PHYSICAL CHEMISTRY 2014

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on Fundamental and Applied Aspects of
Physical Chemistry

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SORPTION OF PERTECHNETATE ONTO ORDERED MESOPOROUS CARBON

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ABSTRACT

Ordered mesoporous carbon (OMC) was used as an adsorbent for the removal of pertechnetate (TcO_4^-) anion. Batch experiments were performed to study the effects of pH and ionic strength of the solution. The adsorption of TcO_4^- is almost pH-independent in very wide pH region (from 4.0 to 10.0). Maximum distribution coefficient, K_d, was obtained at pH 2.0. Adsorption of TcO_4^- was not affected by the ionic strength changes.

INTRODUCTION

The presence of the long-lived radionuclide ^{99}Tc ($t_{1/2}$ = 2.1 x 10^5 y) in the environment comes from anthropogenic nuclear activities such as nuclear power plants, nuclear weapons testing facilities, reprocessing of spent nuclear fuel, nuclear accidents and medical application of ^{99m}Tc. The predominant chemical form of the radionuclide ^{99}Tc is the highly soluble and environmentally mobile TcO_4^- anion, that easily penetrates the ecosystems [1]. Because ^{99}Tc is hazardous to the environment, its effective removal from contaminated groundwater and sediments is considered a primary task.

Removal of TcO_4^- by activated carbons is one of the most efficient processes, because of large specific surface area, porous structure and variety of surface functional groups of activated carbons. Ordered mesoporous carbon belongs to a family of porous materials which attracted great attention in recent years due to its unique features such as high surface area and uniform, large and tunable pore channels. These features make OMC attractive for use in the adsorption processes.

The aim of the present paper was to investigate the influence of the solution pH and ionic strength on the TcO_4^- sorption by the OMC.

EXPERIMENTAL

The OMC was synthesized under acidic conditions as previously described by Momčilović et al [2]. Radioactive technetium (in the form of pertechnetate) was obtained from a ^{99}Mo/^{99m}Tc generator and its radioactive concentration was determined by a Capintec CRC-15 dose calibrator (Capintec, Inc. Ramsey, USA). The resolution of the instrument is 0.001 MBq, i.e. 0.01 □Ci. The measurements of ^{99m}Tc activity in aliquots were made using vial geometry.

Batch experiments were performed at room temperature by mixing 0.04 g of well powdered OMC adsorbent and 10 cm^3 of working $^{99m}TcO_4^-$ solution in closed glass vials. Experimental set-up is presented in Fig. 1. The total $^{99m}TcO_4^-$ radioactive concentration in the working solution was 1.4 MBq cm^{-3} (37.8 mCi cm^{-3}). The initial pH was adjusted by using either 0.1 M HCl or NaOH. The samples were shaken on a laboratory shaker for 60 min at a stirring speed of 200 rpm. After that, the liquid phases were separated from the solid phases by filtration through a microporous membrane filter (0.45 μm). The residual radioactive concentration of $^{99m}TcO_4^-$ in each aliquot was determined with a Capintec CRC-15 dose calibrator.

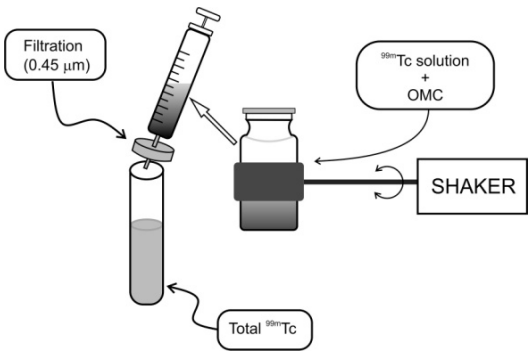

Figure 1. Experimental set-up for the sorption of TcO^{4-} onto the OMC.

The effects of solution pH (2.0-12.0) and ionic strength (0.01 and 0.1 M NaCl) were investigated with respect to the removal efficiency of TcO_4^-. All experiments were carried out in duplicate, and the data obtained were used for analysis.

The removal efficiency (E, %) and the distribution coefficients (K_d, cm^3 g^{-1}) of $^{99m}TcO_4^-$ were calculated using the following equations:

$$E = \frac{\left(A_i - A_f\right)}{A_i} \times 100 \qquad (1)$$

$$K_d = \frac{\left(A_i - A_f\right)}{A_f} \times \frac{V}{m} \qquad (2)$$

where A_i and A_f are the initial and the final $^{99m}TcO_4^-$ radioactive concentrations in the liquid phase (MBq cm^{-3}), V the volume of the solution (cm^3) and m the mass of adsorbent (g).

RESULTS AND DISCUSSION

The effect of the solution pH and ionic strength, I, on the adsorption of TcO_4^- using the OMC was investigated at varying pH values from 2.0 to 12.0. The obtained results are presented in Fig. 2. It is evident that the adsorption of TcO_4^- was pH-dependent, with maximum removal efficiency of 99% at the pH value 2.0. Then, the removal efficiency slightly decreases and reaches the value of 93% at the pH values between 4.0 and 10.0. At the pH values beyond 10.0, the adsorption of TcO_4^- decreased sharply and the removal efficiency of TcO_4^- fell to 72%. The distribution coefficients, presented in the insert in Fig. 2, behaved in a similar manner as a function of solution pH, with the maximum K_d value of 6.6 x 10^3 at the pH 2.0, plateau with the mean K_d of 2.9 x 10^3 at the pH range from 4.0 to 10.0, and the K_d value less than 1 x 10^3 at the pH > 10.0. Under the investigated experimental conditions, E and K_d values are insensitive to ionic strength changes, indicating that the sorption of TcO_4^- was not affected by the presence of competing Cl^- ions.

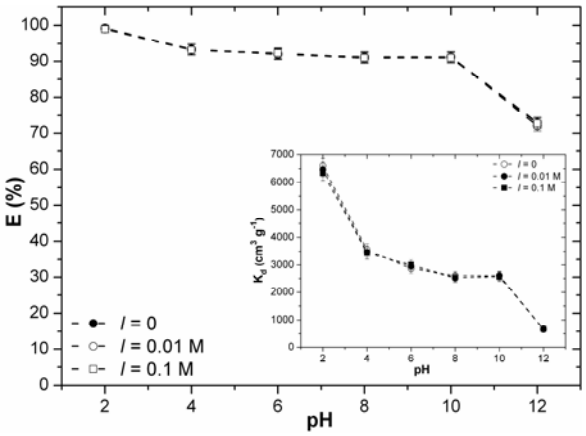

Fig. 2 Effect of pH and ionic strength (0.01 and 0.1 M NaCl) on the sorption of TcO^{4-} by the OMC. Insert: Variation of K_d values with pH and ionic strength.

CONCLUSION

Ordered mesoporous carbon (OMC) was investigated for its sorption capability to remove pertechnetate (TcO_4^-) anions from aqueous solutions.

The conclusions are summarized as follows:

- Maximum E and K_d values of 99% and 6.6 x 10^3 cm^3 g^{-1}, respectively, were obtained at the pH 2.0;
- The adsorption of TcO_4^- is almost pH-independent in very wide pH region (from 4.0 to 10.0);
- The sorption of TcO_4^- was not affected by the presence of competing Cl^- ions.

The obtained results indicate that the TcO_4^- sorption by the OMC is well classed compared to other carbon materials (adsorbents). As a material resistant to aging (to the oxidizing conditions and the presence of water), as well as its good sorption properties, the OMC appears to be an efficient adsorbent for the TcO_4^- removal from aqueous solutions.

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