CHEMICAL ENGINEERING TRANSACTIONS
VOL. 78, 2020
A publication of AIDIC
The Italian Association of Chemical Engineering
Online at www.cetjournal.it
Guest Editors: Jeng Shiun Lim, Nor Alafiza Yunus, Jiří Jaromír Klemeš

ISBN 978-88-95608-76-1; **ISSN** 2283-9216
DOI: 10.3303/CET2078062

Recycled Poly(ethylene terephthalate) as Dye Adsorbent : A Mini-Review

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Poly(ethylene terephthalate) (PET) has become one of the most major post-consumer plastic waste, in addition to poly(ethylene), poly(propylene), poly(styrene) and poly(vinyl chloride). PET bottles are mostly used in packaging of drinking and mineral water as well as soft drinks. This review demonstrates the potential of PET waste as a value added product in dyes adsorption application. Dyes are known to affect human health, aquatic life and the overall ecosystem in adverse way. They need to be removed from wastewater, hence the technologies utilized for such removal are widely investigated. The review presents the different preparation techniques to produce adsorbent from PET waste. Besides that, it discusses the adsorption capacities among the PET adsorbents prepared. Based on the review, it can be concluded that activated carbon produced from PET waste has the potential to be an effective adsorbent for dye adsorption.

1. Introduction

Nowadays, many industries, such as dyestuffs, textile, paper and plastics, use dyes in order to color their products and colouring process consumes substantial volumes of water. As a result, the industries generate a considerable amount of colored wastewater (Crini, 2006). It is estimated that about 10–60 % of reactive dyes are lost during textile dyeing, producing large amounts of colored wastewater (Hessel et al., 2007). The dye-containing wastewater discharged from these industries can adversely affect the aquatic environment by impeding light penetration and, as a consequence, precluding the photosynthesis of aqueous flora (Cardoso et al., 2011). Thus, dyes should be removed in order to save the aquatic environment.

Adsorption is a common practice to remove dyes from wastewater due to its simplicity and high efficiency, as well as the availability of a wide range of adsorbents that can be applied (Reddy et al., 2012). The toxic materials, hazardous ions and dyes from industrial effluents can be removed by adsorption technique without harmful effect to the environment and human health. Among various treatment technologies, adsorption onto activated carbon has proven to be one of the most effective and reliable physicochemical treatment method (Pala and Tokat, 2002). Non-conventional low-cost adsorbents, including natural materials, biosorbents, and waste materials from industry and agriculture have been proposed by several workers. These materials could be used as sorbents for the removal of dyes from solution. Some of the reported sorbents include clay materials such as sepiolite (Santos and Boaventura, 2016) and biosorbents such as water bamboo husk (Lin et al., 2015). Sepiolite is one of the natural clay mineral that often used as the alternative low-cost adsorbent because of its unique physicochemical properties (Tunç et al., 2012). It has a fibrous structure formed by an alteration of blocks and channels that grow up in the fiber direction (Akçay, 2004). The adsorption of azo dyes including Acid Blue 193 (Bilgiç, 2008), Brilliant Yellow (Bingol et al., 2010), Basic Red 46 (Santos and Boaventura, 2008), Direct Blue 85 (Santos and Boaventura, 2008) and Remazol Red B (Uğurlu, 2009), anthraquinone dye including Acid Blue 25 (Han et al., 2014) and reactive dyes including Reactive Blue 21 (Demirbas and Nas, 2009) and Reactive Blue 221 (Alkan et al., 2007) onto sepiolite has been studied.

Different types of plastics have become a major portion of the municipal wastes (Khorram et al., 2017). One of the plastics in high demand is poly(ethylene terephthalate) (PET), which is used as packaging materials for water and beverages (Kang et al., 2017). The non-degradable nature of PET makes the waste increases over years, cause serious problems to the environments. Although PET waste can be easily collected and recycled,

Paper Received: 30/04/2019; Revised: 21/09/2019; Accepted: 04/11/2019
Please cite this article as: Mohd Din S.F., Othman N., Mohamad Z., Che Man S.H., Abd Karim K.J., Hassan A., 2020, Recycled Poly(ethylene terephthalate) as Dye Adsorbent : A Mini-Review, Chemical Engineering Transactions, 78, 367-372 DOI:10.3303/CET2078062

there is still a limited number of alternatives to maximize and diversify the use of PET waste into useful and valuable end products. Currently most of the applications of recycled PET are for construction usage such as pavement (Rahman and Wahab, 2013), concrete (Rahimi et al., 2016), and ECO bricks (Taaffe et al., 2014). Recently, Leng et al. (2019) studied on the production of a sustainable paving material through chemical recycling of PET waste into crumb rubber modified asphalt to improve the storage stability and rheological performance of crumb rubber modified asphalt. Mechanical recycling is one of the practical method available to recycle PET (Raheem et al., 2019). PET possesses good thermal properties, mechanical properties, stability and non-toxicity (Li et al., 2015). Therefore, PET as a matrix to support nanoparticles presents a novel method for using waste resources to reduce environmental burden. Abd-Aziz et al. (2019) reported on review of usage of various plastic wastes as sorbents for environmental pollutant including heavy metal, oil and dye. Based on the review PET waste is promising alternative of low cost adsorbent. Numerous approaches have been reported on reproducing a dye adsorbent from PET waste that was not included in the previous review. This review for the first time focus on PET waste in producing a promising dye adsorbent. The review (i) presents the different preparation techniques to produce adsorbent from PET waste; and ii) discusses the adsorption capacities of the PET adsorbents prepared.

2. PET waste based adsorbent preparation techniques

Various techniques were used to produce dye adsorbent from PET waste. The first step of adsorbent processing involves PET sample preparation stage, where the PET waste (in any form) was shredded into desired sizes and cleaned with distilled water before heated in ethanol solution to remove surface contamination (Khorram et al., 2017). Then, the cleaned PET was dried in the oven prior to further activation or surface treatment.In 2005, Akmil-Basar et al. (2005) studied activated carbons that were prepared from PET waste by chemical activation with sodium hydroxide (NaOH). The ratio of 1:1 recycled PET to NaOH was used. The sample was heated to the activation temperature of 800 ˚C at a rate of 10 ˚C/min under nitrogen (N_2) (100 ml/min) atmosphere and held at the temperature for 1 h. After the activation, the sample was cooled under N_2 flow and washed several times with hot distilled water until filtrate was neutral. Then, the washed sample was dried at 110 ˚C. Another activated carbon was prepared using the same technique from pine sawdust with zinc chloride ($ZnCl_2$,) chemical activation. The activation temperature and $ZnCl_2$/pine sawdust ratio was 500 ˚C and 1:1 respectively (Akmil-Başar et al., 2005). Another method to produce activated carbon was developed by Djahed et al. (2016) which utilized potassium hydroxide (KOH) and combination of the carbonation and activation processes. This technique reduced the cost of the inert gases needed in the production process. The activated carbon utilized in the study was produced through a novel method in which activation and carbonation processes were simultaneously performed and no inert gas was used in the production process. The scanning electron microscopy (SEM) micrograph showed there was 72.63 % pores on the produced activated carbon, which is categorized as mesopores carbon (Djahed et al., 2016). Thus, this adsorbent is appropriate for the adsorption of large molecules such as methylene blue molecules. Al-Mazan et al. (2006) done a study on preparing of activated carbon without the activation process as economical option of producing activated carbons. The activated carbon was prepared by pyrolysis of plasma treated PET waste from drinking bottles. The plasma treatments of PET were carried out in a microwave apparatus prior to carbonization. Square pieces (1 × 1 cm) of PET were placed in the chamber and were outgassed for 5 min to a pressure of less than 0.000266 bar. Then the gas used to produce plasma was admitted up to a pressure of 0.00133 bar and flowed for 5 min. The power supplied to produce the plasma was 200 W, and the PET samples were exposed to plasma for 4 min and 15 min. Four different gases were used as the plasma source. After the plasma treatments, the PET samples were carbonized either at 800 °C or 950 °C based on the experimental conditions. The textural characteristics of the carbon materials obtained after 4 minutes of plasma treatment were very similar to those obtained after 4 h of carbon dioxide at 800 °C activation. This method can be an alternative to avoid the burn off and high energy cost of the activation step in producing activated carbons (Almazan-Almazan et al., 2006).Mesoporous activated carbons with high surface were prepared by Lian et al. (2011) from three polymer wastes which were tire rubber, poly(vinyl chloride) (PVC) and PET. The polymer wastes were carbonized at 600 °C in N_2. The carbonized sample was then activated by ground KOH in a mass ratio of 1:2 under N_2 flow at 850 °C. The activation time was 120 min for tire rubber and 90 min for PVC and PET to obtain a 50 % burn off. The obtained solids were washed with 1 M hydrochloric acid (HCl) solution and distilled water to reduce the ash and decomposed fragments. The activated carbons then dried at 110 °C for 24 h before grounded and sieved. The adsorption/desorption characteristics of two dyes (methylene blue and methyl orange) on the activated carbons were studied as compared to commercial coal-based activated carbon (F400) (Lian et al., 2011).In 2015, Lin et al. studied the composite membrane to adsorb cationic dye using composite membrane made from PET waste and water bamboo husk particles. The chopped PET was dissolved in phenol and tetrachloroethane solvent before

mixed with water bamboo husk particles to form the composite membrane. Water-vapor-induced phase inversion technique was used to prepare the composite membrane. The surface area of composite membrane, pure PET membrane and the pristine bamboo particles is 1.7 m^2/g, 0.5 m^2/g and 1.9 m^2/g, respectively. The SEM micrograph of pure PET membrane and composite membrane showed a porous structure. The composite membrane was thicker (0.077–0.080 mm) and heavier (30.5 mg) as compared to the pure PET membrane with 0.044–0.050 mm of thickness and 15.0 mg of weight for 25 mm disc membrane (Lin et al., 2015).A green and sustainable method of producing graphene from PET waste was studied by El Essawy et al. (2017). Two grams of PET waste was added into 50 mL autoclave reactor and heated to 800 ˚C at a rate of 8 ˚C/min in an electric furnace. The temperature was maintained for 1 hour, then left to cool overnight. The SEM micrograph exhibited a porosity fibres network morphology. The transmission electron microscope (TEM) images showed a few-layers of graphene sheets and graphite deposits. Energy dispersive X-ray (EDX) was used to analyse the elemental composition of graphene which consisted of high amount carbon and traces of oxygen. Powder X-ray diffraction (XRD) and Raman spectroscopy also were used to characterise and prove the production of graphene. The synthesized graphene was thermally stable up 485 °C with the ash content of 2.46 %. Graphene is a form of carbon, with extremely high relative surface area compared to activated carbon. It is lighter for being a material made up of one single atomic layer and it is ecologically friendly (El Essawy et al., 2017). A study on PET composite cation exchanger is another way of recycling PET to form a dye adsorbent. Khan et al. (2013) prepared an unsaturated polyester Ce(IV) phosphate (USPECe(IV)P) composite cation exchanger. The PET waste from soft drink bottles were converted through glycolysis in the presence of a catalyst. One-liter three-necked round bottomed flask (reactor) was used for all glycolysis experiments. The reactor with a thermometer, magnetic stirrer and a reflux condenser was heated up to 100 ˚C and held at that temperature for at least 5 min. The ratio of diethylene glycol to PET used in the glycolysis experiment was 4:1 by weight where the weight of diethylene glycol and PET flakes were 84 g and 21 g, respectively. Glycolysis temperature was set at 190 ˚C and glycolysis time was 2 h with 0.25 % catalyst by weight of PET. Various samples of (USPECe(IV)P) were prepared by mixing different volume ratios of unsaturated polyester in an inorganic Ce(IV) phosphate gel. The adsorbent and dye interaction was confirmed by Fourier-transform infrared spectroscopy (FTIR) (Khan et al., 2013).

3. Adsorption capacity of PET waste based adsorbent

An effective adsorbent for dye adsorption is defined as i) efficient for removal a wide variety of dyes, ii) high capacity and rate of adsorption, iii) high selectivity for different concentrations; and iv) tolerant of a wide range of wastewater parameters (Crini, 2006). The adsorption capacities are also depending on the condition of the waste water (such as concentration, contact time, pH and temperature). The adsorption kinetics and isotherm of the adsorbent produced by PET waste fitted well with the Langmuir and Freundlich isotherm models. Table 1 presents a summary of the adsorption capacity of PET waste based adsorbent prepared from various techniques. The activated carbons prepared by NaOH chemical activation showed that the optimum pH for removal of malachite green dye in solution was in the range of 6-10. The surface area and total volume of the produced activated carbons were 410 m^2/g and 0.86 cm^3/g for PET and 1390 m^2/g and 0.77 cm^3/g for pine sawdust, respectively. Both adsorbents were very effective in all the pH values at low initial dye concentration. However, the dye adsorption was affected by pH at high initial dye concentration. Adsorption percentage increased with increasing initial dye concentration then started to decrease while initial dye concentration increases beyond 500 ppm. For the activated carbon from PET, the equilibrium time for adsorption of 450 mg/L malachite green dye was 90 min while for the activated carbon from pine sawdust, the equilibrium is achieved in about 120 min at malachite green concentration of 100 mg/L (Akmil-Başar et al., 2005).The activated carbon from PET waste produced using inexpensive and efficient process by Djahed and co-workers (2016) yielded an activated carbon with surface area and the total volume of 353 m^2/g and 0.288 cm^3/g, respectively. The maximum adsorption capacities of methylene blue dye was 404 mg/g (Djahed et al., 2016). The removal efficiency decreased from 80 % to 60 % for the range of initial concentration studied from 80 mg/L to 120 mg/L. The equilibrium time for the adsorption was around 20 min for all the initial dye concentration studied with an optimum pH of 10.The activated carbons from PET waste prepared by Lian et al. (2011) exhibited the largest surface area of 2831 m^2/g and pore volume of 1.68 cm^3/g compared to PVC and tire rubber. The activated carbons derived from PVC exhibited relatively lower surface area of 2666 m^2/g but more narrowed pore size distribution of (2–3) x 10^{-9} m. The complex composition and high ash content of tire rubber particles resulted in activated carbons product with low surface area (398.5 m^2/g) and heterogeneous pore width. The high oxygen content of PET-derived activated carbons significantly affected its adsorption to methylene blue and iodine. Due to the remarkable surface area and highly mesoporous structures, activated carbons based on both PET and PVC exhibited much higher adsorption capacities than that of tire rubber and commercial coal-based activated carbon (F400).

Table 1: Adsorption capacity of different dye adsorbent

Type of Dye	Precursor	Preparation/Activation Method	Dye Adsorption Capacity (mg/g)	S_{BET} (m^2/g)
Malachite green	PET waste	Activated carbon with chemical	169	410
	Pine-sawdust	activation using NaOH at 800 ˚C	370	1,390
		(Akmil-basar et al., 2005)		
Methylene blue	PET bottle waste	Activated carbon with chemical activation using KOH at 500 ˚C without inert gas (Djahed et al., 2015)	404	353
Methylene blue	PET waste	Activated carbon with chemical	210	2,831
	PVC waste	activation using KOH at 850 ˚C (Lian	240	2,666
	Tire rubber waste	et al., 2011)	50	398
	F400	-	100	1,003
Iodine	PET waste	Activated carbon with chemical	3,240	2,831
	PVC waste	activation using KOH at 850 ˚C (Lian	2,620	2,666
	Tire rubber waste	et al., 2011)	510	398
	F400	-	1,280	1,003
Methylene blue	Water bamboo husk particles	-	1,020	1.9
	Composite membrane	Water-vapor-induced phase Inversion (Lin et al., 2015)	307	1.7
Methyl violet 2B	Water bamboo husk particles	-	1,085	1.9
	Composite membrane	Water-vapor-induced phase Inversion (Lin et al., 2015)	326	1.7
Methylene blue	PET bottle waste	PET synthesized graphene (El	743	721
Acid blue		Essawy et al., 2017)	710	721
Malachite green	Soft drink waste PET bottle	Glycolysis of PET with presence of catalyst (Khan et al., 2013)	137	-

The carbon formation during thermal decomposition of polymer is a competition between a chain scission to generate volatile species and a condensation process to form carbon structure (Yuan et al., 2008). PET is an aromatic polymer with rich in benzene rings that located in the main chain. Due to the structure, PET is easier to convert into polyhexagonal carbon layers compared with PVC which is aliphatic polymers. The aromatic makes condensation much easier, thus leading to almost simultaneous occurrence of chain scission and condensation. However for PVC and tire rubber, the condensation and aromatization mainly occur in the second and/or third weight loss step in thermal gravimetric curve (San Miguel et al., 2006).The graphene synthesized from PET waste showed excellent adsorption of methylene blue and acid blue 25 with maximum adsorption capacity of 761 mg/g and 642 mg/g, respectively. High surface area of 721 m^2/g allowed maximum adsorption of dye. Thermodynamic parameters demonstrated that adsorption of dye is spontaneous and endothermic in nature. PET waste has a great potential to be transformed into valuable carbon-based nanomaterials due to high content of carbon and negligible amounts of mineral impurities in its chemical constitution (El Essawy et al., 2017).

The performance of porous composite membrane produced from PET waste and bamboo was compared to performance of water bamboo husk particle to adsorb methylene blue and methyl violet 2B done by Lin et al. (2015). The pure PET membrane showed negligible dye adsorption, however the saturated capacities of methylene blue and methyl violet 2B adsorption of composite membrane improved with addition of 30 wt.% water bamboo husk particles. Interaction between the composite membrane and the dyes was attributed by the ion-exchange capacity of the water bamboo husk particles. The composite membrane maintained a good dye removal performance, with dye removal of above 90 % after three consecutive cycles of adsorption and desorption (Lin et al., 2015).

Efficient desorption process is important for an economical adsorption. Desorption process recover the adsorbates and regenerate the adsorbents to be reuse. The mechanism between the adsorbate and adsorbent can also be determined from the desorption studies (Salleh et al., 2011). A mixture of salts and organic solvent needed for combination of various binding mechanisms. Desorption studied by Lin et al. (2015) proved that 1 M potassium thiocyanate (KSCN) in 80 % methanol was an effective desorption solution

for removing the adsorbed cationic dyes from the composite membrane. The composite cation exchanger which prepared to adsorb malachite green was found to be maximum adsorption of 98 % (1.01 mg/g) at pH 8 with equilibrium time of 30-35 min for initial concentration of 50-100 ppm. The extent of removal of malachite green was found to be dependent on adsorbent dose, temperature and time. The process was endothermic and spontaneous process. Kinetic studies showed better applicability of an intraparticle diffusion kinetic model (Khan et al., 2013).

4. Conclusions

In this review, a wide range of techniques used to produce dye adsorbent from PET waste have been presented. One of the most common practice adsorbents is activated carbon. Activated carbons have been the easiest and economical way to produce dye adsorbent with good adsorption capacity with proper chemical and physical treatment. Adsorbent from PET waste has a great potential as an alternative way to replace the conventional activated carbon. Unfortunately, efficient application of particulate adsorbents like activated carbons is still remains serious challenges. For an example, it is difficult to separate and aggregation of small into large particles and loss of activity, thereby leading to short service life, the limited actual re-usage count and unexpected re-adsorption capacity as an adsorbent. Therefore, a suitable design of support materials is needed to widen the potential of PET waste with good adsorption performance and making the process practical for industrial application.

Acknowledgments

The authors would like to thank the Ministry of Higher Education (MOHE), Malaysia under FRGS/1/2019/TK05/UTM/02/15 grant and Universiti Teknologi Malaysia under TDR grant (06G40).

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