Journal of Cleaner Production 149 (2017) 1109–1117

Contents lists available at ScienceDirect

Journal of Cleaner Production

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

The relative performance of microwave regenerated activated carbons on the removal of phenolic pollutants

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ARTICLE INFO

Article history:
Received 30 September 2016
Received in revised form
24 February 2017
Accepted 25 February 2017
Available online 27 February 2017

Keywords:
Regeneration
Microwave
Activated carbon
Phenolic pollutant
Adsorption

ABSTRACT

The adsorption of reagent on the surface of porous material and the regeneration of porous material are significant processes that are widely used in chemical industries. The performance of a granular commercial activated carbon and two activated carbons synthesised from the factory waste tea and demineralised waste tea, were tested during a series of adsorption and regeneration cycles. Phenol and p-nitrophenol (PNP, 4-nitrophenol) were used as the reagents to represent water pollutants. The activated carbon samples were regenerated by the application of a short period of microwave energy (30 s). The influence of the microwave regeneration process on the surface area and pore volumes were investigated with laboratory characterisation techniques. Several cycles of adsorption and regeneration processes were conducted to determine the variation in the adsorption capacity and the characteristics of each porous material. The adsorption and regeneration processes were interpreted in terms of the amount of adsorbed reagent and surface characteristic of the porous material. The materials were found to be good adsorbents for phenol and PNP and the regeneration process worked effectively; broadly maintaining the adsorption capacity over multiple adsorption – regeneration cycles.

1. Introduction

Porous materials are used in a variety of applications such as separation processes, catalysis, tissue engineering and capacitor manufacture (Gurten Inal et al., 2015). Activated carbon is a porous material that is widely used in many of these and other applications. One specific application is the adsorption of different chemicals on activated carbon. Phenolic compounds are water pollutants due to their toxicity. These compounds are listed as “priority pollutants” by the EPA (Environmental Protection Agency). The removals of phenolic compounds are widely and often used as a test in case studies. Phenolic compounds such as phenol (Salvador and Sanchez Jimenez, 1999), phenol and 4-nitrophenol (Salvador and Sanchez Jimenez, 1996), orto-chlorophenol and pentachlorophenol (Rivera-Utrilla et al., 2003), 2,4,5-trichlorobiphenyl (Liu et al., 2007) have been reported. Dabrowski et al. (2005) published an extensive review detailing phenol and its derivatives used in different concentrations in aqueous media for adsorption onto various activated carbons. Other studies have used a wide range of contaminants to assess the performance of different activated carbons; including promethazine and salicylic acid (Çalışkan et al., 2012), methylene blue (Weng and Hsu, 2008), ethanol and acetone (Fang and Lai, 1996), azo dyes such as Reactive Black 5 (Chang et al., 2010), Direct Red 79 (Salvador and Sanchez Jimenez, 1999) and Acid Orange 7 (Qu et al., 2009). Using these reagents the adsorption capacities, the adsorption behaviour and the adsorption kinetics were determined and the related data were also fitted to several models such as Langmuir, Freundlich, Temkin in some of these studies. The activated carbons used in these studies were characterised in terms of by BET surface area, pore size distribution, surface functionality, TG/DTA and SEM analyses.

An activated carbon can be saturated with the reagent during the adsorption process. When the activated carbon reaches its saturation limit, it does not adsorb further reagent. Exhausted activated carbon can be discharged or disposed of in a landfill if it is a low cost material and is within the consent levels. However, activated carbon can be an expensive material and disposal may not be the most sustainable solution. The regeneration of the exhausted material should be considered; to reuse the material and minimise the cost and environmental impact. There are several techniques

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http://dx.doi.org/10.1016/j.jclepro.2017.02.188
0959-6526/

for regeneration of the exhausted activated carbon such as conventional thermal regeneration (San-Miguel et al., 2001), oxidation (González et al., 2002), solvent regeneration (Chinn and King, 1999), biological regeneration (Scholz and Martin, 1998), electrochemical methods (Narbaitz and Cen, 1994) and ultrasonic regeneration (Hamdaoui et al., 2005). Microwave regeneration has been reported in the literature for activated carbon loaded with different reagents; including 2,4,5-trichlorobiphenyl (Liu et al., 2007), phenol (Ania et al., 2004), salicylic acid (Ania et al., 2007), methylene blue (Foo and Hameed, 2012a) and Acid Orange 7 (Quan et al., 2004).

Microwave irradiation has been widely used domestically, in different fields of industry and also in medical applications. Due to the advantages of microwave irradiation (including safety, speed and selective heating) when compared with conventional heating, microwave heating is an emerging technology in the chemical process industries (Yuen and Hameed, 2009). A number of studies have been reported in the existing literature regarding the use of microwave heating to regenerate activated carbons. Carbons are good microwave absorbers due to the interactions of the delocalised π−electrons with the microwaves. Different types of activated carbons have been employed to adsorb different reagents such as phenolic compounds as reported above. The activated carbons saturated with these reagents were regenerated using microwave heating. The efficiency of regeneration in different successive adsorption and regeneration cycles was reported in these studies. The regenerated samples were characterised in terms of the BET surface area, pore size distribution and surface chemistry.

The main objective of this study was to investigate the effect of sequential microwave regeneration of the saturated activated carbon in equilibrium, in terms of the nature of the regenerated materials and the adsorption performance. The other objective was to determine and contrast the adsorption behaviour of phenol and p-nitrophenol (listed as priority pollutants by the EPA) on powdered activated carbon produced from factory waste tea and on a coal based commercial granulated activated carbon material.

2. Experimental

2.1. Materials

Commercial activated carbon (CAC) and activated carbon produced from the factory waste tea were used in the adsorption of phenol and p-nitrophenol. Analytical grades of phenol (Riedel de Haen) and PNP (Sigma) were 99.5% and 99.9%. The waste tea was less than 500 μm in particle size. The activated carbon was prepared from the factory waste tea and also a demineralised sample of the factory waste tea. The commercial activated carbon, which was used for comparative purposes, was a coal based porous material and in granule form.

2.2. Demineralisation of the waste tea

The mineral matter in the raw material and in the activated carbon may play a role as a receptor during the microwave heating process (Yagmur et al., 2008a). The surface area and surface properties of the activated carbon may change significantly in the presence of the mineral matter during the microwave regeneration process.

The waste tea sample (20 g) was mixed in a beaker using 200 ml HCl (0.1 M) solution and the mixture was agitated using a magnetic stirrer at room temperature for 4 h. The suspension was filtered and washed with distilled water until the filtrate was of neutral pH. The solid was then dried in a vacuum oven for 48 h. This demineralisation method was reported in the literature (Vamvuka et al., 2006).

2.3. Production of activated carbon

The production of activated carbon was performed using the natural waste tea (tea factory waste) and demineralised waste tea. The production method previously reported in literature (Yagmur et al., 2008b) was applied. Briefly, the precursor was mixed H_3PO_4 (wt, 85%) to 2/1 acid/precursor ratio by weight. The material was then exposed to microwave energy (MW) for 30 s (in a domestic type microwave oven: Vestel, MDG-620, 900 W; dimensions (mm): 440 × 355 × 259; frequency: 2.45 GHz; wavelength: 12.2 cm; quantum energy of microwave photon: 1×10^{-5} eV; noise level: 58 dB) to enhance the surface area of the final product. The solid mass taken from the microwave oven was carbonised/activated at 450 °C (heating rate: 10 °C/min., from room temperature (≈20 °C) to 450 °C, takes 43 min) in a quartz tube reactor placed in a temperature programmable rotary tube furnace under N_2 flow (0.15 L/min). The sample was held in the furnace at 450 °C for 1 h before cooling down under a nitrogen flow. The sample was kept in hot distilled water (75 °C, 500 ml) overnight (16 h) then washed and filtered. The washing process was repeated until the filtrate became neutral. The washed sample was dried in an oven at 110 °C overnight and then milled at 20,000 rpm for 3 min.

2.4. Adsorption of phenol and p-nitrophenol (PNP)

The phenol and PNP adsorption experiments were carried out at 30 °C in glass vessels. The vessel was covered with a teflon lid to avoid the influence of air during the adsorption process. An orbital shaker (Gerhardt Thermoshaker) was utilised to mix the slurry and the rotation speed of the shaker was 150 rpm during adsorption process. The activated carbon content in the slurry was 2 % wt/wt (9.8 g solution + 0.2 g solid). At the end of adsorption process, the sample slurry was withdrawn using a glass syringe and then immediately filtered. A pressure type syringe holder was used to filter the slurry. A high quality polycarbonate membrane filter was employed for the filtration process. The properties of the reagents are reported in Table 1.

2.5. Microwave regeneration

Prior to microwave regeneration each reagent (at high concentration: 2000 ppm) was adsorbed on the activated carbon. Phenolic compounds represent a serious potential hazard for human health and aquatic life, and that is why phenols have been registered as a priority pollutant by the US Environmental Protection Agency (USEPA, 1985) with a permissible limit of 0.1 mg/L (0.1 ppm) in wastewater (Srivastava et al., 2006). The reagent remaining in the water (unadsorbed) was determined using an ultraviolet spectrophotometer (UV). The activated carbon with the adsorbed reagent was regenerated in the same microwave oven used in the production of activated carbon. The regeneration time was 30 s. The

Table 1
Properties of phenol and PNP.

	Phenol	PNP (4-nitrophenol)
Molecular formula	C_6H_5OH	$C_6H_5NO_3$
Molar mass, g/mol	94.11	139.11
Density, g/cm^3	1.07	1.27
Melting point, °C	40.50	113–114
Boiling point, °C	181.70	279
Solubility in water (20 °C)	83 g/L	11.6 g/L