

Variation of Pertechnetate Ion Adsorption on Some Active Carbons from Mineral Acid Solutions

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The practice of permanently disposing of high level nuclear waste (HLNW) after solidification in glass depends for its safety on the availability of a reliable means of collecting TcO_4^- from the solution leaching out of solidified waste after its underground disposal. One such means is to let the leached TcO_4^- be adsorbed by active carbon, which can be derived from various organic sources. In practical application, the choice of source of material for the active carbon will influence its TcO_4^- adsorbing performance.

The present study is aimed at obtaining basic information to serve in selecting the best source for deriving active carbon to be used for collecting Tc.

Adsorption runs were performed applying the method of experiment described in a previous paper¹⁾, which also outlined the procedure adopted for producing $^{95\text{m}}\text{Tc}$. The experiment was carried out on three samples of active carbon derived from different sources (listed in Table 1). The samples were of commercially marketed grade that is commonly utilized for water treatment.

Table 1 Surface area and pore size of active carbon samples

Sample	Source material	Particle size (mm)	Surface area (m^2/g)*	Pore size (nm)*
A	Coconut shell	0.59-2.00	952	1.5
B	Oil pitch	0.35-0.70	810	1.5
C	Coal	0.51-2.00	880	2.3

*Surface area was determined using a MICROMERITICS ASAP-2400 Automatic Surface Area and Pore Size Analyzer. The samples were previously degassed for 1 h at 200 °C.

Figure 1 presents the time dependence of the distribution coefficient K_d ¹⁾ of Tc adsorption on the active carbon samples. The plots for 0.5 M H₂SO₄ revealed a conspicuous relapse of K_d beyond 7 h. This relapse of Tc adsorption performance can be attributed to displacement of pertechnetate anion, once adsorbed on active carbon, by sulfate anion deriving profusely from sulfuric acid that had been added.

Compared with sample C, samples A and B are seen to have produced higher peak values of K_d , occurring at around 7 h from start of adsorption; at 100 h, the difference in K_d is seen to have diminished to quite a small value. Beyond 100 h, K_d has tended to rise again in the case of samples A and B, but not with sample C. For application to underground collection of Tc at HLNW disposal sites, sample C might be considered the most suitable, for its freedom marked relapse of Tc adsorption performance.

Differences in the surface areas and the pore sizes of the active carbon samples, within the range covered in the present experiment (Table 1), would not appear to influence K_d very much. The relapse of K_d — attributed to desorption displacement of pertechnetate anion by other anion deriving from the added acid — was also noted on adsorption curves obtained with solutions of 0.1 M HNO₃, 0.5 M HCl and with HClO₄ solutions of acidity above 0.1 M (Figs. 2- (a), - (b) and - (c)). On the other hand, runs made with phosphoric acid solution (Fig. 2- (d)) have presented no indication of such relapse through 24 h of adsorption; only after more than 30 h a small sag of K_d is discernible.

The relapse of TcO₄⁻ adsorption performance beyond a certain duration of adsorption has been observed only with acidic solutions of pH below 1. The underground water found in the localities designated for HLNW disposal is known to present pH values of 7 - 9²⁾, so that, for practical purposes of HLNW disposal, such particular behavior of active carbon can be dismissed from consideration.

References

- 1) Ito K., Akiba K.; CYRIC Annual Report 1989, 133.
- 2) Allard B., Kipatsi H., Rydberg J.; UCRL-trans-11340 (1978).

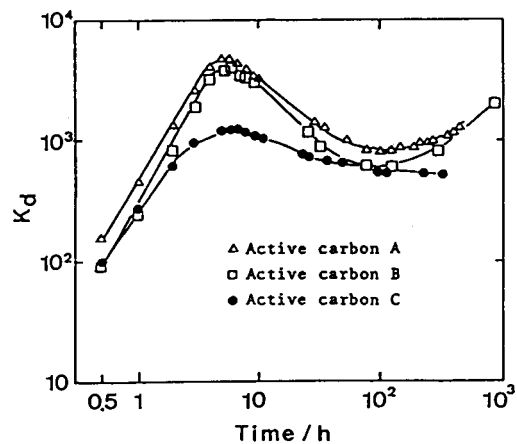


Fig. 1 Time dependence of distribution coefficient K_d of Tc on various active carbon samples. 10 ml TcO_4^- solution containing 0.5 M H_2SO_4 , 0.1 g active carbon.

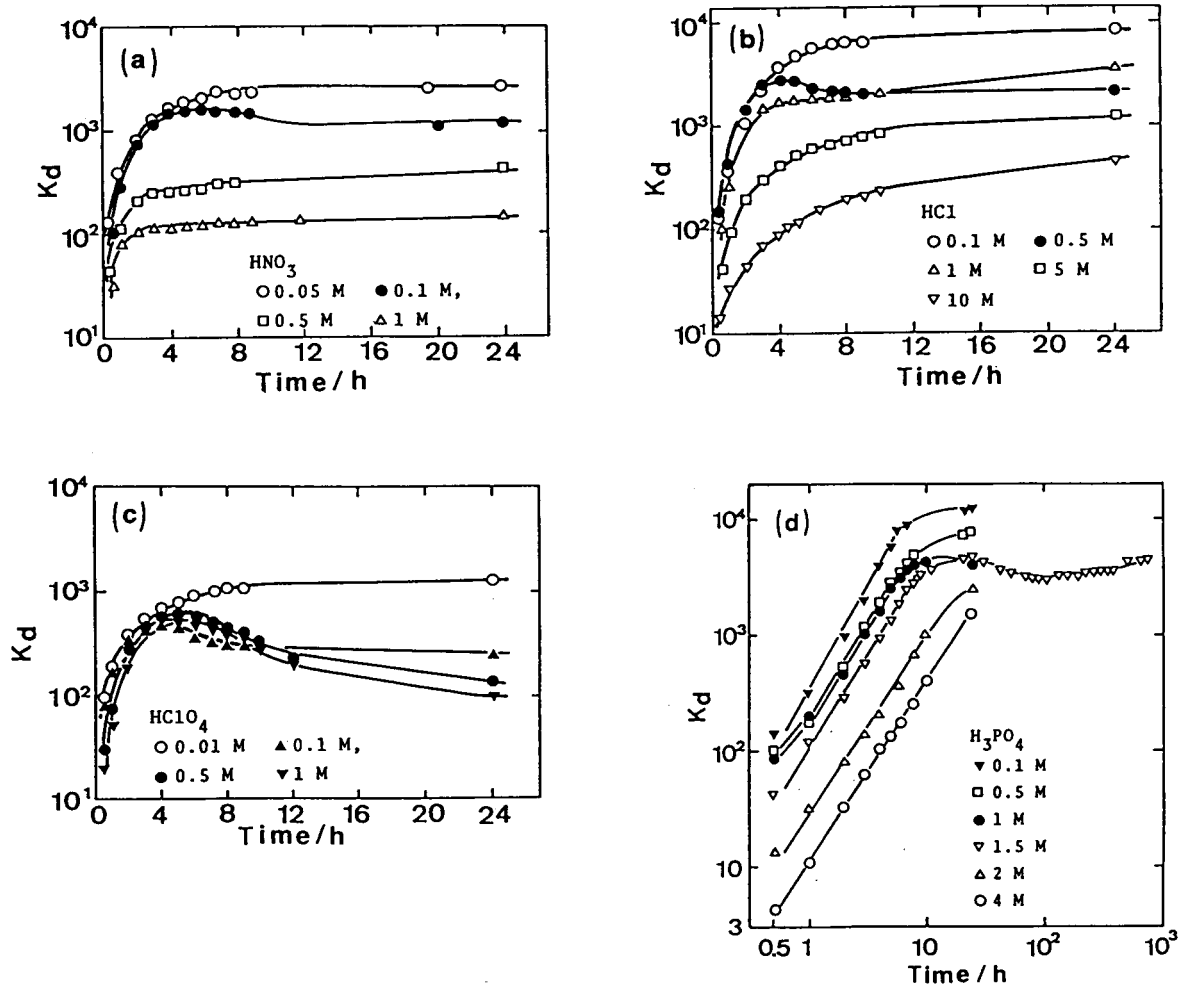


Fig. 2 Time dependence of distribution coefficient K_d of Tc on active carbon A from mineral acid solutions of different concentrations. 10 ml TcO_4^- solution, 0.1 g active carbon A.