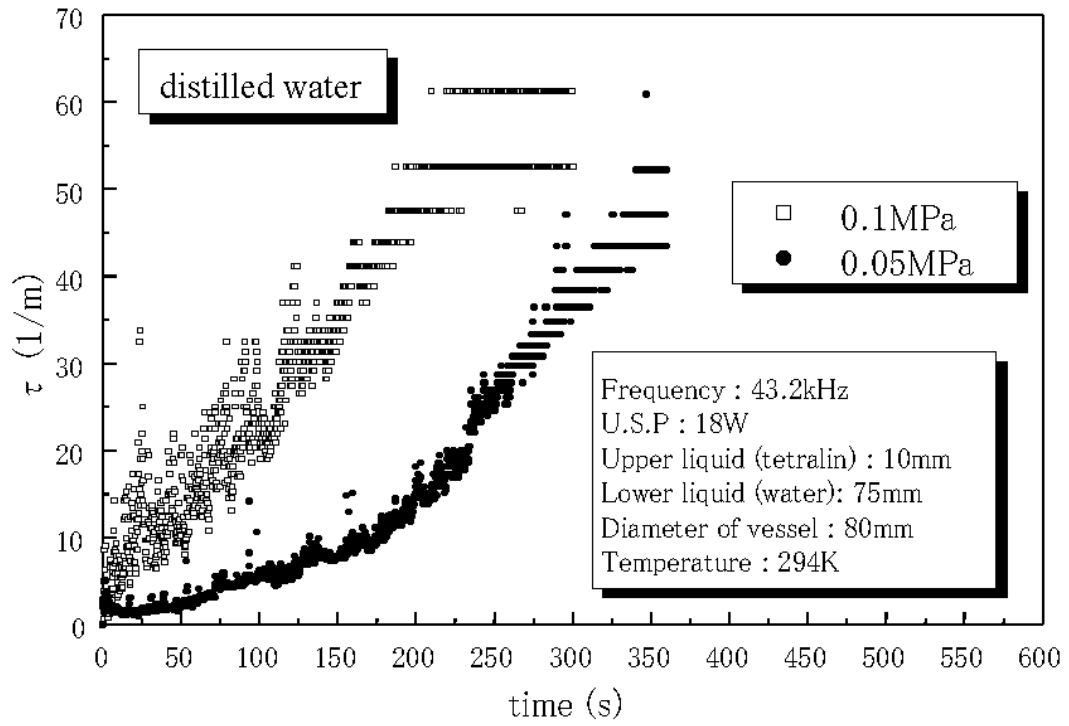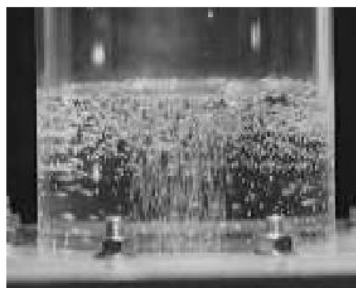Fig. 3. Effect of dissolved gas on turbidity during emulsification.

Fig. 4. Effect of atmospheric pressure on turbidity during emulsification.

(a) Just after irradiation of ultrasound
(exposure time : 1/125s)

(b) 5 min after irradiation of ultrasound
(exposure time : 1/125s)

Frequency : 42.3kHz      Liquid depth : 55mm
Atmospheric pressure in acrylic resin vessel : 0.06MPa

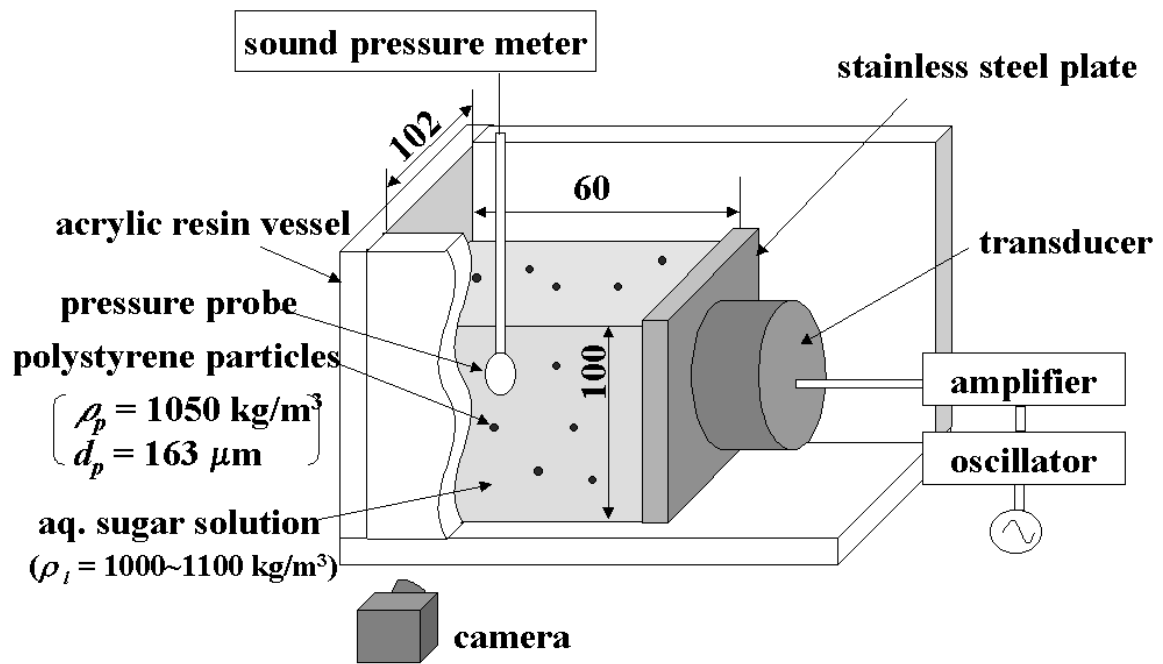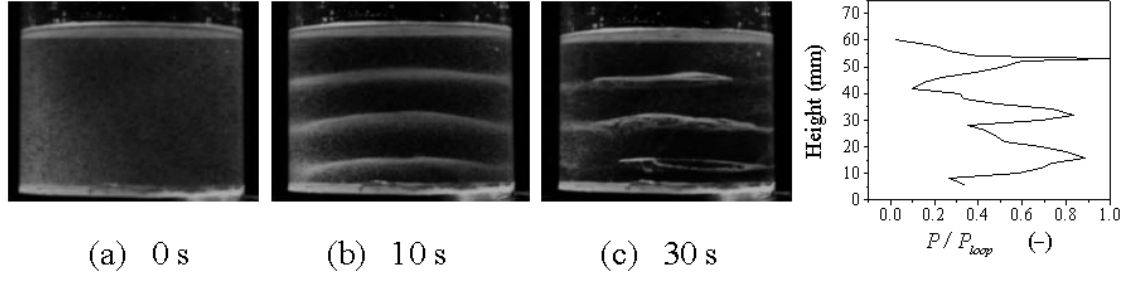Fig. 5.  Motion of cavitation bubbles in water with irradiation of ultrasound.

Fig. 6. Experimental apparatus for ultrasonic separation with horizontal
irradiation of ultrasound.

(a)  0 s          (b)  10 s          (c)  30 s

$f$ = 48.5 kHz ($\lambda$ = 33.6 mm), $E_{av}$ = 5.0 J/m³, $\rho_p = \rho_l$ = 1050 kg/m³, $d_p$ = 163 μm

Fig. 7. The motion of particles when ultrasound is radiated vertically
and observed longitudinal distribution of acoustic pressure.

(a)  0 s          (b)  5 s          (c)  10 s          (d)  60 s

$f$ = 43.3 kHz          $\rho_p$ = 1050 kg/m³          $d_p$ = 163 μm

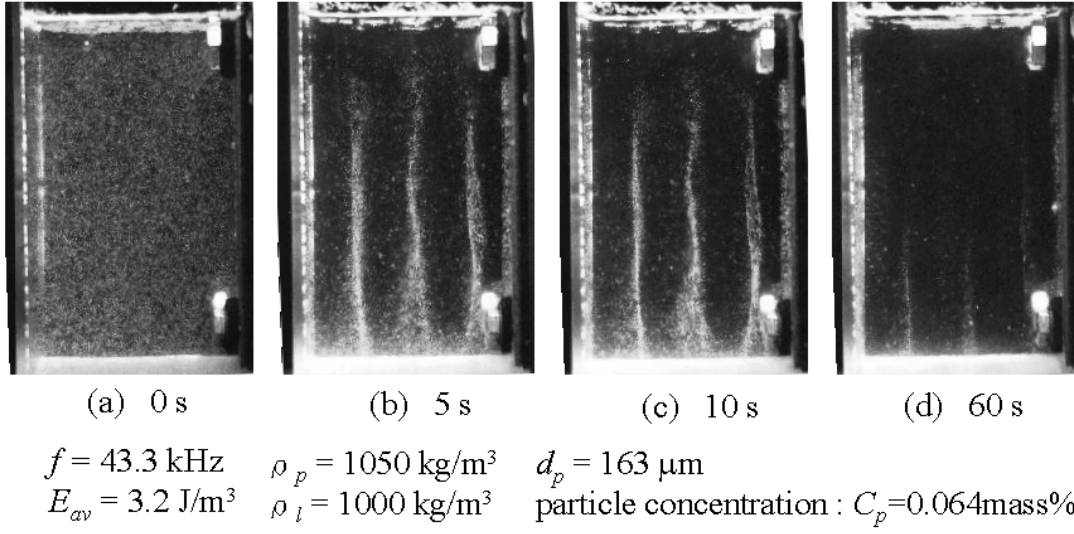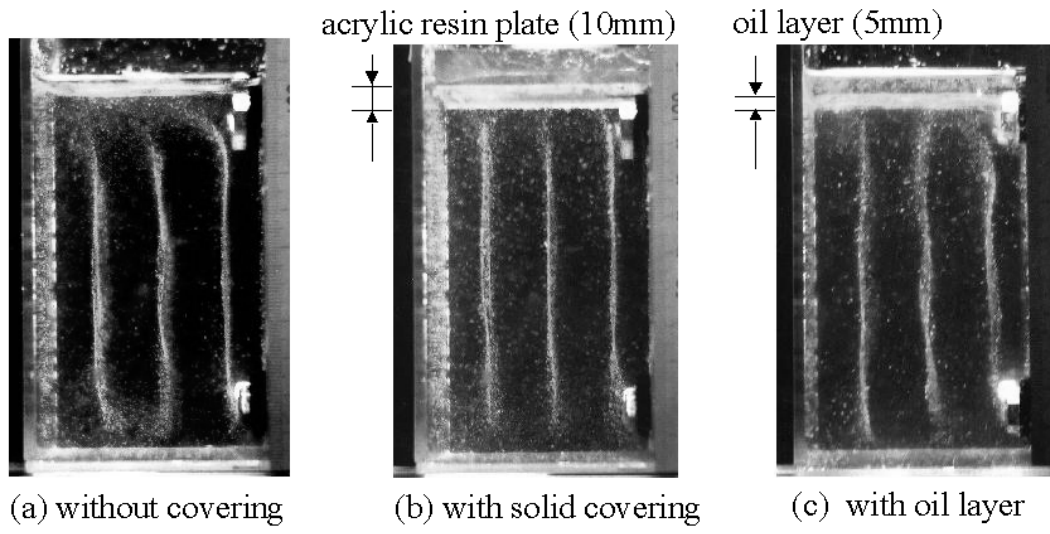$E_{av}$ = 3.2 J/m³          $\rho_l$ = 1000 kg/m³          particle concentration : $C_p$=0.064mass%

Fig. 8. Transitional coagulation of dispersed polystyrene particles in aqueous sugar solution irradiated by 43.3kHz horizontal ultrasound.

acrylic resin plate (10mm)   oil layer (5mm)

(a) without covering   (b) with solid covering   (c) with oil layer

$f \cong 44$ kHz, $E_{av} \cong 3$ J/m$^3$

$\rho_l = 1050$ kg/m$^3$, $\rho_p = 1050$ kg/m$^3$, $d_p = 163$ μm, $C_p = 0.064$ mass%

Fig. 9.  Effect of bath surface on ultrasonic separation of suspended particles in liquid with horizontally irradiated ultrasound .