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Improvement of metal adsorption onto chitosan/*Sargassum* sp. composite sorbent by an innovative ion-imprint technology

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ABSTRACT

Technology for immobilization of biomass has attracted a great interest due to the high sorption capacity of biomass for sequestration of toxic metals from industrial effluents. However, the currently practiced immobilization methods normally reduce the metal sorption capacities. In this study, an innovative ion-imprint technology was developed to overcome the drawback. Copper ion was first imprinted onto the functional groups of chitosan that formed a pellet-typed sorbent through the granulation with *Sargassum* sp.; the imprinted copper ion was chemically detached from the sorbent, leading to the formation of a novel copper ion-imprinted chitosan/*Sargassum* sp. (CICS) composite adsorbent. The copper sorption on CICS was found to be highly pH-dependent and the maximum uptake capacity was achieved at pH 4.7–5.5. The adsorption isotherm study showed the maximum sorption capacity of CICS of 1.08 mmol/g, much higher than the non-imprinted chitosan/*Sargassum* sp. sorbent (NICS) (0.49 mmol/g). The used sorbent was reusable after being regenerated through desorption. The FTIR and XPS studies revealed that the greater sorption of heavy metal was attributed to the large number of primary amine groups available on the surfaces of the ion-imprinted chitosan and the abundant carboxyl groups on *Sargassum* sp.. Finally, an intraparticle surface diffusion controlled model well described the sorption history of the sorbents.

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1. Introduction

Biosorption has been considered as a promising technology for the removal of low-level toxic metals from industrial effluents and natural waters (Volesky, 2007; Mehta and Gaur, 2005; Wang and Chen, 2009). Marine algae have received greater attention because of their high metal biosorption capacity, low cost, and renewable nature. They can effectively remove heavy metal ions with concentrations ranging from few ppm to several hundreds ppm. The maximum metal biosorption capacity ranging from 0.1 to 1.5 mmol/g biosorbent has been reported (Davis et al., 2003; Chen and Yang, 2005, 2006). However, biosorbents often have small size, weak mechanical strength and low density. Leaching of such

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organics as carbohydrates and protein may be serious when some of biosorbents (e.g. raw seaweed) are used. These drawbacks have hindered the applications for treatment of waste metallic streams from various sources.

In recent years, various efforts have been focused on the immobilization of alga biomass, which is a potential way to overcome the disadvantages. Such supporting materials as alginate (Bayramoğlu and Yakup Arıca, 2009; Mata et al., 2009), silica gel (Rangsayatorn et al., 2004), polyacrylamide and polyurethanes (Mehta and Gaur, 2005) were used for the biomass immobilization. However, metal uptake efficiency of immobilized cells is reportedly often much lower than that of raw biomass. The functional groups on biomass for metal binding may become less available due to the immobilization and thus the sorption becomes normally less. For example, the cadmium sorption capacity drops from 98 mg-Cd/g for the *Spirulina platensis* to 71 and 37 mg-Cd/g for the alginate and silica immobilized species, respectively (Rangsayatorn et al., 2004). Mata et al. (2009) reported that the sorption capacity of alginate immobilized *Fucus vesiculosus* for copper was reduced nearly 60% due to immobilization. Synthetic polymers, such as polyacrylamide and polyurethanes, may be used for immobilization of biomass; the high cost and toxicity restrict their applications (Mehta and Gaur, 2005).

Chitosan produced by partial deacetylation of chitin is the second most abundant biopolymer next to cellulose in nature. In addition to the high adsorption capacity for various heavy metals (Bassi et al., 2000; Guibal, 2004), chitosan can easily form hydrogel (Zhao et al., 2007). Thus, chitosan may be a good carrier for immobilization of biomass. However, the chitosan-based hydrogel are often poorly resistant to acid and has weak mechanical strength. In order to overcome the weaknesses, a cross-linking approach has commonly been used; the disadvantage is that the sorption capacity becomes reduced after cross-linking (Hsien and Rorrer, 1997; Ruiz et al., 2000).

In this study, an innovative ion-imprint technology illustrated in Fig. 1 was developed and used for the immobilization of alga biomass with the objectives of achievement of the higher sorption capacity of biomass and prevention of leaching of organics. Metal ion is used as an imprint ion to first "occupy" some of the functional groups (adsorption sites) in immobilization agent that can be used for metal adsorption shown as Step (1). The immobilization agent and biomass are solidified into a pellet shown as Step (2). Finally, elution agents (e.g., EDTA and NaOH) are used to strip the "pre-occupied" metal ions from the functional groups (elution of imprinted metal ions) so that they become available for metal sorption illustrated as Step (3).

The chitosan was selected as an immobilization agent for *Sargassum* sp., which has higher metal uptake capacity and is abundant in many parts of the world (Chen and Yang, 2005, 2006; Mehta and Gaur, 2005). More importantly, it originates from natural living organisms and is not toxicity to human beings. A copper imprinted chitosan/*Sargassum* (CICS) composite sorbent was prepared according to the abovementioned ion-imprint technology. The adsorption properties such as pH effect and adsorption isotherm were studied. The surface characteristics of sorbent and sorption mechanisms were elucidated through Fourier transform infrared spectroscopy (FTIR) and X-ray photoelectron spectroscopy (XPS) analysis.

2. Materials and methods

2.1. Materials

The raw biomass of *Sargassum* sp. was collected from the coast of Singapore. The biomass was first washed with deionized (DI) water, and then dried overnight in an oven at 60 °C. The dried seaweed was ground to fine particles with size below 150 μm. Chitosan (90% deacetylation), acetic acid and copper sulfate were purchased from Sinopharm Chemical Reagent Company (China). Sodium pyrophosphate was provided by

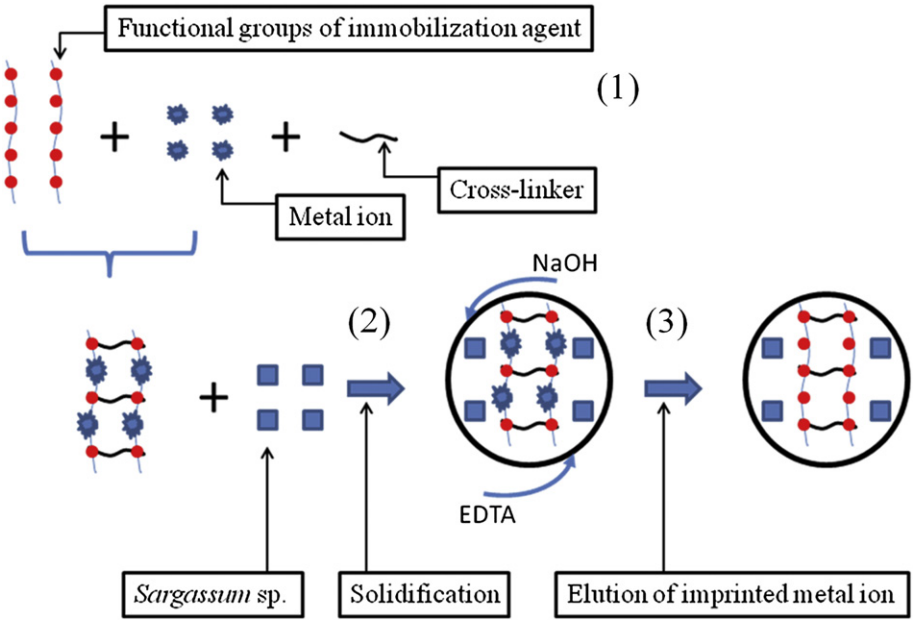

Fig. 1 – Demonstration of fabrication of metal imprinted composite sorbents.

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