



Review

Adsorptive removal of phosphate from water using mesoporous materials: A review

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ABSTRACT

Mesoporous materials have significant potential for use as adsorbents for removal of phosphate from water. The chemical and structural properties of materials greatly affect their capacity and rate in the phosphate adsorption process. This paper reviews recent activities in the development of mesoporous materials as phosphate adsorbents. In particular, it mainly focuses on the synthesis, properties and phosphate removal efficiency of various materials with mesoporosity, including metal-coordinated amino-functionalized silicas, ammonium-functionalized silicas, metal-doped mesoporous silicas, metal oxides, metal sulfate and carbon.

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1. Introduction

Phosphorus is the mineral nutrient which is essential for all living species. However, the excessive presence of phosphorus in water bodies, which is mainly considered as a result of untreated

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sewage effluent and agricultural run-off, causes eutrophication problem in rivers, lakes and seas. Eutrophication induces overgrowth of phytoplankton, thus deteriorating water quality, depopulating aquatic species and accelerating water scarcity (Sellner et al., 2003). According to the Australian Water Quality Guidelines for Fresh and Marine Waters, the total phosphorus contaminant level in rivers and streams is controlled in the range of 0.010–0.100 mg/L; whilst the requirement for lakes and reservoirs is more stringent as 0.005–0.050 mg/L (Australian and New Zealand Environment and Conservation Council and Agriculture and Resource Management Council of Australia and New Zealand, 2000).

Phosphate removal from water has attracted considerable research interest in the last few decades. Until now, a range of methods have been developed, mainly including biological, chemical, and physical treatments (Morse et al., 1998; Wang et al., 2012). Biological methods, *i.e.* the conventional activated sludge process, can achieve nearly 100% removal of phosphate. However they are less effective at trace level, which is ascribed to the fact that the presence of insufficient phosphate lowers metabolism of microorganisms. Furthermore, specific care and strict control are often needed during implementation of biological methods. Chemical treatments using lime, alum, and iron salts have been mostly utilized in phosphate removal, which suffer difficulty in sludge disposal and effluent neutralization. Physical methods, as in the case of reverse osmosis and electrodialysis, have been proven to be either too expensive or inefficient in removal (Rodrigues and da Silva, 2009). In comparison to the aforementioned methods, the adsorption process is promising for phosphate removal (Peleka and Deliyanni, 2009), attributed to its attractive advantages, *e.g.* simple operation, high removal efficiency and fast adsorption rate, especially at low phosphate concentration. Some materials, *e.g.* activated aluminum, have been selected as adsorbents to polish effluent from wastewater treatment facilities (Jiang et al., 2004). The total annual economic cost for the process incorporated with chemical precipitation was approximately 10% lower than that with adsorption (Jiang et al., 2004). However, the adsorption process could reduce phosphate concentration in the discharge to a much lower level, as compared with the chemical method (Jiang et al., 2004). The development of materials with superior adsorption and regeneration capacity could further enhance phosphate removal, minimize sludge disposal and eventually make adsorption cost-effective for practical application. Furthermore, the adsorption is able to be used for not only phosphate removal, but also phosphate recovery (Biswas et al., 2008; Das et al., 2006; Gan et al., 2009; Hongshao and Stanforth, 2001; Huang et al., 2008; Karageorgiou et al., 2007; Kostura et al., 2005; Li et al., 2009; Liao et al., 2006; Ning et al., 2008; Onyango et al., 2007; Papadopoulos et al., 1998; Pengthamkeerati et al., 2008; Pradhan et al., 1998; Ugurlu and Salman, 1998).

In recent years, there has been tremendous interest focusing on the fabrication of mesoporous materials and their potential in different practical applications (Gabaldon et al., 2007; Liu et al., 2000; Melde et al., 2008; Mahmud et al., 2016; Slowing et al., 2007; Vallet-Regi et al., 2008; Vunain et al., 2016; Yamaguchi and Teramae, 2008; Yang et al., 2008; Yeung and Han, 2014; Yu et al., 2016). The International Union of Pure and Applied Chemists (IUPAC) has classified mesopores as the pores with sizes of 2 – 50 nm. Mesoporous materials, especially those with ordered pore systems, endow huge interface to accommodate guest species as well as feasibility for functionalization; which are necessary requirements for excellent adsorbents (Wu and Zhao, 2011). Therefore, this class of materials has been extensively investigated as adsorbents to remove diverse inorganics and organics from water (Bibby and Mercier, 2002; Ho et al., 2003; Yan et al., 2006; Yokoi

et al., 2004; Yoshitake et al., 2003).

Some review papers have been devoted to the fabrication of mesoporous materials and their use as adsorbents to remove ions, *e.g.* arsenate, chromate, selenate and molybdate (Mahmud et al., 2016; Vunain et al., 2016; Walcarius and Mercier, 2010; Yokoi et al., 2012; Yoshitake, 2007; Yu et al., 2016). However, until now, there has been no review on the development of mesoporous materials as adsorbents especially targeting at phosphate removal. This review gives an overview on the development of mesoporous materials as phosphate adsorbents, including functionalized mesoporous silicas, mesoporous metal oxides/hydroxides/sulfate and mesoporous carbon. The mesoporosity of materials covered herein is not limited to that arising from well-ordered pore systems but also non-ordered and intraparticle pores. Synthetic methods, structural and surface properties, and phosphate removal of mesoporous materials are featured. However, the removal of organophosphates by mesoporous materials (Meng et al., 2011; Pan et al., 2006) is not covered.

2. Mesoporous silica

2.1. Organic-functionalized mesoporous silica

The most widely reported mesoporous silicas in phosphate adsorption include MCM-41 (with a non-intersecting hexagonal arrangement of mesopores), MCM-48 (with a cubic arrangement of mesopores), as well as SBA-15 (with a hexagonal arrangement of mesopores) (Beck et al., 1992; Huo et al., 1996; Zhao et al., 1998a, 1998b). Mesoporous silicas hardly removed phosphate from water when used as adsorbents due to no active sites (Choi et al., 2012a; Delaney et al., 2011; Saad et al., 2008; Shin et al., 2004; Kim et al., 2015; Hamoudi and Belkacemi, 2013). In order to improve adsorption capacity, organosilanes terminated with chemically selective ligands were utilized to functionalize mesoporous silicas (Fryxell et al., 2007). Amino (including mono, di, and tri) groups were the commonly selected ones (Hamoudi et al., 2010; Hao et al., 2011; Yokoi et al., 2003; Yoshitake, 2005). Basically, amino-functional groups were immobilized onto mesoporous silicas, which were then coordinated with metal cations or protonated to provide active sites and enable uptake of phosphate (Chouyyok et al., 2010; Hamoudi et al., 2007, 2013; Huang et al., 2013a; Kang et al., 2016; Saad et al., 2007; Zhang et al., 2011a, 2011b). Table 1 summarizes the development of metal-coordinated or protonated amino-functionalized mesoporous silicas as phosphate adsorbents. Seen from the literature, to optimize performance of such adsorbents, it is critical to achieve high loading of functional groups, uniform dispersion of organic moieties and good accessibility of pore openings (Yokoi et al., 2012).

Two strategies, namely post-synthesis grafting and co-condensation, have been adopted to synthesize functionalized mesoporous silicas (Fig. 1) (Yokoi et al., 2012). Post-synthesis grafting is to “graft” functional moieties onto mesoporous silicas after organosilanes condensing with surface silanol groups (Fig. 1a). This strategy has shown a number of advantages, especially its applicability to various types of mesoporous silicas and capability to retain mesostructures of silicas (Zhao and Lu, 1998). However, there have been some demerits specified, one of which is the difficulty in controlling uniform distribution of organic moieties on silicas. In particular, organic ligands would tend to congest near entrances of mesopores and reduce effective pore sizes (Lim and Stein, 1999; Zhao and Lu, 1998).

Co-condensation process (Fig. 1b), also known as one-pot or direct synthetic method, proceeds *via* hydrolysis and subsequent condensation of a gel consisting of a silica precursor, surfactant and organosilane. It is feasible to precisely control quantity of

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