RadChem 2018



Contribution ID: 640

Type: Poster

Activated carbon as an effective pertechnetate adsorbent

Monday, 14 May 2018 18:30 (15 minutes)

Activated carbon (AC) is pores space bounded by the carbon atoms. Pores of AC are directive for its application. It was shown that, surface modified form of carbon-based materials, are promising sorbents for separation of various radionuclides. In our study 5 AC adsorbents (A-E) are prepared from cellulose wool by treating with different solutions and carbonized at the temperatures of (700-800) °C for Tc adsorption. They are characterized by FTIR for organic functional groups determination, HRTEM for microscopic imaging, liquid N₂ sorption for pore size distribution and specific surface area determination, and acid-base titration conduced potentiometrically for surface charge investigation. Surface area of the sample is within the range of (150-510) m²·g⁻¹. Samples C and E possess also mesopores. AC samples have a positive surface charge and point of zero charge lies in the range of pH 6-8.

Tc is a fission product with a long half-life 97 Tc (EC; $4.21 \cdot 10^6$ a), 98 Tc (β^- ; $4.2 \cdot 10^6$ a), 99 Tc (β^- ; $2.1 \cdot 10^5$ a) formed in the thermal neutron fission of 235 U and 99m Tc is the most commonly used diagnostic tool. One option how to remove or control released Tc in the environment, is sorption process. It has been shown that AC has a relatively high efficiency for Tc retaining (99m TcO₄⁻). Adsorption equilibrium of TcO₄⁻ is reached in few minutes except for sample D (60 minutes). Mechanism of TcO₄⁻ adsorption involves: R-C-OH + TcO₄⁻ = R-C-OTcO₃ + OH⁻ or if phenolic or carboxylic groups are present: R-CO…H + TcO₄⁻ = R-C…H…OTcO₃. Tc can bind direct to the C as -C-OTcO₄ or mechanism involves chemical reduction of the soluble TcO₄⁻ to TcO₂ and its adsorption on the surface. Adsorption of TcO₄⁻ on samples A - E occurs at pH of 2.

Presence of anions suppresses adsorption of TcO_4^- . $HCOO^-$, NO_3^- , Br^- , Cl^- , CH_3COO^- , SO_4^{2-} , and ClO_4^- are used to investigate the adsorption competition between TcO_4^- and anions. Competition strongly depends on pH, structure of the anions, and standard absolute molar enthalpy of the hydration. The most significant decrease of TcO_4^- adsorption is observed in presence of NO_3^- and ClO_4^- . It is due to close values of the highest standard absolute molar enthalpies of hydration. Moreover, ClO_4^- has similar tetrahedral structure to TcO_4^- . Following order for sample A shows the suppression of TcO_4^- adsorption from the weakest to the strongest (kJ mol⁻): SO_4^{2-} (-1,099) < $HCOO^-$ (-384) < CH_3COO^- (-374) < Cl^- (-359) < Br^- (-328) < NO_3^- (-316) < ClO_4^- (-205).

In order to investigate desorption of TcO_4^- and thus reusability of AC, simple column desorption experiments are carried out. Desorption solutions consist of anions as in the previous paragraph, respectively, at pH 12. Continuous gamma measurements show that 1 M ClO_4^- has the "strongest" ability to remove TcO_4^- from AC. Adsorptions of TcO_4^- on samples with oxidized and reduced surface are also studied. Immediate adsorption of TcO_4^- makes reduced surface sample preferable to oxidized one. The results obtained in this study improve that activated carbon is an effective sorbent for the separation of the pertechnetate. This work was supported by VEGA Project No. 1/0507/17.

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Session Classification: Poster SEP

Track Classification: Separation Methods, Speciation