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Removal of Lead from Aqueous Solutions by Xerogel Film Supported on Activated Carbon and Silica

M.M.Selim[#], N.A.Youssef [*], K. F. Mahmoud[*] and F. Abdel Samee[*]
Dept. Phys. Chemistry, National Research Center and [*]Faculty of Girls, Ain Shams University, Cairo, Egypt.

CARBON xerogel (X), a kind of novel carbon material with low-density and continuous nano-porous structure that can be controlled and tailored on nanometer scale, has been prepared through the sol–gel polycondensation of resorcinol (R) with formaldehyde (F) followed by drying at ambient pressure and carbonization in inert atmosphere. Batch adsorption experiments were performed to find out the effective lead removal at different metal ion concentrations. Removal of lead from aqueous solutions by adsorption onto xerogel film supported on activated carbon and silica has been performed. The removal of Pb ions from aqueous solutions was studied by batch method. The effects of initial metal concentrations, initial pH, contact time and the quantity of adsorbents were investigated. It was found that carbon xerogel supported on activated carbon exhibited the best adsorption potential for removal of lead ions from solutions.

Keywords : Lead removal,Xerogel, Activated carbon, Silica and Coating.

The presence of heavy metals in the aquatic environment has been of great concern to scientists and engineers because of their increased discharge, toxic nature, and other adverse effects on receiving waters. Unlike most organic pollutants, heavy metals are generally refractory and cannot be degraded or readily detoxified biologically. Hence the safe and effective disposal of wastewater containing heavy metals is always a challenge to industrialists and environmentalists, since cost-effective treatment alternative is not available. In recent years, Pb has been introduced into natural water from a variety of sources such as storage batteries, lead smelting, tetraethyl lead manufacturing, mining, plating, ammunition, and the ceramic glass industries [(1)] .

The permissible limit of lead in drinking water is 0.05 mg l^{-1}[(2)] . The presence of excess lead in drinking water causes diseases such as anemia, encephalopathy, and hepatitis. Lead ions have an affinity for ligands containing thiol and phosphatic groups and they inhibit the biosynthesis of hemi, causing damage both to the kidney and liver; this behavior of lead is similar to that of calcium.

[#]Author for correspondences E-mail mmaselim@yahoo.com

However, Pb can remain immobilized for years, and hence it is difficult to detect the metabolic disorders it causes. The problems connected with heavy metal pollution are curtailed by processes such as precipitation, electrode deposition, ultra-filtration, cementation, selected solvent extraction, activated carbon adsorption, ion exchange, and biological processes[3] . Adsorption is an attractive process, in view of its efficiency and the ease with which it can apply to the treatment of waste water containing heavy metals.

The use of adsorbents such as modified groundnut husk[4], olive stones[5], lignite material, bagasse and fly ash, peanut hull carbon, Fe(III)/Cr(III) sludge, bentonite, water biogas residual slurry, crude coniferous bark, modified sawdust, and sugar beet pulp[6] for the removal of Pb(II) from aqueous solution has been reported.

A carbon gel can be synthesized by the sol–gel polycondensation of resorcinol (R) with formaldehyde (F) using sodium carbonate as a basic catalyst, followed by drying and pyrolysis in an inert atmosphere [7–11] . The resulting nanostructure is very sensitive to the various synthesis and processing conditions. The RF organic gels undergo two main steps during synthesis. The first one is the preparation of the sol mixture, and aging. The second one is the drying of the wet gel. During the preparation of the sol, the most important parameters that affect the properties of the gel are catalyst concentration, the initial gel pH and concentrations of the solids in the gel.

In this paper, systematic laboratory investigations of the removal of Pb (II) from aqueous solutions by adsorption onto xerogel film supported on activated carbon (AC) and silica (Si), were performed.

Materials and Methods

Adsorbate

A stock solution of Pb (II) (1000 ppm) was prepared by dissolving a required quantity Pb $(NO_3)_2$ for Pb (II). Dilute metal ion solutions were prepared as required by further dilution with distilled water Pb $(NO_3)_2$ in deionized water.

Adsorbent

Xerogel film supported on activated carbon and silica

RF gels were prepared according to the method of Pekala[12]. 1.5990 g resorcinol (R), and 0.0308 g sodium carbonate as catalyst (both from Merck), were dissolved in 48 ml distilled water. After homogenizing with 2.2 ml 37% aqueous formaldehyde (F), the pH of the solution was set to 6.0 by adding dilute HNO_3. The solution was stirred for 30 min and poured into activated carbon (ml/gm) which was then cured at 85^{o}C for 7 days. The carbon xerogel was formed by calcination in an inert atmosphere at 800 ^{o}C during 2hr.

Reagents

All the chemicals were of analytical-reagent grade .

Apparatus

Perkin-Elmer 2380 atomic absorption spectrometer was used for determination of lead. The flame type was air acetylene. The pH values of the solutions were adjusted by adding HCl or HNO_3 and controlled with a digital pH-meter (model 231).

Adsorption experiments

Batch adsorption experiments were carried out at room temperature by shaking a series of bottles containing the desired quantity of adsorbent in a predetermined concentration of heavy metal solution. Samples were withdrawn at different time intervals. Supernatant was separated by filtration and analyzed for remaining heavy metal content. Experiments were carried out at initial pH values ranging from 2.8 to 5.8. The initial pH of the solution was adjusted to the desired value either by hydrochloride acid or sodium hydroxide solution. The percent removal of heavy metal from solution was calculated by the following equation:

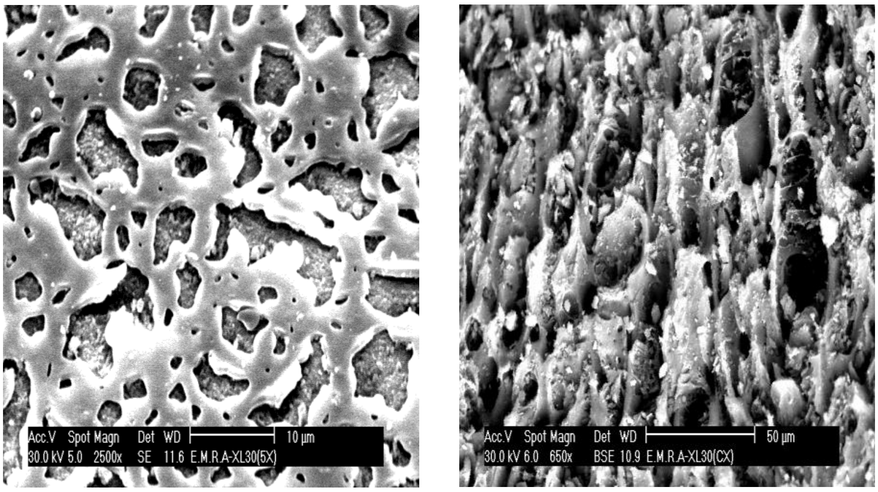
$$\% \text{ Adsorption} = \frac{Co - Ce}{Co} \times 100$$

where C_o is initial concentration of heavy metal, C_e is final concentration of heavy metal.

Results and Discussion

Characteristics of adsorbing material

Scanning electron microscopic photographs of activated carbon, Figure 1 shows the surface texture and porosity of the sample. The availability of pores and internal surface is requisite for an effective adsorbent.



(a) (b)

Fig. 1. Scanning electron micrographs of the two adsorbents, (a) SiX, (b) CX.

Effect of initial concentration of heavy metal

The effect of initial concentration on the percentage removal of lead by xerogel film supported on activated carbon and silica, is shown in Fig. 2. It can be seen from this figure that the percentage removal decreases with the increase in initial concentration.

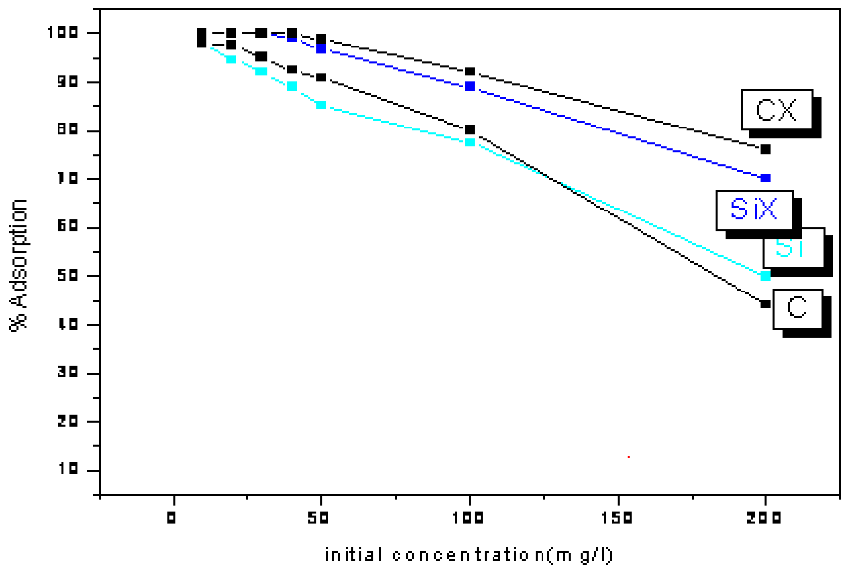


Fig. 2. Effect of initial concentration on adsorption of Pb (II) (contact time 30 min, 0.2 g /20 ml solution, initial pH of solution 5.8, temp., 298 K) .

The effect of initial concentration on the percentage removal of Pb (II) ions by CX , SiX, , AC and Si is shown in Fig. 2. From this figure it is seen that the adsorption of Pb (II) decreased gradually from 100% to 76% for CX, from 100% to 70% for SiX , from 98% to 44 % for AC and from 97% to 50 % for Si by increasing the Pb (II) concentration from 10 mg/l to 200 mg/l, respectively. Sufficient adsorption sites are available at lower initial concentration, but at higher concentration metal ions are greater than adsorption sites. Thus it can be said that removal of lead is highly concentration dependent.

Effect of adsorbent quantity

The effect of adsorbent quantity on removal of Pb (II) is presented in Fig. 3, which indicates that the adsorption increased with increasing CX, SiX, AC and Si quantity up to a certain value and then became almost constant. Therefore, the optimum CX, SiX, AC and Si quantities were selected from 0.20 g / 20 ml of metal ions solution.

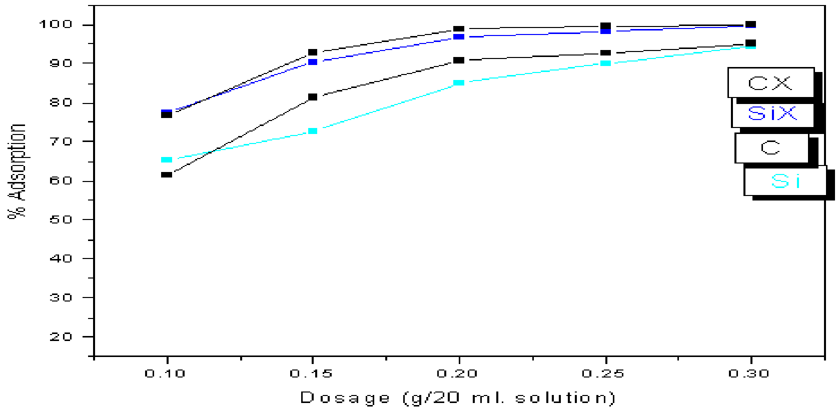


Fig. 3. Effect of Si, C, SiX and CX dosage on adsorption of Pb (II) (initial concentration 50 mg/l, initial pH of solution 5.8, contact time 30 min, temp., 298 K).

Effect of contact time

The removal of lead ions increases with time and attains saturation in about 60 min. Basically, the removal of sorbate is rapid, but it gradually decreases with time until it reaches equilibrium. Figure 4 represents the percent removal of Pb (II) ions versus the contact time for the initial concentration and by using the optimum pH value which was obtained for Pb (II) ions. The Pb (II) ions showed a fast rate of sorption during the first 10 min of the sorbate–sorbent contact and the rate of percent removal becomes almost insignificant due to a quick exhaustion of the adsorption sites. The rate of percent metal removal is higher in the beginning due to a larger surface area of the adsorbent being available for the adsorption of the metals. The two stage sorption mechanism with the first rapid and quantitatively predominant and the second slower and quantitatively insignificant, has been extensively reported in literature(13) .

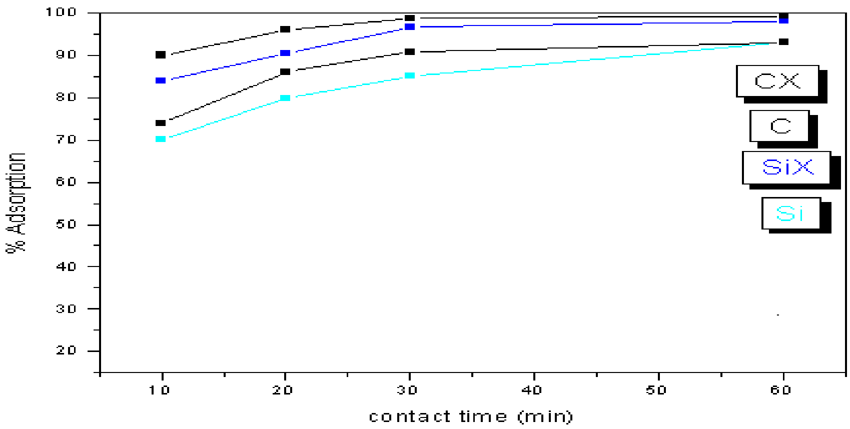


Fig. 4. Effect of contact time on adsorption of Pb (II) (initial concentration 30 mg/l, 0.2g, Si, C, SiX and CX / 20 ml solution, initial pH of solution 5.8, temp., 298 K).

Effect of pH

The effect of initial pH on the adsorption process is presented in Fig. 5. The pH is one of the most important environmental factors influencing not only site dissociation, but also the solution chemistry of the heavy metals: hydrolysis, complexation by organic and/or inorganic ligands, redox reactions, and precipitation are strongly influenced by pH and, on the other hand, strongly influence the adsorption availability of heavy metals(14). As seen in Fig. 5, the adsorption percent of Pb (II) increased with increasing pH and maximum adsorption for Pb (II) was obtained at pH 5.8–which is the initial pH of the solutions. The pH of the solution, which has initial pH 5.8 at the equilibrium conditions, was controlled and found to be 4.8. Therefore, optimum initial pH values of Pb (II) solution was chosen to be 5.8. Thus, adsorption Pb (II) ions on xerogel film supported on activated carbon and, silica was studied at pH 5.8 for subsequent experiments at the equilibrium conditions.

The precipitation of Pb (II) ions may contribute to high uptake of the metal ions at high pH of solutions(15,16). Hence, higher pH values were not studied.

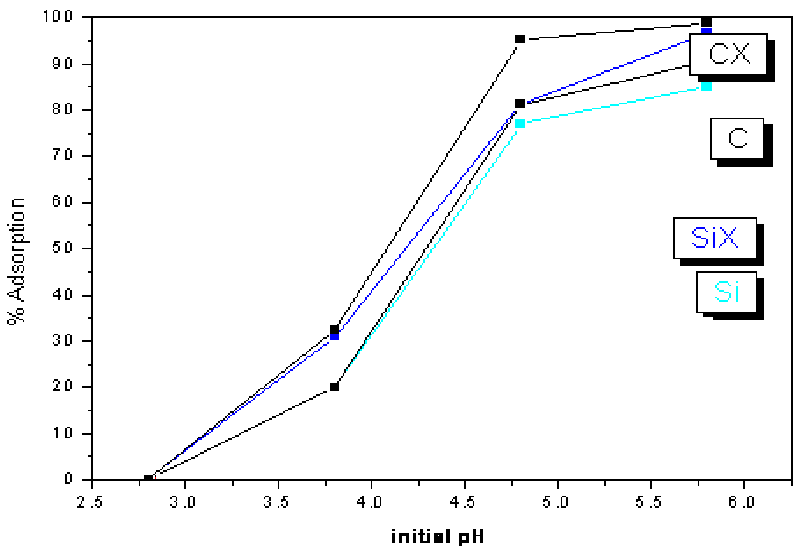


Fig. 5. Effect of initial pH on adsorption of Pb (II) (initial concentration 50 mg/l, 0.2g C/ 20 ml solution, contact time 30 min, temp., 298 K).

Conclusions

The study indicated that carbon xerogel could be prepared from the sol gel polycondensation of resorcinol (R) with formaldehyde (F) followed by drying in an inert atmosphere.

Carbon xerogel film supported on activated carbon and silica exhibited more adsorption capacity of Pb (II) from aqueous solution than activated carbon and silica only.

Adsorption of Pb (II) by CX, SiX and Si has been shown to depend significantly on initial concentration, the pH, contact time, surface area and the amount of CX, SiX, AC and Si .

The CX, SiX and Sican be considered as useful in treatment of wastewater containing Pb (II).

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ازالة الرصاص من محاليل مائية بواسطة فيلم من الزيروجل على الكربون المنشط والسليكا

محمد محمد سليم ، نادية عبد الحكيم يوسف* ، قــوت القلوب فــارس محمــود* و فاطمه عبد السميع*
قسم الكيمياء الفيزيقية - المركز القومى للبحوث و*كلية البنات - جامعة عين شمس – القاهرة – مصر .

تم تحضير كاربون زيروجل له كثافة منخفضة وتركيب مسامى نانوى وذلك بطريقة صول جل عديدى التكثيف للريزورسينول مع الفورمالدهايد ثم جفف عند ضغط جوى وفحم فى جو خامل. اجريت مجموعة من التجارب لايجاد الازالة المؤثرة للرصاص عند تركيزات مختلفة. تم اجراء تجارب ازالة الرصاص من محاليل مائية على فيلم من الزيروجل المحمل على كاربون منشط وعلى سليكا. درس تاثير التركيزات الابتدائية والاس الهيدروجينى الابتدائى ووقت التجربة وكمية المادة الماصة على مدى الازالة. من النتائج التى تم الحصول عليها وجد ان الكاربون المغطى بالزيروجل له اعلى كفاءة ازالة للرصاص من المحاليل.