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An effective process of harvesting *Chlorella* sp. biomass for bioresource by rapid flocculation in a helical tube

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A B S T R A C T

An effective flocculation technique was developed for harvesting microalgal biomass, i.e., *Chlorella* 64.01, by using a home-made helical tube. The flow field characteristics in the tubes were studied by a computational fluid dynamic method. About 90% of cell recovery was achieved using 0.12 g/L diluted algal solution with an 18-s flocculation and 15 min sedimentation. The harvesting efficiency was significantly affected by the flow velocity. For a helical tube with 8 mm diameter, the optimal velocity was found to be 0.83 m/s to flocculate the microalgae. The helix diameter may have an impact on the flocculation due to the secondary flow intensity, and the 30-cm diameter was found to be a suitable choice. The residence time also plays a role in the recovery process as a competing effect between agglomeration and dissociation. Flocculation in a helical tube was proved to be a rapid and an effective approach for harvesting microalgae.

Keywords: Helical tube; Flocculation; Harvesting; Microalgae; Fractal dimension; Velocity gradient

1. Introduction

Microalgae are widely recognized as a promising biofuel source material considering their competitive advantages such as a short growth period and a high content of lipid [1,2]. Separation of microalgae from bulk water is a necessity for follow-up utilization, which remains to be a challenge due to their small size (~2 μm) and similar density as water.

Several commonly used techniques, including flotation [3], gravity sedimentation [4], membrane filtration [5,6], centrifugation [7], magnetic separation [8], and flocculation [9], have been adopted for microalgae harvesting. Flotation represents one promising method for the separation of microalgae *C. vulgaris* from freshwater [10], illustrating the advantages of high biomass recovery, short operation time, no requirement for arable land, and good flexibility [11]. However, an unfavorable factor using flotation method is a relatively high energy consumption. Gravity sedimentation is a facile and economic method for solid–liquid separation, which is widely used to separate microalgae from water. However, sedimentation is usually time-consuming and not suitable for large-scale operation [4]. Membrane filtration can provide high-quality algal biomass without the addition of chemical coagulants [12,13], but usually has the fouling problems [6,14]. Centrifugation is a rapid but expensive harvesting method [7] (approximately 1 MJ/kg energy for algal biomass [15]) and thus not suitable for large-scale separation [1]. Magnetic separation, an emerging technology, has been widely used for solid–liquid

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separation in water treatment. Despite its rapid speed in separation, problems such as postharvest separation, recycling of nanoparticles, and practical applicability, remain unsolved [16]. Overall, flocculation is an ideal candidate for dewatering [17]. Previous flocculation research has been performed in batch or semi-batch mode reach high recovery of up to 98% with suitable flocculants and hydrodynamics [9]. Zhang et al. [4] carried out a series of flocculation experiments and recovered 95.61% of *C. vulgaris* using combined coagulants ($FeCl_3$ and polyacrylamide (PAM)) and a gradually reduced shear (50–30 rpm). Huang et al. [18] synthesized a novel flocculant (Gemini surfactant), which was more effective than cetyltrimethylammonium bromide (CTAB) in the same hydrodynamic conditions.

Flocculation efficiency is determined by agents and hydrodynamics according to flocculation kinetics [19]. When flocculants, including chemical agents, biological flocculants and microorganisms, cause the microalgae cells to attach [20,21], a hydraulic field improves the collision frequency of cells. The hydrodynamics in flocculation reactors are always characterized by a velocity gradient (G) or the turbulent dissipation rate (ε), and they need to be controlled well to enhance collision as well as to avoid breaking the flocs due to excessive turbulence [19]. In addition, residence time (T) also affects the flocculation result [22], and it needs to be adjusted for both technological and economic reasons.

Among several flocculation units with low retention time, helix tube flocculation reactors have been gaining prominence in scientific research, due to their compact size, low cost, and high efficiency [23]. In a forerunner to the present study, Carissimi and Rubio [24] found that the coagulation effect can also be achieved by using pipelines or pipeline connection devices. When using helically coiled tubes to promote flocculation, the direction of flow will change smoothly throughout the unit [25]. The generation of flocs was a dynamic balance of aggregation and detachment, which was influenced by flocculation time and shear intensity [26]. Helical tubes can provide high particle collision efficiency due to the secondary flow induced by centrifugal forces [27]. In recent years, Zhang et al. [28] carried out a series of experiments using a helix tube flocculation reactor following an ejector to harvest microalgae and got a high recovery efficiency (94%).

In the present work, helical tubes were used to flocculate *Chlorella* sp. 64.01. The influence of hydraulic characteristics (G and ε) and residence time on the flocculation behavior of microalgae was investigated in terms of obtaining better process and equipment parameters and improving the efficiency of microalgae harvesting.

2. Materials and methods

2.1. Microalgae cultivation

Chlorella sp. 64.01 from the Freshwater Algae Culture Collection at the Institute of Hydrobiology was grown to a stationary phase in BG-11 medium in an 80 L photobioreactor at 25°C. The illumination intensity was 67.5 μmol/(m^2·s) and the cells were aerated at 16 L/min without pH adjustment. After 5 d, *Chlorella* sp. reached the stationary phase, with a dry cell concentration of 0.12 g/L.

2.2. Flocculation experimental setup

The experimental equipment consisted of a T-cock, pump, ejector, helical tube reactor, and sedimentation tank, in which the T-cock and ejector were used for coagulant and flocculant addition, respectively. Further details are shown in Fig. 1.

2.3. Flocculation experiments

The pH of the microalgae broth was adjusted to 6.5 using HCl and NaOH to get a wider optimum zeta potential band (+3.45 to +8.71 mV) [29]. $FeCl_3$ and cationic PAM (molecular weight 10,000,000; charge density 40%) were used as the coagulant and the flocculant, respectively, and their dosages were set at 40 and 5 mg/L according to the classic jar test. $FeCl_3$ and PAM were prepared in 1 g/L solutions. Flow rates of feedstocks and agents were controlled by adjusting the spindle speed of peristaltic pumps. The flocculated suspension was settled in a tank for 15 min and the supernatant concentration was detected by a WGZ-1A turbidimeter (Shanghai Xinrui Instrument Co., Ltd., China). Cell recovery (R, harvesting efficiency) was calculated using Eq. (1):

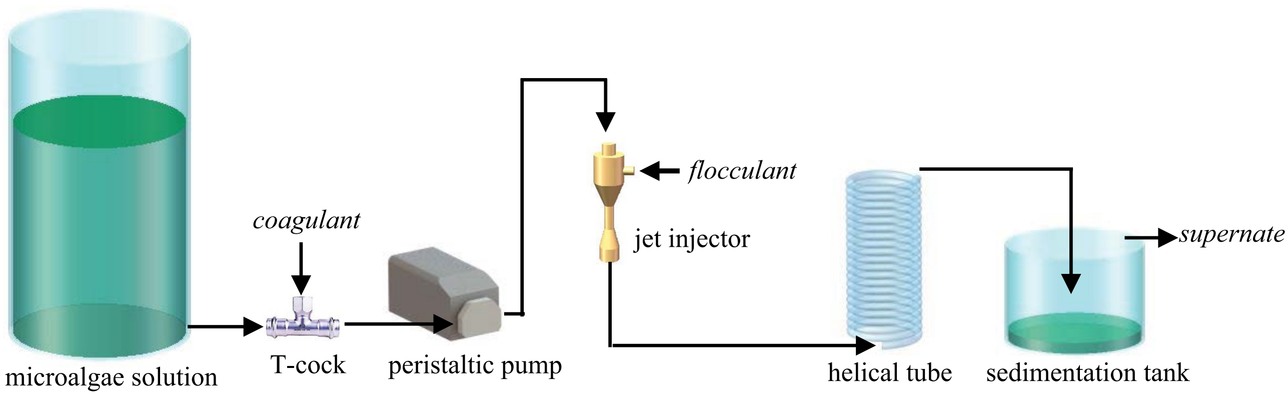

Fig. 1. Flocculation setup using a helical tube.

$$R = \left(1 - \frac{C_s}{C_r}\right) \times 100\% \quad (1)$$

where C_r is the cell concentration of raw microalgal broth and C_s is the supernatant concentration. Experiments were repeated 3 times to estimate the recovery. Besides, the suspension at the outlet of the helical tube was collected and observed using an SVA-7045A stereomicroscope (Cossim Instrument Co., Ltd., Beijing, China) to acquire the areas (A) and perimeters (P) of the flocs. Then, the 2D fractal dimension D_{2f} of the floc can be calculated by the fitted line of A versus P:

$$\ln A = D_{2f} \ln P + C \quad (2)$$

where C is the intercept of the line.

The helical tubes used in this work were 8 mm in diameter. To determine the effect of hydrodynamics on flocculation, the helix diameter (D, shown in Fig. 2) was varied from 10 to 40 cm, and the flow rate was changed from 50 to 200 L/h. In addition, the tube length was altered from 10 to 20 m to investigate the influence of residence time.

2.4. Flow field simulation

To determine the hydraulic characteristics in helical tubes, the computational fluid dynamics method was used to simulate the motion of water without cells. As a stable-state flow in a uniform diameter tube, one helix circle was modeled instead of the whole flocculation tube for saving the calculation source. The three-dimensional structure grids with maximum cell skewness below 0.8 were prepared by the software package ICEM. The software package Ansys Fluent was employed to solve the simulation. The parameters used in the simulation are listed in Table 1.

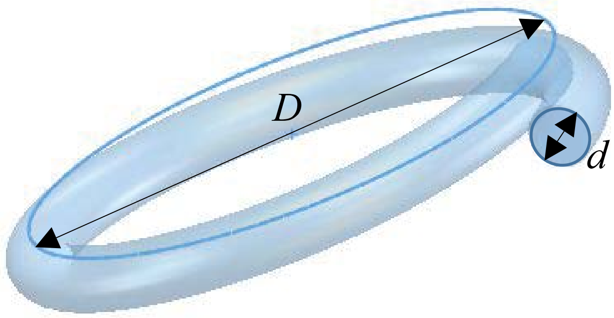

Fig. 2. Helical tube parameters helix diameter (D) and tube diameter (d).

The turbulent dissipation rate (ε) can be given by the simulation results; then, the velocity gradient (G) is calculated using the classical Eq. (3) [30]:

$$G = \sqrt{\frac{\varepsilon}{\upsilon}} \quad (3)$$

where υ is the kinematic viscosity, 1.01×10^{-6} m^2/s for water at 20°C.

3. Results and discussion

Fig. 3 shows the results of flocculation. The cell recovery varied from 80.5% to 90.5% under different conditions, while the flocs fractal dimension differed over the range of 1.67–1.83.

3.1. Effect of flow velocity

When the tube diameter was fixed at 8 mm, different flow rates mean different flow velocities, which determine the velocity gradient to a certain extent. Fig. 3b shows the influence of flow velocity on the flocculation recovery and the fractal dimension of the flocs. When the flow velocity changed from 0.28 to 1.11 m/s, the cell recovery rose to the highest value, 90.5% at 0.83 m/s. This means that the optimal velocity gradient was approximately 700 s^{-1} (shown in Fig. 4). This result is quite different from Zhang's research, with the best G of approximately 75 s^{-1} [31]. This difference is probably due to the differences in cell concentration (0.12 g/L in the present work and 1.5 g/L in Zhang's). Because the rate of flocculation is a function of collision efficiency (α), collision frequency (β), and the particle concentration (n_0) as the expression [32]

$$\text{Rate of flocculation} = \alpha\beta n_0^2 \quad (4)$$

According to the Eq. (4), when the particle concentration is low, it is necessary to increase the G to improve the collision efficiency, so as to improve the flocculation efficiency. On the other hand, it must be noted that α is also related to G. The adhesion efficiency highly depends on the competition between the attraction of particles and the external destructive force such as shear force. Thus, a flow velocity

Table 1
Parameters used in the simulation

Parameters	Values
Turbulence model	RNG κ-ε model was proposed by Yakhot and Orzag. RNG in this model is the abbreviation of "renormalization group".
Wall	Adiabatic no-slip wall, using enhanced wall function method for near wall region
Inlet	Velocity, their values were given as same as the experiments; and turbulent kinetic energies were calculated by 0.16 $Re^{-1/8}$, where Re is Reynolds number
Outlet	Pressure, 0.1 MPa

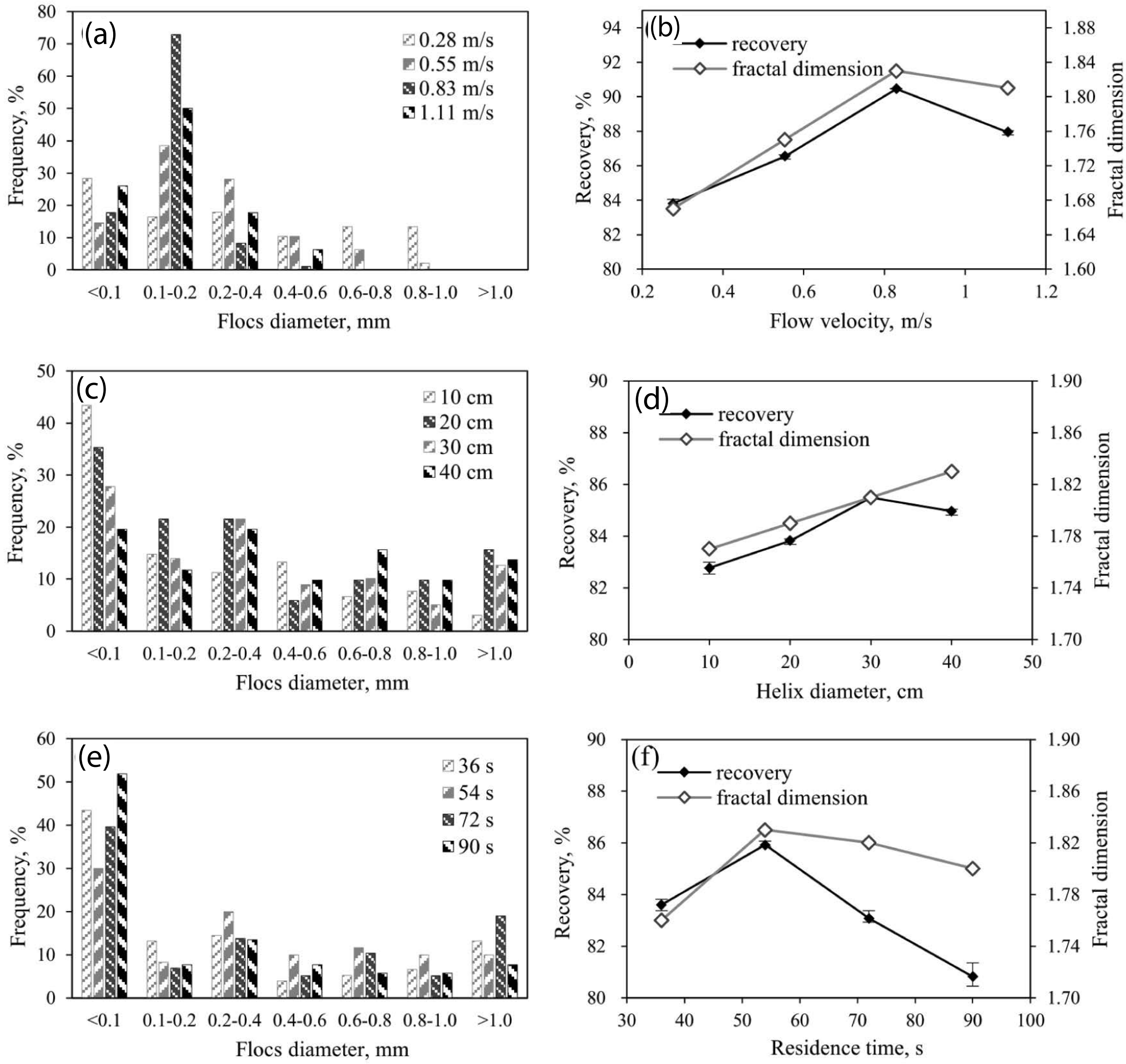

Fig. 3. Effects of (a, b) mean flow velocity, (c, d) helix diameter, and (e, f) residence time on flocs diameter distribution, cell recovery, and the fractal dimensions of the flocs. All the tube diameters are 8 mm. The helix diameter, flow rate, and tube length are fixed at 10 cm, 50 L/h (means a flow velocity of 0.28 m/s), and 15 m, respectively, if they are not an investigated factor.

greater than 0.83 m/s seemed to have negative effect on the flocculation. As shown in Fig. 3a, the flow velocity of 0.83 m/s provided a more uniform flocs size at 0.1–0.2 mm, and the greater flow velocity of 1.11 m/s caused the flocs to break into smaller flocs. Because the lower velocity gradient provides lower collision frequency, a flow velocity smaller than 0.83 m/s also had poor performance, and the size distribution of the flocs tended to be uneven. Thus, flocs at a lower flow velocity have smaller fractal dimension, causing lower floc density and lower settle velocity.

3.2. Effect of helix diameter

As shown in Fig. 3d, cell recovery increased from 82.8% to 85.5% when the helix diameter enlarged from 10 to 30 cm, and increasing the helix diameter further tended to bring a slightly lower recovery. This means that an optimal secondary flow intensity exists for the balance of aggregation and breaking, although helix diameter change resulted in little variation in velocity gradient (Fig. 4). The flocs fractal dimension increased slightly with the enlarging helix diameter. The flocs diameter distribution seemed to be similar, except for the decreased amount of flocs smaller than 0.1 mm, as shown in Fig. 3c.

Helix diameter may affect flocculation via the secondary flow intensity. Fig. 5 shows the flow patterns of the cross-section of helical tubes with different helix diameter. Secondary flows were all observed in the four conditions, with a little variation in velocity magnitude. But the velocity near the center of the vortex (named Dean vortex)

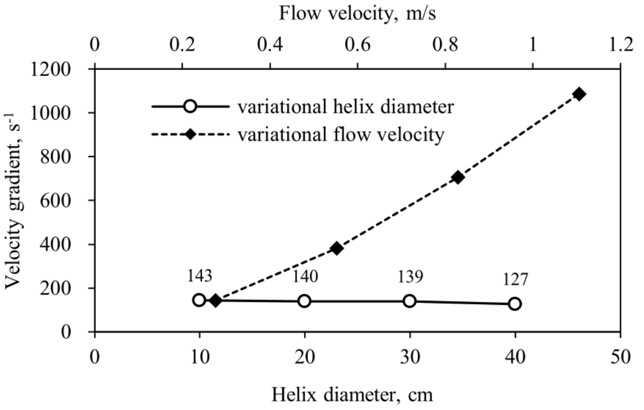

Fig. 4. Effect of flow velocity and helix diameter on velocity gradient.

tended to increase as the diminishing helix diameter, indicating an increasing velocity gradient and energy dissipation. The vortex in the helical tube with a helix diameter of 10 cm (Fig. 4) was almost compressed into a flat shape. That excessive shear may be the reason for bringing a mass of fine flocs. Hence, the helix diameter needs to be controlled to provide a suitable secondary flow.

3.3. Effect of residence time

Tube length determined the residence time when the flow rate was fixed at 50 L/h. Fig. 3f shows that the recovery varied from 80.8% to 85.9% when residence time differed from 36 to 90 s. Both the highest recovery and fractal dimension were seen at 54 s. As shown in Fig. 3e, flocs smaller than 0.1 mm increased for a residence time greater than 54 s, which indicates that the flocs are broken into small fragments due to the excess residence time instead of excessive shear. This is because the fracture of the floc is largely irreversible. Once the flocs break up, it is difficult to re-grow them up to their original size [33]. Comparing the flocculation time from 5 to 30 min [4,21], 54 s in the present work was a very short time. In addition, when the recovery was 90.5% at the flow velocity of 0.83 m/s, as shown in Fig. 3b, the residence time was shortened to 18 s, indicating a more rapid flocculation process.

4. Conclusion

Flocculation in helical tubes was carried out to harvest *Chlorella*. The cell recovery of an algal solution with a dry biomass concentration of 0.12 g/L could reach 90.5% under the conditions of tube diameter, flow velocity, helix diameter, and residence time of 8 mm, 0.83 m/s, 10 cm, and 18 s, respectively. Flow velocity and residence time influenced the harvesting efficiency greatly due to the balance between collision-aggregation and breaking. The helix diameter had no obvious effect in the investigated range. This study also indicated the significance of operational optimization, and the results can provide technical support and theoretical guidance for large-scale microalgae harvesting in the future.

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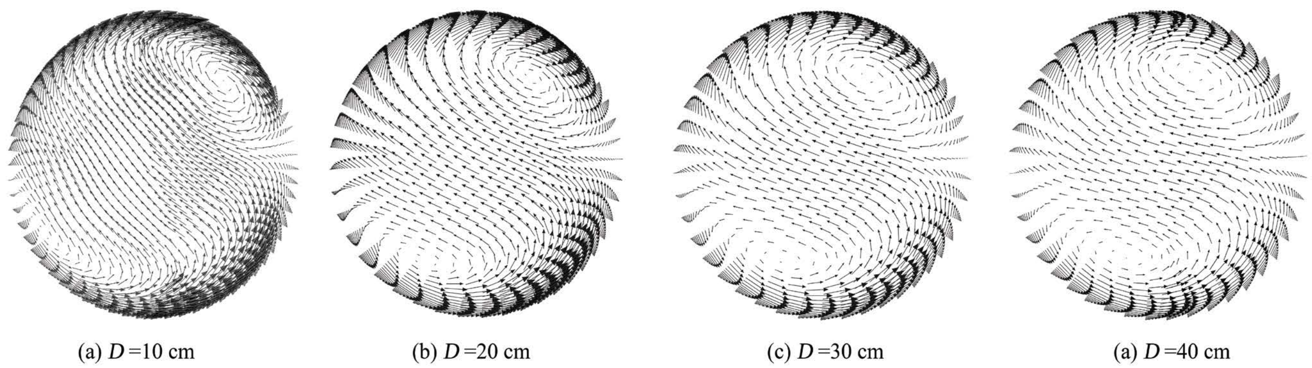

Fig. 5. The velocity vectors on the cross-section of helical tubes with different helix diameters (*D*).

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