Title	Flotation of blue-green algae using methylated egg ovalbumin
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Citation	Chemical Engineering Journal, 155(1-2), 49-54 https://doi.org/10.1016/j.cej.2009.06.011
Issue Date	2009-12-01
Doc URL	http://hdl.handle.net/2115/39591
Type	article (author version)
File Information	CEJ-D-07-00971_authorversion.pdf

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Flotation of Blue-green Algae Using Methylated Egg Ovalbumin

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Abstract

Removal of blue-green algae by dispersed gas flotation was conducted. Methylated ovalbumin (MeOA) was used as frother and flocculant, which is a biodegradable substance. The continuous flotation experiments were conducted at different feed mass flow rate of the blue-green algae cells and MeOA. The operating variables were the mass flow rate of blue-green algae cell and MeOA, the initial concentration of the cells and MeOA, and superficial gas velocity. The results showed that the mass flow rate of MeOA was most dominant variable affected to the removal efficiency and that the removal efficiency achieved ca. 0.85 when a ratio of the mass flow rate of MeOA and that of the cells was over 0.3. A proposed flotation model considering the adsorptions of MeOA to the cells, MeOA to bubble surface and the cells bearing with MeOA to bubble surface was applied to explain the experimental removal efficiency. The experimental and the calculated removal efficiency agreed well within error 19 %, indicating that the proposed model was valid fundamentally.

Keywords: flotation, methylated egg ovalbumin, blue-green algae, removal, clarification

1. Introduction

Blue-green algae are kinds of freshwater phytoplankton in surface water and are of concern due to abilities to produce taste and odor compounds, which have hepatotoxic, cytotoxic and dermatotoxic behaviors, being harmful to animal and human. Therefore, removal of blue-green algae cells has become important to save healthy water environments used as water resources.

Recently removal of these harmful blue-green algae cells has become important process for keeping clean water environment. Several attempts for removal of blue-green algae have been reported [1-4]. Among them, dissolved air flotation (DAF) method [5-9] has been a promising technique for more effective treatment of blue-green algae removal. The apparatus for DAF, however, have many mechanically parts and its operation is not ease. On the other hand, flotation by dispersed gas have some advantages: low energy requirements, a little mechanical parts in the apparatus, no-requirement of tedious treatments such as desorption or addition of any other chemicals and extending to a continuous operation with ease, although removal efficiency for the flotation is a few lower than that of DAF. The authors suppose that combination of flocculation of blue-green algae cells and flotation of its floc will be more effective.

Flocculants mostly accepted are chemically synthesized high molecular weight polymers and alum. These flocculants are environmentally undesirable, because chemical synthesized flocculants especially remain in natural environments for long period of time without degradation to less harmful form. Recently the use of extracellular polymers produced by some bacteria have been attempted to remove suspended solid as bioflocculant [10-17]. Such bioflocculants, however, have to be separated or purified from culture media by intricate treatment, which makes them expensive for practical application.

In the present study, the authors conducted flotation experiments for removal of

blue-green algae by using methylated egg ovalbumin (MeOA), which is a biodegradable surfactant proposed by our previous study [18]. The aims of the present study are clarification of the effective operating variables on the continuous flotation of blue-green algae and verification of the proposed flotation model.

2. Flotation model

The present model assumed the following points on the basis of observations and the preliminary experiments; (i) the volumetric flow rate of the liquid within the foam is negligible small than the volumetric flow rate of the drain liquid, and (ii) the adsorption densities of MeOA on the blue-green algae and on the bubble surface are much smaller than the saturated adsorption densities on these, respectively. The detail explanations of the present model are described in follows.

According to the mass balance about bubble dispersed phase within the column, the next relationship is obtained:

$$V \frac{dC_s}{dt} = w_{si}C_{si} - w_bC_s - w_fC_f \quad (1)$$

Where, C_{si} , C_f and C_s represent the initial concentration of feed solution, the bulk liquid concentration and the foamate (the collapsed foam liquid) concentration for blue-green algae, respectively. w_{si} , w_b and w_f represent the volumetric flow rate of the feed, the drain and liquid within the foam, respectively. V is the volume of the liquid within the column. The term, w_fC_f , in the right hand side of Eq. (1) correspond to the overhead mass flow rate of blue-green algae and can be expressed as:

$$w_fC_f = w_fC_s + S_bX \quad (2)$$

Where, S_b and X represent the production rate of bubble surface area and adsorption density of blue-green algae on bubble surface, respectively. By substituting Eq. (2) into Eq (1), Eq. (1) is rewritten as:

$$V \frac{dC_s}{dt} = w_{si}C_{si} - (w_b + w_f)C_s - S_b X \quad (3)$$

Judging from the preliminary experiments, the value of w_f was regarded as very small value than that of w_b . Then, Eq. (3) can be expressed approximately as:

$$V \frac{dC_s}{dt} = w_{si}C_{si} - w_b C_s - S_b X \quad (4)$$

From Eq. (4), the following relationship in the steady state within the column liquid can be obtained as:

$$w_{si}C_{si} = w_b C_s + S_b X \quad (5)$$

The removal efficiency, η , of blue-green algae is defined as:

$$\eta \equiv \frac{w_{si}C_{si} - w_b C_s}{w_{si}C_{si}} \quad (6)$$

The adsorption density, X , of blue-green algae can be expressed as:

$$X = N\theta = \frac{NKC_s}{1 + KC_s} \quad (7)$$

Where, K , N and θ represent the equilibrium adsorption constant, the number of available site for adsorption of blue-green algae onto bubble surface and the coverage fraction of bubble surface by blue-green algae, respectively. The authors employed Langmuir adsorption isotherm as the adsorption equilibrium relationship. N can be expressed as:

$$N = N_m \theta_b \quad (8)$$

Where, N_m and θ_b are the saturated adsorption density of blue-green algae and the coverage fraction of MeOA adsorbed on bubble surface, respectively. Moreover, adsorption density, X_b , of MeOA on bubble surface and θ_b can be expressed as:

$$X_b = \frac{N_b K_b C_p}{1 + K_b C_p} \quad (9)$$

$$\theta_b \equiv \frac{X_b}{N_b} = \frac{K_b C_p}{1 + K_b C_p} \quad (10)$$

Where, C_p , K_b and N_b represent the equilibrium adsorption concentration, the equilibrium adsorption constant and the saturated adsorption density of MeOA, respectively. MeOA molecule can also adsorb onto blue-green algae surface, then, the adsorption equilibrium relationship can be expressed as in the same fashion:

$$X_s = \frac{N_s K_s C_p}{1 + K_s C_p} \quad (11)$$

$$\theta_s \equiv \frac{X_s}{N_s} = \frac{K_s C_p}{1 + K_s C_p} \quad (12)$$

Where, K_s , N_s , X_s and θ_s represent the equilibrium adsorption constant, the saturated adsorption density, the adsorption density of MeOA on blue-green algae and the coverage fraction of MeOA adsorbed on blue-green algae surface, respectively. In the present system, all, MeOA, blue-green algae cells and bubble surface present in the same liquid. Therefore, the equilibrium concentrations of MeOA involved in both the adsorption of

MeOA on cells surface and that on bubble surface cannot be classified. In Eqs. (9) and (11), Langmuir adsorption isotherm is employed for adsorption equilibrium relationship between MeOA and bubble surface, and, MeOA and blue-green algae surface. In the present conditions, X_s and X_b can be assumed to be much smaller than N_s and N_b , respectively. This assumption makes Eqs. (9) and (11) simply as follows:

$$X_s = N_s K_s C_p \quad (13)$$

$$X_b = N_b K_b C_p \quad (14)$$

On the other hand, at the steady state, the mass balance of MeOA within the column can be expressed as:

$$w_{pi} C_{pi} = w_b C_p + w_{si} C_{si} X_s + S_b X_b \quad (15)$$

C_{pi} and w_{pi} represent the concentration of MeOA feed solution and the volumetric flow rate of MeOA feed solution. By substituting X_s and X_b expressed as Eqs. (13) and (14) into Eq. (15), C_p can be solved as:

$$C_p = \frac{w_{pi} C_{pi}}{w_b + w_{si} C_{si} N_s K_s + S_b N_b K_b} \quad (16)$$

From Eqs. (5), (7), (8), (10) and (16), the following relationship concerning to X is derived as:

$$X^2 - \frac{w_b / K + N_m S_b \theta_b + w_{si} C_{si}}{S_b} X + \frac{N_m \theta_b w_{si} C_{si}}{S_b} = 0 \quad (17)$$

By solving Eq. (17) in terms of X , X is expressed as:

$$X = \frac{(w_b + KN_m S_b \theta_b + Kw_{si} C_{si}) - \sqrt{(w_b + KN_m S_b \theta_b + Kw_{si} C_{si})^2 - 4K^2 N_m S_b \theta_b w_{si} C_{si}}}{2K^2 N_m S_b \theta_b w_{si} C_{si}} \quad (18)$$

From Eqs. (5) and (6), the removal efficiency, η , of blue-green algae is rewritten as:

$$\eta = \frac{S_b X}{w_{si} C_{si}} \quad (19)$$

From Eqs. (18) and (19), the calculated value of η can be estimated by determination of K , K_s , N_s , and N_m by least square method, and the presented model can be verified with the experimental data.

3. Experimental

3.1 Materials

3.1.1 Blue-green algae

Blue-green algae used in the present study were collected from Lake Ohnuma, Oshima, Hokkaido, Japan. The cells were washed with distilled water and were used for flotation experiments. The cell diameter distribution is shown in Fig. 1 and was measured by laser scattering particle size analyzer (HORIBA LA-300, Japan).

3.1.2 Methylated egg albumin

Preparation method of methylated egg albumin (MeOA) was almost the same manner in the previous study [18]. Egg ovalbumin (OA) was methylated according to the method reported by Fraenkel-Conrat and Olcott [19]. An aqueous solution of OA (ca. 10 g/L) was prepared, and a 0.1 M HCl solution was added. At pH 4.6 (isoelectronic point of OA), OA precipitated, and the mixture was centrifuged at 3000 rpm for 20 min. The precipitated OA was washed with methyl alcohol and dispersed in a 100-fold volume of methyl alcohol containing 0.05 M HCl. This solution was stirred for 24 h at room temperature. The methylated OA (MeOA) was collected in a centrifuge at 3000 rpm for 20 min and then washed with distilled water. The MeOA was dialyzed for 24 h to remove methyl alcohol and HCl. The degree of methylation was determined from the change in the number of carboxylic groups before and after methylation by a potentiometric titration [18]. In this study, MeOA with methylation degrees of 88 % was used.

3.2 Experimental setup and procedures

3.2.1 Flotation experimental setup

A schematic diagram of the experimental setup is shown in Fig. 2. The experimental setup used in this study was mostly same as that used in the previous study [20]. A bubble column of 4.4 cm in inside diameter and 0.4 m in height was employed. The column was made of transparent acrylic resin. Sintered glass filter (10-15 μm mean-pore size) was installed as a gas distributor at the bottom of the column. Liquid feed and drain were installed at the center and the bottom of the column, respectively. Pressure taps for measuring gas holdup, ε_G , in the column were installed along the wall at intervals of 25 cm. From the value of the measured ε_G , the surface area production rate, S_b , of bubble with in the column was estimated by the following equation [21, 22].

$$S_b = 6 A \varepsilon_G (1 - \varepsilon_G)^{4.65} \{ (4/225) (\rho_L - \rho_G)^2 g^2 / (\mu_L \rho_L) \}^{1/3}, \quad (20)$$

Where, A , g , ρ_G , ρ_L and μ_L are the cross sectional area of the bubble column, gravitational acceleration, gas density, liquid density and liquid viscosity, respectively.

3.2.2 Flotation experimental procedure

Suspended solution of blue-green algae and MeOA solution were prepared at the desired concentration, respectively. The ionic strength of these was controlled at 0.005 M with NaCl. The column was filled with 0.005 M NaCl solution, then, air was supplied from air compressor and was dispersed as bubble through the gas distributor. After confirmation of stable bubbly flow of air within the column, MeOA solution was supplied from the liquid feed at the center of the column by using a quantitative pump. The

suspended solution of blue-green algae was fed from the top of the column. The feed volumetric flow rate was $20 \text{ cm}^3/\text{min}$. To keep the liquid level within the column constant, the volume of the solution within the column as much as MeOA solution and the suspension was drained from the drain tap at the bottom of the column. The drained liquid was sampled at desired time and the concentration of blue-green algae was measured by turbidimetry at 700 nm.

3.2.3 Determination of adsorption parameters for MeOA

Two adsorption parameters in Eq (9), the adsorption equilibrium constant, K_b , and the saturated surface density, N_b , for MeOA onto bubble surface were determined by continuous foam separation experiment. The experimental setup for foam separation and the procedure was almost the same as our previous study [23].

4. Results and Discussion

4.1. Effectiveness of adding MeOA

Fig. 3 shows a typical cell concentration profile within the column. The open triangle and the open circle correspond to the data obtained with or without feeding MeOA, respectively. The solid line is calculated value from the following equation.

$$C_s = \frac{w_{si}C_{si}}{w_b} \left\{ 1 - \exp\left(-\frac{w_{si}C_{si}}{V}t\right) \right\} \quad (21)$$

Eq. (21) can be obtained by solving Eq. (4) with the initial condition: $C_s = C_{si}$ at $t = 0$. As seen in Fig. 3, the concentration profile without MeOA (open circle) followed the expected value from Eq. (21). This result indicates that the mixing condition in the present system is regarded as a complete mixing and supports the assumption of neglecting w_f in Eq. (3). On the other hand, the concentration profile with MeOA (open triangle) is rather lower than that without MeOA. This result indicates that blue-green algae cells were removed from the column by adding MeOA.

4.2. Influence of mass flow rate of cells and MeOA and superficial gas velocity rate on cell removal efficiency

Fig. 4 shows the removal efficiency, η , as functions of the ratio of mass flow rate of MeOA and blue-green algae as a typical result. The superficial gas velocity did not affect significantly the removal efficiency. On the other hand, the ratio, $(C_{pi}W_{pi})/(C_{si}W_{si})$, of the both mass flow rate affected the removal efficiency up to ca. 0.4. In the range of $(C_{pi}W_{pi})/(C_{si}W_{si}) > 0.4$, the value of η was mostly constant, although some scatters are

observed. This indicates that the dosage of MeOA should be enough to remove the cells. Influence of $C_{pi}W_{pi}$ on η is shown in Fig. 5. In case of this figure, the $C_{si}W_{si}$ value is constant (ca. 4.0×10^{-3} g/min). As seen in Fig. 5, the value of $C_{pi}W_{pi}$ affected strongly on η . This reason can be explained by the functions of MeOA molecule as flother and flocculant.

4.3. Verification of the proposed model

The adsorption parameters, K_b and N_b , for MeOA adsorbed onto bubble surface were determined by a continuous foam separation method [23]. The adsorption equilibrium relationship could be expressed by Langmuir adsorption isotherm. The Langmuir plot is shown in Fig. 6. X in the coordinate of figure 6 correspond to $W_{f0}(C_{f0} - C_b)/S_b$, in which W_{f0} and C_{f0} represent the volumetric flow rate of the liquid in the foam within the column and the concentration of MeOA in the foam liquid at liquid-foam interface, and C_b is the equilibrium concentration of MeOA within the column, respectively. By measuring the axial distributions of the volumetric flow rate of the liquid in foam and the concentration of MeOA in the foam liquid, W_{f0} and C_{f0} were determined by the extrapolation graphically, respectively [23]. From the values of the intercept and the slope, K_b and N_b were determined as $14.9 \text{ dm}^3/\text{g}$ and $1.26 \times 10^{-2} \text{ g/m}^2$, respectively.

Fig. 7 shows the comparison of the experimental value of η and the calculated value of η . The estimated adsorption parameters are summarized in Table 1. The data shown in Fig. 7 includes the data of Fig. 4, in addition, data for $U_g = 1.42 \times 10^{-2}$ and $1.10 \times 10^{-2} \text{ cm/s}$ are shown in Fig. 7. The experimental conditions for all data in Fig. 7 of $C_{pi}W_{pi}$ and $C_{si}W_{si}$ were varied from 1.8×10^{-4} to $6.42 \times 10^{-3} \text{ g/min}$, and, from 6.31×10^{-4} to $1.22 \times 10^{-2} \text{ g/min}$, respectively. The both agree well within error 19 %. These parameters were estimated by a least squares regression except K_b and N_b . As seen in Fig. 7, the proposed flotation mechanism in this study was valid fundamentally for the removal process of

blue-green algae by using MeOA as frother and flocculant.

5. Conclusions

The flotation experiments for removal of blue-green algae were conducted with the methylated egg ovalbumin (MeOA). The flotation model was proposed, which was considered of the adsorption equilibrium relations of MeOA-bubble surface, MeOA-cells surface and cells-bubble surface. From the experimental results, the mass flow rate of MeOA mainly affected the removal efficiency. The proposed model was verified with the experimental data and agreed well the experimental results within 19 % error.

Acknowledgement

The authors gratefully acknowledge Mr. K. Tanowaki, former students of the department of marine bioresources chemistry, Hokkaido University, for his help in the experiments.

Notation

A	= the cross sectional area of bubble column	$[m^2]$
C_s	= the bulk concentration of blue-green algae within the column	$[kg/m^3]$
C_{si}	= the feed concentration of blue-green algae	$[kg/m^3]$
C_f	= the collapsed foam liquid concentration of blue-green algae	$[kg/m^3]$
C_p	= the equilibrium concentration of MeOA	$[kg/m^3]$
C_{pi}	= the concentration of the feed solution for MeOA	$[kg/m^3]$
ε	= gas holdup	$[-]$
g	= gravitational acceleration	$[m/s^2]$
η	= the removal efficiency for blue-green algae by flotation	$[-]$
K	= the equilibrium adsorption constant of blue-green algae adsorbed on bubble surface	$[m^3/kg]$
K_b	= the equilibrium adsorption constant of MeOA adsorbed on bubble surface	$[m^3/kg]$
K_s	= the equilibrium adsorption constant of MeOA adsorbed on blue-green algae	$[m^3/kg]$
μ_L	= liquid viscosity	$[kg/(m \ s)]$
N	= the number of available site for adsorption of blue-green algae on bubble surface	$[kg/m^2]$
N_b	= the saturated adsorption density of MeOA adsorbed on bubble surface	$[kg/m^2]$
N_m	= the saturated adsorption density of blue-green algae on bubble surface	$[kg/m^2]$

N_s	= the saturated adsorption density of MeOA adsorbed on blue-green algae	[kg/kg-cell]
θ	= the coverage fraction of bubble surface by blue-green algae	[-]
θ_b	= the coverage fraction of MeOA on bubble surface	[-]
θ_s	= the coverage fraction of MeOA on blue-green algae surface	[-]
ρ_L	= liquid density	[kg/m ³]
ρ_G	= gas density	[kg/m ³]
S_b	= the production rate of bubble surface area within the column	[m ² /s]
t	= time	[s]
U_g	= the superficial gas velocity	[m/s]
V	= the bulk liquid volume within the column	[m ³]
w_{si}	= the volumetric flow rate of the feed liquid	[m ³ /s]
w_b	= the volumetric flow rate of the brain liquid	[m ³ /s]
w_f	= the volumetric flow rate of the liquid within foam	[m ³ /s]
w_{pi}	= the volumetric flow rate of the feed MeOA solution	[m ³ /s]
X	= the adsorption density of blue-green algae on bubble surface	[kg/m ²]
X_b	= the adsorption density of MeOA on bubble surface	[kg/m ²]
X_s	= the adsorption density of MeOA adsorbed on blue-green algae	[kg/kg-cell]

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Figures and table captions

Fig. 1. Size distribution of blue-green algae cells used in the flotation experiments.

Fig. 2. Schematic diagram of experimental setup for blue-green algae removal: (1) bubble column, (2) gas distributor, (3) pressure tap, (4) flow meter, (5) needle valve, (6) air compressor, (7) liquid feed pump, (8) pressure/voltage transducer (9) amplifier, (10) volt meter, (11) liquid flow meter.

Fig. 3. Typical time course of cell concentration profiles within the column with MeOA (triangle) and without MeOA (circle), respectively. The superficial gas velocity is 5.37×10^{-2} cm/s.

Fig. 4. Effect of the ratio of the mass flow rate, $C_{pi}W_{pi}$, of MeOA and the mass flow rate, $C_{si}W_{si}$, of blue-green algae cell on the flotation efficiency, η .

Fig. 5. Effect of the mass flow rate, $C_{pi}W_{pi}$, of MeOA on the flotation efficiency, η . The superficial gas velocity for each symbols are same as Fig. 4.

Fig. 6. Langmuir plot of MeOA for determination of the adsorption parameter, K_b and N_b .

Fig. 7. Comparison of the values of the experimental η and that of the calculated η .

Table 1 Adsorption parameters for MeOA and blue-green algae

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K [dm ³ /g]	N_m [g/m ²]	K_s [dm ³ /g]	N_s [g/m ²]	K_b [dm ³ /g]	N_b [g/m ²]
1.15	1.63	1.08	1.08	14.9	1.26×10 ⁻²