음파를 이용한 Ca-Alginate Bead의 연속생산에 관한 연구

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Continuous Production of Uniform Size Ca-Alginate Beads by Soundwave-Induced Vibration

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INTRODUCTION

Many works on entrapment of biocatalysts in hydrogel has been reported[1,2,3,4]. Although the immobilization technique has been used extensively in biological research, the continuous production and size control are major impediment to wider uses[2]. High production capacity and formation of uniform size beads are essential for the employment of immobilized biocatalysts in industrial applications. Reported methods for continuous and mass production of alginate beads have been application of a atomizer[3], a electric field[6] and a vibrator[2]. These methods require expensive apparatuses, delicate control schemes or both, thus inappropriate for wide use. This study proposes a mass production method of alginate beads with the improved size and shape control at low apparatus cost with easy-to acquire apparatus. This process is based on the phenomenon of breaking up of alginate jet flow by soundwave-induced vibration by a horn type speaker. Application of a periodic vibration breaks up the power-law fluid jet and enables the high-speed production of uniformly sized droplets.

MATERIALS AND METHODS

Apparatus

Components of immobilization vessel consisted of brass, these were of fastening plate, rubber membrane, cylindrical vessel support, and two different bottom plates with nozzle diameters of 0.8mm and 1.0mm, respectively. Various concentration of sodium alginate solutions were used(1.0, 1.5 and 2.0 percent by weight)Schematic diagram of the breakup apparatus is shown in Fig. 1 and the enlarged configuration of immobilization vessel is shown in Fig. 2. The sine wave from the signal-generator(oscillator) was amplified and was converted into sound by horn type speaker. Rubber membrane was resonated with the sound produced by sine wave. This membrane delivered vibration to alginate solution and the vibration induced the breakup of alginate solution into droplets. We varied the types and thickness of membrane, and the optimum membrane was found to be 0.4mm-thick natural rubber. The membrane was fastened with 60% elongation in all directions and was fixed between the top plate and vessel support.

Bead Production

The alginate solution was pumped into the vessel at a constant flow rate by a peristaltic pump. The production rate was determined by measuring the volume of alginate solution flowing out from the immobilization nozzle in unit time. The sodium-alginate droplets were dripped into a stirred 0.2M $CaCl_2$ solution, located 0.6m below the bottom plate, and solidified to form calcium-alginate beads for 12hrs. Viscosity and surface tension were measured as the rheological properties of the solution at 30°C. The forming of the alginate droplets from the immobilization vessel was visually inspected by a stroboscope with the frequency controller. The flow rate and frequency were varied to find the optimal condition for the production of uniformly sized beads. The 30 samples of beads were taken from each experimental conditions, and then the shape and diameter of beads were measured by low powered microscope.

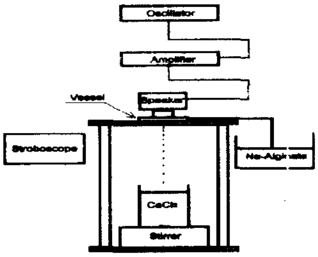

Fig 1. Schematic diagram of beads producing apparatus.

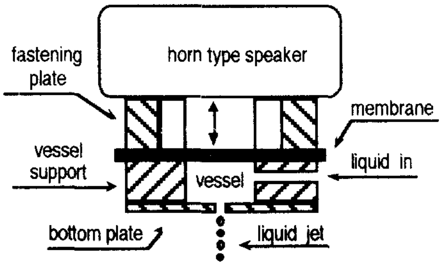

Fig 2. Enlarged configuration of the immobilization vessel

Theoretical Background

Attempts were made to mathematically explain the forming of droplets through the nozzle. Weber[8] expanded the Rayleigh's theory by including the viscosity effect of Non-newtonian fluid such as sodium alginate solution. According to Weber[8], the optimum frequency for breakup was given by;

$$f_b = \frac{u_j}{\pi d \sqrt{2(1+3V_i)}} \quad \text{with} \quad V = \frac{\mu}{\sqrt{\sigma \rho d}}$$

We investigated the agreement between Weber's theory[8] and the experimental results of our study on the formation of droplets by sound wave.

RESULTS AND DISCUSSION

When vibration was not applied to the membrane with alginate solution constantly pumped into the vessel, fluid jet solution through the nozzle formed droplets of irregular shape and size. The phenomena were due to unwanted disturbances from the bottom plate nozzle edge and the interaction between flow rate and viscosity of solution. Proposed by Weber[8], the major disturbance to fluid jet breakup was a flow rate of alginate solution. When the membrane was vibrated by sound wave, the fluid jet was resonated with the frequency of vibration. Droplets of uniform size and shape were formed at certain frequency ranges. The phenomena indicated that the application of vibration would reduce the influence of disturbances above mentioned. Fig. 3 is the photograph of droplet formation of 2%(w/w) alginate solution through the nozzle(diameter=1.0mm). Fig. 4 shows the calcium alginate beads solidified in CaCl2 solution(0.2M), with the average mean diameter of 1.8mm.

Fig. 5 and Fig. 6 show the relationship between breakup frequency and flow rate of jet fluid at two different nozzle diameters varying the concentration of sodium alginate solution. The vertical bar in Fig. 5 and 6 represent the frequency ranges for the formation of uniform droplets at each experimental conditions. At transient frequencies, which was at the both end of droplets forming frequency ranges, the jet breakup was irregular and no breakup was observed outside of either end of droplets forming frequency ranges.

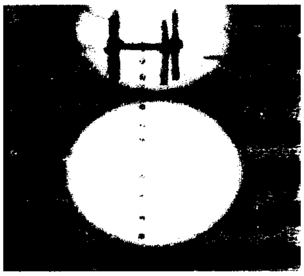

Fig. 3 Photograph of droplets forming of 2wt% alginate - solution.

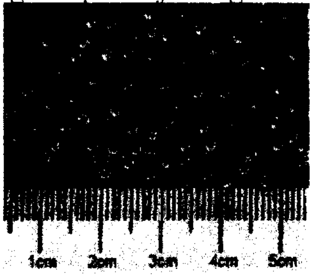

Fig. 4 Photograph of Alginate beads(2wt% sodium alginate)

As the sodium-alginate concentration was increased, the reduction of droplets forming-frequency ranges was observed with the same nozzle diameter and the same flow rate of alginate solution. This resulted from the increase in the viscosity of sodium alginate solution as the increase of the concentration of alginate solution. But the concentration change of alginate solution contributes little to the surface tension. The viscosity of the sodium alginate solution was found to be more important rheological property than the surface tension. Breakup frequency increased as the fluid flow rate increased, and decreased as the viscosity of alginate solution increased. The nozzle diameter was inversely proportional to the droplets-forming frequency at the constant flow rate because the nozzle with larger diameter formed the relatively larger droplets and the formation of large droplets at constant flow rate implied the reduction of breakup frequency .The breakup frequency ranges for the production of uniform size droplets were expanded as the fluid flow rate was increased. The linear lines in Fig. 5 and 6 represented the calculated relationships between breakup frequency and flow rate by Weber[8]. The experimental results of breakup frequency agreed well with the optimum frequency which was estimated by Weber's theory[8] at 1.0mm of nozzle diameter but was shown a minor difference at 0.8mm of nozzle diameter. These figures shows that the theory of Weber[8] is in good accordance with the results of our experiments and the method of applying a vibration by soundwave is practical for the production of uniformly sized droplets. The size of beads was measured and the results are shown in Table 1.

Table 1. describes the relationship among flow rate, frequency and bead size. By varying nozzle diameter, flow rate and frequency, diameter of the beads could be controlled on the range of 1.50~3.50mm. The uniformity of bead size were dependent on the

flow rate and breakup frequency, and the uniformity of bead shape was significantly improved by adjusting the breakup frequency and reducing the shock at the surface of stirred $CaCl_2$ solution.

Table 1. Relationship among flow rate, frequency, beads size at different bottom plate nozzle diameter.

d0=0.8mm				d0=1.0mm			
Q (L/h)	f (s^{-1})	l (cm)	d_s (mm)	Q (L/h)	f (s^{-1})	l (cm)	d_s (mm)
4.08	85	2.65	2.32	4.08	41	3.51	3.16
	326	0.69	1.72		141	1.02	2.03
4.80	109	2.43	2.25	4.80	53	3.20	2.81
	502	0.53	1.63		190	0.89	1.80
5.88	223	1.45	1.92	5.88	62	3.35	2.97
	672	0.48	1.58		245	0.85	1.90

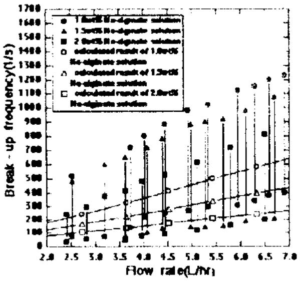

Fig.5 Relationship between droplets forming frequency and flow rate at different sodium alginate conc. (nozzle diameter=0.8mm)

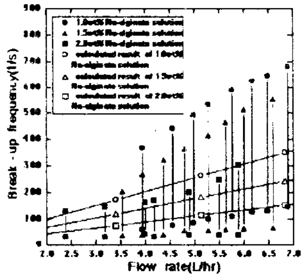

Fig. 6 Relationship between droplets forming frequency and flow rate at different sodium alginate conc. (nozzle diameter=1.0mm)

NOTATION

u_j=jet velocity(m/s)
λ = wavelength[m]
ρ = liquid density[kg/m^3)
Vi=viscosity number(dimensionless)
μ = liquid viscosity [Pa· s]
σ = liquid surface tension [N/m]

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