



Can biofilm affect alum sludge adsorption: An engineering scope in a novel biofilm reactor for wastewater treatment



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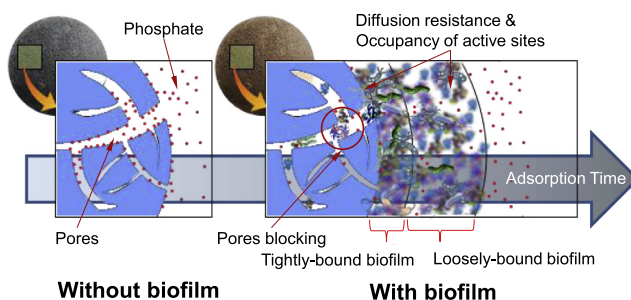
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HIGHLIGHTS

- The activated sludge posed severe deterioration to the alum sludge's adsorption.
- Suspended sludge hindered the intra-particle diffusion by blocking the pores.
- Biofilm occupied the active sites and brought extra diffusion resistance.
- The activated sludge in the GBR is expected to reduce the adsorption rate by 35–40%.
- Loosely-bound biofilm is not welcome and regular change of alum sludge is in demand.

GRAPHICAL ABSTRACT



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ABSTRACT

Dewatered alum sludge, a kind of water treatment residual, has been intensively studied as an effective adsorbent/substrate for phosphorus (P) removal from wastewater in constructed wetland (CW). Recently, a “Green Bio-sorption Reactor (GBR)” has been developed by introducing the alum sludge-based CW into conventional activated sludge system. Accordingly, some concerns about the P-adsorption rate of alum sludge appeared due to the different conditions in CW and in activated sludge system. In the present study, the investigations were conducted to explore the impact of activated sludge and natural organic matter (NOM) on P-adsorption behavior by alum sludge for providing technical support for the GBR. The results revealed that the activated sludge posed an adverse effect on the alum sludge's adsorption rate. The interference of activated sludge was mainly derived from pores blocking (by suspended activated sludge and its secretions), occupancy of the surface-active sites (via tightly-bound biofilm) and diffusion resistance increase (via loosely-bound biofilm). The overall P adsorption reduction in the GBR is expected to be 35–40%. More significantly, the loosely-bound biofilm should be removed while the tightly-bound biofilm holds some benefit for nitrogen removal. Contradicted by previous studies, NOM in the present study showed minor deterioration to the adsorption of alum sludge.

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1. Introduction

The anthropogenic activities, industrialization and the rough plan of water utilization have posed unprecedented issues for

wastewater treatment. One of them is the emerging contaminants, such as the endocrine disruptors, pharmaceutical and personal care products [1,2], which are highly toxic, recalcitrant and persistent in a long term period. Accordingly, this severe situation promoted the development of novel processes to replace or supplement the conventional biological treatment processes [3]. So far, various novel technologies including advanced oxidation

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processes, membrane reactor and its variants, ultrafiltration processes etc. have been proposed, studied and employed in practice. However, the disappointing matter is associated with the energy-intensive and high-technique requirement in both design and operation of these technologies [4].

By the contrast consideration, adsorption is a high-efficient and well-established process. For a long time, the exploration of low-cost adsorbents from by-products or waste materials of various industries and application of them into wastewater treatment have attracted international attention [5–8]. Alum sludge, a kind of dewatered drinking water treatment residual, has been successfully applied in constructed wetland (alum sludge-based constructed wetland, AICW) as matrix and concurrently adsorbing phosphorus (P) along with probably other pollutants [9–11]. The predicted P adsorption capacity of alum sludge is robust and higher than most of other typical low-cost adsorbents (Table S1, Supplementary Information (SI)). Application of alum sludge in the lab- and pilot-scale CW demonstrated its suitability as a carrier for biofilm development and, particularly, as an adsorbent for the satisfied performance of P immobilization [10].

Most recently, the AICW has been successfully integrated into conventional activated sludge system to form a Green Biosorption Reactor (GBR) to enhance the nutrients removal [12,13]. Comparatively, the scenario of alum sludge in GBR (i.e. the AICW being embedded into the aeration tank of the activated sludge process) is different from it as a substrate in CW. Specifically, the alum sludge in the GBR is surrounded by the suspended activated sludge (SAS) in the aeration tank. In a lab-scale GBR study [13], a considerable deterioration of P removal was observed as a consequence that a thick layer of activated sludge wrapped the alum sludge (Fig. 1). The literature survey showed that similar situation was reported. Arias et al. [14] stated that the adsorption capacity of calcite in the full-scale application in biological wastewater treatment was tenfold lower than estimated in the laboratory and the formation of biofilms on the calcite could probably be the culprit. Moreover, Widjaja et al. [15] ever reported the adsorption capacity of activated carbon decreased after adding it into aeration tank of biological wastewater treatment.

Despite the SAS flowing freely around the alum sludge particles, there are two types of biofilms (Fig. 1), the loosely-bound biofilm and the tightly-bound biofilm. The former refers to the outer and thick (in millimeter) layer that can be easily flushed away from the surface of the alum sludge while the later implies the inner (in micrometer) and tight adhesion layer. Therefore, the present study was initiated to explore the insight into the possible

influencing factors on alum sludge's adsorption behavior in GBR from an engineering scope. Specifically, the impacts of SAS, biofilm and natural organic matter (NOM) on alum sludge's P-adsorption in GBR were experimentally studied and discussed. It is expected that the current study can provide some insight into the actual P-adsorption capacity of alum sludge in a GBR.

2. Materials and methods

2.1. Alum sludge in this study

Dewatered alum sludge cakes were collected from Ballymore Eustace Water Treatment Plant, Co. Dublin, Ireland, with the aluminum mass content of $29.7 \pm 13.3\%$ [16] and other chemical components reported in Yang et al. [17]. The collected large chunk of alum sludge cakes was then broken into around $2 \times 2 \times 2$ cm particles (termed as virgin alum sludge) (Fig. S1-a, SI), and rinsed for several times with deionized water to remove the loosely-attached scraps before used in the experiments.

2.2. Biofilm cultivation

2.2.1. Tightly-bound biofilm

Initially, the tightly-bound biofilm cultivation on the surface of the alum sludge was conducted in a polyethylene column with diameter and height of 12 cm and 70 cm, respectively. A 5 cm-depth gravel was layered at the bottom as support layer followed by 65 cm-height prepared alum sludge on the top, which gave a liquid volume of 3.3 L. An air diffuser was buried in the support layer to constantly supply oxygen for the growth of the microorganisms. 15.4 L/d of the artificial solution, consisted of 200 mg COD/L (sodium acetate), 40 mg $\text{NH}_4^+\text{-N/L}$ (ammonium chloride), 0.05 mg P/L (monopotassium phosphate, [18]) and necessary trace elements [19], was continuously introduced into the column from the bottom with a peristaltic pump and the effluent overflowed from the top. The column was operated for 30 days while the biofilm was observed as depicted in Fig. S1-b (SI). The alum sludge with tightly-bound biofilm in the column was ready for further test.

2.2.2. Loosely-bound biofilm

The loosely-bound biofilm was cultivated by submerging four cages of alum sludge (162 ± 3 g, Fig. S1-c, SI) into a lab-scale aeration tank of a simulated GBR [13]. After three days, a layer of loosely-bound biofilm in 0.5 cm-thickness was formed on the alum

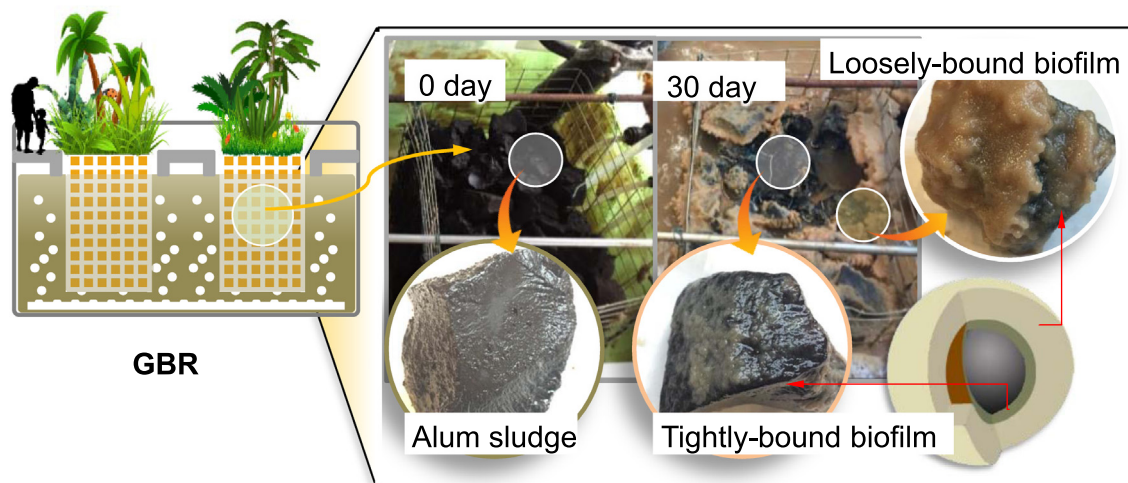


Fig. 1. Observation and schematic model of alum sludge covered by biofilm in AICW of an experimental GBR.

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