ID: 5033

Retention of Toxic Metal in Solution by Marl Treated by Heat at Different Temperatures

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ABSTRACT

In this study, two marl samples recovered from the bedrock of the Draria Nuclear Research Centers site aquifer (CRND) were heat-treated in an oven at 200°C and 400°C for four hours. This treatment was carried out to study its effect on the retention capacity of MPM-200 and MPM-400 samples when treated with a solution containing Uranium. The marl samples were characterized by X-Ray Fluorescence (XRF), Brunauer–Emmett–Teller (BET), particle size distribution and FTIR analysis. The elemental XRF analysis shows that the major compounds are Silica, Calcium, Aluminum, Iron and Potassium. BET analysis established the predominance of the meso-porous character associated with a specific surface area value equal to 17.55 m^2/g for MPM-200 and 17.88 m^2/g for MPM-400. The results of the particle size analysis of the MPM-200 and MPM-400 samples show that the heat treatment of our marl samples did not affect their particle sizes. The heat treatment applied to the marl samples caused a slight progressive decrease in the intensity of the FTIR transmittance band signal for both samples MPM200 and MPM400 compared to the signal obtained for the raw marl. The various adsorption experiments were monitored by varying the mass of adsorbent, the volume of solution, the contact time, the initial pH, the initial Uranium concentration and the stirring speed. An optimum adsorption capacity (3.5 mg/g for MPM-200 and 2.8 mg/g for MPM-400) was obtained after 60 min with a sample mass of 80 mg, a volume of 30 ml, an initial solution concentration of 10 mg/L, an initial pH equal to 2 and a stirring speed of 180 rpm. The kinetics of Uranium adsorption by our MPM-200 and MPM-400 adsorbents follow the pseudo-second-order model, with a coefficient of determination R^2=0.9782 for MPM-200 and R^2=0.9977 for MPM-400. The diffusion process is controlled simultaneously by intra-particle diffusion and liquid film diffusion. The isotherm with the best correlation for MPM-200 is the Freundlish model, followed by the Sips model and finally the two Langmuir and Toth models. For MPM-400, the best model is Sips, followed by Toth, then Langmuir and finally Freundlish, but with very little difference in the values obtained for R^2, R^2_{adj}. The thermodynamic study revealed that the process of Uranium adsorption by MPM-200 and MPM-400 is thermodynamically feasible, spontaneous, and endothermic, with the existence of a slight order.

Keywords: adsorption, marl, heat treatment, Uranium, isotherms

1 Introduction

The use of natural, low-cost materials for the retention of toxic metals [1-5] is becoming an increasingly important part of the methods used to treat solutions containing toxic metals. Their use in the treatment of solutions containing Uranium, as a toxic element, is well documented in the literature [6]. Previous work on the use of untreated Plaisancian marl as a retention material for the treatment of solutions containing toxic heavy metals such as copper and zinc has produced acceptable results. [7-8] In this study, two marl samples, one heat-treated in an oven at 200°C and the other at 400°C for four hours, will be used as adsorbents for Uranium in a synthetic solution. The first part of this study will characterize our two marl samples by determining their particle size, identifying existing functional groups, performing qualitative and quantitative X-ray fluorescence analysis and determining specific surface areas and porosities. In the second part, we will study the retention of synthetic Uranium solutions using the two adsorbents, and compare the results obtained. This will be done by an optimization study of the operating conditions followed by a kinetic study, then a study of the diffusion models and retention tests by applying the Langmuir Freundlish,

DOI: 10.21467/abstracts.170 ISBN: 978-81-970666-1-0 (eBook)

Toth and Sips models with a non-linear treatment of the data obtained. Finally, a thermodynamic study will be carried out for each case.

2 Experimental

The Marne Plaisancien Medium (MPM) sample was obtained by mixing other samples collected from aquifer substratum of the watershed of wadi El-Ghoula (Al-Achour locality in southeast Algiers). The mixed sample was dried at room temperature, sieved through an 80 micron and homogenized before use. To make a heat treatment, a sufficient quantities of samples are placed in a muffle furnace heated to 200°C (sample named MPM-200) and then to 400°C (sample named MP-M400) for a period of four hours for each sample. The particle size distribution was performed by using a granulometer from Frish Analysette model. Fourier transform infrared spectroscopy (FTIR) from Perkin-Elmer was used to identify the functional groups. The nitrogen adsorption-desorption isotherms at 77 K were carried out using a Micromeritics ASAP 2010 surface area analyzer. The elemental composition of adsorbent was analyzed by X-Fluorecence. The adsorption experiments of Uranium (VI) onto Marl samples (MPM-200 and MPM-400) were conducted in batch.

3 Results and discussion

Heat treatment of the marl samples had no significant effect on particle size distribution. Heat treatment resulted in a slight decrease in mass contents for all elements, and especially for major elements such as Si, Ca, Al, Fe and K. The specific surface area by BET results for MPM-200 and MPM-400 are equal repectively to 17.55 m^2/g and 17.88 m^2/g. The heat treatment applied to the marl samples caused a slight progressive decrease in the intensity of the FTIR transmittance band signal for both samples MPM-200 and MPM-400 compared to the signal obtained for the raw marl. The comparison of the two-parameter models and those with three parameters gives for the case of MPM-200, the Freundlish isotherm as the best correlation of our experimental results, followed by the Sips model and finally the two Langmuir and Toth models (Freundlish > Sips > Langmuir and Toth). For the case of MPM-400, the best model is Sips followed by Toth then Langmuir and Freundlish (Sips > Toth > Langmuir and Freundlish). The ΔH values obtained for MPM-200 and MPM-400 are positive, which means that the Uranium adsorption is endothermic. The positive values of ΔS reflect the existence of a slight order or the existence of enhanced disorder. Negative values of ΔG indicate that the Uranium adsorption process in heat-treated marls at different temperatures (200°C, 400°C) is thermodynamically feasible and spontaneous.

4 Conclusion

Two marl samples recovered from the bedrock of the CRND site aquifer were heat-treated in an oven at 200°C and 400°C for four hours. This treatment was carried out to study its effect on the retention capacity of MPM200 and MPM400 samples when treated with a solution containing Uranium. This study comprises two main parts: the first part concerns the characterization of the two materials, and the second part concerns the study of Uranium adsorption. The results of the particle size analysis of MPM200 and MPM400 show that the heat treatment of our marl samples did not affect their particle sizes. Elemental XRF analysis of our marl samples shows that the major compounds are Silica, Calcium, Aluminium, Iron and Potassium. Heat treatment caused a slight decrease in the mass contents of these elements. BET analysis established the predominance of the meso-porous character associated with a specific surface value equal to 17.55 m^2/g for MPM200 and 17.88 m^2/g for MPM400. The kinetics of Uranium adsorption by MPM200 and MPM400 follow the pseudo-second-order model. The diffusion process is controlled simultaneously by intra-particle diffusion and liquid film diffusion. The isotherm with the best correlation for MPM200 is the Freundlich model, followed by the Sips model and finally the two Langmuir and Toth models. In the case of MPM400, the best model is the Sips model, followed by the Toth model, then Langmuir and finally Freundlich, but with very little difference in the values obtained for R^2, R^2_{adj}. The thermodynamic study revealed that the process of Uranium adsorption by MPM200 and MPM400 is thermodynamically feasible,

ISBN: 978-81-970666-1-0
Series: AIJR Abstracts
DOI: 10.21467/abstracts.170

spontaneous, endothermic, with the existence of a slight order.

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ISBN: 978-81-970666-1-0

Series: AIJR Abstracts

DOI: 10.21467/abstracts.170