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Magnetophoretic removal of microalgae from fishpond water: Feasibility of high gradient and low gradient magnetic separation

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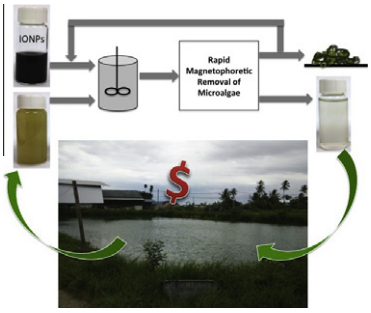
HIGHLIGHTS

- Rapid magnetophoretic separation of mixed strains of microalgae from fishpond water is feasible.
- The concentration of surface functionalized IONPs affect the microalgae removal efficiency.
- LGMS and HGMS systems achieve high separation efficiency.
- Kinetic behavior of both LGMS and HGMS systems are compared.
- LGMS system more cost effective for small scale fishfarm water treatment.

GRAPHICAL ABSTRACT



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ABSTRACT

Microalgae blooms in commercial fish production ponds resulting in a deficit in the overall oxygen budget have posed serious challenges to aquaculture industry. In this study, we demonstrate the feasibility of rapid microalgae separation in real-time from fishpond water by magnetophoresis. By relying on the magneto-shape anisotropy of rod-liked iron oxide magnetic nanoparticle (IONPs), overall separation efficiency of microalgae cells up to 90% can be achieved in less than 3 min. The IONPs employed, with a saturation magnetization at 113.8 emu/g, are surface functionalized with cationic polyelectrolyte that promotes the attachment of these particles onto microalgae cells via electrostatic interaction. Kinetic of magnetophoretic separation process was monitored by suspension opacity measurements based upon a custom built light dependent resistor (LDR setup) sensor. Whereas, the overall separation efficiency of microalgae cells is determined spectrophotometrically at 685 nm wavelength. Performance of both high gradient magnetic separation (HGMS) with $\nabla B > 1000$ T/m and low gradient magnetic separation (LGMS) with $\nabla B < 80$ T/m were tested with varying particle concentration (50–500 mg/l) and the results obtained were interpreted in term of cooperative magnetophoresis theory. Cost analysis was conducted to verify the feasibility for large scale implementation of LGMS system with the cost involved at $0.13 for every one meter cube of treated fishpond water.

1. Introduction

Aquaculture and fish farming is one of the solutions for the worldwide decline of fisheries stocks of either marine or freshwater fish and also to fulfill the demand of world's growing population [1]. One major problem that plagues the freshwater fish

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farm is microalgae blooms that occur in every fishpond during summer or tropical country, such as Malaysia. Microalgae will naturally grow in fishpond water because of the presence of nutrients, such as nitrogen, phosphorus, carbon source [2,3], which originated from the fish excretion, excess fish food and decaying organic matter. Most of those nutrients promote the growth of microalgae are in organic matter form and they can be quantified as Chemical Oxygen Demand (COD) with an ideal value for fishpond at less than 50 mg/l [4]. Ironically, the growth of the microalgae naturally is beneficial for the removal of the excess nutrient in the water to avoid nutrient overloading as well as reducing the COD level. However, this benefit is diminished once the microalgae start to grow excessively. For a typical freshwater fishpond, the microalgae will keep growing as long as there are nutrient to substantiate its growth. High microalgae concentration beneficial as oxygen source through photosynthesis [5] and also provides shades for fishes from the sunlight. However, the high concentration of microalgae will be disastrous, as their huge amount will exhaust the oxygen supply through respiration and releasing carbon dioxide during nighttime. Fish may be killed overnight through suffocation [6] when dissolved oxygen (DO) is less than 2 mg/l [7]. In most cases, DO in fish pond should be maintained at least 4 mg/l all the time [4]. At extreme high nutrient level eutrophication will occur [7]. The nutrient will promote excessive growth of algae bloom in the presence of sunlight. The sunlight will be blocked totally by the dense algae bloom to form a death zone beneath the bloom with drastic oxygen depletion and hence deteriorate the fish production.

Conventionally, there are multiple well-practiced methods to maintain the microalgae population within a small fishpond [8]. Effective management of microalgae growth can be achieved naturally via few methods, such as, growing aquatic plants around the fishpond to consume the nutrient and starving the microalgae [9,10], avoid over feeding and using high quality food to ensure complete digestion of the food [11], and using barley straw to control algae growth in pond [12]. Besides the natural treatment, by using the algaecides chemical, which contain simazine, chelated copper and potassium permanganate, is also able to kill the algae but it is harmful to living organisms and environment [13]. The quick death of all the microalgae may increase ammonium concentration and decrease dissolve oxygen in water and hence it is not favorable. Nevertheless, most of the standard practices involved for microalgae removal were labor intensive and had limited efficiency [14]. Since microalgae biomass can be employed as third generation biofuel [15] and other useful products, like nutrients in form of polyunsaturated fatty acid (PUFA) [16–18] or pigment [19,20] with a robust removal technique without direct annihilation of microalgae might be economically more attractive.

There are several microalgae separation methods which have been developed to meet high microalgae separation efficiency. The most common microalgae separation methods are filtration, centrifugation, flocculation and settling and ion exchange [21,22]. Flocculation and settling is a versatile method, which is suitable to process large quantity of biomass, but it is time consuming [23]. Filtration method has recorded high separation efficiency, however, this method is quite costly with the problem of blocking and fouling [21,22]. Centrifugation is the most reliable method but it is expensive and consumed large amount of energy during operation and is not suitable to handle enormous quantity of effluents [24]. In addition to all the separation techniques discussed so far, magnetic separation of microalgae from water resources is a relatively new concept which was introduced in 1970s [25,24]. This method was recently revisited by us [26] and others [27,28] due to its attractive advantages such as high permeation fluxes, high removal efficiency, small land area utilization and no clogging and fouling problems [29]. Moreover, magnetic separation process can also be perform directly on raw samples that contain suspended solid material due to its ease in capturing the targeted samples by using surface functionalized magnetic particles [30].

In order to achieve magnetic separation of biological cell, tagging the cell surface with a paramagnetic dipole moment is necessary since most cells are irresponsive to applied magnetic force [31]. Microalgae cells membrane surface are negatively charged because of the present of lipids, proteins and sugars, which have functional groups like –SH, –OH and –COOH. Deprotonation of those ligands will give a net negative charge on cell surface at natural pH of water [22,32–34]. While for the magnetite, it is negative by charge when disperse in deionized water [35], with isoelectric point between 6.30 and 6.85 [36–38]. Under this scenario, we need a binder to immobilize the magnetic nanoparticles onto the microalgae cells. The binder that is normally employed to serve this purpose is a positively charged polyelectrolyte, where it can be adsorbed on the nanoparticle surface [26] through direct method or link to the negative charged cell surface indirectly through [30] electrostatic interaction [35]. After tagging the microalgae cells with magnetic nanoparticles, cells can be separated magnetophoretically through either low gradient magnetic separation (LGMS), without magnetized matrix or high gradient magnetic separation (HGMS) [30], which contains magnetized matrix [39].

In this work, we illustrated the engineering feasibility of LGMS and HGMS in harvesting microalgae from fishpond water by direct method, which is tagging surface functionalized iron oxide nanoparticle (IONPs) onto the microalgae cells surface. Furthermore, we compared the separation efficiency between LGMS and HGMS at various IONPs concentration corresponding to different kinetic behaviors. Optical light intensity sensing system (LDR setup) is employed to measure the overall separation efficiency and quantify its kinetic, while the UV–Vis spectrometer is conducted to measure the specific cell separation efficiency. Cost analysis on HGMS system for microalgae separation from fishpond water is conducted to provide a guideline for different system setup and design preferences.

2. Experimental methods

2.1. Materials

Rod-liked iron oxide magnetic nanoparticle (IONPs) were obtained from Toda America, Inc. The 35 wt.% very low molecular weight poly(diallyldimethylammonium chloride) (PDDA) in water with molecular weight, M_w < 100,000 g/mol was obtained from Sigma–Aldrich, Inc. Deionized water used was obtained by reverse osmosis and further treated by the Milli-Q Plus system (Millipore) to 18 MΩcm resistivity.

2.2. Characteristic of microalgae in fishpond water

Fishpond water sample was collected from fish farm of Aik Lee Fishery, which is located at Sungai Bakau, Parit Buntar, Perak, Malaysia. The samples were brought back to the laboratory for analysis. Microalgae cells species were observed under Olympus CX41RF microscope equipped with Image Pro Express 4.0.1 imaging software. Chemical Oxygen demand (COD) of the samples was measured spectrophotometrically by the DR 5000™ UV–Vis Spectrophotometer with the use of High Range Plus COD Reagent from HACH Company, USA. The pH of our fishpond water was measured by using Eutech CyberScan pH 1500.

2.3. Nanoparticles attachment to microalgae

In this work, rod-like IONPs was used with physical dimension of ~20 nm in diameter and 300 nm in length respectively [35]. The