



# The relationship between bubble motion and particle flocculation pattern under 20-kHz-ultrasound radiation in water



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## HIGHLIGHTS

- Dispersed particles with sub-millimeter and millimeter diameters in water were flocculated by irradiating kHz-band ultrasound.
- The flocculating patterns varied with the components and concentration of the dissolved gases.
- We evaluated the forces acting on the particle flocculation by visualizing both the particle flocculation and liquid motion.
- The particle flocculation was dominantly influenced by the acoustic radiation force acting on the ACOBs.
- We also discussed the motion of the acoustic cavitation-oriented bubble adhering to the particle surface.

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## ABSTRACT

Dispersed particles with sub-millimeter and millimeter diameters in water were flocculated by a kHz-band-ultrasound radiation. This phenomenon was caused by the acoustic cavitation-oriented bubbles (ACOBs). The flocculating pattern varied with the components and concentration of the dissolved gases. The relationship between the gas conditions and the particle flocculation pattern is discussed along with the estimation of the force balance acting on the particle flocculation. First, the particle flocculation was visualized. The liquid motion in the vessel was then measured via particle image velocimetry. The flocculation patterns were mainly classified into two types: spherical flocculation and chain-like flocculation. Spherical flocculation occurred when air-dissolution water was used, and chain-like flocculation occurred when CO<sub>2</sub> concentration-controlled water was used. Both patterns were dominantly influenced by the acoustic radiation force acting on the ACOBs. At the occurrence of spherical flocculation, the ratio of the acoustic radiation force to the other forces was greater than that at the occurrence of chain-like flocculation. We analyzed the motions of ACOBs and particles by high-speed visualization to clarify the interaction between the particles and ACOBs. The ACOBs adhering to the particle surface in CO<sub>2</sub> concentration-controlled water oscillated with the irradiation frequency and synchronized with other ACOBs. The member particles of the chain-like flocculation were tenaciously held by the Bjerknes force. In contrast, in the air-saturated water, the diameter of the spherically flocculated particle swarm oscillated due to the unstable motion of ACOBs. We suspect that the diameters of the spherically flocculated particle swarm fluctuated.

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## 1. Introduction

Ultrasound technologies such as ultrasonic cleaning (Lamminen et al., 2004), ultrasonic atomizing (Barreras et al., 2002), ultrasonic separation (Kapishnikov et al., 2006; Kirpalani and Suzuki, 2011) and particle manipulation (Yamakoshi and Noguchi, 1998;

Kozuka et al., 1998) have been applied in various industrial processes due to their advantages of non-contact and simple apparatuses. MHz-band ultrasound, which possesses high directionality, is usually used for the particle manipulation technologies, which are summarized as follows. King (1934) established a theory of the acoustic radiation force acting on a rigid spherical sphere. This theory was improved by many researchers (e.g., Nyborg, 1967; Yosioka and Kawasima, 1955; Hasegawa and Yosioka, 1969). Kamakura et al. (1995) constructed a mathematical model of acoustic streaming induced by focused 5-MHz-ultrasound irradiation.

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## Nomenclature

$a$	radius [mm]
$B$	position of bubble
$c$	sound speed [m/s]
$C_D$	drag coefficient
$D$	diameter [mm]
$f$	radiation frequency [kHz]
$F(\varepsilon)$	function of porosity
$F_{ac}$	acoustic radiation force [ $\mu\text{N}$ ]
$F_B$	buoyancy force [ $\mu\text{N}$ ]
$F_D$	drag force [ $\mu\text{N}$ ]
$F_g$	gravity force [ $\mu\text{N}$ ]
$g$	acceleration of gravity [ $\text{m/s}^2$ ]
$k$	wave number [1/m]
$m$	mass [mg]
$N$	the number of particle/bubble
$P$	position of particle
$Re$	Reynolds number
$t$	time [s]
$\Delta t$	time interval [s]
$S$	projected area [ $\text{mm}^2$ ]

$U$	composition velocity [mm/s]
$v_t$	terminal velocity [mm/s]
$V$	volume [ $\text{mm}^3$ ]

### Greek letters

$\gamma_{ratio}$	packing ratio
$\varepsilon$	porosity
$\lambda$	wavelength [mm]
$\mu$	viscosity [kg/m/s]
$\rho$	density [ $\text{kg/m}^3$ ]
$\phi$	phase [degrees]

### Subscripts

$b$	bubble
$n$	particle/bubble number
$p$	particle
$ps$	particle swarm
$w$	water

tion in water. Kozuka et al. (1997) demonstrated their original manipulation technique for fine (16- $\mu\text{m}$ -dia.) aluminum particles by MHz-band-ultrasound radiation into water.

The high directionality and high frequency of ultrasound technologies are a double-edged sword. The ultrasonic manipulation technologies using a MHz-band usually use the acoustic radiation force acting directly on the targets; hence the diameters of target particles must be smaller than the wavelength; e.g., the diameters of manipulable particles are less than several micrometers in the case of water. These technologies thus bring the limitations of manipulable particle diameters less than several micrometers (from the high frequency) and the limitations of a small manipulable range in space (from the high directionality).

In contrast, there have been very few studies on particle manipulation through kHz-band ultrasound, and their findings can be summarized as follows. Ochiai et al. (2014) proposed a three-dimensional particle manipulation technique through a 40-kHz-ultrasonic transducers array in air. Ohta et al. (2007) reported the behavior of 50–150- $\mu\text{m}$ -dia. aluminum particles in water under horizontal irradiation by 20–100-kHz ultrasound. Ohta and Nakano (2008) classified the flocculation patterns into four types: band, point, particle clump, and non-flocculation. However, the knowledge and understanding of the separation/manipulation technologies using kHz-band ultrasound that could lead to industrial applications are still insufficient.

In the present study, with the goal of eventual industrial use, we used kHz-band ultrasound (which has received little attention for the past few decades) to reveal the flocculation mechanism. Mizushima et al. (2013) reported a very interesting phenomenon in which particles with sub-mm-to-mm diameters dispersed in non-degassed purified water are flocculated into a spherical particle swarm by kHz-band-ultrasound radiation. This particle flocculation never occurred under degassed purified water. Acoustic cavitation-oriented bubbles (ACOBs) adhering to a particle surface moved the particle itself into the particle swarm.

Muramatsu et al. (2015) investigated the characteristics of a spherically flocculated particle swarm (SFPS) and proposed a novel particle separation technique performed according to particle diameters. We observed a new and marvelous phenomenon in which the flocculation pattern varies with the components and

the concentration of the dissolved gases in water. We could categorize the flocculation patterns into two main types: a spherical pattern (purified water in which air dissolves) and a chain-like pattern (purified water in which  $\text{CO}_2$  dissolves). By experimentally revealing the roles and mechanisms of the dissolved gas components and concentration herein, we establish a fundamental technology for the industrial application of this new and characteristic phenomenon.

The purpose of the present study was to clarify the flocculation mechanisms by evaluating the forces acting on the particles during the flocculation processes under the precise control of the gas components and concentration in purified water. We focused on four types of dominant forces: (a) the gravity force, (b) the buoyancy, (c) the drag force, and (d) the acoustic radiation force.

First, we developed a system that precisely controlled the gaseous components and concentration. Second, we measured the particle flocculation patterns by the shadow graph method and evaluated the liquid motion in the vessel by the particle image velocimetry (PIV) technique. Last, we investigated the influence of ACOBs on the particles by analyzing the motion of the ACOBs adhering to the particle surface, as well as the motion of the particles. Based on the forces acting on the particles and ACOBs observed in the above-described analyses, we discuss the mechanisms of the particle flocculation patterns and the interaction between ACOBs and flocculated particles.

## 2. Experimental setups

### 2.1. Processes for degassing, and controlling $\text{CO}_2$ dissolution

To investigate the influences of the gas components and concentration on the particle flocculation patterns and processes, we used five types of purified-water-based solutions in the present study: (1) air-saturated water (GC1), (2)  $\text{CO}_2$ -concentration-controlled water (GC2), (3) degassed water (GC3), (4)  $\text{CO}_2$ -dissolved water without degassing process (GC4), and (5)  $\text{CO}_2$ -saturated water (GC5). Since the ACOBs' behavior is significantly influenced by the surrounding environment of temperature, pressure, dissolved gas components and concentration, we conducted all experiments in a room controlled for temperature, pressure

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