Preparation of multitracer solutions from an Au target by means of a supported liquid membrane

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The transport of Au(III) and 19 other radioactive metal ions through a tributyl phosphate(TBP) - decalin membrane supported on a microporous polytetrafluoroethylene sheet is studied in order to establish a procedure for preparing multitracers from a gold target irradiated by high-energy heavy ions. It is found that Au(III) ions are transported with the highest rate from the feed solution of 3 mol dm^{-3} HCl into distilled water through the membrane, leaving the other metal ions in the feed solution as tracers. The multitracer thus obtained is in a carrier- and salt-free state. Recovery of radioactive Tc and Re from the aqua-regia distillate by the supported liquid membrane method is also investigated.

Separation of metal ions by supported liquid membranes consists of three steps: (i) complex formation of metal ions with extractants at the surface of membrane, (ii) transport of the extracted species to the opposite side of the membrane, and (iii) dissociation and liberation of the metal ions at the opposite interface. Separation by means of supported liquid membranes is considered to be one of the most useful methods for separating radioactive nuclides, because of: (i) the simplicity of operation, (ii) the feasibility of concentrating radioactive nuclides into a small volume of solution, and (iii) the necessity of a far smaller amount of extractant than in conventional solvent extraction, resulting in little organic radioactive waste.

Here we describe separation of Au from the Au-target solutions by means of a TBP-decalin membrane supported on a microporous polytetrafluoroethylene sheet.[1,2] Recovery of radioactive Tc and Re from the aqua-regia distillate by the supported liquid membrane method was also investigated.[3]

Hydrochloric acid and HNO_3 solutions containing Au(III) and various kinds of radioactive nuclides were used as feed solutions. Distilled water was used as stripping solutions. The microporous sheet used as the support for TBP-decalin was polytetrafluoroethylene with an average pore size of 0.45 μm, 75% porosity, and 0.08 mm thickness. A supported liquid membrane with a diameter of 24 mm was prepared by immersing the sheet in a TBP-decalin solution for at least 3 days. A Teflon vessel with the membrane at the bottom containing distilled water as a stripping solution was placed in a dilute HCl feed solution. The feed solution in the outer vessel was stirred with a magnetic stirrer at room temperature. A portion of the circulating stripping solution was trapped in a Teflon vessel set on the top of an intrinsic Ge detector for γ-ray measurements (Fig. 1). Measurements of 600–1200 s duration were automatically repeated for 200–300 min, and the obtained γ-ray spectra were recorded on a magnetic disk. For the permeation of Tc and Re, the distillate neutralized at various pH was used as a feed solution. Distilled water or 0.5 mol dm^{-3} $NaHCO_3$ was used as a strip solution. Measurement of 5000 s duration for the strip solution was automatically repeated.

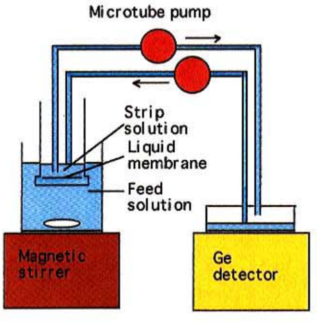

Fig. 1. Permeation apparatus.

Figure 2 shows the transport of Au(III) ions from feed solutions of various concentrations of HCl. A feed solution of 3 mol dm^{-3} HCl gave the highest transport rate, though the difference in the initial transport rates was small among the HCl feed solutions in the 1–6 mol dm^{-3} concentration range. More than 90% of the Au(III) ions were transported to the stripping solution within 100 min. On the other hand, the initial transport rates of Au(III) from feed solutions of 0.1 and 8 mol dm^{-3} HCl were smaller than those described above, though transport from these two feed solutions was observed to increase with time.

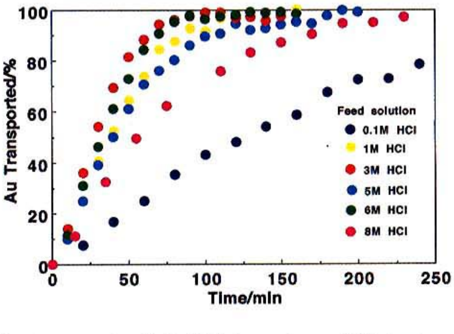

Fig. 2. The transport of Au(III) ions from HCl feed solutions to distilled water through a membrane of TBP-decalin.

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Radioactive nuclides of Sc, Fe, Zr, Nb, Te, and Hf were also found in the stripping solution after 200 min of permeation from a feed solution of 8 mol dm^{-3} HCl. The amounts of transported nuclides decreased along with a decrease in the acid concentration of the feed solution. No radioactivity due to these nuclides was detected for feed solutions of 0.1 and 1 mol dm^{-3} HCl, and only small amounts of Fe, Te, and Hf were transported to the stripping solution from a 3 mol dm^{-3} HCl feed solution. Since the 3 mol dm^{-3} HCl feed solution was found to realize a fast elimination of Au(III) ions, and to cause only a small loss in the above-mentioned nuclides during permeation, 3 mol dm^{-3} HCl is concluded to be the most suitable feed solution for preparing a multitracer solution from a gold target.

For the transport of Au(III) ions from feed solutions of HNO_3, it was found that the transport reached saturation after 70, 80, and 90% of Au(III) ions had been transported from feed solutions of 2, 1 and 0.2 mol dm^{-3} HNO_3, respectively. The transport rates of Au(III) ions from the HNO_3 feed solutions are smaller than those from 1–8 mol dm^{-3} HCl feed solutions. The radioactivities of nuclides other than Au were negligible in the stripping solutions with HNO_3 feed solutions.

On the basis of the experimental results described above, the establishment of a practical procedure for separating a multitracer from an acid solution of an Au target was attempted. As described above, Au(III) ions can be transported from both HCl and HNO_3 feed solutions. Therefore, permeation from an aqua-regia solution was examined as the final practical procedure. In this case, 4.8 g of Au target was dissolved in aqua regia. This solution was used for permeation after dilution with distilled water. In order to avoid saturation, permeation was carried out with a continuous supply of fresh distilled water to the stripping-solution compartment. Figure 3 shows the decrease of Au in the feed solution with the permeation time. About 99% of the Au(III) ions were removed from the feed solution within 70 h. Only peaks due to the radioactive Au were detected in the stripping solution, showing selective permeation of the Au(III) ions.

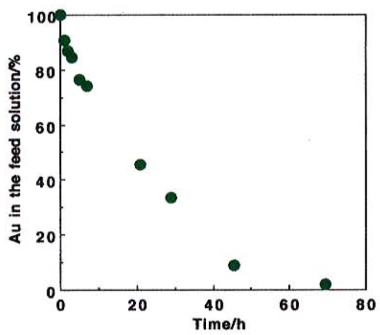

Fig. 3. Transport of Au(III) ions (4.8 g) from a feed solution of 1.8 mol dm^{-3} in HCl and 0.7 mol dm^{-3} in HNO_3 to distilled water through a membrane of TBP-decalin.

Radioactive Tc(VII) and Re(VII) were transported from feed solutions having a pH range between 0 and 1.36 to distilled water or a 0.5 mol dm^{-3} $NaHCO_3$ solution by means of the TBP-decalin membrane. The $NaHCO_3$ solution was shown to be better than distilled water as a strip solution giving higher rates of transport. Figure 4 shows dependence of the transport of Tc on pH of the feed solutions. Only a slight difference in the initial rate of Tc transport was observed for the feed solutions at pH ranging from 0–1.36. However, the transport of Tc from the feed solution at pH 0 showed signs of saturation soon after the start of permeation. Transport of Tc from the feed solution at pH 0.54 increased gradually with time. In the permeation using the feed solution at pH 0.74, the strip solution was replaced by fresh 0.5 mol dm^{-3} $NaHCO_3$ solution 24 h after the start of transport. After replacement, transport yield was enhanced as shown in Figure. The transport behavior of Re is similar to that of Tc; however, the amount of Re transported at a given time is smaller than that of Tc. It was found that the pH of the feed solutions exerts only a slight influence on the initial transport of Tc and Re but that the amount of Tc and Re transported from a feed solution at lower pH reaches saturation at an earlier stage of permeation. Therefore, a feed solution at around pH 1 is suitable for the transport of Tc and Re.

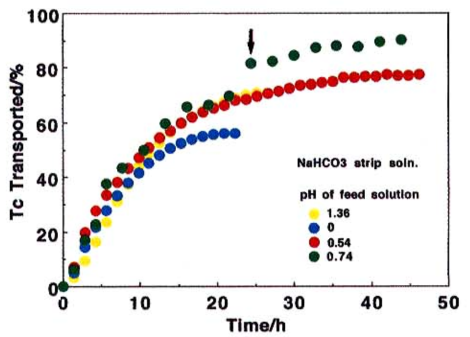

Fig. 4. pH dependence of the transport of Tc(VII) through the membrane of TBP-decalin to 0.5 mol dm^{-3} $NaHCO_3$ solution. An arrow indicates replacement of the strip solution with a fresh one in the permeation using the feed solution at pH 0.74.

References

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