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Recovery of phosphorus via harvesting phosphorus-accumulating granular sludge in sequencing batch airlift reactor

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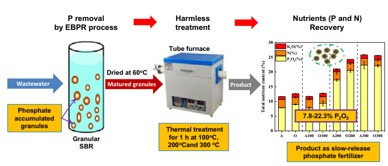
HIGHLIGHTS

- The matured phosphate-accumulating granules cultivated in SBAR system.
- An efficient method was proposed to recover P via thermal treatment of granule.
- The total P in thermal treated sludge is over 50% of total nutrients (N, P and K).
- P release rate was negatively correlated with thermal treatment temperature.

GRAPHICAL ABSTRACT



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ABSTRACT

A novel approach was developed for phosphorus recovery from wastewater through thermal treatment of matured phosphorus-accumulating granular sludge cultivated in sequencing batch airlift reactor (SBAR) system. Results showed that SBAR system had stable performances, in which COD, total phosphorus (TP) and total nitrogen (TN) removal efficiencies were stabilized at 80%, 89% and 86%, respectively. The matured granules were gathered from SBAR reactor and heated at relatively low temperature (100 °C, 200 °C, 300 °C). The total P content in thermal treated granular sludge was more than half of total nutrient. Furthermore, the phosphorus release rate for treated granules was negatively correlated with thermal treatment temperature. These results demonstrated that the granules harvested from SBAR system followed with thermal pre-treatment could probably be applied as excellent slow-release phosphorus fertilizer. Hence, low temperature treatment of phosphate-accumulating granules is efficient for phosphorus recovery from wastewater, which is likely to promote the application of granulation technology.

1. Introduction

Phosphorus (P) is essential to living organisms because of its vital role in numerous key molecules, including DNA, RNA and ATP (Elser, 2012). Yearly, about 22 megaton of P was extracted from mined phosphate ores (Reijnders, 2014) and approximate 90% of global phosphate demand is mostly for the production of agricultural fertilizers (Dawson and Hilton, 2011). However, phosphate ore is a finite, non-renewable resource existing only at the earth's crust (Liu et al., 2008). It has been anticipated that the world's reserves will last only for around 125 years if the demand for the phosphate fertilizers continues increasing at the current rate of 2.5–3% per year (Cordell and White, 2011). In contrast, eutrophication caused by the discharge of excess P from wastew-

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ater and agricultural runoff into aquatic systems is a global environmental problem (Zou and Wang, 2016). Hence, the P concentration for protected waters in North America and Europe consents limited at 10 μg/L and 50 μg/L, respectively (Keeley et al., 2016). Moreover, increasingly stringent effluent standard for wastewater treatment plants (WWTPs) are implemented by many national governments around the world. In China, the P concentration in the effluent of WWTPs is required to meet 0.5 mg/L according to the grade I-A of the "Discharge Standard of Pollutants for Municipal Wastewater Treatment Plant" (GB 18918-2002) (Zhang et al., 2016). Thus, the removal and recovery of P from wastewater would not only prevent eutrophication but also be utilized as a critical strategy to recycle P resource for fertilizer production.

Enhanced biological phosphorus removal (EBPR) processes are widely applied for the removal of phosphorus from wastewater via enrichment of polyphosphate-accumulating organisms (PAOs) at alternating anaerobic and aerobic/anoxic conditions (Mino et al., 1998; Oehmen et al., 2007). From the last decade, aerobic granular sludge was successfully cultivated in sequencing batch airlift reactor (SBAR) and is believed to be a promising technology in wastewater treatment (Beun et al., 2002; de Kreuk et al., 2007; Liu and Tay, 2004). Aerobic granules are self-immobilized aggregates of microorganisms and can be utilized to simultaneously remove organic matter, nitrogen and phosphorus when the biosystem functioned as alternative anaerobic and aerobic conditions (Bassin et al., 2012; de Kreuk et al., 2005). In addition, some studies proved that aerobic granules used in the EBPR system tend to form inorganic phosphate precipitates such as $Ca_7Mg_2P_6O_{24}$ (Zhou et al., 2013) and ($Ca_5(PO_4)_3(OH)$) (Mañas et al., 2011) as an internal core. In fact, Huang et al. (2015) and Lang et al. (2015) demonstrated that the P content of aerobic granular sludge was ranged between 5 and 9% (m/m), which was even higher than that (*ca.* 3–7%, m/m) of activated sludge cultivated in EBPR system (Baetens, 2001). Therefore, phosphorus recovery through harvesting excess granules in EBPR system is of tremendous potential. Moreover, compared with conventional activated sludge, the lower growth yield (ca. 0.2 $gVSS/gCOD_{removed}$) of aerobic granular sludge (also named phosphate-accumulating granular sludge) can minimize the detrimental environment effects when using methods such as incineration and landfill to treat excess sludge (Liu et al., 2005). Furthermore, excess granules containing high P can be possibly applied as a precious source for the production of phosphorus fertilizer.

To date, thermal treatment (e.g., gasification, pyrolysis, hydrothermal liquefaction, and hydrothermal carbonization) of activated sludge has attracted growing attention on an increasingly international scale by removing hazardous components, reducing sludge volume, producing energy and generating valuable products (Bridle and Pritchard, 2004; Huang and Tang, 2016). It is well known that thermal pre-treatment of excess sludge ranged from 50 °C to 70 °C could significantly enhance sludge hydrolysis rate and methane production during anaerobic digestion (Ge et al., 2010). Moreover, Val del Río et al. (2011) tested the effect of thermal pre-treatment (ranged from 20 °C to 210 °C) on anaerobic digestion for aerobic granular sludge. However, there are few reports on P recovery of granular sludge as phosphorus fertilizer when using thermal treatment as an alternative technology, although excessive phosphorus should be enriched in granules.

In order to develop a sustainable and valuable approach to treat excess granular sludge, this study was aimed: 1) to cultivate phosphate-accumulating granular sludge in SBAR system; 2) to deal with excess granular sludge with thermal treatment at different temperature; 3) to investigate the physic-chemical properties of thermal treated granular sludge.

2. Materials and methods

2.1. Reactor set-up

A laboratory scale sequencing batch airlift reactor (SBAR) was used for this study. The SBAR was designed for a working volume of 4.5 L with internal diameter of 10 cm and total height of 60 cm. The effluent was withdrawn from the middle of the reactor which ensures a volumetric exchange ratio of 50%. The wastewater was fed into the reactor from the bottom of the reactor. The SBAR was operated automatically under anaerobic and aerobic conditions alternatively with 6 h per cycle, consisting of 1 min feeding, 60 min anaerobic phase, 284 min aerobic phase, 10 min settling and 5 min discharging. Nitrogen gas and air were supplied into the reactor by fine bubble diffuser for anaerobic and aerobic phases. Gas flowmeters and electric valves were used to control the gas flowrate and thus the superficial air velocities (SAV) in anaerobic and aerobic phase were controlled at 1.2 and 2.5 cm/s, respectively. The seed sludge was crushed aerobic granules produced by a lab-scale aerobic granular sludge reactor operating with a mixed liquor suspended solid (MLSS) concentration of 3.5 g/L.

2.2. Synthetic wastewater

The reactor was fed with a synthetic wastewater having a mixed organic carbon source: sodium propionate and sodium acetate, each of them contributed 50% of the total COD (800 mg/L). The other components of the synthetic wastewater (except for trace element solution) were (mg/L): NH_4Cl (60), KH_2PO_4 (20), $K_2HPO_4 \cdot 3H_2O$ (35), $NaHCO_3$ (50), $CaCl_2$ (23). Additionally, 1 mL/L of a trace element solution was added, which contained the following (in g/L): $MgSO_4 \cdot 7H_2O$ (11.2), $FeSO_4 \cdot 7H_2O$ (4.6), $CoCl_2 \cdot 6H_2O$ (0.05), $MnSO_4 \cdot H_2O$ (0.05), $CuSO_4 \cdot 5H_2O$ (0.05), $NiCl_2 \cdot 6H_2O$ (0.05).

2.3. Thermal treatment of aerobic granular sludge

To investigate the feasibility of P recovery, aerobic granular sludge was gathered from SBAR at the end of anaerobic and aerobic phases, then immediately washed three times with distilled water and dried at 60 °C for 24 h. The dried sludge taken from the end of anaerobic and aerobic phase were labeled as A and O, respectively. Afterward, a certain amount of dried sludge was thermal treated in a tube furnace under nitrogen protection for 1 h at 100 °C, 200 °C, 300 °C, respectively. The samples which were taken from the end of anaerobic and aerobic phase and were heated at 100 °C, 200 °C, 300 °C were named correspondingly A100, O100, A200, O200, A300 and O300. Total nitrogen (TN), total phosphorus (TP), potassium content and organic matter content were measured to investigate the total nutrient content of treated sludge. After sulfuric acid-hydrogen peroxide digestion, TN and TP were measured using Kjeldahl method and spectrophotometry method, respectively. Total potassium content was determined using atomic absorption spectrometry while the content of organic matter for treated sludge was analyzed using potassium dichromate volumetric method.

2.4. Phosphorus release experiments

Batch tests were conducted in 100 ml conical flasks. 0.1 g granular sludge samples after thermal treated under different conditions were added into 50 ml distilled water respectively. The conical flasks were placed on a shaker at 150 rpm and 25 °C. Samples were taken at different time intervals and immediately filtrated through 0.45 μm membrane filters. Finally, TP of each sample was measured.