Sorption of Technetium on Activated Carbon

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Ito K., Yachidate A. and Akiba K.
Research Institute of Mineral Dressing and Metallurgy, Tohoku University

Technetium-99 with a long half-life (2.12×10^5 y) is one of the most dangerous nuclides after 1000 y of nuclear waste storage. Once the waste eventually comes in contact with water, ^{99}Tc dissolves in the water and migrates away from the repository. Neretnieks$^{1)}$ has reported that the alpha particles from the waste radiolyze the water forming hydrogen gas and hydrogen peroxide. The hydrogen gas is less reactive and diffuses away, while the hydrogen peroxide is a strong oxidizing agent.

In order to obtain a better understanding of sorption of Tc ion in the presence of hydrogen peroxide, the present paper deals with the sorption rate of Tc ion by carbonaceous materials.

Technetium-95 was produced by the ^{95}Mo(d,2n)^{95m}Tc reaction$^{2)}$, by using an AVF Cyclotron at the Cyclotron and Radioisotope Center, Tohoku University. Pertechnetate ion of carrier-free ^{95m}Tc was extracted by a tetraphenylarsonium chloride-chloroform solution. The carrier-free $^{95m}TcO_4^-$ ion was stripped into 8 M HNO_3 .

The batch equilibrium methods were utilized in the sorption studies. Under oxic condition, a sorbent was equilibrated with a 50 ml solution containing $^{95m}TcO_4^-$ ion at the pH range 7-9. Being allowed to equilibrate for 24 h, the sorbent and the liquid phase were separated by centrifugation at 25,000 G.

The sorption rate of Tc ion on carbonaceous materials under oxic condition is shown in Table 1. The sorption by charcoal and anthracite was 15% and 9%, respectively, however the rate by activated carbon was 85%. The activated carbon was found to sorb pertechnetate ion very much. Table 2 shows the effect of source materials of activated carbon on the sorption of pertechnetate ion under oxic solution. The sorption rates by bone char and the activated carbon from sawdust were lower than the other activated carbons from coal, petroleum and coconut husks. An activation method of the bone char is carbonization, and the activated carbon from sawdust is carbonization with zinc chloride. The activated carbon from coal, petroleum and coconut husk are produced with oxidizing agents such as steam, air, carbon dioxide at elevated temperature. These activated carbon seem to have specific ability for the sorption of pertechnetate ion.

The variation in distribution coefficient of Tc ion with changing the ratio of V/m is shown in Fig. 1, where V is the solution volume and m is the weight of the activated carbon. The K_d values were very influenced by the concentration of coexisting salts. The K_d values under oxidizing condition were almost constant (10^4) in the regions from the V/m ratio from 6 to 1000. The K_d values under reducing condition decrease with an increase in the ratio from 7 to 25, and slightly increase from 25 to 170. The sorption rates of Tc ion under reducing condition have been published elsewhere.3) The sorption mechanisms of Tc ion under oxidizing and reducing condition were different from each other. Hassler4) showed that various substances such as permanganates and chromates, undergo reduction by the activated carbon during sorption. These substances are no longer soluble after such a chemical change and the mechanism could follow one of several paths.

1) The substance may be first sorbed and then undergo reduction.
2) The reduction and sorption may constitute integral parts of an over-all phenomenon.
3) The reduction may occur first and convert the substance into an insoluble form; in which case, the role of the carbon would be a purely mechanical one of sweeping a solid precipitate from the liquid.

From these suggestion, it appears that the sorption of Tc ion under oxidizing condition proceeds through mechanism 2, and the sorption under reducing condition proceeds through mechanism 3.

References

1) Neretnieks I., Nucl. Technol. 62 (1983) 110.
2) Chiloshi G. et al., IL NUOVO CIMENTO 19 (1979) 1121.
3) Ito K. and Kanno T., CYRIC Annual Report (1985) 62.
4) Hassler J. W., "Activated Carbon", p. 226, Chemical Pub. Co., Inc., New York (1963).

Table 1. Sorption of technetium ion by coke, coal, charcoal and activated carbon under oxic condition.

Material	pH	Sorption rate (%)
Coke (coal)	8.7	3.7
Coke (petroleum)	7.9	2.4
Coal	7.3	1.7
Anthracite	7.1	9.2
Charcoal (wood)	9.5	15.0
Activated carbon (coconut husk)	9.5	85.0

Activated carbon 2 g, 0.1 M NO_3^-.

Table 2. Effect of starting material of activated carbon on sorption of technetium ion under oxic condition.

Starting materials	pH	Sorption rate (%)
Coal (Thurumi, GL-30)	8.7	94.5
Coal (Diahope-006, acid)	8.0	89.3
Coal (Diahope-006, acid), wash	8.1	87.3
Coal (Diahope-006)	11.2	91.6
Coal (Diahope-006), wash	10.5	90.2
Petroleum (ABAC-MP)	10.5	94.2
Petroleum (ABAC-MP), wash	10.5	94.7
Charcoal bone	8.2	28.1
Sawdust (PS-D)	8.8	63.2
Coconut husk (Diasorb, W10-30)	10.1	93.7
Coconut husk (Diasorb, W10-30), wash	9.8	93.5
Coconut husk (Diasorb, F-100)	8.7	88.3

Activated carbon: 2 g, 0.1 M NO_3^-.

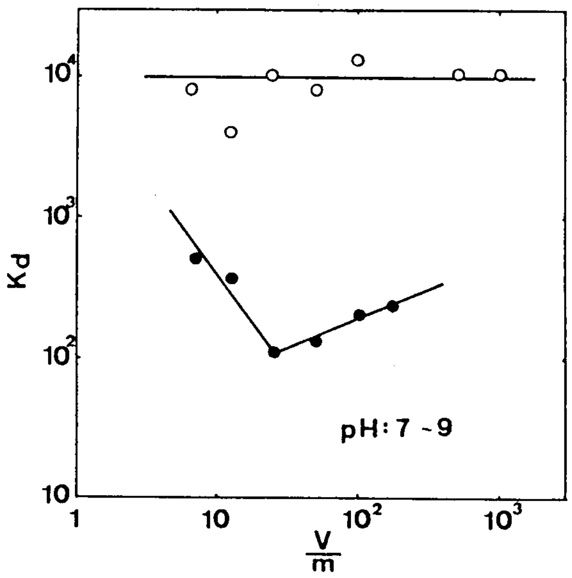

Fig. 1. Effect of the ratio of the liquid volume and the sorbent weight on the distribution coefficient, open: oxidizing condition (0.1 M H_2O_2 in the absence of NO_3^- ion), closed: reducing condition (0.1 M $NaBH_4$ solution in the presence of 0.1 M NO_3^- ion).