Journal of Environmental Management 181 (2016) 522–535

Contents lists available at ScienceDirect

Journal of Environmental Management

journal homepage: www.elsevier.com/locate/jenvman

Research article

Preparation of high-quality activated carbon from polyethyleneterephthalate (PET) bottle waste. Its use in the removal of pollutants in aqueous solution

Raúl Mendoza-Carrasco, Eduardo M. Cuerda-Correa, María F. Alexandre-Franco, Carmen Fernández-González, Vicente Gómez-Serrano*

Department of Organic and Inorganic Chemistry, Faculty of Sciences, University of Extremadura, Avda. de Elvas, s/n, E-06006, Badajoz, Spain

ARTICLE INFO

Article history:
Received 14 July 2015
Received in revised form
29 June 2016
Accepted 30 June 2016

Keywords:
Polyethyleneterephthalate (PET)
Activated carbon
Physico-chemical characterization
p-Nitrophenol
Ferric ion
Adsorption

ABSTRACT

A waste-treats-waste approach has been used for the removal of two common pollutants, namely *p*-nitrophenol and/or Fe(III) from aqueous solution. Polyethyleneterephthalate (PET) from bottle waste has been used as the precursor for the preparation of activated carbons (ACs) by physical activation with steam and chemical activation with potassium hydroxide under controlled heating conditions and atmospheres. The resulting ACs were characterized in terms of chemical composition, porous texture and surface chemistry, and morphology. Selected ACs were tested as adsorbents for the removal of the aforementioned pollutants in aqueous solution. For comparison purposes, a commercial AC was also used. In general, the yield of the process of preparation of ACs is lower than 10% with steam and between 24.62 and 32.07% with potassium hydroxide. ACs possess a very high carbon content and a very low ash content. The BET surface areas reach 1235 $m^2\ g^{-1}$ with steam and 1002 $m^2\ g^{-1}$ with potassium hydroxide at most. Also, the degrees of development of micro- and mesoporosity are markedly larger with steam. Conversely, the development of macroporosity is much larger with potassium hydroxide. The PET-derived ACs exhibit a better adsorption behavior towards *p*-nitrophenol than the commercial AC, both in terms of adsorption rate and adsorption capacity. On the contrary, the commercial AC acts as a better adsorbent of Fe(III) ions. As compared to separately, the simultaneous presence of both solutes in the adsorptive solution scarcely affects the adsorption process except for equilibrium for Fe(III).

1. Introduction

PET is a widely-used polymer in the food industry because of its inertness towards food, transparency, difficulty to be holed, negligible permeability to CO_2 and malleability (Parra et al., 2004a). Due to its excellent properties, the consumption of PET has increased markedly in the last few years (Europe, 2004). As a consequence, large amounts of PET wastes are produced worldwide every year. As an average, plastic materials take approximately 180 years to degrade due to their inertness. Thus, plastic wastes are not likely to be assimilated back into nature. Hence, the global amount of PET waste has increased dramatically in the last decades. Furthermore, their presence in inappropriate places is clearly perceptible to the population and has a high visual impact and the discharge of plastic waste, including PET, represents a serious challenge for waste and environmental management (Dias et al., 2007).

For several years, incineration in landfill sites has been a common practice in the treatment process of PET residues (Saha and Ghoshal, 2005). However, this procedure may also adversely affect the environment as it takes place with air consumption and release of heat and volatile compounds (Yasuhara et al., 2002). These latter cause severe damages to the atmosphere due to the emission of ammonia, sulfur dioxide, organic acids with corrosive properties, dioxins, etc (Katami et al., 2002). Furthermore, the combustion of some of the additives included in the formulation of PET gives rise to the generation of ashes that may modify the composition and pH of the soil.

Hence, the search for new processes to valorize PET wastes as well as to avoid the generation of hazardous materials is interesting from both, the theoretical and applied standpoints (Park and Gupta, 2015). For instance, a few years ago Wang et al. (2012) reported on

---

* Corresponding author.
E-mail address: vgomez@unex.es (V. Gómez-Serrano).

http://dx.doi.org/10.1016/j.jenvman.2016.06.070
0301-4797/

the possibility of obtaining chelating fibers prepared with waste PET fibers for fast removal of heavy metal ions from water. Another alternative would be the use of the spare PET bottles as the raw material in the preparation of carbon-based materials as, for instance, carbon microspheres (Lee et al., 2005; Pol et al., 2009), carbon nanotubes (Bazargan and McKay, 2012) or activated carbons (Esfandiari et al., 2012; Kartel et al., 2006; Laszlo et al., 2000; Laszlo and Szucs, 2001; Laszlo et al., 2001). In fact, PET appears as an excellent candidate to be used as precursor for the preparation of activated carbons since it has a high content of carbon and negligible amounts of inorganic matter in its chemical constitution (Blazso, 1997; Domingo-Garcia et al., 2010; Marzec et al., 1999; Nakagawa et al., 2004). The experimental methods and conditions of preparation of ACs from PET used in earlier studies are summarized in Table 1, which also lists the values of textural parameters for the best samples reported.

Activated carbon is a carbonaceous material characterized by its well-developed porous structure and surface chemistry. These properties of AC depend on the starting material and the method used in its preparation, including the activating agent and the operational conditions. In the case of ACs prepared using PET as the raw material, the method of chemical activation has been proved to be more effective than the method of physical activation to prepare ACs with a remarkable porosity (Almazan-Almazan et al., 2010; Esfandiari et al., 2012; Fernandez-Morales et al., 2005; Parra et al., 2004c; Ruiz-Fernandez et al., 2011; Strachowski and Bystrzejewski, 2015). These textural properties, together with the amphoteric nature of the surface of the adsorbents, make activated carbons prepared from PET waste suitable for application in industrial sorption processes.

Industrial activity generates a large volume of effluents containing hazardous species that need to be treated before being delivered into the environment. Phenol and its derivatives are considered as severe toxic pollutants in the environment. These chemicals are introduced into surface water from a wide variety of industrial effluents, domestic wastewaters, and agricultural or chemical spills (Lin and Juang, 2009). This kind of products exhibit low biodegradability, which make them markedly dangerous for the environment (Boukhoubza et al., 2009). Many phenolic compounds are recognized as toxic or even act as carcinogens (Kavlock et al., 1991). They have relatively high solubility in water and their presence causes unpleasant taste and fetid odor. The exposure of humans or animals to certain phenolic compounds, including nitrophenols, may cause serious health effects (Mwangi et al., 2014). The origin of nitrophenols is entirely anthropogenic, that is, humans are solely responsible for the presence of these chemicals in the environment.

Due to its large industrial production and environmental pollution potential, *p*-nitrophenol can be considered the most important among all of the mononitrophenols (Herrera-Melian et al., 2012). *p*-nitrophenol can enter the environment during the production of dyes, pesticides and drugs (Cotoruelo et al., 2012). According to the Environmental Protection Agency (EPA), the limit of *p*-nitrophenol should not exceed 100 mg/L, due to its toxic nature as it is easily adsorbed by the respiratory tract and digestive tract and skin (Kishino and Kobayshi, 1996). The presence of *p*-nitrophenol has been detected in the urine of people who were unaware of having been exposed to this chemical. Hence, the implementation of processes aimed at removing *p*-nitrophenol from aqueous solutions is a topic of current interest for the scientific community. A wide variety of methods have been proposed to remove *p*-nitrophenol from wastewaters, including advanced oxidation process (AOPs), sonolysis, extraction, and adsorption (see (Bastami and Entezari, 2012) and references therein).

On the other hand, iron is present in the water environment for purely natural causes and due to anthropogenic action related to their applications. It is the metal with the greatest number of industrial applications. It is an essential element for humans, but if iron exceeds the amount required by the body, it accumulates in the liver, pancreas, spleen and heart, which can damage these vital organs. Furthermore, Fe(II) and Fe(III) ions as well as many iron compounds can react with oxygen to produce oxygen radicals and peroxides, a group of chemicals that may cause severe damages in DNA (Papanikolaou and Pantopoulos, 2005). The iron removal is usually carried out mainly in water purification treatments. A commonly used process for the removal of Fe(II) from aqueous solution has been crystallization or oxidation of Fe(II) to Fe(III) followed by chemical precipitation. However, this procedure does not generally allow strict regulatory requirements to be met (Tahir and Rauf, 2004).

From all the above exposed, it becomes evident that it is appropriate to remove contaminants such as *p*-nitrophenol and iron from water environment. In this connection, adsorption technology is widely applied for the decontamination of waters from these compounds. Activated carbons (ACs) are conventionally employed as powerful adsorbents in many industrial applications.

Furthermore, an adequate bibliographic review clearly shows that the preparation of adsorbent materials that may be useful in the simultaneous removal of organic and inorganic substances in aqueous solution is a topic that has been investigated to a relatively small extent to the date, despite most natural systems are multicomponent ones. The most widely used adsorbents are layered alumino-silicates (e.g., bentonites or ogano-bentonites (Jovic-Jovicic et al., 2014; Lee et al., 2002; Ma and Zhu, 2006; Zhu et al., 2006) and illite (Polubesova and Nir, 1999)), magnetic materials (He et al., 2013; Wang et al., 2014), hybrid materials (Choi et al., 2011, 2009; Jin et al., 2011; Suchithra et al., 2013) and carbonaceous adsorbents (Diaz-Flores et al., 2009; Li and Helmreich, 2014). Hence, the preparation of adsorbent materials exhibiting high affinity towards organic and inorganic pollutants simultaneously is a

Table 1
Textural properties of the activated carbons prepared from PET by physical activation methods. Values reported in the literature.

Activating agent	Carbonization temperature/ °C	Activation temperature/ °C	Treatment time/ min	S_{BET}/ $m^2\ g^{-1}$	V_{mi}/ $cm^3\ g^{-1}$	V_{me}/ $cm^3\ g^{-1}$	V_T/ $cm^3\ g^{-1}$	Reference
CO_2	800	975	240	790	–	–	0.666	Esfandiari et al. (2012)
Steam	700	900	90	1254	–	–	–	Laszlo et al. (1999)
H_2SO_4/steam	–	800	5–8	950	0.36	–	0.60	Sych et al. (2006)
Steam	750	900	–	1190	–	–	0.62	Laszlo et al. (2001)
Steam	750	900	90	1443	0.65	–	0.70	Podkoscielny and Laszlo (2007)
H_2SO_4/steam	–	825	5–8	1030	0.36	–	0.60	Kartel et al. (2006)
CO_2	–	925	–	2468	0.90	0.236	1.087	Parra et al. (2004b)
CO_2	800	975	240	2010	–	–	0.93	Ali et al. (2011)
Steam	750	900	90	1170	0.425	–	0.625	Laszlo and Szucs (2001)