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Study of ultrasonic pitting mechanism on starch particle
スターチ粒子に対する超音波ピッティング機構の検討

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1. Introduction

We are studying the effects of ultrasonic cavitation on microscopic samples (bacteria, algae, microcapsules, etc.). It was previously reported that one of the causes of destruction of such samples is mechanical resonance due to cavitation bubbles [1]. Observations of the samples after ultrasonic irradiation revealed the presence of numerous ruptured or deformed samples. In addition, it was observed that small holes (pits) were formed on the surfaces of some samples. It is well known that bubbles that are small enough for the sample collapse by sensing asymmetry and generate microjets. This effect of pit formation by microjets is referred to as ultrasonic pitting.

In this study, we investigated the ultrasonic pitting effect on microscopic samples. The number of pits formed on the sample surface was measured via scanning electron microscopy (SEM) after ultrasonic irradiation at various frequencies (26–3600 kHz). Furthermore, the influence of the addition of a surfactant on the ultrasonic pitting behavior was examined for 1600 kHz ultrasonic waves, which displayed the highest pitting ability.

2. Experimental

2.1. Samples

Potato starch particles (Wako Pure Chemical Industries Ltd., Japan) with a particle size distribution of approximately 10–100 μm were used in this study. The particles were typically spherical, with ellipsoidal particles also present for diameters exceeding 20 μm. Suspensions of the starch particles (0.088 wt%) in purified water were used in the ultrasonic irradiation experiments. To determine the influence of surfactant addition, sodium dodecyl sulfate (SDS; Wako Pure Chemical Industries Ltd., Japan) was added to the starch suspension at concentrations of 0.01, 0.1, 0.5, 1.0, and 10 mM.

2.2. Sonication

Fig. 1 presents a schematic diagram of the experimental apparatus. A stainless-steel cylindrical sample tank with an inner diameter of 48 mm was installed on top of an ultrasonic generator (QUAVA mini, Kaijo Corporation, Japan). The ultrasonic generator was composed of an oscillator unit and a transducer unit, where the latter contained an ultrasonic transducer with a diameter of 30 mm. The ultrasonic frequencies used were 26, 430, 1600, and 3600 kHz. Samples with a volume of 100 mL were sonicated for 4 min. The acoustic power was measured via the calorimetric method and kept constant at 10 ± 1 W. The sample temperature was maintained at 20 ± 1°C by circulating cooling water inside the sample tank.

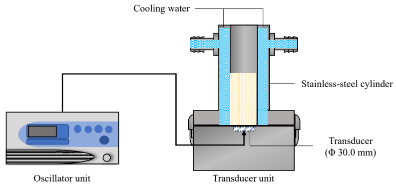

Fig. 1 Schematic diagram of experimental apparatus.

2.3. Pitting analysis

Following ultrasonic irradiation, the starch particles were collected by centrifugation and dried, and the particle surface was examined via SEM (TM3030Plus, Hitachi High-Technologies, Japan). The pits formed on the starch particle surface were counted from the SEM images. It has been reported that the number of pits is dependent on the size of the starch particles [2]. Therefore, in this study, starch particles with a diameter of 10–20 μm were used to evaluate the pitting effect. All experiments were repeated three times and the results were averaged to ensure reproducibility.

3. Results and discussion

3.1. Influence of ultrasonic frequency

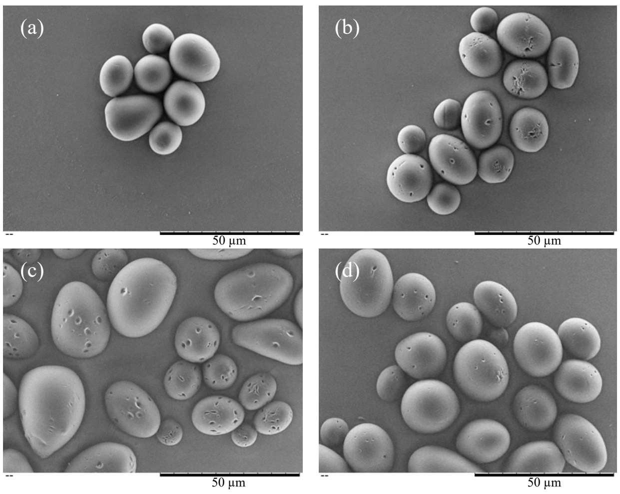

Fig. 2 SEM images of starch particle surfaces after ultrasonic irradiation at various frequencies: (a) 26 kHz, (b) 430 kHz, (c) 1600 kHz, and (d) 3600 kHz.

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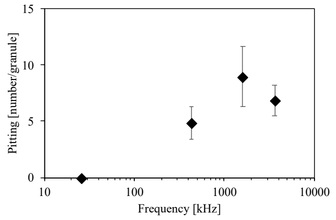

Fig. 3 Number of pits formed on the surfaces of 10–20 μm starch particles at various frequencies.

Fig. 2 presents SEM images showing the pits formed on the starch particles at various ultrasonic frequencies. **Fig. 3** shows a plot of the average number of pits per granule formed on the 10–20 μm starch particles with respect to frequency. The maximum number of pits was observed at 1600 kHz. This is thought to be attributable to the increased asymmetry of bubbles due to the reduction in bubble size [2]. It is known that the bubble size decreases with increasing frequency. Pit formation by microjets occurs when the cavitation bubbles are smaller than the target [3]. Therefore, the diameter of the bubbles responsible for forming pits on the starch particles examined in this study is considered to be equal to or smaller than the particle diameter of 10–20 μm. As a typical bubble size, we used the resonant bubble diameter calculated from the Minnaert equation. At 26 kHz, the resonant bubble diameter is 250 μm, which is much larger than the diameter of the starch particles, explaining the lack of pit formation at this frequency. The resonant bubble diameters at 430 and 1600 kHz are 15 and 4 μm, respectively. As these are smaller than the diameter of the evaluated starch particles, it is considered that under these conditions the bubble oscillation became asymmetric, resulting in the generation of microjets. This is considered to account for the increasing number of pits observed as the frequency was increased to 1600 kHz.

In contrast, at 3600 kHz, the number of pits decreased compared to 1600 kHz. Increasing the frequency leads to a decrease in the microjet velocity [4], upon which the pitting ability depends [5]. Therefore, it is considered that the number of bubbles capable of forming pits decreased at 3600 kHz, thereby decreasing the number of pits observed compared to 1600 kHz.

3.2. Influence of surfactant addition

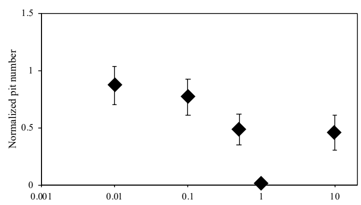

Fig. 4 Normalized number of pits formed on the surfaces of 10–20 μm starch particles at various SDS concentrations.

Fig. 4 shows the dependence of the number of pits formed at 1600 kHz, normalized to the number of pits formed in pure water, on the SDS concentration. The normalized pit number decreased up to an SDS concentration of 1.0 mM, and then increased in the presence of 10 mM SDS. Almost no pits were formed at an SDS concentration of 1.0 mM. These changes in the number of pits are considered to be attributable to the effects of SDS on bubbles and starch particles.

It has been reported that SDS adsorbs to bubbles and inhibits bubble coalescence, thereby lowering bubble size distribution [6]. Lowering the bubble size leads to a reduction in microjet stress [7]. Therefore, it is considered that the addition of SDS reduced the number of bubbles capable of pit formation.

Furthermore, the adsorption of SDS to starch particles leads to electrostatic repulsion between the particles and cavitation bubbles, thereby preventing the cavitation bubbles from approaching the particles [7]. Consequently, the cavitation bubbles cannot sense the asymmetry with respect to the starch particles. Thus, in addition to the decreased number of bubbles capable of pit formation, repulsion between the bubbles and the starch particles may be a factor in the reduced number of pits observed in the presence of SDS.

At high concentrations (10 mM), the excess SDS acts to counteract these effects [8], causing the observed number of pits to increase.

The obtained results indicate that the relative sizes of the bubbles and target particles play a crucial role in ultrasonic pitting, in addition to external factors such as solute addition.

References

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