Bioresource Technology 267 (2018) 341–346

Contents lists available at ScienceDirect

Bioresource Technology

journal homepage: www.elsevier.com/locate/biortech

Buoy-bead flotation harvesting of the microalgae *Chlorella vulgaris* using surface-layered polymeric microspheres: A novel approach

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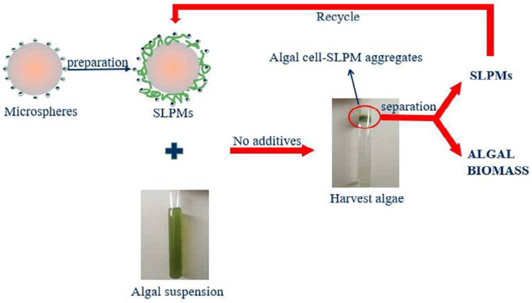
GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:
Microalgae harvesting
Buoy-bead flotation
Surface-layered polymeric microspheres (SLPMs)
Harvesting efficiency

ABSTRACT

To improve microalgae harvesting efficiency and to reduce the addition of chemicals in the buoy-bead flotation process, a novel buoy-bead flotation approach has been developed for harvesting *Chlorella vulgaris*, using surface-layered polymeric microspheres (SLPMs). Next, the detachment of microalgae cell-SLPM aggregates and the reusability of SLPMs were investigated. The experimental results showed that a maximum harvesting efficiency of 98.43% was achieved at a SLPM dosage of 0.7 g/L and a pH of 9, and harvesting efficiency quickly decreased with increasing ionic strength. A detachment efficiency of 78.46% and a concentration factor of 19.56 were achieved at an ionic strength of 700 mM and a mixing speed of 3000 rpm without changing the pH. Reused SLPMs can still reach an efficiency of 72.13% after five cycles. The presented results show that this method can potentially be applied for large-scale microalgae harvesting.

1. Introduction

Microalgae are single-cell organisms with a wide range of uses, such as for water treatment (Sukačová et al., 2015), health supplements and animal feed (Bakar et al., 2015), and as a sustainable feedstock for biofuels (Yen et al., 2013). They can be grown in both seawater and wastewater (Hamid et al., 2014), thereby reducing land usage (Zhu et al., 2013). The estimated biofuel yield from microalgae is 10–300 times higher than that from oleaginous crops (Liu et al., 2012) and compared to terrestrial crops, microalgae achieve higher growth rates and photosynthesis rates (Quinn and Davis, 2015). Despite these advantages, due to their small size (density similar to that of water and low biomass concentrations), microalgae harvesting is relatively difficult, and the use of microalgal biofuels is therefore challenging from an economic point of view (Zhu et al., 2016). The estimated cost of energy for microalgae biomass harvesting is 20.00–30.00% of the total product

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https://doi.org/10.1016/j.biortech.2018.07.065
Received 30 May 2018; Received in revised form 11 July 2018; Accepted 12 July 2018

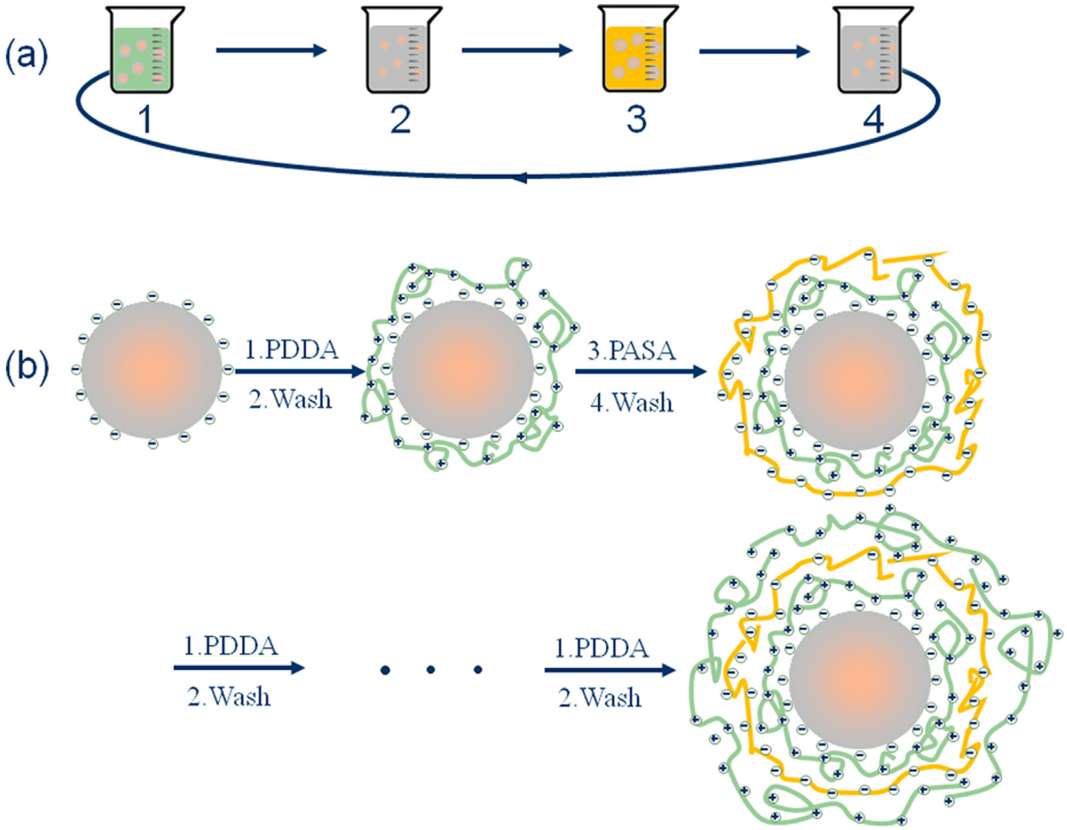

Fig. 1. (a) Schematic illustration of the multilayer film adsorption process. Steps 1 and 3 represent the adsorption of a polycation and polyanion, respectively, and Steps 2 and 4 are washing steps. (b) Simplified molecular drawing of adsorption steps, depicting film adsorption, starting with a negatively charged substrate and ending with the positively charged PDDA. Counterions are omitted for clarity. The polyion conformation and layer interpenetration are an idealization of the surface charge reversal with each adsorption step.

cost (Mata et al., 2010), making it necessary to develop a cost-effective method for microalgae harvesting.

Centrifugation (Chen et al., 2015), filtration (Nurra et al., 2014), flocculation (Lananan et al., 2016; Ummalyma et al., 2017), sedimentation (Depraetere et al., 2015), and flotation (Kandasamy and Shaleh, 2018) are common harvesting systems applied for microalgae separation. However, high energy costs and low efficiency limit their large-scale commercial application (Zhu et al., 2017). In addition to these traditional harvesting methods, ballasted flotation via low-density ballasted agents has recently emerged and is considered a time-saving and energy-efficient method (Jarvis et al., 2009). The underlying mechanism of this process is the attachment of algal cells onto ballasted agents. Once these microalgae-ballasted agent flocs have been harvested, the ballasted agents can be separated from the algal biomass and recycled. Compared to traditional dissolved air flotation, Ometto et al. (2014), who employed the ballasted agents at 300 mg/L with chemical coagulant doses of 5 mg/L, reported that ballasted dissolved air flotation can achieve a removal efficiency of > 90.00%, while significantly saving energy. In another study, Zou et al. (2018) achieved a harvesting efficiency of 92.47% within 3 min at pH 7 when low-density microspheres of 550 mg/L were applied to harvest *Chlorella vulgaris* (*C. vulgaris*) via chitosan. Although there are several studies on this topic, this technology is still in the early stage and needs further development.

Clearly, buoy-bead flotation is a promising method due to its low operating costs, low energy requirements, and high harvesting efficiency. However, the process typically requires the addition of chemicals to promote the attachment of algal cells onto microspheres (Ometto et al., 2014; Zou et al., 2018). Microsphere surfaces and algal cells are typically both negatively charged, resulting in low harvesting efficiency without the addition of chemicals (Xu et al., 2018) such as ferric chloride, aluminum sulfate, and chitosan (Ometto et al., 2014). However, the addition of these chemicals results in biomass contamination, while also contributing to the operating costs (Laamanen et al., 2016). In addition, the buoy-bead flotation process can only harvest microalgae cell-microsphere aggregates, which need to be further separated to obtain the final microalgae biomass and to recycle the microspheres. However, to our knowledge, only few studies have investigated this separation process.

In this study, to improve the harvesting efficiency as well as to reduce the addition of chemicals, positively charged surface-layered polymeric microspheres (SLPMs) were prepared. A buoy-bead flotation process was investigated for the harvesting of *Chlorella vulgaris*, using low-cost SLPMs without any chemical additives. The effects of operation parameters (SLPM dosage, pH, and ionic strength) on the harvesting efficiency of buoy-bead flotation were also studied. In addition, the effect of mixing speed on the detachment efficiency of the aggregates and the reusability of the SLPMs after aggregate detachment were evaluated.

2. Materials and methods

2.1. Preparation of surface-layered polymeric microspheres (SLPMs)

In this study, the low-cost hollow microspheres (Shanghai, Mingbo New Material Technology CO., Ltd. China) were composed of sodium borosilicate glass with a diameter of 50–60 μm. Prior to use, the microspheres were cleaned, degreased by ethanol, and carefully washed with deionized water, followed by drying at 25 °C. Multilayer films of organic compounds on solid surfaces have been extensively studied because they allow the production of multicomposite molecular