Journal of the Taiwan Institute of Chemical Engineers 45 (2014) 2610–2614

Contents lists available at ScienceDirect

Journal of the Taiwan Institute of Chemical Engineers

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

Short communication

Fungi aerobic granules and use of Fe(III)-treated granules for biosorption of antimony(V)

Chunli Wan [a], Li Wang [a], Duu-Jong Lee [a,b,c,*], Qinlan Zhang [a], Jieni Li [a], Xiang Liu [a]

[a] Department of Environmental Science and Engineering, Fudan University, Shanghai 200433, China
[b] Department of Chemical Engineering, National Taiwan University of Science and Technology, Taipei 106, Taiwan
[c] Department of Chemical Engineering, National Taiwan University, Taipei 106, Taiwan

ARTICLE INFO

Article history:
Received 9 March 2014
Received in revised form 24 June 2014
Accepted 24 June 2014
Available online 24 July 2014

Keywords:
Adsorption
Antimony
Granules
Modification

ABSTRACT

This study cultivated fungi aerobic granules in sequencing batch reactor with glucose as carbon source at pH 4.5, which were mainly composed of single fungus *Geotrichum fragrans*. Iron precipitate was then produced with 0.1 M $FeCl_3$ onto the granules surface, accompanied with removal of Ca, Mg, K and Na from the biomass matrix. The Fe(III)-treated granules had 4.3-folds higher adsorption capacity of antimony(V) than the original granules, peaked at pH 3.4 at a Langmuir maximum capacity of 111 mg/g of dry mass. Fe(III) treatment was for the first time applied to convert fungi aerobic granules to cost-effective biosorbent for Sb(V).

1. Introduction

Aerobic granules are an emergent biotechnology for treatments of wastewater containing high-strength wastewaters containing organics, nitrogen, phosphorus and toxic substances [1–7]. During long-term operation, aerobic granules frequently encountered in filamentous overgrowth which converted the bacterial granules into filamentous granules [8]. Filamentous aerobic granules are loose in structure and low in density, so their overgrowth can lead to easy biomass washout from the reactor [9]. Efforts were made to prevent filamentous overgrowth in aerobic granular process.

Antimony (Sb) is a metalloid widely applied in flame retardants, metal alloy, plastics, semiconductors and therapeutic agents against protozoan disease and is often released in an uncontrolled manner into the environments [10]. Biosorption is a low-cost, highly efficient and environmental friendly remediation process for heavy metal-laden wastewaters [11,12]. The peer-reviewed studies noted that biomass performed notable adsorption capacity on multifarious metals, such as Cd(II) [13,14], Cr(III) and Pb(II) [15–17]. Untreated and treated freshwater cyanobacteria *Microcystis* biomass were used to adsorb Sb(III) and Sb(V) from waters [18].

Methods were proposed to modify the surfaces of adsorbent for promoting their performance of adsorptive removal of target heavy metals [19]. Iron doping onto biomass is proposed effective to promote its adsorption capacity on Cr(VI) and As(V) [20–22]. Applying Fe(III)-treatment to enhance adsorption capability of aerobic granules on heavy metals removal is rarely reported. Wang et al. [23] modified the surfaces of bacterial granules by applying Fe(III) and showed that the Fe-treatment significantly increased the adsorption quantities of Sb(V) from waters.

Conversely, owing to the loose structure of the filamentous granules and the endless growth in size for entangled mycelia in granules, this biomass may have high potential as biosorbents for hazardous substances from waters. This work cultivated filamentous aerobic granules from glucose medium at acidic environment. The microbial analysis revealed that the cultivated granules were composed of a few fungi. The cultivated granules were modified using Fe(III) treatment with 0.1 M $FeCl_3$ for 24 h. The treated granules were used for adsorption tests of Sb(V). The modified granules were characterized and their adsorption behaviors were experimentally revealed.

2. Materials and methods

2.1. Granule cultivation

Four identical SBRs were installed (G1–G3, G5). The reactors were five columns (180 cm × 5 cm) with a working volume of 2.0 l.

* Corresponding author at: Department of Chemical Engineering, National Taiwan University of Science and Technology, Taipei 106, Taiwan.
Tel.: +886 2 33663028; fax: +886 2 23623040.
E-mail address: djleetw@yahoo.com.tw (D.-J. Lee).

http://dx.doi.org/10.1016/j.jtice.2014.06.019
1876-1070/

The seed sludge was at suspended solids (SS) of 3000 ± 200 mg/l. The synthetic influent contained NH_4Cl (0.2 g/l), KH_2PO_4 (0.2 g/l), $NaHCO_3$ (0.013 g/l), $MgSO_4{\cdot}7H_2O$ (0.025 g/l), $CaCl_2$ (0.03 g/l), $FeSO_4{\cdot}5H_2O$ (0.02 g/l), and peptone (0.04 g/l). The SBRs were operated at 4-h cycles with air pumped to the diffuser at column bottom at a volumetric flow rate of 5 l min^{-1}. Each cycle consisted of 3-min of filling, 227-min of aeration and settling, 5-min of effluent decanting, 5-min idling. All the chemicals were purchased from Sinopharm Chemical Reagent Co., Ltd (Shanghai, China).

Fungal community was analyzed by 18S rRNA gene-based clone libraries. Genemic DNA (50 μl) was extracted by Fungi Genomic DNA Isolation Kit (Sangon Biotech Inc., Shanghai, China). PCR primers NS7 and NS8 were used to amplify the variable V8–V9 region fungal 18S rRNA gene. The PCR mixture of 50 μl contained: 5 μl of 10 × LA Taq buffer, 8 μl of 2.5 mmol dNTP; 1.5 μl of each primer (20 μmol/l), 2.5 U of LA Taq, 1 ng of fungal DNA template. The PCR amplification was conducted as follows: 94 °C for 10 min; 30 cycles of 94 °C for 1 min, 55 °C for 1 min, 72 °C for 90 s; final extension at 72 °C for 10 min. The PCR products and DNA extract were confirmed by 1.0% agarose gel in TAE buffer stained with ethidium bromide. The primers were also purchased from Takara Co., Ltd as follow: NS7: 5′-ATAACAGGTCTGTGATGC-3′, NS8: 5′-CGCAGGTTCACCTACGGA-3′. The non-labeled PCR products were purified using agarose gel electrophoresis. The purified PCR products were ligated to vector pMD-18T, and then transformed to competent cells DH5α. Finally, the positive colonies were randomly picked for sequence. The operational taxonomic unit (OUT) analysis was taken by Sequencher 4.2. The sequences were subjected to BLAST analysis against the database of NCBI (KF861033–KF861039). All the PCR chemicals were purchased from Takara Co., Ltd (Dalian, China), while the PCR primers were synthesized by Sangon Co., Ltd (Shanghai, China).

2.2. Fe(III) treatment

The Fe(III) treatment was conducted by introducing 25 g (wet basis) granules to 250 ml, 0.1 M $FeCl_3$ solution for 24 h at 35 °C and pH 2.0. The suspension was continuously shaken on a commercial shaker. After 24 h, the granules were collected and repeatedly washed using ddH_2O until no residual Fe^{3+} ions were detected in the supernatant. The treated granules were stored in tap water at 4 °C.

2.3. Adsorption studies

Standard stock solution of Sb(V) (1000 mg/l) was prepared by dissolving $KSb(OH)_6$ into the deionized water. Solutions at different concentrations were prepared by diluting the standard stock solution at prescribed proportions. The pH of solution were adjusted by adding 0.1 M NaOH.

One gram of untreated granules or Fe(III)-treated granules (wet basis) were mixed with 50 ml of 20, 60 or 100 mg/l Sb solution in 100-ml conical flasks. The pH of suspensions adjusted by adding 0.1 M HCl or 0.1 M NaOH, with the flasks shaken at 175 rpm and 35 °C. The supernatant was sampled at fixed time intervals for measurement. Preliminary tests revealed that the adsorption loss to flasks or sampling were negligible. The adsorption quantity, q_t (mg/g) and the removal rate, r (%), were calculated as follows: $q_t = (C_0 - C_e)v/m$ and $r = (C_0 - C_e)/C_0$, where C_0 and C_e is the initial concentration and equilibrium concentration of antimony (mg/l), v is the volume of solution (l) and m (g) is the dry weight of untreated or treated granules.

2.4. Other analyses

The metals of samples and the concentration of antimony in the solution were analyzed by dissolution and inductively coupled plasma atomic emission spectroscope (ICP-AES, Hitachi 4010, Japan). The granules were firstly freeze-dried under −60 °C and then 0.1 g of granule sample was put into a cuvette loading with 3 ml of HNO_3. The mixture was heated to 150 °C for 1 h and 300 °C for 10 min using an electric heating plate. Then, the mixture was placed in a muffle furnace with 600 °C for 2 h. After that, 10 ml of ddH_2O was added in cuvette, and the mixture was centrifuged at 10,000 × g for 10 min. 8 ml of supernatant and solution samples were analyzed using Perkin Elmer inductively coupled plasma optical emission spectrometry (ICP-OES) (PE OPTIMA7000DV, Perkin Elmer, Waltham, MA, USA).

3. Results and discussion

3.1. Cultivated granules and Fe treatment.

The cultivated granules had white appearance and irregular surfaces (Fig. 1A). The granule size ranged 1–10 mm in 5 d cultivation. The fugal communities of granules were analyzed by 18S rDNA clone libraries. Six filamentous fungi dominated granules were randomly picked from reactors and each granular sample was analyzed by clone libraries. As shown in Fig. 2, majority of granules had only one fungus, *Geotrichum fragrans*, and at most three kinds of fungi composing the granule. Independent bacterial analyses revealed no bacterial strains existed in these granules. Restated, this study cultivated pure fungi granules.

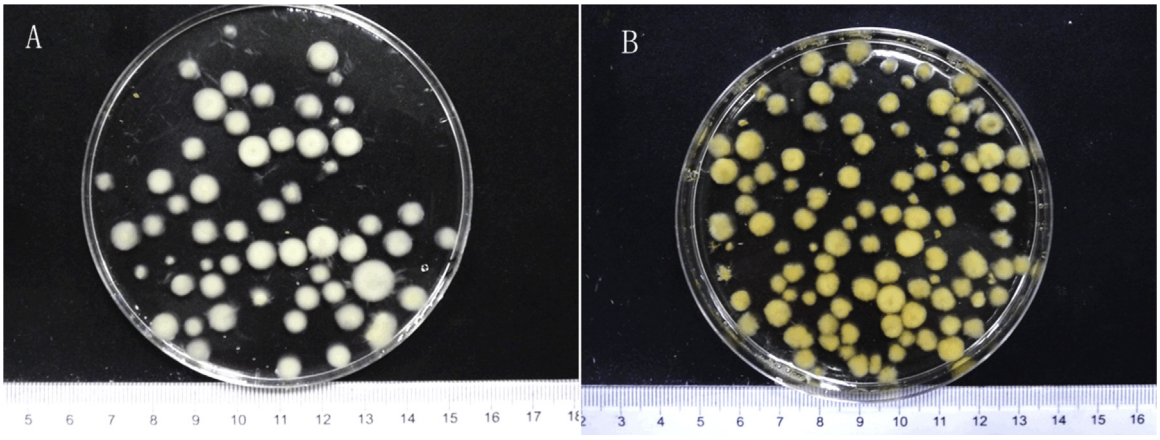

Fig. 1. Morphology of granules. (A) Original granules; (B) Fe-dosed granules. (For interpretation of the references to color near the citation of this figure, the reader is referred to the web version of the article.)