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Research article

Granulation of drinking water treatment residuals as applicable media for phosphorus removal

Xiuqing Li, Jun Cui, Yuansheng Pei*

The Key Laboratory of Water and Sediment Sciences, Ministry of Education, School of Environment, Beijing Normal University, Beijing 100875, PR China

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ABSTRACT

Recycling drinking water treatment residuals (DWTR) show promise as a strategy for phosphorus (P) removal; however, powdered DWTR is not an ideal practical medium due to clogging. This study granulates DWTR by entrapping powdered DWTR in alginate beads. Results show that granular DWTR has an appreciable amount of mesopores along with a Brunauer-Emmett-Teller (BET) surface area of 43.8 m^2/g and total pore volume of 0.049 cm^3/g. Most metals (e.g., Al, Ba, Be, Cd, Co, Cr, Mn, Ni, Pb, and Zn) in granular DWTR became more stable and granular DWTR could be considered non-hazardous material. Further analysis indicates that the granular DWTR has strong P adsorption capability with a maximum adsorption capacity of 19.70 mg/g as estimated by the Langmuir model. Good P adsorption may be attributed to the formation of Fe-PO_4 and Al-PO_4 associated with the amorphous state of enormous iron and aluminum in granular DWTR. More importantly, granular DWTR exhibits good mechanical stability and maintained its shape with weight loss below 12.49% after three recycling rounds. Overall, granular DWTR appears to serve as better media for phosphorus removal in water treatment structures such as wetlands.

1. Introduction

Excessive phosphorous (P) can induce eutrophication in receiving waters. P removal is thus an effective strategy for eutrophication control. Multiple technologies, including biological treatment (Praveen and Loh, 2016; Xiao et al., 2016), chemical precipitation (Huang et al., 2015), electrocoagulation (Mores et al., 2016), and adsorption (Eljamal et al., 2016; Wang et al., 2016a; Zhu et al., 2016), have been developed for P removal. Adsorption has attracted considerable attention from scientists and managers due to its excellent properties including ease of operation, highly efficient pollutant removal, and cost-effectiveness. Recently, some industrial wastes have been developed as alternative adsorbents for P removal (Biswas et al., 2008; Haddad et al., 2015; Jellali et al., 2011; Wang et al., 2011; Yang et al., 2014). Among them, drinking water treatment residuals (DWTR) is a promising adsorbent to remove P from contaminated water at a low cost.

DWTR is an inevitable and non-hazardous byproduct of drinking water treatment processes that occupy about 4–7% of the total water produced (Sun et al., 2015). Okuda et al. (2014) reported that global daily DWTR production is around 10,000 tons. Large quantities of DWTR are often disposed of as waste or in landfills. In recent years, recycling of DWTR has attracted international interest; it is an ideal adsorbent for various contaminants, including P (Wang et al., 2011), arsenic (Nagar et al., 2010), perchlorate (Makris et al., 2006), hydrogen sulfide (Wang and Pei, 2012), boron (Irawan et al., 2011), mercury (Hovsepyan and Bonzongo, 2009), selenium (Ippolito et al., 2009), cobalt (Jiao et al., 2017), lead (Zhou and Haynes, 2011), chromium (Zhou and Haynes, 2011), copper (Lin et al., 2014), nickel (Chiang et al., 2012), zinc (Chiang et al., 2012), glyphosate (Hu et al., 2011), and chlorpyrifos (Zhao et al., 2013). This is mainly due to high content of amorphous Al or Fe in DWTR, as aluminum or iron salt is commonly used as a coagulant in water purification. For P pollution control, DWTR typically serves as the main substrate in constructed wetlands to remove P from wastewater (Babatunde and Zhao, 2009; Babatunde et al., 2009, 2011; Bai et al., 2014; Zhao et al., 2011a, 2011b, 2009) and as an amendment for *in situ* remediation of P-contaminated sediments or soils (Agyin-Birikorang and O'Connor, 2009; Wang et al., 2013; Wang and Pei, 2013). The beneficial applications of DWTR presumably offer a win-win strategy for waste management.

DWTR is commonly reused in powder form, which may

* Corresponding author.
E-mail address: yspei@bnu.edu.cn (Y. Pei).

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accelerate filtration system clogging due to lower hydraulic conductivity. Generally, a media size of 5–20 mm is used in filtration systems such as constructed wetlands (Vymazal and Kröpfelová, 2008). To satisfy industrial use demand, powdered DWTR must be transformed into a granular form with high mechanical strength. However, few studies have addressed DWTR granulation. Common technologies, including pelleting and sintering (Zou et al., 2012), wood loading (Soleimanifar et al., 2016), and gel entrapment (Jung et al., 2016), have been developed to convert powdered DWTR into a granular form. Granular DWTR, which is created by wood loading (Soleimanifar et al., 2016) and gel entrapment (Jung et al., 2016), has been used in urban stormwater runoff (Soleimanifar et al., 2016) and industrial wastewater treatment processes (Jung et al., 2016). These granular DWTR improve the hydraulic conductivity of filtration systems and greatly slow down clogging. In addition, DWTR-ceramsite obtained from the pelleting and sintering process is often used as a carrier to attach microorganisms in the biofilter (Zou et al., 2012). The development of granular DWTR could expand its applications. Moreover, it is essential to maintain high P adsorption capacities and mechanical stability in granular DWTR. Though granular DWTR produced through the gel entrapment process is an efficient adsorbent (Jung et al., 2016), mechanical stability of the granules has yet to be reported. Therefore, it is highly desirable to develop granular DWTR with high P adsorption capacity and mechanical stability.

In this study, sodium alginate was used as a supporter to encapsulate powdered DWTR into porous structured alginate beads in a granular form. Sodium alginate exhibits gel properties and can be easily fabricated into stable beads by crosslinking with various polyvalent cations, such as Ca^{2+}, Ba^{2+}, and Fe^{3+}. It is a preferred carrier for constructing granular adsorbents. In fact, alginate beads have been used to entrap and immobilize waste metal hydroxide (Escudero et al., 2009), acid mine drainage sludge (Lee et al., 2015), and active carbon (Lin et al., 2005). In addition, the DWTR-alginate beads show excellent adsorption capacity for fluoride in treating industrial wastewater (Jung et al., 2016). These studies demonstrate that sodium alginate can be a supporter for granular adsorbent formation.

The purposes of this study are threefold: (1) to prepare granular DWTR by encapsulating powdered DWTR into alginate beads and characterizing their morphologies using BET, FTIR, and XRD; (2) to investigate the P adsorption properties of granular DWTR, including kinetics, equilibrium of adsorption, and influences of initial solution pH, and to clarify the interaction of P with granular DWTR via SEM-EDS and XPS analysis; and (3) to evaluate the reusable and stable properties of granular DWTR.

2. Materials and methods

2.1. Materials

DWTR was sampled from a drinking water treatment plant in Beijing that used Fe and Al salts as coagulants. Then, the DWTR was air-dried naturally, milled in a crusher, and passed through 125-μm sieves. Sodium alginate and $FeCl_3 \cdot 6H_2O$ were applied to prepare the granular DWTR; KH_2PO_4 was used to prepare the P-containing solution. NaOH and HCl in 0.1 mol/L were used to adjust the initial pH of the P-containing solution. All chemical reagents employed in this study were analytical grade without further purification, purchased from Sinopharm Chemical Reagent Co., Ltd., China. The deionized water used in the experiments was obtained from Milli-Q System of Millipore (Milford, MA, USA).

2.2. Granular DWTR preparation

Granular DWTR was prepared based on the method of Jung et al. (2016) with slight modifications. Briefly, powdered DWTR (10 g) was added into a 100-mL 2% (w/v) sodium alginate solution prepared with deionized water. Then, the mixture was stirred well to obtain a homogeneous dispersion. After that, the mixture was added dropwise into a 2% (w/v) $FeCl_3$ solution with a syringe, and granular DWTR formed. The spheres in the $FeCl_3$ solution were left to stand for 4 h, after which the spheres were collected and washed with deionized water. Finally, the spheres were naturally air-dried and used in the following experiments.

2.3. Heavy metal lability

2.3.1. Toxicity characteristic leaching procedure

The toxicity of the powdered and granular DWTR samples was determined using the toxicity characteristic leaching procedure (TCLP). In the leaching test, 20 mL of acetic acid-sodium acetate solution (pH 4.93 ± 0.05) was applied as the leaching fluid to a 1.00 g sample and stirred at 150 rpm for 18 h. After that, the leachates were filtered through 0.45-μm cellulose acetate membrane, and metal contents were determined by either inductively coupled plasma atomic emission spectrometry (ICP-AES; ULTIMA, JY, France) or inductively coupled plasma mass spectrometry (ICP-MS; NexION 300, Perkin Elmer, Shelton, CT, USA).

2.3.2. In vitro bioaccessibility (IVBA) procedure

Briefly, 1.0 g samples were added to 100 mL of 0.4 M glycine buffered solution, which was adjusted to pH of 1.5 ± 0.5 at 37 °C with trace-metal grade HCl. Then, the samples were incubated at 37 °C for 1 h at 30 rpm. After incubation, the system pH was measured. If the pH variation was not within ±0.5 pH units of the starting pH, the test was repeated. If the same pH outcome was obtained in the second test, the results were considered qualified. Next, 10 mL of extract was collected and filtered through 0.45-μm cellulose acetate membrane, and heavy metal content was measured using ICP-AES or ICP-MS. Bioaccessibility was expressed as the ratio of extracted heavy metal to the total heavy metal in the sample.

2.4. P adsorption

2.4.1. Adsorption kinetics

Granular DWTR (1.0 g) was placed into 250 mL conical flasks. Then, 100 mL of a solution containing 0.01 mol/L KCl and KH_2PO_4 concentrations of 100, 500, and 1000 mg/L (corresponding to respective P concentrations of 22.79, 113.97, and 227.94 mg/L) at pH 6 were added to the conical flasks. P concentrations were selected based on various wastewater concentrations and helped to fully explain P adsorption behaviors onto granular DWTR. The conical flasks were placed in a shaking incubator at 100 rpm and 25 °C for P adsorption. After shaking for 2, 4, 6, 8, 10, 12, 24, 48, and 72 h, the supernatants were sampled for residual P concentration analysis according to the ammonium molybdate spectrometry method. All experiments were conducted in triplicate. The P adsorption capacities of powdered DWTR and alginate beads under initial KH_2PO_4 concentrations of 100, 500, and 1000 mg/L and a contact time of 72 h are shown in Table S1. Results indicate that the P adsorption capacity was attributed to DWTR.

2.4.2. Adsorption isotherm

The experiments were similar to adsorption kinetics. The initial P concentrations were 0.31, 3.1, 15.5, 31, 155, and 310 mg/L. The shaking time was fixed at the equilibrium time obtained from the