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New carbon adsorbent from polymer waste for effective removal of mercury from water

Ivanka G. Stoycheva, Boyko G. Tsyntsarki*, Bilyana N. Petrova,
Temenuzhka K. Budinova, Nartzislav V. Petrov

Laboratory Chemistry of Solid Fuels, Institute of Organic Chemistry with Centre of Phytochemistry, Bulgarian Academy of Sciences, Acad. G. Bonchev, BL. 9, Sofia 1113, Bulgaria, Tel. +359 2 9606145; Fax: +359 870 02 25; emails: racheva89@abv.bg (I.G. Stoycheva), boykotsyntsarski@abv.bg (B.G. Tsyntsarki), bnp@abv.bg (B.N. Petrova), goriva@orgchm.bas.bg (T.K. Budinova), nartzi@abv.bg (N.V. Petrov)

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ABSTRACT

Adsorption of mercury ions on activated carbon, prepared from polymer waste product, polyolefin wax (POW), was studied. The structure and surface properties of the sample are characterized by different methods—N_2 adsorption, IR spectroscopy, determination of surface oxygen groups. Adsorption studies of mercury(II) from aqueous solution on synthesized activated carbon are investigated under various conditions—agitation time, metal ion concentration, adsorbent quantity, pH. It was established that the adsorption follows both Langmuir and Freundlich isotherms. The activated carbon obtained from POW demonstrates high adsorption capacity toward mercury ions (196.1 mg/g). The results show that Hg^{2+} removal from water solutions increases with the increase of pH from 2 to 5, and slightly decreases at pH > 6.

Keywords: Polymer waste; Activated carbon; Adsorption; Mercury

1. Introduction

Water pollution is one of the most challenging problems nowadays. In the last years, the scientists all over the world continue their efforts to solve water management problems, caused by industry, transport, energy, and municipal wastes. The pollution effects cause under pricing and overuse of energy, water, etc. The working pollution control installations rarely operate at full efficiency, due to poor maintenance and operating conditions. Industry generates a large volume of effluents containing hazardous species. The heavy metals are among the most toxic species present in industrial effluents, whereas Hg is considered to be one of the most toxic metals. Mercury is the only common metal liquid at ambient temperature. Once Hg enters the food chain, large accumulation of mercury compounds was detected in humans and animals. The toxicity of Hg depends strongly on its oxidation state.

Hg has adverse effects on the central nervous system and bones, pulmonary and kidney functions, and it damages cell chromosomes [1–4]. The limit for Hg(II) in surface waters is 10 $\mu g/dm^3$, and in drinking water—1 $\mu g/cm^3$ [5]. In humans, mercury compounds can bind and deactivate recycling processes of amino acids, proteins, and certain enzyme groups, resulting in several types of ailments that in many cases may be fatal. It is also well known that mercury is present in

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*Corresponding author.

trace quantities in coals [6,7], which after burning of coal in power plants and heat generators is discharged in streams rain water. Methyl mercury, which is the most toxic of all the mercury compounds, is also formed under microbial degradation of elemental mercury dissolved in oxygenated waters and settled at the bottom of the lakes and streams and accumulated in fish. Thus, it is essential that potable water be free of any dissolved mercury salts. Other major sources of mercury pollution in the aquatic environment are industries such as chloralkali, pulp paper, paint, rubber processing, oil refining, electrical, and fertilizer productions [1,5]. Numerous methods are available for removal of mercury, including membrane separation [8,9], chemical precipitation [10,11], coagulation [12], photodegradation [13], reverse osmosis [14], biodegradation [15–17]. However, these methods are associated with high operational cost and incomplete removal, and they require large amounts of energy and chemicals.

The adsorption technique is generally considered to be a promising method and it has been successfully applied for removal of mercury from water solution [5,18,41]. The attention of the researchers is focused especially on various novel and cheap adsorbents, which would be able to remove the pollutants from contaminated wastewater.

Activated carbons are widely used as adsorbents in technologies related with pollution abatement, pharmaceutical and food industries, due to their highly porous structure and large adsorption capacity. In the last years, many reports have appeared, dedicated on the preparation of activated carbons from various cheap and alternative precursors—coal, agricultural by-products, and other biomass materials, polymer materials, etc. [5,18–45]. Namasivayam [28,32] has recently reviewed different sources of biomass for producing activated carbon, suitable for removal of mercury. Many reports have appeared dealing with preparation of activated carbons derived from rice husks, coconut shell, and peanut hull carbon, and they have been successfully employed for the removal of mercury from aqueous solution. Other authors applied activated carbon obtained from antibiotic waste for the removal of mercury. The results are very promising [31].

In last years, the using of activated carbons, obtained from other waste materials has been extended [24,26,29,30,33–35,37–41], but till now the application of different polymers wastes is not developed, independently that they are obtained at various polymer technologies in a big amount. Polymer wastes are very important raw materials for production of activated carbons due to their availability, and often with low price as by-product waste materials. The thermochemical conversion of polymer wastes is a promising way to produce energy as well as activated carbons with good adsorption properties and insignificant content of mineral matter.

In this paper, we will focus on the investigation on the adsorptive properties toward mercury(II) of novel low-cost activated carbon, synthesized by pyrolysis in the presence of water vapor from polymer waste—polyolefin wax (POW), a by-product from production of polyethylene—which have not found suitable application till now. This work is intended to bring environmental contribution, connected not only with Hg removal from water, but also with valorization of polymer wastes by converting them to valuable materials like adsorbents.

2. Experimental

2.1. Adsorbent preparation

The POW sample (waste product of polyethylene production at low pressure—from Burgas petroleum plant, Bulgaria) has melting point around 115˚C, and average molecular mass of 1,100. When heating in air at 360˚C it decomposes (without solid residue) to low-molecular products, which evaporate.

Two hundred grams of POW was heated up to 115˚C until melting 98%. H_2SO_4 was added by drops with continuous stirring, and the mixture was heated up to 160˚C. The obtained solid product was washed with water, dried at 150˚C, and carbonized at 600˚C.

POW carbonizate was subjected to water steam activation at 800˚C for 60 min. The obtained solid product is denoted as POW-AC.

2.2. Surface measurements

Textural characterization was carried out by measuring N_2 adsorption isotherms at −196˚C using Micrometrics ASAP 2010 M porosimeter. Prior to the adsorption measurements, the samples were outgassed under vacuum at 300˚C overnight, to remove any adsorbed moisture and gases. The isotherms were used to calculate specific surface area S_{BET} and total pore volume V_t. The pore size distribution, micropore and mesopore volumes were obtained by applying DFT model to N_2 adsorption data, assuming a slit-shaped pore geometry [46].

The carbon samples were analyzed using FTIR spectrometer Bruker IFS 113 V with resolution 1 cm^{-1} and 64 scans. The samples were mixed with potassium bromide and the mixture was pressed into pellets to be used in the analysis.

2.3. Determination of oxygen groups

The content of oxygen-containing functional groups with acidic character on the carbon surface was determined applying the Boehm method by neutralization with bases of increasing strength: $NaHCO_3$, Na_2CO_3, NaOH, and sodium ethoxide. Four portions of the sample with mass 0.5 g were introduced to four sealed flasks with 0.05 N 100 cm^3 of different base solutions ($NaHCO_3$, Na_2CO_3, NaOH, and sodium ethoxide). The suspensions were shaken at least 16 h (960 min), and then filtered. The excess of bases remaining in the solutions was determined from back-titration after adding an excess of standard HCl water solution. It was assumed that $NaHCO_3$ was capable of neutralizing all carboxylic groups, Na_2CO_3-carboxylic and lactonic groups, NaOH-carboxylic, lactonic, and phenolic groups, and sodium ethoxide—all acidic groups [47]. The basic sites were determined by titration with 0.05 N HCl [48]. The procedure is the same as above mentioned, as back-titration of the excess of HCl was performed with 0.05 N NaOH water solution.

2.4. pH determination

The following procedure was carried out: 4.0 g of carbon was put into a 250 cm^3 beaker and 100 cm^3 of distilled water was added. The beaker was covered with a watch glass and heated to a boiling temperature for 5 min. The mixture was then set aside and the supernatant liquid was poured off at 60°C. The decanted portion was cooled down to room temperature and measured to the nearest 0.1 pH value [49].

2.5. Adsorption capacity

The adsorption capacity of the carbons toward mercury was determined by the following procedure: 0.01 g portions of the activated carbon were added to the aqueous solutions (50 cm^3) of $HgCl_2$ with different concentrations—from 10 to 40 mg/dm^3. The initial and equilibrium concentrations of the mercury in the solution were determined by UV spectrophotometer Pharo 300 with rhodamine 6G, according to the procedure described elsewhere [50].

The effect of pH on Hg(II) removal was studied using 0.01 g activated carbon and 20 mg/dm^3 water solution of $HgCl_2$. The pH is adjusted by HCl and NaOH water solutions.

3. Results and discussion

3.1. Chemical characterization of the samples

Chemical analysis of the carbonizate and activated carbon is presented in Table 1. The results show that the activation with water vapor leads to slight increase of the carbon and oxygen content. The samples have very low ash content and demonstrate high hydrogen content, whereas sulfur content diminish two times during the process of activation with water vapor.

3.2. Oxygen functional group content

The chemical character of the surface was investigated by determining the amount of oxygen functional groups, and by pH measurements and IR spectra of the samples.

The oxygen-containing functional groups are very important specific characteristics of the activated carbon, because they have strong effect on the adsorption properties. The identification and quantification of the oxygen groups for the investigated carbon sample are shown in Table 2. The Boehm titration reveals that various oxygen-containing groups with different chemical properties are present on the activated carbon surface.

A large amount of carbonyl groups are detected on the surface of activated carbon prepared from waste POW by activation with water vapor. In the same time, only insignificant amount of strong acidic lactone-like binding structures are identified. The activation with water vapor leads to considerable increase of the amount of basic groups, which is in a good agreement with the pH value. It should be mentioned that this distribution of the amount of different oxygen groups is logical, because the application of

Table 1
Chemical characterization of the samples

Sample	Ash^d, wt.%	C^{daf}, wt.%	H^{daf}, wt.%	S^{daf}, wt.%	O^{daf}, wt.%
POW carbonizate	0.1	87.1	3.7	1.0	8.2
POW-AC	0.1	87.4	3.5	0.5	8.6

Notes: d—dry basis; daf—dry ash-free basis.

Table 2
Quantification of oxygen groups on carbon surface (mequiv/g)

Sample	$NaHCO_3$	Na_2CO_3	NaOH	NaOEt	Basic groups	pH
POW carbonizate	0.120	0.250	0.340	0.900	0.120	4.61
POW-AC	BDL	0.100	0.220	2.100	0.660	8.31

Note: BDL—below detection limit.

the process of steam pyrolysis as activation method leads to obtaining of activated carbon with basic character of the surface.

3.3. IR spectra of the carbonizate and activated carbon

IR spectra of the samples (carbonizate and activated carbon) are presented in Fig. 1. Stretch vibration of associated –OH groups (3,400–3,230 cm^{-1}) are detected as they dominate in the initial sample (carbonizate), which is in agreement with the results of the determination of OH-groups according to the Boehm method. C–H stretching vibrations in the region of 3,000–2,800 cm^{-1} are related to aliphatic structures. The band at 1,704 cm^{-1} could be related to the stretching of C=O in linear aliphatic aldehydes, ketones, and carboxyls. The bands around 1,600 cm^{-1} cannot be interpreted unequivocally. They could be due to: (i) aromatic ring stretching couples to highly conjugated carbonyl groups (C=O); (ii) stretch vibrations of C=C bonds in aromatic structures; (iii) OH groups. The bands in the region of 1,360–1,150 cm^{-1} are due to C=O in complex ethers and ring structures [51–53]. The results of the performed IR spectroscopy analysis confirm that various oxygen groups with acidic and basic character are formed on the surface of activated carbon during pyrolysis.

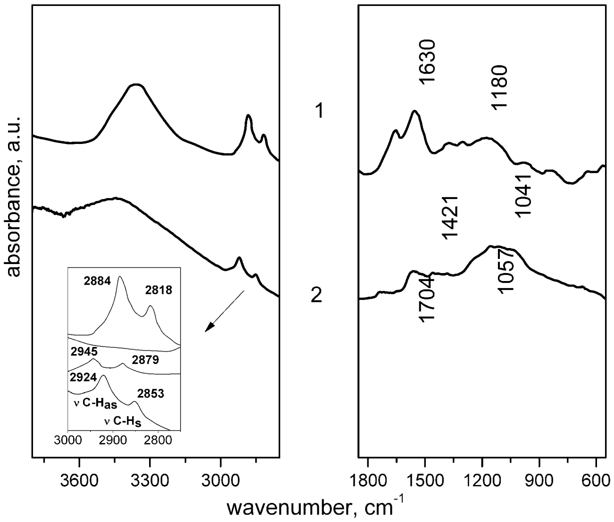

Fig. 1. IR spectra of the carbonizate (1) and activated carbon (2), obtained by water vapor activation.

3.4. Textural parameters

The porosity has a strong effect on the adsorption properties of activated carbon. Textural characterization was carried out by measuring the N_2 adsorption isotherms at −196˚C

Data in Table 3 give detailed characteristics of the pore structure of activated carbon. Analysis of nitrogen adsorption data using several equations indicates that the pyrolysis of POW in the presence of water vapor leads to formation of moderate surface area $S_{BET} = 513\ m^2/g$ and micropore volume of 0.270 cm^3/g. The nitrogen adsorption isotherm of activated carbon obtained from POW is presented in Fig. 2.

The part of the isotherm in the range of the low relatively pressures (a steep increase with a tendency for saturation) is typical for microporous adsorbents. The N_2 adsorption isotherm obtained corresponds to IV type according to Brunauer et al. (low pressure) [54]. Type IV isotherms are obtained for solids containing pores also in mesopore range. Typically, the shape of the Type IV isotherms follows the same path as the type II at lower relative pressure, and these adsorption isotherms have led to the development of the theory of capillary condensation. In principle, it is assumed that along the initial part of the isotherm, that adsorption is restricted to a thin layer on the pore walls until capillary condensation begins in the smallest pores.

4. Adsorption of mercury ions from water

4.1. Adsorption kinetics

The kinetic curves in Fig. 3 show the adsorption of Hg(II) on the synthesized activated carbon from solution with metal concentrations of 10–40 mg/dm^3. Metal ion adsorption increases sharply at a short contact time. The adsorption process is quite fast and efficiently illustrates that the adsorption of metal ions

Table 3
BET surface area and pore volume of the activated carbon sample determined by N_2 adsorption

BET (m^2/g)	V_{tot} (cm^3/g)	V_{micro} (cm^3/g)	V_{meso} (cm^3/g)	V_{macro} (cm^3/g)	I_2 (mg/g)
513	0.600	0.270	0.140	0.190	600

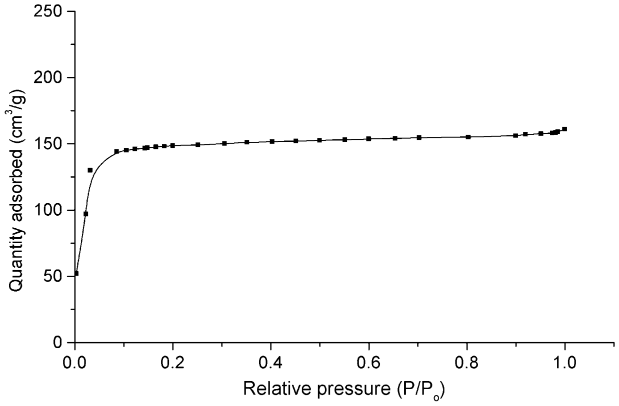

Fig. 2. N_2 adsorption of the activated carbon from POW.

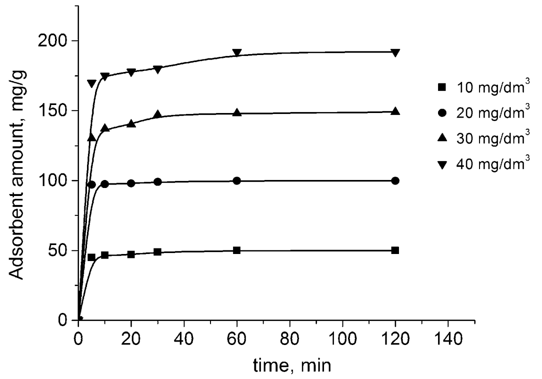

Fig. 3. Effect of treatment time and initial Hg(II) concentration on the adsorption of Hg(II). Conditions: activated carbon amount, 10 mg/50 cm^3; Hg(II) concentration: 10, 20, 30, and 40 mg/dm^3.

is completed almost on the whole surface of activated carbon.

Furthermore, the adsorption capacity increases slowly and the adsorption equilibrium is established within 20 min. The removal curves are smooth and continuous, leading to saturation, suggesting the possibility of formation of monolayer coverage of Hg(II) on the surface of the adsorbent [55]. However, limited functional groups and saturation of the relatively less available adsorption sites could lead the delay of the adsorption process.

4.2. Effect of carbon adsorbent amount

Fig. 4 shows the removal of Hg(II) as a function of carbon adsorbent amount. Increasing carbon quantity leads to increase removal of Hg(II). Quantitative (complete) removal of Hg(II) from 50 cm^3 with concentrations 20 and 40 mg/dm^3 $HgCl_2$ was established when using 15 and 30 mg of the activated carbon, respectively.

4.3. Adsorption isotherms

The isotherm was obtained using a similar procedure as for the adsorption experiments previously mentioned. The adsorption capacity was investigated for different initial concentration (10–40 mg/dm^3) for Hg^{2+} for 60 min (Fig. 5).

The adsorption isotherm for Hg^{2+} was described by the well-known adsorption isotherm models of Langmuir and Freundlich. The Langmuir model isotherm based on monolayer adsorption is expressed as follows [56]:

$$C_e/q_e = C_e/q_m + 1/b\,q_m \qquad (1)$$

where q_e is the equilibrium metal ion concentration on the adsorbent (mg/g), C_e—the e equilibrium metal ion concentration in the solution (mg/dm^3), q_m—the

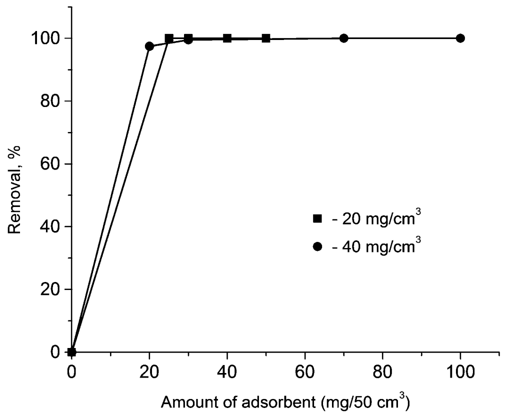

Fig. 4. Effect of carbon amount on adsorption of Hg(II). Conditions: time of treatment 60 min.

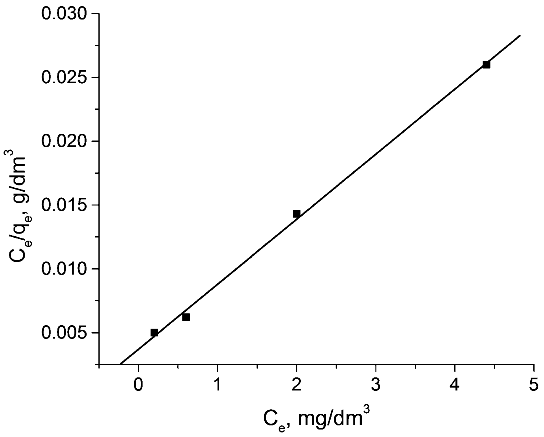

Fig. 5. Langmuir plot for mercury(II) adsorption on activated carbon. Conditions: Hg(II) 10–40 mg/cm^3, 60 min contact time; activated carbon amount, 10 mg/50 cm$^{3.}$

monolayer adsorption capacity of the adsobent (mg/g), and b is the Langmuir adsorption equilibrium constant (dm^3/mg) related to the free energy and the affinity of adsorption.

Freundlich isotherm based on sorption on a heterogeneous surface is expressed as follows:

$$\log q_e = \log k_f + (1/n)\log C_e \tag{2}$$

where k_f is a constant related to the adsorption capacity and $1/n$ is an empirical parameter corresponding to adsorption intensity, which depends on material homogeneity (Fig. 6).

The values of the applied equations models are presented in Table 4.

Surface area, occupied by Hg(II) ions is $S^{2+}_{Hg} = 23.12$ m^2/g, calculated according to the following equation [57]:

$$S^{2+}_{Hg} = q_m \times N_a \times a_{Hg} / A_{Hg} \tag{3}$$

where q_m—adsorption capacity, mg/g; N_a—Avogadro number, $N_a = 6.022 \times 10^{23}$ particles (atoms, molecules or ions) in 1 mol; a_{Hg}—surface, occupied by one Hg ion, nm^2, $a_{Hg} = 3.14\ r^2_{Hg}$, r_{Hg} is the Goldsmith ionic radius, $r_{Hg} = 0.112$ nm [27,58]; A_{Hg}—atomic weight of Hg, $A_{Hg} = 200.6$.

Relative part of the activated carbon surface, occupied by Hg(II) ions is $S^{2+}_{Hg}/S_{tot} = 4.57\%$.

The linear plots of the applied model equations show that they satisfactorily describe the adsorption process of mercury ions. The parameters of the equations are presented in Table 4.

Some theoretical parameters in the Langmuir equation are calculated from the experimental data and are presented in Table 5. The surface area of Hg(II) ions is calculated on the base of Goldsmith ionic radius of Hg [27,58], which is 0.112 nm. It is evident (see Eq. (3) and the calculations) that the relative area (23.12 m^2/g) of the surface of polyolefin-based activated carbon, occupied by mercury(II) ion is 4.57% of the total surface area (513 m^2/g).

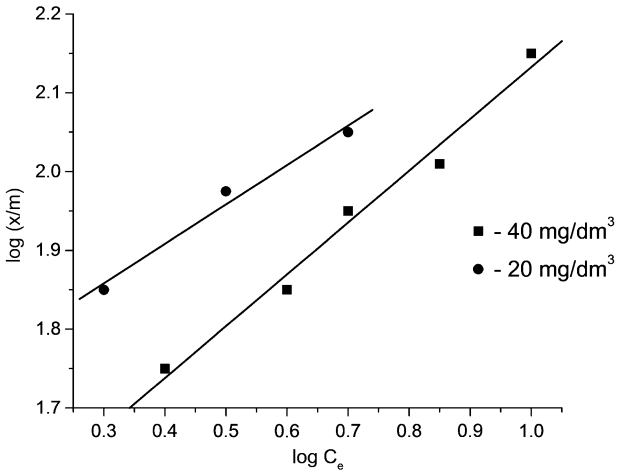

Fig. 6. Freundlich isotherm of Hg(II) adsorption. Conditions: activated carbon amount 0.02–0.1 mg/dm^3, time of treatment 60 min.

Table 4
Data for mercury(II) ion adsorption obtained from Langmuir and Freundlich plot

Model	Parameters	R^2
Langmuir	$q_m = 196.1$; $b = 1.36$	0.9990
Freundlich	$C_e = 20$ mg/dm^3; $k_f = 52$; $n = 2$	0.9974
	$C_e = 40$ mg/dm^3; $k_f = 30.14$; $n = 1.52$	0.9913

A comparison of the adsorption capacities of the prepared activated carbon from POW (196.1 mg/g) with these of other reported carbons, obtained from waste materials, shows that it is much higher than other adsorbents (Table 5).

The results indicate that only carbonized acrylic textile fibers have higher adsorption capacity, but this material is very expensive in comparison with carbons obtained from waste polymer materials, such as activated carbon from POW.

5. Effect of pH on adsorption

In the metal ions adsorption process, the pH value of the solution affects two aspects: metal ion species distribution and adsorbent surface charge, since

Table 5
Hg adsorption capacity of various adsorbents obtained from Langmuir model

Adsorbent type	q_m (mg/g)	Refs.
Pyrolyzed polymer waste	196.1	This study
Graphite oxide/chitosan composite	187.0	[20]
Sludge-derived AC	175.0	[25]
AC from furfural	174.0	[27]
Coirpith activated carbon	154.0	[28]
Walnut shell activated carbon	151.5	[29]
Mix-ZC	147.1	[30]
AC from antibiotic waste	129.0	[31]
Peanut hull carbon	110.0	[32]
Coal adsorbent (Some)	105.0	[33]
Indian almond activated carbon	94.4	[34]
Coal adsorbent (Mengen)	92.2	[33]
Multi-walled carbon nanotubes	89.0	[26]
Commercial granular activated carbon (Merck)	69.4	[32]
Coal adsorbent (Seyitomer)	56.0	[33]
Sago waste activated carbon	55.6	[35]
Coal adsorbent Bolluca	37.0	[33]
Carbon aerogel	34.9	[36]
Bamboo leaf powde	31.0	[24]
Commercial activated carbon	12.4	[37]
Waste rubber	4.0	[38]
Fuller's earth	1.1	[39]
Silico-calcic ash	4.9	[40]
Acrylic textile-based carbon fibers	210.0	[41]

protons can be adsorbed or released [27,59,60]. This behavior depends on the functional groups present on the surface of adsorbent, which in turn determine the acidity constant [60]. Thus, pH value of the solution affects the ionization equilibrium of functional groups on the adsorbent. The type of the species of Hg(II) in water solution depends strongly on pH. The calculation of the stability constant show that in the presence of Cl^-, at $pH > 4.0$, the dominant form is $Hg(OH)_2$ and at $pH < 4.0$ mercury is mainly in $HgCl_2$ form [38]. The effect of pH of the solution on the extent of adsorption can be seen in Fig. 7.

The pH is adjusted by addition of hydrochloric acid and alkali base. It is evident from Fig. 7 that the activated carbon is effective for the quantitative removal of Hg(II) over pH range 5.0–7.0. This shows that the dominant species in the range of highest sorption efficiency are probably $Hg(OH)_2$, retained in the pores of the carbon. The influence of pH on the Hg(II) adsorption may be explained as follows: at low pH values, both the adsorbent and the adsorbate are positively charged, and there is electrostatic repulsion between them. Besides, the H^+ ions, which are available in high concentrations at lower pH values of the solution, compete with positively charged Hg(II) ions for the adsorption sites, thus also hampering the adsorption of Hg(II). Different oxygen-containing groups are determined on the carbon surface, and they also form complexes with the mercury(II) [61]. Having in mind that strong acidic groups are not detected on the carbon surface, we imply that OH-groups and sulfur-containing groups, which are

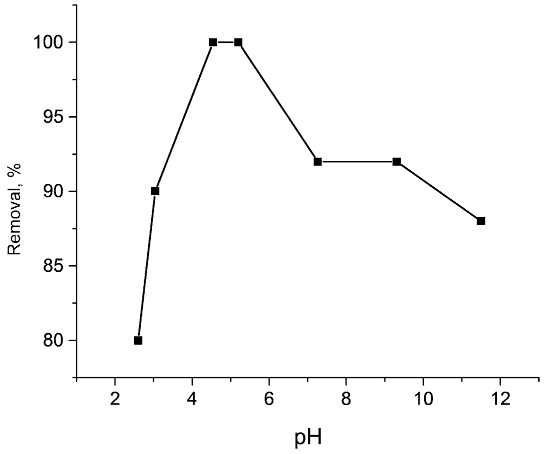

Fig. 7. Effect of pH on Hg removal. Conditions: time of treatment—60 min, activated carbon amount—10 mg/dm^3, Hg(II) concentration—20 mg/dm^3.

formed during the chemical treatment with sulfuric acid, may form the following complexes:

$$C_xOH^{2+} + Hg^{2+} \leftrightarrow C_xOHg^{2+} + 2H^+ \quad (4)$$

$$2C_xOH + \; + Hg^{2+} \leftrightarrow (C_xO)_2Hg^{2+} + 2H^+ \quad (5)$$

$$C_xSO_3H + Hg^{2+} \leftrightarrow (C_xSO_3)_2Hg^{2+} + 2H^+ \quad (6)$$

This is one of the reason that this activated carbon with moderate surface area show high adsorptive ability toward mercury ions, in comparison with other activated carbons with higher surface area—AC from furfural (1,000 m^2/g), activated carbon from antibiotic waste (1,260 m^2/g) [27,31]. Therefore, there are two main factors which determine the high adsorption ability—surface area and surface groups. This investigation illustrates that the oxygen and sulfur-containing groups play important role in the adsorption process as they form different complex compounds with Hg(II) ions.

6. Desorption measurements

The forces acting between the carbon surface and Hg(II) ions are of two main types: physical and chemical, and they give rise to physisorption and chemisorptions, respectively. A study of the character of the adsorption of Hg(II) ions on activated carbon surface is carried out. For this purpose, we tried to desorb Hg(II) ions from metal-loaded adsorbent: (i) with hot water; (ii) with KI. The results show that a small fraction (6%) of the Hg(II) ions are weakly adsorbed in unstable quasi-chemical complexes on the carbon surface area, which are recovered as a result of the water boiling. The rest of the ions are chemically adsorbed and retained in the pores of the carbon. We have some successful desorption and regeneration experiments, with 80% recovery using 2% water solution of KI.

7. Conclusions

Novel adsorbent with very low ash content was synthesized from polymer waste by chemical and physical activation with water vapor. The low-cost adsorbent has good surface characteristics and it is highly effective for removal of Hg(II) from aqueous solution. Activated carbon exhibits high capacity (196.1 mg/g) to adsorb mercury ions from aqueous solution.

The adsorption of Hg(II) follows both Langmuir and Freundlich isotherms. The pH value of the solution influences the mercury adsorption. It increases at pH > 4. Surface functional groups on the carbon surface area are an important factor in mercury adsorption. Their content determines the amount of mercury ions adsorbed. Stable chemical bonds (chemisorptions) are formed between Hg(II) ions and functional groups on the carbon surface, and only a small fraction of the Hg(II) ions are connected by physical adsorption. The surface properties strongly depend on the preparation conditions.

Our investigations show that the waste polymer material POW could be used for production of effective adsorbent for mercury in water solutions. In this way, both valorization of polymer wastes and removal of toxic mercury will be completed.

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