

平成 9 年度入学 大学院博士後期課程 物質生産工学専攻 材料物理工学講座

氏 名 柳田 裕隆 ( Hiroataka YANAGIDA )

論文題目 : 超音波場の時空間的制御によるソノケミストリの反応制御の研究

英訳題目 : Control of sonochemical reaction by control of sonication condition

- **Abstract**

In this thesis, the relationship between the efficiency of sonochemical reactions and various sonication conditions have been studied in detail. A mechanism of multi-bubble sonochemical reactions shall be proposed based on the kinetics of the cavitation bubbles. Clarification of this relation and mechanism is of great use to control the sonochemical reaction by sonication condition. In order to obtain quantitative and reproducible results, it is essential to measure and control the sound field quantitatively. To achieve this, the spatial distribution of the sound wave was visualized by the schlieren method, and the sound pressure was determined by Ramann-Nath parameter. The chemical reactions induced by sonication were monitored by the sonochemical luminescence of luminol solution. These methods enable us to measure the sound field and reaction field without disturbing the sound field.

By using these methods, the sound field and reaction field were compared in various configurations. It has been found that:

- 1) Turning off the sound even for a very short period caused a decrease in the luminescence efficiency.
- 2) Traveling wave has higher threshold sound pressure than standing wave.
- 3) When a concave transducer is used, the luminescence is observed on the converging side only. The diverging side shows no luminescence at sound pressure twice as high as that of the converging side.
- 4) Superposing two sound waves produces stronger luminescence than the sum of the luminescence induced by each wave alone.

These results show that the sonochemical reaction is not determined by the sound pressure alone, but strongly depends on the sound wave forms. In order to obtain high efficiency, important condition seems to be to concentrate bubbles in pressure antinode region and keep them there sufficiently long. Since the acoustic streaming would interfere with the aggregation of the bubbles into the pressure antinode, it may suppress the sonochemical luminescence. This expectation has been confirmed by experiment, which shows that the streaming velocity of 15 cm/sec in the water makes the threshold sound pressure double. In order to get further insight into the effect of the spatial and temporal sound wave shape, the growth and decay process of the cavitation bubbles are studied in detail by measuring the transient response of the luminescence. The results suggest the following model; Cavitation bubbles are created at nuclei such as small impurity particles which present in water, and grow in two steps. In the first step, small bubble created at nuclei gradually grows. Then the second

step starts, probably when the bubble detaches from the impurity. In the second step, the bubbles concentrate in the pressure antinode and grow rather rapidly. Finally the bubble collapses, and the resulting fragments are reused as the seeds for the second step. The first step takes  $10 \text{ msec} \sim 1 \text{ sec}$  and the second step needs  $1 \sim 50 \text{ msec}$ . When the sound field is turned off, the bubbles in the second step dissolve in a few seconds, but bubbles in the first step remain for about 1 min. This model can explain most of the observed effects of the sound wave shape. For example, the burst wave has lower efficiency than the continuous wave because the short off period of the sound causes rapid decay of the bubbles in the second growth step. The traveling and diverging waves can not concentrate the bubbles, and consequently have low efficiency. If two perpendicular sound waves are superposed, bubbles are concentrated in narrower regions and reused more efficiently than the single wave case, and the luminescence intensity is strongly enhanced.

To summarize, it has been found that the spatial and temporal shapes of the sound waves have very strong effects on the efficiency of the sonochemical reaction. The mechanism of these effects were discussed by using a model of the formation, growth and decay of the cavitation bubbles. In order to achieve high efficiency, it is important to confine many bubbles near the pressure antinode, and keep them there for sufficiently long time (for example 10 msec).