CIRCULATION TIME DISTRIBUTION AND VOLUME OF MIXING REGIONS IN HIGHLY VISCOUS XANTHAN GUM SOLUTION IN A STIRRED VESSEL

HITOSHI FUNAHASHI, HIROAKI HARADA AND HISAHARU TAGUCHI

Department of Fermentation Technology, Osaka University, Suita 565

TOSHIOMI YOSHIDA

International Center of Cooperative Research in Biotechnology, Osaka University, Suita 565

Key Words: Agitation, Non Newtonian, Xanthan Gum, Circulation Time, Mixing, Mixing Region

The distribution of circulation time of a tracer particle and the volume of mixing regions were investigated using a pseudoplastic solution at various concentrations of xanthan gum up to 4.6% in a stirred tank with a turbine impeller having six flat blades. When the relative frequency/ N of each circulation time was plotted against $N\theta$ (N , agitation speed; θ , circulation time) at various agitation speeds, the distribution curve for solutions of 1.8% (flow behavior index 0.12 and consistency coefficient $26.4 \text{ Pa} \cdot \text{s}^{0.12}$) or more xanthan gum shifted to the side of larger values of $N\theta$ with increasing agitation speed, while distribution curves of the same shape were obtained in water or a diluted solution (0.9% xanthan gum). The mean circulation time also increased with increasing agitation speed. This phenomenon was confirmed by measuring the volumes of two mixing regions, the micromixing and the macromixing regions. These volumes increased with increases in the agitation speed, resulting in decreases of the stagnant region.

Introduction

The shear stress is an important factor in the microbial production of xanthan gum.³⁾ Xanthan gum solution behaves as a pseudoplastic fluid.²⁾ In a fermentor, the culture broth of this solution becomes viscous during cultivation and homogeneous mixing of the broth becomes difficult. The intensity of mixing in a stirred tank and the residence time of cells in the region of high shear stress may be important for the microbial production of xanthan gum. In a viscous fluid, the intensity of shear is high only in the region close to the impeller, and decreases further away from that region.⁵⁾ This phenomenon of non-homogeneity is significant in a pseudoplastic fluid. To assess the production of xanthan gum, it is important to investigate the circulation of a volumetric element among the regions of high shear stress, low shear stress, and stagnancy. Circulation time distributions in fluids with low viscosity have been studied.^{4,6,10,11)} It was suggested that the logarithmic regulatory distribution may fit for fluids with low viscosity. In continuous culture of high cell concentration of yeast¹⁾ and in mycelial growth,^{7,8)} good results of simulation have been obtained by use of models in which the circulation time distribution was considered. However, the circulation time distribution in a

highly viscous non-Newtonian fluid has not been investigated. Such information is needed to find proper operating conditions or to simulate the production of a highly viscous product such as xanthan gum.

This paper describes findings about the circulation time distribution in a pseudoplastic fluid, xanthan gum solution, in a stirred tank with a turbine impeller having six flat blades. We found that the xanthan gum concentration, the agitation speed, and the aeration affected the circulation time distribution, variation in the volume of each flow region, and the mean circulation time.

1. Materials and Methods

1.1 Fermentor and impeller

The fermentor used was of a 10-l capacity and an inner diameter of 200 mm (B.E. Marubishi Co., Ltd., MODEL MD500). The total liquid volume was 6 l. In the case of aeration, air was supplied at 0.10 l/s. The tank was equipped with three baffle plates. The impeller used was a turbine impeller with six flat blades.³⁾ The diameter of the impeller, the height of the blade and the width of the blade were 100, 20 and 25 mm, respectively. The diameter of the disk was 66.7 mm. The impeller was set at half of the liquid height. The agitation speed was from 5 to 20 r.p.s.

1.2 Preparation of xanthan gum solution

The xanthan gum used was a commercial prepara-

Received September 19, 1986. Correspondence concerning this article should be addressed to H. Funahashi.

tion (Taiyo Kagaku Co., Ltd.) and xanthan gum solution prepared was of concentrations less than 4.6%. A method previously described³⁾ was used for the measurement of xanthan gum concentration. The values of consistency coefficient and flow behavior index are shown in Table 1. The consistency coefficient and the flow behavior index were estimated by use of a Brookfield-type viscometer (Tokyo Keiki Seizousyo) at 30°C. The values of the flow behavior index were about 0.12 in all solutions of 0.9% or more xanthan gum. Hence the xanthan gum solutions used in this investigation showed pseudoplastic behavior.

1.3 Circulation time distribution

The inside of the stirred tank could not be seen, because xanthan gum solution is turbid. The circulation time was measured by the method of Mukataka *et al.*,⁶⁾ which involves use of a magnetic particle as the tracer. A diagram of the instrument is shown in Fig. 1. A copper wire was coiled 240 times around the stirred tank at the height of the impeller. As a tracer, a cork sphere was used in which a cobalt magnet was embedded. Its specific gravity and diameter were adjusted to about 1 and 5 mm, respectively. Mukataka *et al.*⁶⁾ reported that the size of the tracer does not affect the circulation time in the case of a tracer of 5 mm diameter in a stirred tank of 134 mm inner diameter. When the tracer passed across the coil of copper, it induced a pulse of electric current. The pulses were introduced into a micro-computer (NEC PC9800). The time between two pulses was measured to estimate the circulation time at the minimum unit of 10 msec and the number of measurements for each condition was from 5,000 to 12,000 circulation times.

Figure 2 shows the circulation time distribution in 0.9% xanthan gum solution without aeration, and at 5 r.p.s. agitation speed. The form of the distribution curve agreed well with the results reported by Yuge *et al.*¹¹⁾ Moreover, the mean circulation time calculated from this result also agreed well with the result of Yuge *et al.*¹⁰⁾ at the same apparent viscosity as calculated from the mean shear rate in a stirred tank according to Metzner *et al.*⁵⁾ We concluded that the circulation time with xanthan gum solution could be measured by this method.

1.4 Volume of mixing regions

We assumed by observation of the flow pattern that the whole volume of the stirred tank was divided into three regions. The first was the micromixing region, dominated by radial flow near the impeller. The second was the macromixing region, the region of slowly circulating flow. The third was the stagnant region, in which the fluid stagnated.

The volume of each region was calculated by analysis of the dispersion rate of a tracer, NaOH, obtained from the pH change after injection of 0.2 kg/l NaOH solution at a point near the impeller.¹¹⁾

Table 1. Consistency coefficient and flow behavior index of the power law equation at various concentrations of xanthan gum

Xanthan gum concentration [%]	Consistency coefficient [Pa · s ⁿ]*	Flow behavior index [—]
0.9	8.8	0.10
1.8	26.4	0.12
2.7	40.7	0.11
3.9	63.8	0.12
4.6	80.1	0.12

* *n* is the value of flow behavior index in each xanthan gum solution.

Fig. 1. Diagram of instrument for measurement of circulation time and volume of mixing regions.

Fig. 2. Circulation time distribution of a particle in 0.9% xanthan gum solution without aeration when relative frequency was plotted against circulation time.

The experiments were carried out as follows. (1) Xanthan gum solution containing 1% NaCl was prepared to stabilize the viscosity and the initial pH was adjusted at 6.0. (2) One or two ml of 0.2 kg/l NaOH solution was injected near the impeller, which rotated at a constant agitation speed. (3) The pH near the impeller was measured for about 20 min and the

NaOH concentration was estimated by means of a calibration curve that correlated NaOH concentration to pH in a solution of the same constituents. In this case, the time constant of the pH measurement system was 0.38 sec, which is short enough to neglect the delay of the pH measurement system. An example of the response curves is shown in Fig. 3. This curve was obtained when a stagnant region existed in the stirred tank. The curve is shown divided into three sections, **a**, **b** and **c**. They represent the pH change by the injected tracer as it dispersed in the micromixing region, as it dispersed mainly from the micromixing region into the macromixing region, and as it dispersed from both the micromixing and macromixing regions into the stagnant region, respectively. The tracer concentration, C^* , and the volume in the mixing region, V^* , are calculated by the tracer concentration at the point of transferral from section **b** to **c** and by the amount of tracer added. Assuming that the tracer in both the micromixing and macromixing regions was homogeneous and that the dispersion of the tracer into the stagnant region could be neglected during the earlier measurement, the mass balance equations in section **b** can be given as follows:¹¹⁾

$$-V_{\text{micro}}(dC_{\text{micro}}/dt) = q(C_{\text{micro}} - C_{\text{macro}}) \quad (1)$$

$$-V_{\text{macro}}(dC_{\text{macro}}/dt) = q(C_{\text{macro}} - C_{\text{micro}}) \quad (2)$$

To solve the equations simultaneously and to substitute the values of C^* and V^* :

$$\ln(C_{\text{micro}}/C^* - 1) = \ln(V^*/V_{\text{micro}} - 1) - q(1/V_{\text{micro}} + 1/V_{\text{macro}})t \quad (3)$$

When the term on the left-hand side of the equation, $\ln(C_{\text{micro}}/C^* - 1)$, was plotted against t [min], V_{micro} could be obtained from the value of the intercept on the vertical axis. If there was no stagnant region in the stirred tank, V^* was equal to the total liquid volume, V , and the value of V_{micro} can be obtained by substituting V into V^* of Eq. (3). Finally, the value of V_{macro} was estimated from $(V^* - V_{\text{micro}})$ by use of the estimated values of V_{micro} and V^* .

2. Results and Discussion

2.1 Circulation time distribution

1) Circulation time distribution without aeration

The circulation time distribution in water and in 0.9%, 1.8% and 3.9% xanthan gum solutions without aeration at various agitation speeds are shown in Figs. 4, 5, 6 and 7, respectively. $N\theta$ is a dimensionless term representing the circulation time; it stands for the number of revolutions of the impeller during one circulation of a tracer.⁹⁾ $R(\theta)/N$ [s] was plotted against $N\theta$. $R(\theta)$ is a relative frequency defined by the equation

Fig. 3. An example of plotting $(C_{\text{micro}}/C_{\text{final}} - 1)$ against time.

Fig. 4. Circulation time distribution of a particle in water without aeration. Relative frequency/ N was plotted against $N\theta$.

Fig. 5. Circulation time distribution of a particle in 0.9% xanthan gum solution without aeration.

Fig. 6. Circulation time distribution of a particle in 1.8% xanthan gum solution without aeration.

Fig. 7. Circulation time distribution of a particle in 3.9% xanthan gum solution without aeration.

$$\int_0^{\infty} R(\theta) d\theta = 1.$$

It was suggested⁹⁾ that an identical distribution curve of circulation time may be obtained at various agitation speeds in less viscous fluids, because circulation time decreases as agitation speed increases. It was confirmed for the distribution curves of circulation time in less viscous fluids such as water or 0.9% xanthan gum solution (Figs. 4 and 5) that they were almost independent of agitation speed, as suggested elsewhere.⁹⁾ However, the proportion of longer circulation times increased with increasing agitation speed in highly viscous fluids, and the distribution curves shifted to the side of larger values of $N\theta$ with increasing agitation speed (Figs. 6 and 7). That is, the circulation time of a particle in high-concentration xanthan gum solution does not decrease in proportion to increases in the agitation speed, and the mean residence time in the macromixing region do not shorten with increases in the agitation speed. This result suggests the existence of a stagnant region in the stirred tank, and it is speculated that the volumes of the mixing regions increased, reducing the volume of the stagnant region with increasing agitation speed at a high concentration of xanthan gum. This phenomenon might become obvious with increases in the fluid viscosity.

The distribution curves for the less viscous solutions tested had two peaks, and movement of the fluid was observed at the free surface of the solution in the stirred tank at every agitation speed tested. The curve for the 1.8% xanthan gum solution had two peaks at agitation speeds of 11.7 r.p.s. or more. This is the critical agitation speed at which the free surface began to move, indicating that the stagnant region no longer existed. From these results, we speculate that the peak at the smaller value of $N\theta$ may correspond to circulation within the micromixing region and the other within the macromixing region. The distribution of circulation time in 3.9% xanthan gum solution had only one peak.

2) Circulation time distribution with aeration The circulation time distribution in the 1.8% xanthan gum solution with aeration is shown in Fig. 8. Results were similar to those without aeration. However, the proportion of longer circulation time with aeration was greater than that without aeration at the same agitation speed. This was observed at every xanthan gum concentration tested. Perhaps the frequency of the tracer transfer from the micromixing to the macromixing region increases and the frequency from the macromixing to the micromixing region decreases because of air bubbles from the aeration.

3) Mean circulation time The mean circulation time in a less viscous fluid decreases with increasing agitation speed, because the velocity of the tracer increases with increasing agitation speed.¹⁰⁾ The effects of the agitation speed on the mean circulation time are shown in Fig. 9. The mean circulation time for water and for 0.9% xanthan gum solution decreased with increasing agitation speed, as reported before.¹⁰⁾ When the xanthan gum concentration was 1.8% or more, the mean circulation time increased as the agitation speed increased provided that it was less than the critical agitation speed. The mean circulation time decreased as the agitation speed increased beyond the critical value. This phenomenon was also observed with aeration at concentrations of 1.8% or more xanthan gum. The cause of the phenomenon may be that the moving distance of the tracer increases as the agitation speed increases, because of an increase in the volume of the two mixing regions at higher agitation speeds. To clarify this point, the change in the volumes of the micromixing and macromixing regions was investigated at various agitation speeds and xanthan gum concentrations.

2.2 Volume of mixing regions

The effects of the agitation speed on the volumes of the micromixing and macromixing regions are shown in Fig. 10 for 1.8% xanthan gum solution. The volume of both mixing regions increased with increasing agitation speed below a critical value. Above a critical agitation speed, the volume of the micromixing region continued to increase with increasing agitation speed until it reached the total liquid volume, while the volume of the macromixing region decreased. This phenomenon was observed in every xanthan gum solution of concentrations of 1.8% or more, both with and without aeration. These results suggest that the increase in mean circulation time with increasing agitation speed might be caused by increases in the volume of each mixing region as long as the agitation speed was below the critical value. The critical value shifted to a larger value with increasing xanthan gum concentration, because of increases in the viscosity resistance. Perhaps the shift of the distribution curves to the side of larger $N\theta$ values in highly viscous fluids

Fig. 8. Circulation time distribution in 1.8% xanthan gum solution with aeration.

Fig. 9. Effects of agitation speed on mean circulation time in water, and 0.9%, 1.8% and 4.6% xanthan gum solutions.

Fig. 10. Effects of agitation speed on volume of each mixing region with and without aeration. Xanthan gum concentration was 1.8%.

was caused by increases in the volume of the two mixing regions with increases in the agitation speed.

Consequently, the volume of each mixing region—the micromixing region, the macromixing region and the stagnant region—should be considered when a model for the prediction of the circulation time distribution is constructed.

Conclusion

The circulation time distribution and the volume of mixing regions were investigated at various xanthan

gum concentrations up to 4.6% in a stirred tank with a turbine impeller having six flat blades. The results obtained in this investigation are summarized as follows:

1. The circulation time at a high agitation speed could be measured by the present method using a magnetic tracer and a micro-computer.

2. The circulation time distribution in a less viscous fluid had two peaks when the agitation was strong enough to have no stagnant region in the stirred tank.

3. The following results were observed in xanthan gum solution of a concentration of 1.8% or more, both with and without aeration. The distribution curve plotted against $N\theta$ shifted to the side of larger value of $N\theta$ with increasing agitation speed as long as a stagnant region existed in the stirred tank. The critical agitation speed for disappearance of the stagnant region became larger with increasing fluid viscosity.

4. The mean circulation time increased with increasing agitation speed when the stagnant region existed.

5. The results obtained for the volume of each mixing region suggested that the above-mentioned phenomena might be caused by an increase in the volume of two mixing regions, the micromixing and the macromixing regions, with increasing agitation speed.

Therefore, the relationship between the circulation time distribution and the volumes of the mixing regions should be considered for construction of a model to predict the circulation time distribution in a highly viscous pseudoplastic fluid.

Nomenclature

C_{final}	= tracer concentration when tracer is dispersed homogeneously throughout vessel	[kg/l]
C_{macro}	= tracer concentration in macromixing region	[kg/l]
C_{micro}	= tracer concentration in micromixing region	[kg/l]
C^*	= tracer concentration in mixing region when tracer is dispersed homogeneously in micromixing and macromixing regions	[kg/l]
N	= agitation speed	[1/s]
q	= exchange rate between micromixing and macromixing regions	[l/s]
$R(\theta)$	= relative frequency	[—]
t	= time	[min]
V	= total liquid volume in stirred tank	[l]
V_{macro}	= volume of macromixing region	[l]
V_{micro}	= volume of micromixing region	[l]
V^*	= volume of micromixing and macromixing regions	[l]
θ	= circulation time	[s]

Literature Cited

- 1) Bajpai, R. K. and M. Reuss: *Can. J. Chem. Eng.*, **60**, 384 (1982).
- 2) Elliott, J. H.: "Extracellular Microbial Polysaccharides" (ACS Symposium Series No. 45, P. A. Sanford and A. Laskin, eds.), p. 144, American Chemical Society, Washington, D. C. (1977).
- 3) Funahashi, H., M. Maehara, H. Taguchi and T. Yoshida: *J. Chem. Eng. Japan*, **20**, 16 (1987).
- 4) Inoue, I. and K. Sato: *Kagaku Kōgaku*, **33**, 293 (1969).
- 5) Metzner, A. B. and J. S. Taylor: *AIChE J.*, **6**, 109 (1969).
- 6) Mukataka, S., H. Kataoka and J. Takahashi: *Kagaku Kogaku Ronbunshu*, **2**, 628 (1976).
- 7) Mukataka, S., H. Kataoka and J. Takahashi: *Kagaku Kogaku Ronbunshu*, **3**, 416 (1977).
- 8) Mukataka, S., H. Kataoka and J. Takahashi: *Kagaku Kogaku Ronbunshu*, **4**, 34 (1978).
- 9) Sato, K. and I. Inoue: *Kagaku Kōgaku*, **37**, 937 (1973).
- 10) Yuge, K. and E. Oshima: *Kagaku Kōgaku*, **33**, 898 (1969).
- 11) Yuge, K. and E. Oshima: *Kagaku Kōgaku*, **34**, 439 (1970).

EMULSION PHASE EXPANSION AND SEDIMENTATION VELOCITY IN FLUIDIZED BEDS OF FINE PARTICLES

TAKAMI KAI, AKIKO IWAKIRI AND TAKESHIGE TAKAHASHI

Department of Chemical Engineering, Kagoshima University, Kagoshima 890

Key Words: Fluidization, Fluidized Bed, Bed Expansion, Minimum Fluidization Velocity, Sedimentation

In fluidized beds of fine particles, minimum fluidization velocity U_{mf} , voidage of the emulsion phase ε_e and sedimentation velocity U_s were measured for various gas-powder systems. For both U_{mf} and U_s , the relationship between velocity and voidage was experimentally determined on the basis of the Richardson-Zaki equation. The voidage of the emulsion phase was correlated by a dimensionless parameter taking into account the effect of gas density. By using this correlation and the relationship between velocity and voidage obtained in this study, U_s was expressed as a function of U_{mf} . The calculated U_s from U_{mf} agreed with the experimental data.

Introduction

Many industrial fluidized catalytic reactors use fine powder catalysts less than 100 μm in diameter, since good fluidization and good gas-solid contact can be easily established in this size range. Beds of fine, low-density particles homogeneously expand between minimum fluidization and minimum bubbling points. When the beds are fully fluidized, voidage of the emulsion phase is also higher than that of settled beds or that at minimum fluidization. The extent of expansion of the emulsion phase is an important factor⁶⁾ in the prediction of the bed's performance as a chemical reactor and in the estimation of the apparent viscosity of the emulsion phase, which relates to bubble size and fluidization quality.

The purpose of this study is to investigate the expansion behavior of the emulsion phase and to reveal the influence of the properties of gas and particles on the expansion. We measured minimum fluidization velocity U_{mf} , sedimentation velocity U_s and the voidage of the emulsion phase ε_e for various

gas-powder systems.

1. Experimental

The physical properties of the particles used in this study are given in **Table 1**. All powders are within Group A of the Geldart³⁾ classification when air is used as the fluidizing medium. The particles were moistened in the pores to decrease the influence of electrostatic charges.^{7,10)}

The experiments were carried out in an acrylic resin column 5.5 cm in diameter and 2.2 m in height under ambient conditions. The settled bed height above a gas distributor was 0.6 m. The distributor was made of sintered brass and was 3 mm thick. Eight gases were used as the fluidizing gas: argon, oxygen, helium, nitrogen, carbon dioxide, ethylene, methane and hydrogen.

The minimum fluidization velocity was determined from the usual plot of pressure drop against superficial gas velocity. The expansion behavior of the emulsion phase was investigated by using the bed collapse technique.^{12,15)} After the gas velocity had been set at about 0.1 m s^{-1} and the bed had been well fluidized, the gas supply was suddenly stopped ($t=0$)

Received September 26, 1986. Correspondence concerning this article should be addressed to T. Kai.