



## A simple technology for phosphorus recovery using acid-treated concrete sludge



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### ARTICLE INFO

#### Article history:

Received 31 December 2015

Received in revised form 30 March 2016

Accepted 31 March 2016

Available online 1 April 2016

#### Keywords:

Acid-treated concrete sludge

Anaerobic sludge digestion liquor

Bifunctional adsorption-aggregation agent

HCl-soaking

Phosphorus recovery

Wastewater treatment

### ABSTRACT

A simple technology for phosphorus (P) recovery from aqueous solution has been developed using acid-treated concrete sludge (A-CS) as a bifunctional adsorption-aggregation agent. Dried particles of concrete sludge (CS), an alkaline waste containing hydrated cement and fine aggregates, were soaked in 1.3 M HCl at a concentration of 0.1 g/mL for 60 min. The HCl-soaking treatment solubilized alkaline substances such as  $\text{Ca}(\text{OH})_2$  and  $\text{CaCO}_3$ , thereby releasing approximately 87%  $\text{Ca}^{2+}$  from the CS particles to the acid solution. When A-CS was added to 500-mL synthetic anaerobic sludge digestion liquor containing 273 mg/L of phosphate ( $\text{P}_i$ ) at the Ca/P molar ratio of 2.5, A-CS showed 20 times higher  $\text{P}_i$  removal efficiency than that of untreated CS particles. Although A-CS could precipitate 72%  $\text{P}_i$  in 5-min free sedimentation, the  $\text{Ca}^{2+}$ -rich liquid fraction of A-CS alone led to the precipitation of only 48%  $\text{P}_i$ . This suggests that the solid fraction of A-CS can serve as an auxiliary aggregation agent. When  $\text{P}_i$  recovery was examined using anaerobic sludge digestion liquor from a full-scale wastewater treatment, A-CS could recover 96%  $\text{P}_i$  at the Ca/P molar ratio of 2.5. Citrate-soluble  $\text{P}_2\text{O}_5$  accounted for 19% of the dry weight of the recovered  $\text{P}_i$  product. The levels of heavy metals such as As, Cd, Pb, Ni, and Cr in the recovered  $\text{P}_i$  product were much below their regulatory standards for fertilizers.

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### 1. Introduction

Phosphorus (P) is an essential element for all living organisms. It is an indispensable component of cell structure and plays a key role in cellular metabolism [4]. Since P cannot be replaced by any other element in biochemical processes, humans ultimately rely on P availability. Today, P is mostly obtained from mined rock phosphate ( $\text{P}_i$ ) [3]. While high-grade rock  $\text{P}_i$  is depleting on a global scale, the inefficient use of  $\text{P}_i$  leads to the eutrophication problem in natural bodies of water [8,10]. Considerable attention has been paid to P recycling from wastewater to mitigate the anthropogenic mobilization of P [13].

To fully realize P recycling, it is essential to develop a simple, cost-effective technology for P recovery. Previously, we have

reported a novel technique for P recovery from aqueous solution using amorphous calcium silicate hydrates (A-CSHs) [8]. A-CSHs were chemically synthesized using widely-available, inexpensive materials such as siliceous shale and  $\text{Ca}(\text{OH})_2$ . NMR analysis revealed that the A-CSHs consisted of short-length silicate polymers, linked to each other through ion bindings with  $\text{Ca}^{2+}$ . It has also been demonstrated that A-CSHs can serve not only as a  $\text{P}_i$  adsorbent but also as an aggregation agent in aqueous solution [7].  $\text{P}_i$  removed by A-CSHs exhibited better settleability, filterability, and dewaterability than  $\text{P}_i$  precipitated with conventional  $\text{CaCl}_2$  and  $\text{Ca}(\text{OH})_2$ . In addition, no significant carbonate inhibition was observed in  $\text{P}_i$  removal by A-CSHs.

Concrete sludge (CS) is an alkaline waste material consisting of hydrated cement, fine aggregates, and water that is available at construction sites where more concrete materials are generated than required. Conventionally, CS has been used as a landfill material after solid-liquid separation followed by neutralization with a strong acid [11]. However, since this is costly and wasteful, it is desirable to develop an alternative technology that can contribute to the valorization of CS for the cement and construction industry [1,11].

**Abbreviations:** P, phosphorus;  $\text{P}_i$ , phosphate; A-CSHs, amorphous calcium silicate hydrates; NMR, nuclear magnetic resonance; CS, concrete sludge; A-CS, acid-treated concrete sludge; WWTP, wastewater treatment plant; XRF, X-ray fluorescence; P-XRD, powder X-ray diffractometry; C- $\text{P}_2\text{O}_5$ , citrate-soluble phosphate; ACP, amorphous calcium phosphate.

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In the present study, we developed a simple P recovery technology from aqueous solution using acid-treated CS (A-CS) as a bifunctional adsorption-aggregation agent. When CS particles were soaked in HCl, their ability to remove  $P_i$  from aqueous solution was markedly enhanced. The  $P_i$  recoverability of A-CS was confirmed in laboratory experiments using anaerobic sludge digestion liquor from a full-scale wastewater treatment plant (WWTP).

## 2. Materials and methods

### 2.1. Acid treatment of CS

Unused CS was taken from a ready-mix concrete plant, washed with water, and dewatered using a filter press (AK175, Kyokuto Sangyo Co., Ltd., Tokyo, Japan). The filter cake was subjected to air drying at room temperature and ground using a mortar and pestle to obtain CS particles. The CS particles were soaked in 1.3 M HCl at a concentration of 0.1 g/mL for 60 min to form A-CS slurry. During the HCl-soaking treatment, the mixture was continuously stirred by a magnetic stirrer at 600 rpm at room temperature. The pH of the mixture was monitored using a digital pH meter (DT-1023, Able Co., Tokyo, Japan). Samples were taken from the mixture at various time intervals and filtered through a 0.22- $\mu$ m-pore-size Millex-GV filter (Merck Millipore, MA, USA). The concentration of  $Ca^{2+}$  in the filtrate was determined using the Dia-color Liquid Ca kit (Toyobo Co. Ltd., Osaka, Japan) in accordance with the manufacturer's instructions. When required, the A-CS slurry was separated into solid and liquid fractions by centrifugation at 2000g at room temperature for 10 min. To study the effect of the surface electric charge of particles on  $P_i$  settleability, 5 mL A-CS slurry was mixed with 100 mL 1 M NaCl. The mixture was stirred by a magnetic stirrer at 200 rpm at room temperature for 30 min. Then, solids were recovered from the mixture by centrifugation at 2000g at room temperature for 10 min before being used for  $P_i$  settling experiments.

### 2.2. Characterization of CS particles

The chemical composition of CS particles before and after acid treatment was determined using an X-ray fluorescence (XRF) spectrometer (ZSX100e, Rigaku Co., Tokyo, Japan). Acid-soaked samples were dried at 100 °C for 12 h prior to analyses. Powder X-ray diffraction (P-XRD) analysis was performed using a RINT-2100 diffractometer (Rigaku). XRD patterns were recorded using Ni-filtered  $Cu K\alpha$  radiation (step size, 0.02°; scan range,  $2\theta = 10\text{--}60^\circ$ ). Electron microscopy was performed at 1.5 kV using a JSM-7600F scanning electron microscopy (SEM) system (JEOL Ltd., Tokyo, Japan). Before the P-XRD and electron microscopic analyses, acid-soaked samples were dried overnight under vacuum.

### 2.3. $P_i$ recovery experiments

$P_i$  recovery experiments were performed using a synthetic anaerobic sludge digestion liquor consisting of 392 mg of  $KH_2PO_4$ , 1.89 g of  $NH_4Cl$ , and 3.36 g of  $NaHCO_3$  per liter of deionized water

[8]. The initial pH of the synthetic anaerobic digestion liquor was approximately 7.6. When required, the pH was adjusted to 8.0, 8.5, 9.0, or 9.5 using 1 M NaOH. Either 0.5 g of untreated CS particles or 5 mL of A-CS slurry (0.1 g CS /mL) was added to 500 mL of synthetic anaerobic digestion liquor with a Ca/P molar ratio of 2.5 in a 500 mL Pyrex conical beaker. The reaction mixture was stirred by a magnetic stirrer at 200 rpm at room temperature. The pH of the mixture was continuously monitored using a digital pH meter as mentioned above. Samples were taken at various time intervals and filtered through a 0.22- $\mu$ m-pore-size Millex-GV filter (Merck Millipore). The filtrate  $P_i$  was determined by the molybdenum blue method described by Murphy and Riley [6]. Total P was determined as  $P_i$  after autoclaving a sample at 121 °C for 30 min.  $P_i$  removal experiments were also carried out using  $CaCl_2$  at the same  $Ca^{2+}$  concentration as that of the liquid fraction of A-CS slurry.

After 60 min of  $P_i$  removal, the reaction mixture was transferred to a 500-mL graduated cylinder of 5.5 cm diameter to assess the settleability of the removed  $P_i$ . After 5 min of free sedimentation, 400 mL of the supernatant (80% of the original volume) was gently removed using a siphon. The total P concentration in the supernatant was determined as described above. Settleability was evaluated by determining the percentage of total P that remained in the cylinder after siphoning the supernatant. Settleability was also evaluated by visual examination.

$P_i$  recovery experiments were also carried out using anaerobic sludge digestion liquor, which was rejected water from a membrane-type solid-liquid separator for digested sludge at a full-scale WWTP. Approximately 2.0 L of the anaerobic sludge digestion liquor was fed into a 3.0-L acrylic batch reactor. The initial pH of the anaerobic sludge digestion liquor was adjusted to 8.5 after A-CS slurry was added at a Ca/P molar ratio at 2.5. The reaction mixture was continuously stirred by a mechanical agitator (PSH540-001P, Oriental Motor Co., Ltd., Tokyo, Japan) at 200 rpm.

After 60 min of  $P_i$  removal, the reaction mixture was transferred to a 2.0-L graduated cylinder of 8.0 cm diameter and subjected to 20 min of free sedimentation. After siphoning 1.4 L of the supernatant, the remaining 0.6 L of slurry was filtered through a Whatman No. 3 filter paper (GE Healthcare Life Sciences, NJ, USA).  $P_i$  recovered by the A-CS slurry was dried at 105 °C for 24 h. Citrate-soluble P (C- $P_2O_5$ ) was determined as  $P_i$  solubilized by 2% citric acid [5]. Arsenic (As) content was determined by an atomic absorption spectrophotometer (SpectraAA 220, Agilent Technologies Inc., CA, USA). Cadmium (Cd), lead (Pb), and nickel (Ni) contents were determined by an inductively coupled plasma (ICP) mass spectrometer (7500cx, Agilent Technologies Inc.). Chromium (Cr) content was determined by an ICP optical emission spectrometer (Spectroblue, Spectro, Germany).

## 3. Results

### 3.1. Chemical characterization of CS particles

The chemical composition of the CS particles was determined by XRF analysis (Table 1). The main components of the CS particles were  $SiO_2$  (19.3 mass%) and CaO (41.0 mass%). The CS particles also

**Table 1**  
Chemical composition of CS particles and other solid samples.<sup>a</sup>

Sample	Mass% on a dry weight basis							
	MgO	Al <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	P <sub>2</sub> O <sub>5</sub>	SO <sub>3</sub>	CaO	Fe <sub>2</sub> O <sub>3</sub>	Others
CS particles	1.5	6.3	19.3	0.2	1.6	41.0	2.3	27.8
Solid fraction of A-CS slurry	0.7	16.2	50.8	0.3	0.5	12.6	4.8	14.0
Recovered $P_i$ product	0.5	7.3	18.4	22.3	0.3	33.0	2.6	15.6

<sup>a</sup> Values represent the means for three independent experiments.

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