Journal of Chromatography, 114 (1975) 389–395
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CHROM. 8466

COLUMN CHROMATOGRAPHIC SEPARATION OF RADIOACTIVE TELLURATE, TELLURITE, IODIDE AND IODATE BY ACTIVE CHARCOAL

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(Received May 21st, 1975)

SUMMARY

The adsorption behaviour of trace amounts of tellurate, tellurite, iodide and iodate on active charcoal has been studied by a radioactive-tracer method; on the basis of the results, a column chromatographic separation of these ions from each other was devised. By passing the sample solution (at pH 5.6) through an active charcoal column, all but tellurate are retained. Iodate, tellurite and iodide are then eluted successively with water (pH 7.0), 0.1 *N* hydrochloric acid and 1 *N* sodium hydroxide, respectively. The method can be applied to the preparation of radioiodine from a neutron-irradiated tellurate target.

INTRODUCTION

When tellurium is irradiated with neutrons, various kinds of radioactive tellurium isotopes are produced, including ^{131m}Te and ^{131}Te, the parents of ^{131}I: this nuclear reaction is therefore used for the practical production of large amounts of ^{131}I. The separation of ^{131}I from the irradiated tellurium compound is usually performed by distillation (see, for example, refs. 1 and 2) or by solvent extraction (see, for example, refs. 3–5). Recently, however, Tóth[6] used a platinum black column for the adsorption separation of ^{131}I from a dilute sulphuric acid solution of neutron-irradiated magnesium tellurate; after elution of radioactive tellurate, ^{131}I was desorbed by a solution containing sodium hydroxide and sodium sulphite.

In this paper, the use of active charcoal as adsorbent is described. The use of active charcoal in radiochemical separation has hitherto been studied mainly from the view-point of decontamination problems, and Ikeda and Mitsubayashi[7] have reported the adsorption and desorption behaviour of some fission-produced radionuclides in various regions of pH. The present paper shows that systematic chromatographic separation of Te*(VI), Te*(IV), I^{*-} and $I^*O_3^-$ can be achieved by using a column of active charcoal.

EXPERIMENTAL

Materials

Active charcoal. Powdered palm-shell coal of chemical grade was used for the batch experiments. In the column experiments, the palm shell coal used (Tsurumicoal HC-50) was of size 80–200 mesh.

Te(VI) and Te*(IV).* Telluric acid (H_6TeO_6) was irradiated with neutrons in the TRIGA-II reactor at Rikkyo University; some 20% of radiotellurium is produced in the form of Te*(IV) as a result of the hot-atom effect. The separation and purification of Te*(VI) and Te*(IV) were carried out by the anion-exchange method of Inarida[8]. Irradiated H_6TeO_6 was dissolved in 4 *N* hydrochloric acid, and the solution was passed through a column (4 × 1 cm I.D.) of Diaion SA 100 resin (Cl^-; 100–200 mesh). The Te*(VI) passed through the column, while Te*(IV) and ^{131}I were retained; the Te*(IV) was subsequently eluted with 0.1 *N* hydrochloric acid.

Radioiodide ($^{131}I^-$). Commercial $Na^{131}I$ in a dilute solution of sodium carbonate was used.

Radioiodate ($^{131}IO_3^-$). The above-mentioned $^{131}I^-$ solution was oxidized with chlorine in a separatory funnel; excess of chlorine was eliminated by shaking the solution several times with carbon tetrachloride.

Measurement of activity

The activity was measured with a well-type NaI(Tl) scintillation counter connected with a 400-channel pulse-height analyzer.

Determination of distribution coefficients

A 15-ml portion of an aqueous solution of radioactive tellurium or iodine [Te*(VI), Te*(IV), I^{*-}, $I^*O_3^-$], the pH or acidity of which was adjusted with hydrochloric acid or sodium hydroxide solution, was placed in a centrifuge tube, and 0.5 g of powdered active charcoal were added. The solution was then shaken for 10 min, and, after centrifugal separation, the supernatant solution was filtered, and the active charcoal was collected on filter paper. This charcoal was wrapped, together with the filter paper, in thin polyvinylidene film and packed in a polyethylene tube, and its activity (A_1) was measured. A 5-ml protion of the filtrate was taken in a polyethylene tube, and its activity (A_2) was measured. The distribution coefficient (*D*) was then calculated from the expression

$$D\,(\mathrm{ml/g}) = \frac{A_1/0.5}{A_2/5} = \frac{10A_1}{A_2}$$

Separation by the column method

The active charcoal powder (0.5 g) was packed in a glass column (10 × 1 cm I.D.) and conditioned with hydrochloric acid or sodium hydroxide solution of the appropriate concentration, the flow-rate being adjusted to about 1 ml/min; each 1-ml or 5-ml portion of effluent was collected in a polyethylene counting tube, and its activity was measured. Each radionuclide in the effluent was identified with use of a 400-channel pulse-height analyzer.

Identification of the chemical forms of the species in each fraction was carried out as follows. The Te*(VI) and Te*(IV) were identified by their adsorptive behaviour during anion exchange (Te that passes through the column in 4 *N* hydrochloric acid solution is Te(VI), and that which is eluted with 0.1 *N* hydrochloric acid is Te(IV)). The I^{*-} and $I^*O_3^-$ were identified by chromatography on Tōyō No. 50 filter paper, with 1-butanol–water–acetone (1 : 1 : 10) as developing solvent; development was for 20 cm (1 h): R_F values were 0.50 for I^{*-}, ≈0 for $I^*O_3^-$ and 0.85 for I_2^*.

RESULTS AND DISCUSSION

Distribution coefficients

The distribution coefficients obtained for each species are shown in Fig. 1, from which it can be seen that Te*(VI) usually has a very low distribution coefficient, whereas Te*(IV) is adsorbed to a considerable extent at pH values greater than 4 as well as at acid concentrations higher than 1 *N*. On the other hand, I^{*-} is strongly adsorbed, showing distribution coefficients almost as high as 10^4 in the neutral or acid region. For $I^*O_3^-$, the distribution coefficients are lower by factors of 0.1 to 0.01 than those for I^{*-} in the pH region 3 to 9. At acidities above 0.01 *N*, the values fluctuate around the corresponding values for I^{*-}, and, moreover, are less reproducible, suggesting that $I^*O_3^-$ becomes unstable in acid solution.

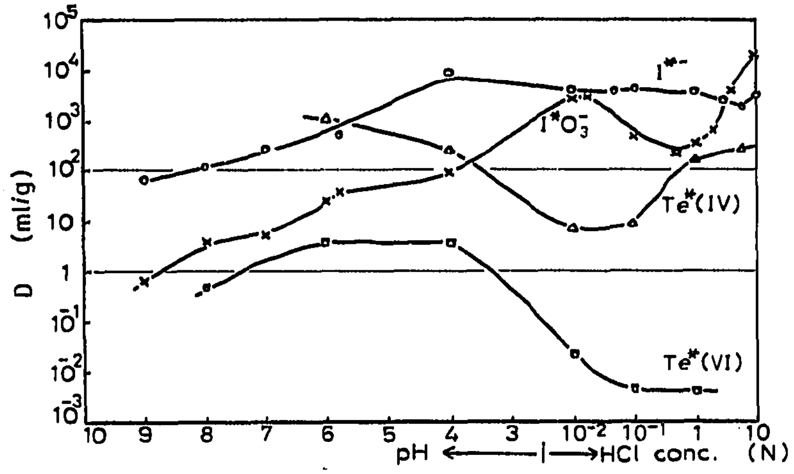

Fig. 1. Distribution coefficients at various pH.

Behaviour of $I^*O_3^-$ in acid solution

Further studies on the unstable behaviour of $I^*O_3^-$ in acid solution were carried out by paper chromatography.

Carrier-free $I^*O_3^-$ in 4 *N* hydrochloric acid was developed under the conditions stated above, giving the result shown in Fig. 2a. When $I^*O_3^-$ together with I^{*-} (each carrier-free) in 4 *N* hydrochloric acid was developed, the result was as shown in Fig. 2b. To identify each peak, individual developments of $I^*O_3^-$, I^{*-} or I_2^* were carried out; each characteristic peak appeared at the position shown in Fig. 2c. It is noteworthy that there is no activity at the origin in Figs. 2a and 2b, which shows

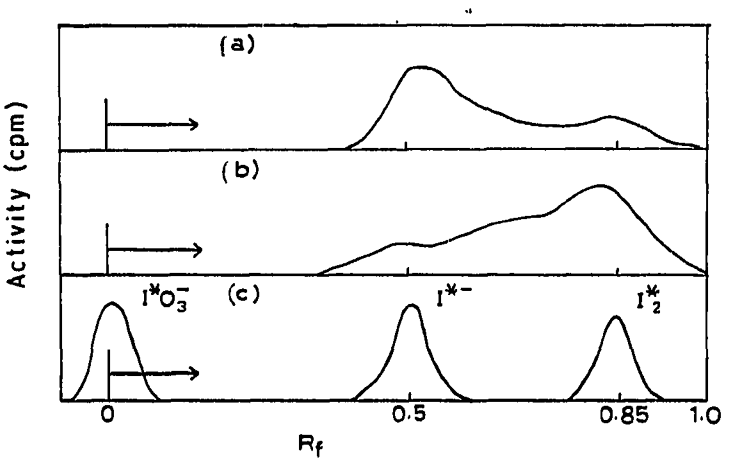

Fig. 2. Behaviour of radioiodine during paper chromatography: (a) $I^*O_3^-$ in 4 *N* hydrochloric acid; (b) I^{*-} and $I^*O_3^-$ in 4 *N* hydrochloric acid; (c) characteristic peaks for I_2^*, I^{*-} and $I^*O_3^-$.

that $I^*O_3^-$ is unstable in hydrochloric acid solution and is converted into I^{*-} and I_2^*, presumably according to such reactions as follows:

$$I^*O_3^- + 6\,Cl^- + 6\,H^+ \rightarrow I^{*-} + 3\,Cl_2 + 3\,H_2O$$
$$2\,I^*O_3^- + 10\,Cl^- + 12\,H^+ \rightarrow I_2^* + 5\,Cl_2 + 6\,H_2O$$
$$I^*O_3^- + 5\,I^{*-} + 6\,H^+ \rightarrow 3\,I_2^* + 3\,H_2O$$

Separation of Te(VI), Te*(IV), I^{*-} and $I^*O_3^-$*

On the basis of the difference in distribution coefficients shown in Fig. 1, the following procedure was devised for the column chromatographic separation of Te*(VI), Te*(IV), I^{*-} and $I^*O_3^-$ from each other. The separation experiments were carried out with neutron-irradiated telluric acid.

Procedure. The irradiated sample is dissolved in water (pH 5.6), and the solution is poured on to an active charcoal column. The Te*(VI) passes through the column, while Te*(IV), I^{*-} and $I^*O_3^-$ are adsorbed. After washing the column with water (pH 5.6), $I^*O_3^-$ is eluted with water (pH 7.0, adjusted by adding dilute sodium hydroxide solution). The Te*(IV) is then eluted with 50 ml of 0.1 *N* hydrochloric acid. Finally, I^{*-} is eluted with 50 ml of 1 *N* sodium hydroxide solution. The results are shown in Fig. 3, from which it can be seen that the recovery of each species is almost quantitative.

Separation of Te(VI), Te*(IV) and I^{*-}*

When it is only necessary to separate Te*(IV) and Te*(VI) (for example, in studies on the hot-atom chemistry of radiotellurium) or when the separation of radioiodine from radiotellurium is required regardless of its chemical form, the following procedure is simple and adequate.

Procedure. The solution containing Te*(VI), Te*(IV), I^{*-} and $I^*O_3^-$ is made 4 *N* in hydrochloric acid, then poured on to the charcoal column; the column is then washed with 10 ml of 4 *N* hydrochloric acid. Only Te*(VI) is eluted. Next, 20 ml of 0.1 *N* hydrochloric acid are passed through the column to elute Te*(IV), the column

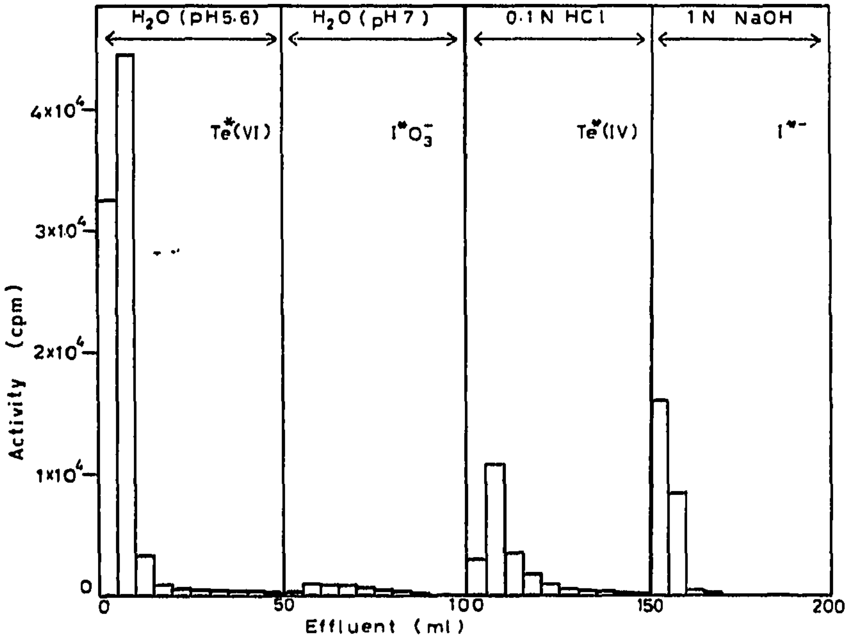

Fig. 3. Separation of Te*(VI), Te*(IV), I^{*-} and $I^*O_3^-$.

is washed with 10 ml of water, and total radioiodine is eluted (as I^{*-}) with 10 ml of 0.1 *N* sodium hydroxide.

An example of the elution curve is shown in Fig. 4; separation of Te*(VI), Te*(IV) and I^{*-} is sharp and quantitative.

Separation of I^{*-} *and* $I^*O_3^-$

The separation of I^{*-} and $I^*O_3^-$ is often necessary, for instance, in the hot-

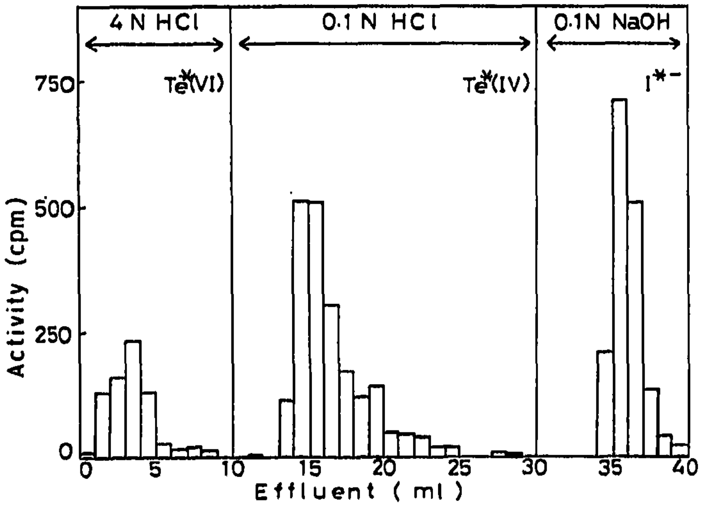

Fig. 4. Separation of Te*(VI), Te*(IV) and I^{*-}.

atom chemistry of radioiodine; both ions can be readily separated by the following procedure.

Procedure. The pH of the sample solution is adjusted to 5, then the solution is passed through the charcoal column to adsorb both I^{*-} and $I^{*}O_3^-$. The $I^{*}O_3^-$ is first eluted with 80 ml of water of pH 7, then I^{*-} is desorbed by elution with 20 ml of 1 *N* sodium hydroxide.

A clean-cut separation can be achieved, with recovery higher than 95% for each ion in the presence of carriers; carrier-free separation is also possible. An example is shown in Fig. 5.

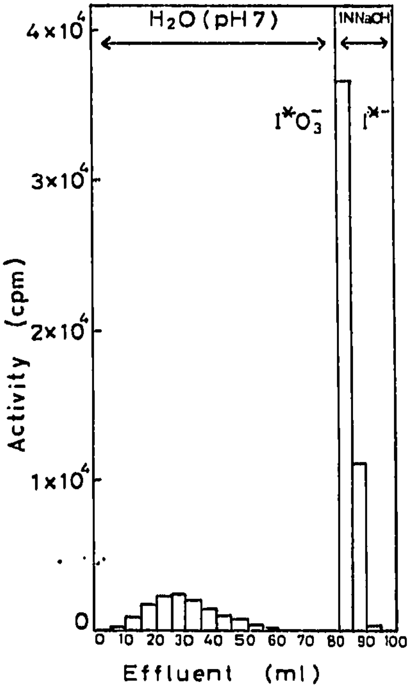

Fig. 5. Separation of I^{*-} and $I^{*}O_3^-$.

CONCLUSION

It is evident that the systematic separation of Te*(VI), Te*(IV), I^{*-} and $I^{*}O_3^-$ from each other can be achieved by use of the active charcoal column. The method can be applied to hot-atom chemical studies of radiotellurium or radioiodine, as well as to the preparation of carrier-free radioiodine from irradiated tellurium salts. Charcoal has the advantages that it is inexpensive and highly stable towards radiation.

It should also be noted that the elution behaviour of Te*(VI) and Te*(IV) from charcoal is closely similar to that reported by Inarida[8] for an anion-exchange resin; it is therefore suggested that the adsorption and desorption processes of these species are of an ionic nature.

ACKNOWLEDGEMENT

Part of the expense of this work was covered by a grant from the Ministry of Education of Japan, for which the authors' thanks are due.

REFERENCES

1 M. Levy, A. S. Keston and S. Udenfriend, *J. Amer. Chem. Soc.*, 70 (1948) 2289.
2 K. Taugböl and K. Samsahl, *Nucleonics*, 13, No. 4 (1955) 69.
3 K. Kimura, N. Ikeda, M. Inarida and H. Kawanishi, *Proc. Jap. Isotope Conf., 1st, Tokyo, 1956*, Japan Atomic Industrial Forum, Tokyo, 1957, p. 180.
4 M. Inarida, *Nippon Kagaku Zasshi (J. Chem. Soc. Japan, Pure Chem. Sect.)*, 79 (1958) 721.
5 B. Zmbova and J. Bzenić, *Anal. Chim. Acta*, 59 (1972) 315.
6 G. Tóth, *Radiochem. Radioanal. Lett.*, 7 (1971) 57.
7 N. Ikeda and T. Mitsubayashi, *Radioisotopes*, 10 (1961) 245.
8 M. Inarida, *Nippon Kagaku Zasshi (J. Chem. Soc. Japan, Pure Chem. Sect.)*, 80 (1959) 399.