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Study on Effect of Toluene-Acid Treatments of Recycled Carbon Black from Waste Tyres: Physico-Chemical Analyses and Adsorption Performance

(Kajian Kesan Rawatan Asid Toluena Karbon Hitam Kitar Semula daripada Bahan Buangan Tayar: Analisis Fiziko-Kimia dan Prestasi Penjerapan)

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ABSTRACT

Recycled carbon black (rCB) produced by pyrolysis has a low value because it contains high levels of impurities, such as sulfur, nitrogen, and oxygen. Various treatments have been proposed using chemicals to purify and improve the properties of rCB. In this study, rCB was treated with toluene (rCB-T), followed by subsequent treatment using acids HCl (rCB-T-HCl), HNO_3 (rCB-T-HNO_3), and HCl-HNO_3 (rCB-T-HCl-HNO_3). The treated rCB samples were characterized using CHNS analyser, scanning electron microscope, BET analyser, zeta potential, Fourier transform infrared spectroscopy, and Raman spectroscopy. The adsorption of methylene blue dye onto the rCB samples was also investigated to study the effectiveness of the treatments. Treatment with toluene alone was insufficient to increase the carbon content and surface area of the rCB. Subsequent treatment of rCB with acids, especially HNO_3, significantly increases the carbon content, surface area, surface functional groups, and surface charge of the rCB. This results in an increased adsorption capacity of the rCB, from 6.04 mg/g to 46.51 mg/g for the rCB-HNO_3 and 54.80 mg/g for the rCB-T-HCl-HNO_3.

Keywords: Adsorption; BET; carbon black; toluene; zeta potential

ABSTRAK

Karbon hitam (rCB) kitar semula yang dihasilkan oleh pirolisis mempunyai nilai yang rendah kerana ia mengandungi tahap kekotoran yang tinggi, seperti sulfur, nitrogen dan oksigen. Pelbagai rawatan telah dicadangkan menggunakan bahan kimia untuk membersih dan menambahbaik sifat rCB. Dalam kajian ini, rCB telah dirawat dengan toluena (rCB-T), diikuti dengan rawatan seterusnya menggunakan asid HCl (rCB-T-HCl), HNO_3 (rCB-T-HNO_3) dan HCl-HNO3 (rCB-T-HCl-HNO_3). Sampel rCB yang dirawat telah dicirikan menggunakan penganalisis CHNS, pengimbasan mikroskop elektron, penganalisis BET, potensi zeta, spektroskopi inframerah transformasi Fourier dan spektroskopi Raman. Penjerapan pewarna biru metilena pada sampel rCB juga dikaji untuk melihat keberkesanan rawatan. Rawatan dengan toluena sahaja tidak mencukupi untuk meningkatkan kandungan karbon dan luas permukaan rCB. Rawatan seterusnya rCB dengan asid, terutamanya HNO_3 meningkatkan kandungan karbon, luas permukaan, kumpulan fungsi permukaan dan cas permukaan rCB dengan ketara. Ini menghasilkan peningkatan kapasiti penjerapan rCB, daripada 6.04 mg/g kepada 46.51 mg/g untuk rCB-HNO_3 dan 54.80 mg/g untuk rCB-T-HCl-HNO3.

Kata kunci: BET; karbon hitam; keupayaan zeta; penjerapan; toluena

Introduction

The increasing production of rubber tyres for automobiles is leading to a growing amount of difficult-to-dispose-of waste. According to the World Business Council for Sustainable Development, the number of waste tyres produced each year, which can be attributed to the increasing number of vehicles on the road, as well as the longer lifespan of tyres (Mountjoy, Hasthanayake & Freeman 2015). Waste tyres are tyres that are no longer suitable for their original purpose or for any other purpose. Waste tyres have significant potential for energy recovery and the reduction of CO_2 emissions. They have a higher calorific value than coal, and the rubber in tyres is a valuable raw material that can be recycled (Juma et al. 2007).

Generally, tyre content is made up of 60-65 wt% styrene butadiene rubber (SBR) and nitrile butadiene rubber (NBR), 25-35 wt% commercial carbon black (N330 & N220), and other additives, such as accelerators and fillers (Costa et al. 2022). In the manufacturing process of tires, an irreversible reaction between the elastomer, sulfur, and other chemicals forms a three-dimensional chemical network through vulcanization (Martínez et al. 2013). This network is very resistant to breaking down, so it can be difficult to separate the rubber from other materials in the tyre.

There are several ways to dispose waste tyres, including pyrolysis, incineration, reclamation, landfilling, and gasification (Dabic-Miletic, Simic & Karagoz 2021; Torretta et al. 2015). Of these methods, pyrolysis is the most environmentally friendly option, as it minimizes waste and allows for energy recovery. Pyrolysis is a thermochemical process that breaks down chemical bonds in the absence of oxygen at high temperatures (Torretta et al. 2015). This process can be used to produce a variety of products, including fuel oil, gas, carbon black, steel wire and ash (Costa et al. 2022; Martínez et al. 2013). Approximately, 35% of recycle carbon black (rCB) can be generated from the pyrolysis pf waste tyres. However, compared to commercial CB, rCB has a lower carbon content and a higher ash and inorganic content, such as sulphur. Moreover, some metals can be found in used tyres; most of these metals are concentrated in the solid residue (Galvagno et al. 2002; Roy et al. 1994). Besides, commercial CB has better particle size distribution and lesser aggregation due to its surface modification for better dispersion in rubber during the processing of tyre (Galli 1982). Therefore, various efforts are being made to improve its properties, such as activation by CO_2 at elevated temperature (Choi et al. 2014) and demineralization by acids (Dong et al. 2017).

There are studies reported on the treatment of rCB using inorganic acids, such as hydrochloric acid, nitric acid, and sulfuric acid to remove residue rubber, volatile inorganic materials, and heavy metal free (Shah et al. 2006). Studies found that acid-based method using sulfuric acid (H_2SO4) and sodium hydroxide (NaOH) can reduce ash and heavy metal contents of rCB (Chaala, Darmstadt & Roy 1996). Besides, acid-alkaline treatment of rCB using nitric acid (HNO_3) and NaOH can effectively remove heavy metals, especially vanadium and nickel (Dong et al. 2017). Later, acid-alkaline treatment of rCB using hydrochloric acid (HCl) and NaOH was proven to be effective in producing treated rCB with low impurity content (Martínez et al. 2019). Besides, the capability of removing heavy metals from rCB, HCl and HNO_3 were also proven able to oxidize the rCB and introduce oxygenated surface functional groups onto the rCB (Iraola-Arregui, Van Der Gryp & Görgens 2018; Sugatri et al. 2017).

In this study, rCB was treated with toluene, followed by subsequent acid treatment using HCl and HNO_3. The effects of the two acids, individually and in combination, were investigated in term elementary composition (CHNS), morphology, particle size, surface area, and Zeta potential. The treated rCBs was also used to study their adsorption performance towards methylene blue adsorption as an indicator of the oxidation of the rCB by the acids.

Materials and Methods

Materials

The pyrolytic recycled carbon black (rCB) was provided by Sun Rubber Industry Sdn. Bhd. Toluene (EMSURE ACS), hydrochloric acid (HCl, 37% EMSURE, ACS), nitric acid (HNO_3,65% EMSURE, ACS) were purchase from Merck Millipore (Burlington, MA, USA). Methylene blue (ACS reagent) was purchased from Sigma Aldrich (Saint Louis, MO, USA). PTFE 0.2 μm filter paper and Nylon 0.2 μm filter paper. Ethanol (Absolute for analysis EMSURE, ACS).

Treatments of rCB

Firstly, the rCB was washed by toluene to remove residues (oils, greases, and rubber resin). Briefly, 3 g of rCB powder was sonicated in 300 mL of toluene for 1-min and stirred at 500 rpm at room temperature overnight. The mixture was then filtered through PTFE paper with 0.22 μm pore size, rinsed with an ethanol, and dried in a convection oven (AP60, Froilabo, France) at 80 °C for 24 h. The produced rCB sample is denoted as rCB-T. The rCB-T powder was subjected to the HCl and HNO_3 treatments as described in Figure 1. Briefly, 2 g of rCB-T powder was added into the acid solution. The mixture was then subjected to a 1-minute sonication process and continued to be stirred at 500 rpm at a desired temperature using a magnetic stirrer for a specific time. Details on the acid treatment conditions are listed in Table 1. Upon the completion of the treatment, the treated rCB powder was separated from the liquid solution by filtering the mixture using the nylon paper with 0.22 µm pore size. The collected rCB was rinsed with distilled water for several times until it achieved pH 6, followed by drying in the oven at 60 °C for 24 h. The produced rCB-T sample is denoted accordingly as shown in Table 1. In this study, the effect of different acids (HCl and HNO_3), and subsequent treatment using HNO_3 after HCl was investigated.

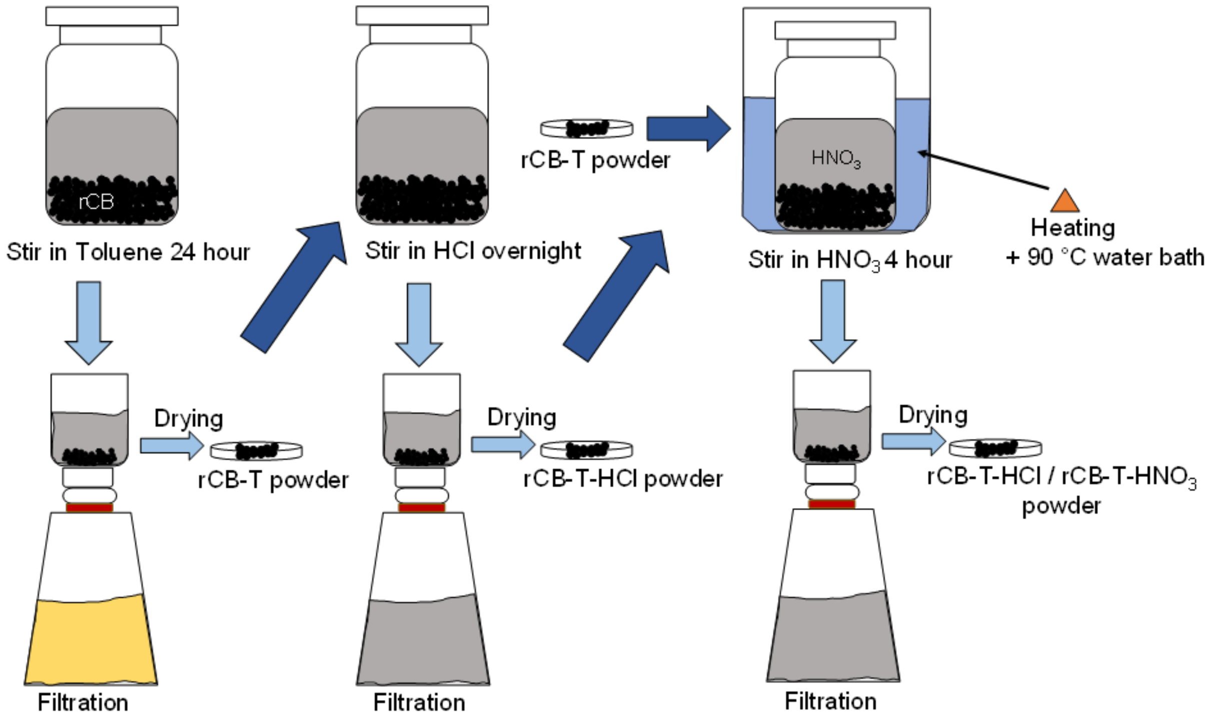

FIGURE 1. Treatment procedures of rCB with toluene, hydrochloric acid, and nitric acid

TABLE 1. Treatment parameters for rCB using toluene and acids

Sample	Reagent	Parameter
rCB-T	Toluene	24 h (Room Temperature)
rCB-T-HCl	10 v/v % HCl	Overnight (Room temperature)
rCB-T-HCl-HNO_3	3 v/v % HNO_3	4 h 90 °C (water bath)
rCB-T-HNO_3	3 v/v % HNO_3	4 h 90 °C (water bath)

CHARACTERIZATIONS OF rCB SAMPLES

The physical and chemical properties of all treated rCB samples were analyzed using various instruments. Elementary (CHNS) analysis of the treated rCB sample were conducted using (FlashSmart-CHNS, Thermo Scientific). Morphology of the rCB samples was analyzed using a field emission scanning electron microscope (FESEM, FEI Quanta 400F) of 10,000× magnification and the particle size of each rCB sample was measured at 50,000× magnification. BET surface area and pore characteristics of the rCB were determined by multi-point N_2 adsorption at 250 °C for 4 h using a Micromeritics ASAP-2020 apparatus. Zeta potential of the treated-rCB samples was determined by using a Zetasizer (Malvern, Nano ZS). The measurements were conducted at pH 5.0 using NaCl as electrolyte. The chemical functional groups of the rCB samples were analysed using an Attenuated Total Reflectance (ATR) mode infrared spectrometer (Bruker Alpha, Ettlingen). Raman analysis was performed using a Raman micro-spectrometer (µRaman-Ci, Technospex). The instrument was configured to use a 532-nm laser, a laser power of 5 mW, and an acquisition time of 5 seconds. The I_D and I_G ratio of the rCB samples was estimated to determine the effect of rCB treatment on the graphitic and amorphous structure.

ADSORPTION OF METHYLENE BLUE BY THE TREATED RCB

Methylene blue (MB) was used as adsorbate and dye indicator to represent surface functionalities of the treated rCB. The obtained results were reported in terms of adsorption kinetics and maximum capacity. A stock solution of MB (50 mg/L) was prepared. The absorbance of MB was measured at 660 nm using a UV-vis spectrophotometer (Jenway 7315). A standard calibration curve of MB was constructed using a series of MB solution with different concentrations according to Beer–Lambert's law. The linear correlation between MB concentration and absorbance unit was established using standard solutions prepared by multiple dilution of the original MB solution.

The adsorption capacity (q_t) of the rCB samples was calculated as described by Equation (1) (Sajab et al. 2023):

$$q_t = \frac{C_o - C_t}{W} \times V \tag{1}$$

where C_o is the initial concentration of MB; C_t is the concentration at time t; V is the volume (L) of MB solution used; and W is the sample weight (g) of rCB.

A 0.05 g sample powder was stirred with 50 mL of 50 mg/L MB solution at 25 °C for 40 min. Five milliliters of the solution were collected at 5, 10, 15, 20, 25, 30, 35, and 40 min, filtered using a syringe and nylon pipe, and the absorbance was measured at 660 nm using a UV-vis spectrophotometer.

RESULTS AND DISCUSSION

PHYSICAL AND CHEMICAL PROPERTIES OF TREATED rCB SAMPLES

The elementary analysis of the rCB before and after treatment is presented in Table 2. After toluene treatment, the carbon content of rCB decreased from 72.0% to 68.8%. A strong odor and yellowish liquid were also observed after toluene treatment. These observations suggest that toluene treatment removed contaminants, such as organic residues, that were still present in rCB (Ren et al. 2011). Subsequent HCl treatment of rCB-T decreased the carbon content from 68.8% to 60.8% and decreased the sulfur content from 2.6% to 1.0%. However, treatment of both rCB-T-HCl and rCB-T with HNO_3 increased the carbon content to 89.0% and 73.7%, respectively. The increase in carbon content indicates the removal of non-carbon phases with acid (Shah et al. 2006). The sulfur content of both samples decreased from 2.6% to 1.0% and 2.6% to 0.8%, respectively, while the nitrogen content increased to 0.4% after HNO_3 treatment. The decrease in sulfur content is due to the oxidation of sulfur, which makes it more soluble in water. The increase in nitrogen content is due to the introduction of nitrogen functional groups during the oxidation process.

TABLE 2. Elementary (CHNS) analysis of rCB samples before and after treatments

Sample	Element (%)			
	Carbon	Hydrogen	Sulfur	Nitrogen
rCB	72.0	0.9	2.6	0.0
rCB-T	68.8	0.8	2.6	0.0
rCB-T-HCl	60.8	0.8	1.0	0.0
rCB-T-HNO_3	73.7	0.8	0.8	0.4
rCB-T-HCl-HNO_3	89.0	0.8	0.8	0.4

The results of BET analyses on the rCB samples are shown in Table 3. Treatment with toluene increased the surface area of the sample but slightly decreased the total pore volume and average pore size. The toluene treatment created new surface areas on the rCB by the removal of contaminants on the rCB. This is consistent with the CHNS results, which showed that the carbon content did not change much after the toluene treatment. Treatment with HCl and HNO_3 resulted in a significant increase, which is approximately 8.6% in the surface area of the rCB sample. This is likely due to the oxidation of the rCB surface by the acids, which created new surface areas. However, treatment with HNO_3 significantly reduced the pore volume of the rCB. This can be attributed to the collapse of the pores due to the erosive effect by the acid (Allwar, Hartati & Fatimah 2017; Selbes et al. 2015; Yang et al. 2021). All rCB samples had lower surface areas than commercial high abrasion furnace (HAF) grade carbon black N330, which is widely used in rubber products (Mikulova et al. 2013).

Figure 2 shows FESEM images of the rCB samples. The rCB, rCB-T, and rCB-T-HCl samples are more aggregated than the rCB-T-HCl-HNO_3 and rCB-T-HNO_3 samples, which are in looser aggregates. This is likely due to the removal of contaminants and rubber residues by HNO_3, as discussed in the CHNS results (Jiang et al. 2022).

TABLE 3. The BET analysis of rCB samples before and after treatments

Sample	BET surface area (m^2/g)	Total pore volume (cm^3/g)	Average pore size (Å)
rCB	55.10	0.39	283.80
rCB-T	63.80	0.32	198.40
rCB-T-HCl	69.30	0.33	188.80
rCB-T-HNO_3	69.40	0.14	80.30
rCB-T-HCl-HNO_3	68.40	0.36	207.40
Commercial CB (N330)	73.50	0.58	317.10

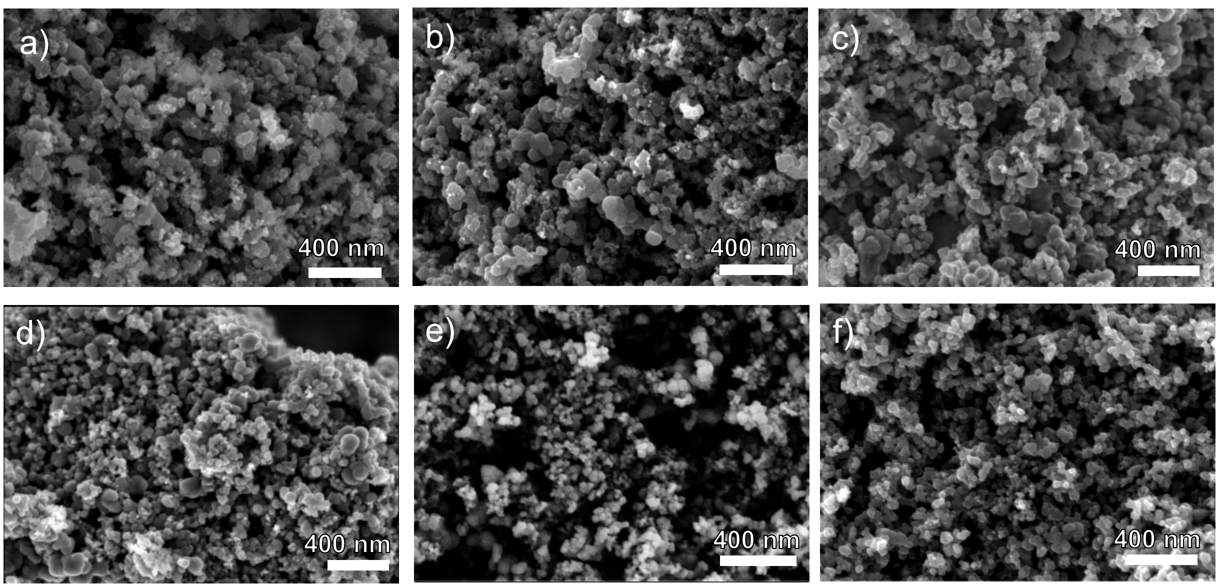

FIGURE 2. FESEM images of the rCB samples before and after treatment: a) rCB, b) rCB-T, c) rCB-T-HCl, d) rCB-T-HCl-HNO_3, e) rCB-T-HNO_3, and f) N330

Figure 3(a) shows the FTIR spectra of the rCB samples. All rCB samples show typical absorption bands at 790 cm^{-1}, 2671 cm^{-1}, and 2888 cm^{-1}, which can be ascribed to C-H bending, aliphatic C-H stretching, and aliphatic CH_2 stretching, respectively, (Gomez-Serrano et al. 1996; Zhang et al. 2018). The absorbance band at 1079 cm^{-1} can be assigned to S=O stretching sulfoxide functional group. As can be seen from Figure 4, the treatment with toluene had little effect on the intensity of this band, indicating that toluene treatment did not remove much of the rubber residues in the rCB that contain sulfur as a vulcanizing agent. However, subsequent treatments using acids (HCl and HNO_3) after toluene had significantly decreased the intensity of the band, suggesting the removal of rubber residues from the rCB samples. This is consistent with the sulfur content of the rCB samples shown in Table 2. An absorption band around 2068 cm^{-1} is observed for the rCB-T-HNO_3, indicating the presence of strong C-H bending of aromatic compounds (Chen & Wu 2004). The presence of absorption bands at 1786 cm^{-1} and 1688 cm^{-1} in the FTIR spectra of rCB-T-HNO_3 and rCB-T-HCl-HNO_3 indicates the introduction of the functional group of C=O stretching secondary amide. The absorption bands 1603 cm^{-1} and 1511 cm^{-1} further proven the introduction of strong stretching ketone and N-O groups (Chen & Wu 2004). Treatment of rCB using HNO_3 allowed the introduction of important acidic functional groups, such as, lactones, phenols, ketones, and carboxylic acids (Cardona-Uribe, Betancur & Martínez 2021).

Figure 3(b) shows the Raman spectra of the treated rCB samples. The presence of D band and G band at 1355 cm^{-1} and 1590 cm^{-1}, respectively, indicates the amorphous and graphitic structures of the rCB sample. Continuous treatment of rCB with toluene, HCl, and HNO_3 remain unchanged the I_D/I_G ratio of the samples indicates that there were no changes in the graphitic structure and no defects were introduced after treatment. However, the removal of the sample's graphitic structure caused by the treatment of rCB-T with HNO_3 further supported the BET results' findings that the pore volume and pore size had decreased (Table 3).

Figure 4 shows the zeta potential values of the rCB samples. The toluene treatment increased the zeta potential of rCB from -19.90 to -25.50. However, the HCl treatment of rCB-T only increased the zeta potential to -20.5, suggesting that HCl treatment did not introduce much functional group to the rCB-T sample. In contrast, both samples treated with HNO_3, rCB-T-HCl-HNO3 and rCB-T-HNO_3, showed a significant enhancement in the zeta potential to -31.80 and -33.80, respectively. This is due to the introduction of functional groups (such as ketones and carboxylic acids) on the surface, as proven by the FTIR results, which increases the number of negatively charged sites on the carbon surface (Park et al. 2007).

ADSORPTION KINETICS AND MAXIMUM ADSORPTION CAPACITY

Figure 5 shows the adsorption performance of the rCB samples. The experiments were conducted up to 40 min to determine the maximum adsorption capacity of the rCB samples. As can be seen in Figure 5, treatment with toluene (rCB-T) increased the maximum adsorption capacity from 6.04 to 9.40 mg/g, while treatment with HCl decreased the adsorption capacity to 3.02 mg/g. In addition, treatments with acids on the rCB-T (rCB-HNO_3 and rCB-T-HCl-HNO_3) significantly increased the adsorption capacity to 46.51 mg/g and 54.80 mg/g, respectively. These observations are consistent with the BET surface area and zeta potential results shown in Table 3 and Figure 4, respectively.

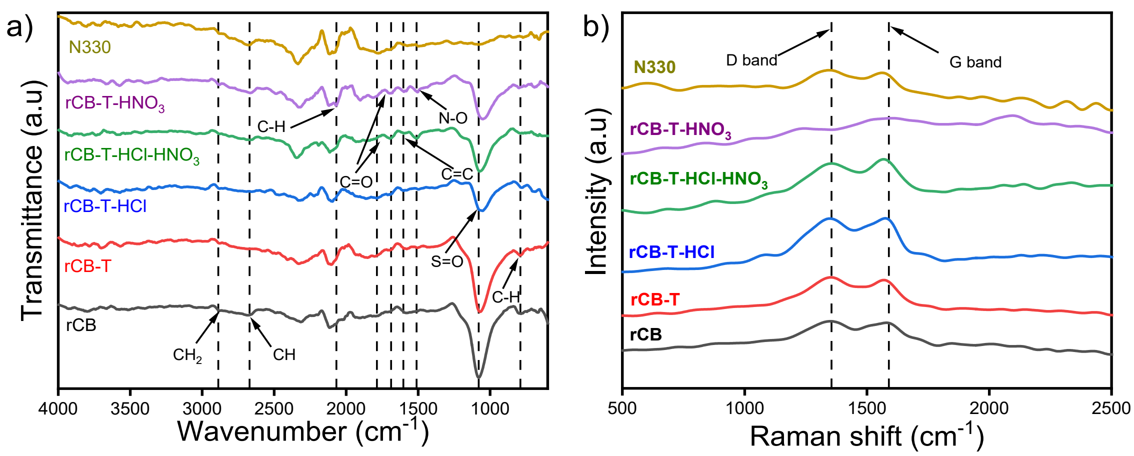

FIGURE 3. a) FTIR and b) Raman spectra of the rCB, rCB-T-rCB-T-HCl, rCB-T-HCl-HNO_3, rCB-T-HNO_3 and N330

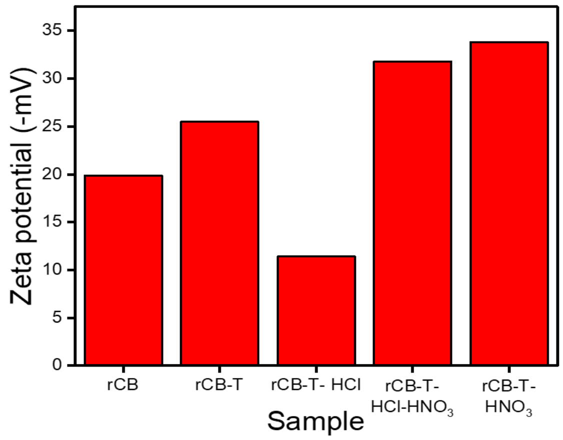

FIGURE 4. Zeta potential of the rCB samples measured at pH 5

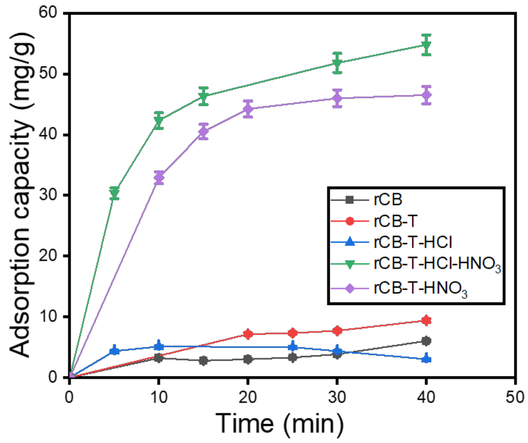

FIGURE 5. Maximum capacity adsorption of rCB treated with MB after 40 min

CONCLUSION

This study proposes a feasible method for treating and purifying rCB using toluene, HCl, and HNO_3. Toluene treatment is effective in removing organic contaminants, such as oils and greases. However, subsequent treatment of rCB-T with HCl is ineffective in purifying the rCB sample. HNO_3 treatment is found to be effective in increasing the carbon content, surface area, and zeta potential of the rCB due to the removal of impurities and rubber residue. This results in a greater adsorption capacity towards MB dye. Overall, the results of this study suggest that the treatment of toluene-treated rCB with HCl and HNO_3 is an effective way to improve the physical and chemical properties of rCB. The treated rCB can be potentially used as adsorbent for the purification of wastewater.

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