APPLICATION OF TAGUCHI METHOD FOR OPTIMIZING THE ADSORPTION OF LEAD IONS ON NANOCOMPOSITE SILICA AEROGEL ACTIVATED CARBON

Fariba Tadayon
Department of Chemistry,
Islamic Azad University, North Tehran Branch, IRAN
f_tadayon@iau-tnb.ac.ir

Shiva Motahar
Department of Chemistry,
Islamic Azad University,
North Tehran Branch,
IRAN.

M.Hosseini
Department of chemistry,
Islamic Azad University,
Omidieh Branch,
IRAN.

ABSTRACT

Using the Taguchi method, this study analyzes the optimum conditions for removal of lead from aqueous solution in batch method by Nanocomposite Silica Aerogel Activated Carbon (SA–AC), which is prepared using the sol gel method. The controllable factors used in this study consisted of the following: (1) pH of the solution(A); (2) adsorption temperature(B); (3) lead initial concentration(C); (4) adsorbent dosage(D); (5) contact time (F).The effects of each factor were studied at four levels on the removal efficiency of lead from aqueous solution.L_{16} orthogonal array (OA) has been used for experimental design. Concentrations of lead were assessed by flame atomic absorption spectrophotometer. The statistical analysis revealed that the most important factors contributing to the removal efficiency are pH of the solution and adsorption temperature. The study shows that the Taguchi's method is suitable to optimize the experiments for total lead ions removal. The total optimum adsorptive removal of leadions were obtained with C_0 = 10 mgL^{-1}, T = 65°C, pH = 6, m = 0.16 g and t = 6 h.

Keywords: *Lead; Adsorption; Nanocomposite; Taguchi; Silica aerogel*

INTRODUCTION

Heavy metal pollution has become a serious health concern in recent years. Continuous exposure to low levels of heavy metals may result in bioaccumulation and resulting health consequences in humans(1).Pb(II) ion is one of the most poisonous heavy metal ions that accumulates in muscles, bones, kidney and brain tissues and has the potential to cause various disorders, and is regarded as the priority controlled pollutant in many countries (2).Lead contamination is mainly due to industries related to lead batteries, phosphate fertilizer, electronic, wood production, paint, oil, metal, and also combustion of fossil fuel, automobile emissions, mining activity, forest fires, sewage wastewater, etc.(3).A number of techniques have been developed for removing lead pollutant from aqueous effluents. These methods include precipitation, electroplating, evaporation, ion exchange, membrane separation, coagulation, floatation, reverse osmosis, membrane filtration, solvent extraction, adsorption and various biological processes (4-7).

Most of these processes are not widely acceptable owing to their high costs, low efficiency, disposal of sludge, inapplicability to a wide range of pollutants (8).The adsorption process is arguably one of the more popular methods for the removal of heavy-metal ions because of its simplicity, convenience, and high removal efficiency (9).Recently, applications of nano particles for the removal of pollutants have attracted substantial interest in the scientific community because of their special properties (10). Nanoparticle sexhibit good adsorption efficiency especially due to higher surface area and greater active sites for interaction with metallic species. Furthermore, adsorbents with specific functional groups have been developed to improve the adsorption capacity(11).Silica aerogel nano particles, a new kind of nanometer-sized material, are widely used in this field for remove metals(12,13).

Silica aerogels obtained through the low-temperature sol–gel process and dried under supercritical conditions are highly porous solids showing large specific surface areas (up to $1000m^2g^{-1}$), an extraordinarily large surface-to-volume ratio(~2×109m^{-1}) and low densities (0.003–0.35 gcm^{-3})(14,15).In this literature was used silica aerogel activated carbon (SA-AC) and its application for adsorption of Pb(II) ion. Adsorbtion of lead ion in SA-AC must be optimization. It is essential to develop a robust method to rank and screen controllable factors upon a thorough investigation of their main and interaction effects.

Taguchi's optimization technique is a unique and powerful optimization discipline that allows optimization with minimum number of experiments. The Taguchi experimental design reduces cost, Improves quality, and provides robust design solutions. This method has evolved into an established approach for analyzing interaction effects when ranking and screening various controllable factors. Moreover, Taguchi method is applicable to solving a variety of problems involving continuous, discrete and qualitative design variables (16,17).Taguchi method is capable of establishing an optimal design configuration, even when significant interaction exists between and among the control variables(18).This method can also be applied to designing factorial experiments and analyzing their outcomes(19).Therefore, we investigate for the first time the remove of lead ions on nanocomposite SA-AC was optimized using Taguchi method.

EXPERIMENTAL

Materials and Methods

Concentration of the lead ion solutions was performed by using A Varian model spectra AA-240(Mulgrara, Victoria, Australia).An infrared spectrum was obtained using a Fourier transform infrared spectrometer (FT-IR, Perkin-Elmer, spectrum 100) and scanning electron microscopy (SEM-EDX, XL30 and Philips Netherland). The pH-measurements of metal ions and buffer solutions were carried out by an Orion 420.

All the reagents used were of analytical grade. Double distilled water was used throughout. Laboratory glassware was kept overnight in a 10% (v/v) HNO_3 solution and then rinsed with deionized double distilled water. The nitrate salt of Pb(II) analytical grade (Merck) were used without further purification. More dilute solutions were prepared daily from $1000mgL^{-1}$stock solutions.

Tetramethylortho silicate (TMOS) and Methyltrimethoxysilane (MTMS) from Aldrich and another reagents used in the present study were of analytical grade from E.merk. pH adjustments were performed with HCl and NaOH solutions.

Adsorbents synthesis

Silica wet gel was prepared with 5.02 mL of TMOS as starting material. The TMOS was diluted with 44.2 mL of methanol, then 1.5 mL of MTMS was added and 1.2 mL NH_4F(0.1 $mol.L^{-1}$)solution was added to mixture finally. Then the mixture was stirred at 20°C for 30 minutes. After the intermediate product was homogenisation by hemogeniser (20000 rpm) and during the homogenisation 0.033 g activated carbon (AC) was added to mixture. The mixture was poured into a Teflon beaker where the sol aged into hydrogel within about 10 min. After gelation, the gel was left for one day. Subsequently, the product was thoroughly immersed into methanol for 24h and dried at room temperature for 5 days and finally the SA–ACadsorbent was obtained.

RESULTS AND DISCUSSION

Characterization of adsorbents

Fig .1 (a, b), shows the SEM micrographs of the SA and SA–AC.

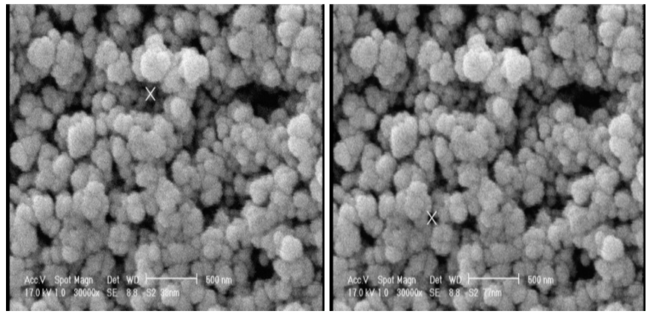

Fig. 1.*The SEM image of SA(left) and SA–AC nanoparticles(right).*

The FTIR spectra of the SA and nanocomposite SA–AC are shown in Fig.2. The broad absorption band in the region 3450-3200 cm^{-1} and band at 1638 cm^{-1} respectively to the adsorbed water and surface silanol groups [20], the silica aerogel exhibits bands in the 1250–1050 cm^{-1} region and 800 cm^{-1} easily attributed to the Si–O–Si asymmetric and symmetric stretching vibrations of the silica network respectively. Besides, the infrared spectrum of the silica aerogel shows a weak C–H symmetric deformation peak at 1384 cm^{-1} that corresponds to residual non-hydrolyzedmethoxy groups on the surface of the gel. This peak is not observed for silica aerogel. Peaks at around 1600 cm^{-1} on both graphs correspond to O–H stretching, provided that they are sharper in the case of silica aerogel, which contains more O–H groups. These two peaks are the weakest in the SA–AC sample [21].The above FTIR data indicates that strong interaction exists on the interface of silica aerogel and SA–AC, and silica aerogel is successfully modified by activated carbon.

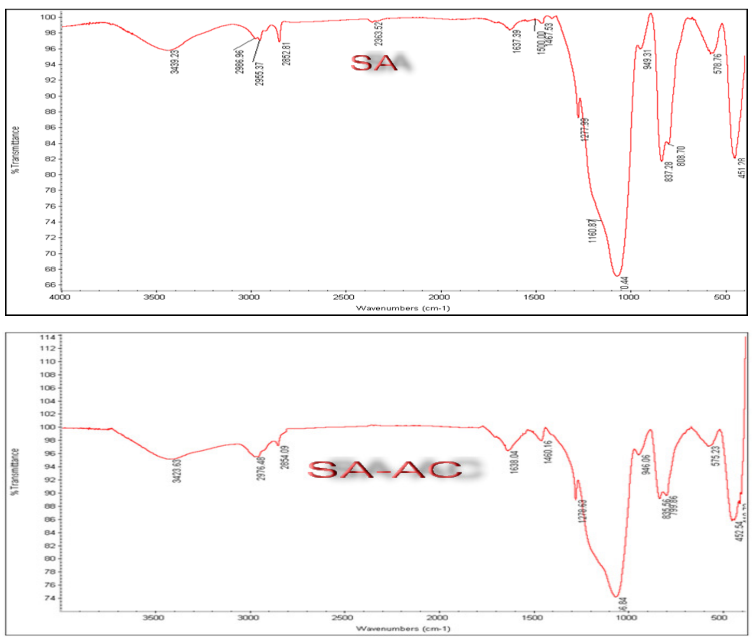

Fig. 2. *FTIR absorption spectra of SA, SA–AC*

Taguchi Design

Taguchi's OA analysis is used to produce the best parameters for the optimum design process, with the least number of experiments. In this work, the effect of five important factors including: pH, temperature, lead initial concentration, adsorbent dosage and contact time were selected and each factor at four levels on the removal efficiency of aqueous solutions was studied using Taguchi's method. The used level setting values of the main factors(A-F) and the $OA_{16}(4^5)$ matrix employed to assign the considered factors are shown in the Table 1.

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Table 1. Coding of factors and levels of the orthogonal test

Factors	Levels			
	1	2	3	4
A: pH	3	4	5	6
B:Temperature(ºC)	25	45	55	65
C: Lead initial concentration (mg L^{-1})	2	10	20	30
D: Adsorbent dosage(g)	0.04	0.08	0.12	0.16
F:Contact time(h)	2	4	6	8

The optimum responses calculated using the following expression:

$$Y_{opt}=\frac{T}{N}+(A-\frac{T}{N})+(B-\frac{T}{N})+(C-\frac{T}{N})+(F-\frac{T}{N}) \quad (1)$$

Where T is the grand total of all results, N is the total number of results; Y_{opt} is the response under the optimum conditions, A, B, C, D and F are the mean responses of the pH, temperature, lead initial concentration, adsorbent dosage and contact time at optimum levels, respectively. The results show that pH and temperature offered the main contribution to the adsorbtion efficiency. By contrast, the other parameters offered secondary contribution. The average of responses for each factor at different levels were also calculated to probe into the effect of each factor and were plotted in Fig. 3-7 to screen the optimum levels.

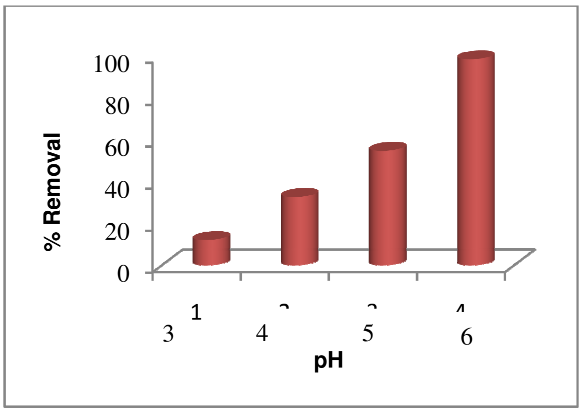

Fig.3.Effect of pH on adsorption of lead(II) ions.

Fig. 3show that with the increase of pH, the amount of removal percentage increased significantly. The adsorption increases as the pH increases due to the increase of the silica surface negative charges favoring the electrostatic attraction between the two entities with opposite charges, Si-O^- and Pb^{2+}.At low pH, these groups are protonated and result in repulsion with the lead cations minimizing the %removal of the SA-AC.

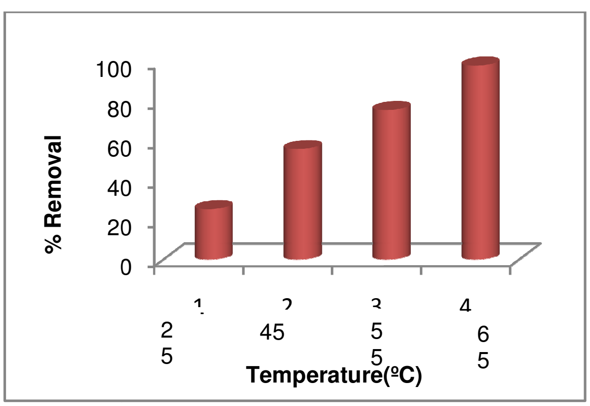

Fig.4.Effect of temperature on adsorption of lead(II)ions.

Fig. 4 shows the recovery obtained at different temperatures from 25 °C to 65 °C. The results obtained indicate that 65ºC temperature is sufficient to complete remove.

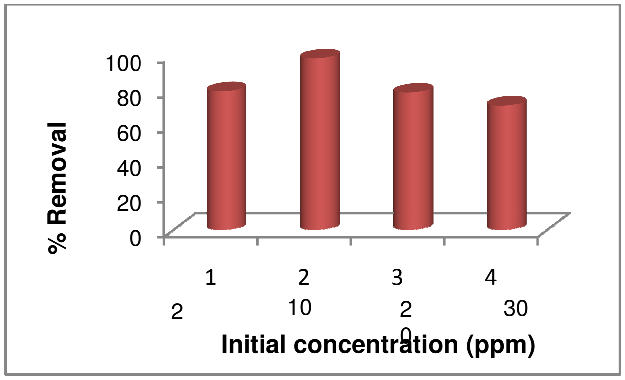

Fig.5.*Effect of initial lead(II) concentration on adsorption.*

Fig. 5 shows the recovery obtained at different initial lead(II) concentration. Ion removal percentage increases when the initial ion concentration decreases. At low ion concentrations the ratio of surface active sites to the total metal ions in the solution is high and hence all metal ions may interact with the adsorbent and be removed from the solution. Best results were obtained with the lead initial concentration was achieved within 10 mgL^{-1} .

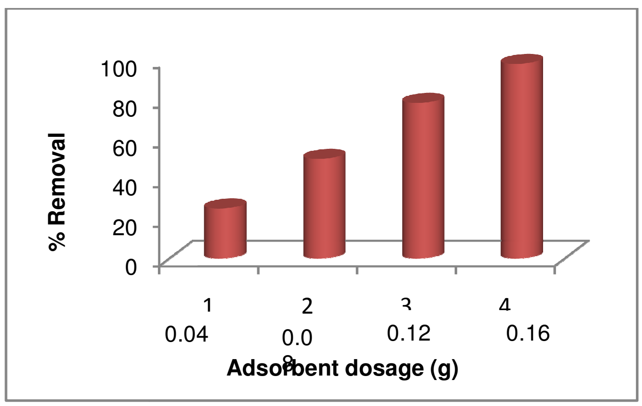

Fig.6.*Effect of adsorbent dose on lead(II) adsorption.*

The effect of the quantity of SA–AC on the recovery of Pb(II) was investigated and the results are shown in Fig. 6.The removal of Pb(II) was found to increase with an increase in adsorbent dosage from 0.04 and 0.16 g. Maximum uptake was obtained at adsorbent dose of0.16 g.

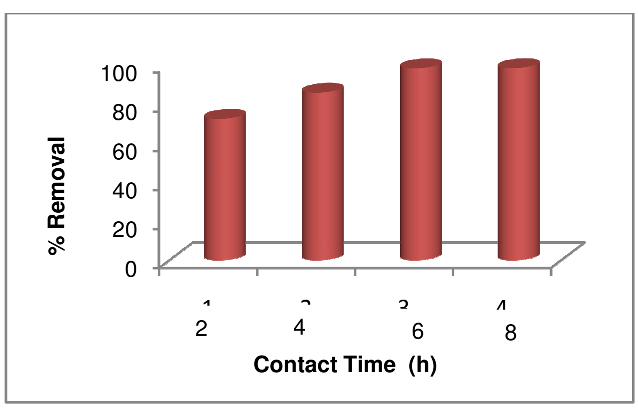

Fig.7.*Effect of adsorbed quantity of lead(II) with time.*

The sorption of Pb(II) by SA–AC at various contact times ranging from 2 to 8 h was performed. As shown in Fig. 7, the removal rate of Pb(II) was completed after almost 6h.According to these results, it was set a contact time of 6 h in order to ensure that equilibrium conditions are reached.

The optimum conditions tabulated in Table 2.As shown in Fig. 3-7, pH and temperature offered the main contribution to the adsorbtion efficiency. By contrast, the other parameters offered secondary contribution.

Table 2. Optimum conditions for adsorbtion of lead on nanocomposite SA-AC

Factor	levels
A: pH	6
B:Temperature (ºC)	65
C: Lead initial concentration (mg L^{-1})	10
D: Adsorbent dosage (g)	0.16
F:Contact time (h)	6

CONCLUSION

This paper demonstrated the optimization of various batch parameters for the removal of lead ions from aqueous solutions by nanocomposite SA–AC using Taguchi's optimization methodology. An OA(4^5) matrix was applied to study the effect of five factors(pH, temperature, lead initial concentration, adsorbent dosage and contact time)on adsorbtion efficiency of aqueous solutions. The results show that pH of the solution and temperature has significant effect on the adsorbtion efficiency. The highest % removal obtained were 98.08% for lead ions.

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