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Review article

Harvesting microalgae by magnetic separation: A review

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ABSTRACT

Magnetic separation has been utilized for the removal of microalgae for nearly forty years. Due to its advantages compared to traditional harvesting methods, magnetophoretic harvesting of microalgal cells has received much attention in recent years. In this context, synthesized magnetic particles for microalgae harvesting are summarized in this review. In addition, the particle–cell interaction and factors influencing the separation process are discussed as well as the feasibility of its scale-up applications using a magnetic separator. Furthermore, the downstream techniques including the extraction of desired products and the reuse of the culture medium and magnetic particles are also assessed. Finally, the current challenges are outlined and future directions to achieve efficient and economic magnetic harvesting of microalgae are discussed.

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Contents

1. Introduction

Over the past few decades, microalgae have received intensive basic research and applied research to sustain biofuel production [1–3]. Microalgae are a potential feedstock for the production of transportation fuels due to several advantages, such as the ability to be cultivated on barren land, a high growth rate, and higher lipid content compared to other feedstocks [4,5]. In addition, microalgae have the potential to accumulate high-value substances such as omega-3 fatty acids, vitamins, and antibiotics, as well as antioxidants for food supplements, animal feed, and pharmaceuticals [6–8]. However, the commercialization

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of microalgae-based products is still limited, especially for biofuel production. This is mainly due to technological and economic limitations in the production process [9–11]. The key stages in the production process are cultivation, biomass harvesting, extraction of the desired components, and production of the desired substances [12,13]. The harvesting step, in particular, is determined to be energy intensive and the sustainability of harvesting techniques is sometimes limited by the energy requirement [14]. In addition, the cost of harvesting step is usually high and contributes to 20–30% of the total cost of the process [15–17]. Harvesting microalgae at low cost and with a positive energy balance is significant for the production of algal biofuels [14].

Overviews of microalgae harvesting methods have been included in multiple reviews [18–22]. Algal cells can be harvested by various methods, including centrifugation, sedimentation, flocculation, filtration, flotation, or by a combination of these methods. However, there is no harvesting method that is considered superior, no one that is suited to all algal species. Current harvesting methods have various disadvantages which include high cost, high energy consumption, or the requirement for a time-consuming process [19,22]. Therefore, it is necessary to develop a reliable and cost-effective approach for the industrial-scale production of algal based products.

Magnetic separation is a simple separation process. For the removal of magnetic contaminants, it simply requires a magnetic separator while for the recovery of a desired product, selective magnetic adsorbents are necessary. The separation is achieved based on the intrinsic paramagnetic movement of the magnetic particle tagged products in the response to the magnetic field [23,24]. Due to its advantages, which include simple operation, low energy consumption, and low cost, it has been widely applied in diverse industries [23]. Magnetic separation has been demonstrated to be effective and reliable in applications such as kaolin decolorization, wastewater treatment in steel factories and power plants, enrichment of ores-mineral beneficiation, the removal of specific elements in the food industry, and the removal of arsenic and metals in water treatment [23,25,26]. In addition to industrial applications, magnetic solutions have been used in many biochemical processes, such as protein and DNA purification, drug targeting and delivery, biocatalysis, and diagnostics [27–29]. Furthermore, by tagging non-magnetic target cells with magnetic beads, the target cells can be rapidly isolated from the medium using very gentle conditions [24]. Due to this principle, magnetic separation can be utilized for the harvesting of microalgal cells.

Recently, different types of magnetic particles have been synthesized and have shown potential when utilized for the separation of microalgae. In this review, the research progress of the magnetic separation of microalgae is reviewed, and the challenges and further directions are discussed.

2. Characteristics of algal broth

The harvesting of microalgae faces three major challenges. First is the dilute nature of the algal broth, which is typically less than 0.5 g/L in commercial production systems [18]. Therefore, large volumes of broth need to be handled to recover the algal biomass [18]. Second, algal cells are small; they typically range from 2 μm to 20 μm, which makes the harvesting difficult via some general techniques, such as filtration [30]. Third, these small cells generally have an electronegative surface charge at a wide pH range [19]. The aggregative of algal cells is difficult due to the electrostatic repulsion effect between algal cells and the cells are generally stably suspended in the broth, which further increases the difficulty in harvesting [31]. In addition, the variety in size, shape, and motility among different algal species makes it difficult to develop a single technique that is suitable for the recovery of all species [19].

The magnetic separation process is based on the interaction between the algal cells and particles. Therefore, the surface characteristics of algal cells are important for the magnetic recovery efficiency (RE). It has been reported in multiple studies that the majority of algal species have a negative surface charge over a wide pH range [32–36]. This is mainly due to the functional groups present in the proteins, lipids, and sugars on the surface of algal cells [37]. Fourier transform infrared (FTIR) spectra analysis indicated that there are abundant –COOH and –OH groups on the surface of *Chlorella* sp. [38,39]. Furthermore, atomic force microscopy (AFM) analysis revealed that the surfaces of algal cells are not smooth, rather they contain groove-like indentations, and striated and sphere-like mound structures; in *Chlorella ellipsoidea*, these mound heights ranged from −60 to 60 nm [38,40].

3. Magnetic particles for the separation of microalgae

Magnetic particles have been used for algal separation for almost forty years. They were initially used for the removal of harmful algae from lakes [41,42]. Due to their advantages and potential, in recent years, magnetic particles have received much attention and stimulated research efforts for microalgae separation. Various types of magnetic particles have been synthesized and studied for the recovery of algal cells.

3.1. Naked magnetic particles

It is well-known that naked magnetite is effective for the removal of microalgal biomass [41]. Recently, these naked magnetic particles have been synthesized using various methods and successfully applied for the harvesting of both freshwater and marine microalgae. For example, Fe_3O_4 particles synthesized by chemical co-precipitation with an average diameter of approximately 10 nm and an isoelectric point of approximately 7 (suspended in deionized water), were efficient in harvesting the freshwater algal species *Botryococcus braunii* and *C. ellipsoidea*, and the marine species *Nannochloropsis maritima* [34,43]. In addition, a new agent with a broad size range of 0.15–20 μm and an isoelectric point at pH 6.2 was prepared using ferrous sulfate as a precursor using assisted microwave treatment. High RE was achieved using naked iron oxide magnetic microparticles (IOMMs). The synthetic process was much simpler and cheaper than more commonly used approaches [33]. It has been confirmed that naked magnetite has ion exchange characteristics and the separation is primarily based on the electrostatic interactions between the magnetite and the algal cells [33,34,43]. In addition, Fe ions released from the IOMMs surface may act as flocculating agents and benefit the harvesting process [33]. However, the released ions may increase the metal content in the harvested algal cells and this can influence the downstream refining of algal biomass. For example, the Fe can poison the catalysts for desulfurization and decrease the gasoline yields [44]. Therefore, the choice of magnetic adsorbent should consider the potential influence on the downstream process.

3.2. Surface functionalized magnetic particles

Due to the negative surface charge of algal cells, a positive charge on the surface of the particles improves separation. As previously mentioned, the surface charge of naked magnetite is pH-based with an isoelectric point around neutral conditions [45]. Therefore, functionalizing the surface of the naked particles with cationic groups is an effective method to enhance the RE. The tagging of a polyelectrolyte is commonly achieved using one of two strategies: either the “attached-to” or the “immobilized-on” strategy [46]. The “attached-to” strategy is based on first coating the cells with a polymer binder and then attaching the magnetic particles. In the “immobilized-on” approach, the naked magnetic particles are first surface functionalized with a polyelectrolyte and then bound to the algal cells [39]. However, naked particles usually have poor dispersibility and aggregate into large particle clusters due to magnetostatic and Van der Waals functions. A lower RE was achieved with an equal dosage of particles in the “attached-to” approach compared with that of the

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