CHEMISTRY LETTERS, pp. 149-152, 1991.

Preparation of a Radioactive Multitracer Solution from Gold Foil Irradiated by 135 MeV/nucleon ^{14}N Ions

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An irradiation facility and a radiochemical procedure for preparing a radioactive multitracer solution are reported. Gold foil was irradiated with 135 MeV/nucleon ^{14}N-ion beam. The foil was dissolved in aqua regia and precipitation of gold as metal with hydrogen peroxide yielded a carrier- and salt-free multitracer solution.

Radioactive nuclides are extensively used as tracers in science, technology, and medicine. The tracers are usually utilized individually. Combined use is limited[1,2] except for ^{3}H and ^{14}C common in biochemical studies, although simultaneous use of a number of radioactive tracers is preferable in many cases. It enables us not only to acquire efficiently data for various elements but also to determine the characteristic behavior of different elements under an identical experimental condition. The advance in solid-state detectors, counting circuits, and computers has made it possible to measure and analyze complex γ-ray spectra of many radioactive nuclides. On the other hand, no procedure has been established yet to produce a multitracer solution ready for application to various fields of science. Although fission of heavy elements such as uranium results in many radioactive nuclides, they are not suitable as a multitracer because most of the nuclides undergo a series of β-decays to change their atomic number.

Recently, a ring cyclotron (Riken Ring Cyclotron, RRC) was constructed in our institute, which can accelerate heavy ions up to 135 MeV/nucleon. The nuclear reactions induced by heavy ions of this energy range result in simultaneous production of a large number of radioactive nuclides. We designed and constructed an irradiation facility for rapid and safe transportation of irradiated targets and also developed a convenient and reliable radiochemical procedure to prepare a multitracer solution free from both carriers and salts.

Figure 1(a) is a schematic view of the irradiation facility, the falling ball irradiation system. The transport tube passes through the floor of the irradiation room down into a hood in the hot laboratory. The chamber and the transport tube can be

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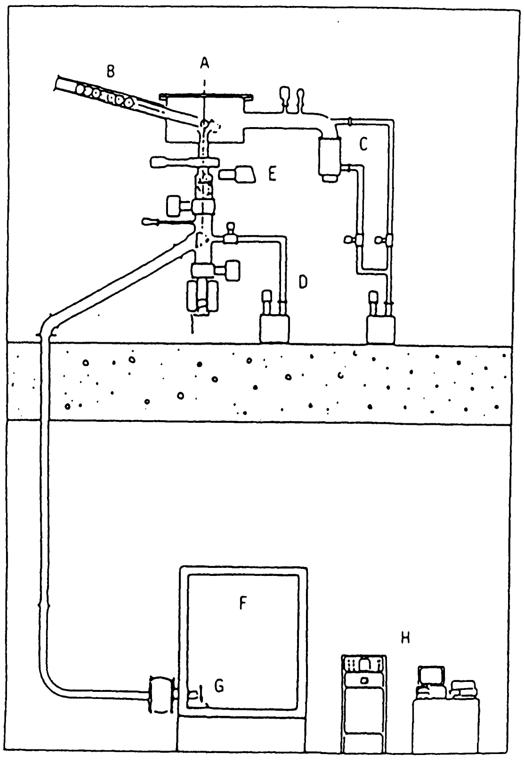

(a)

(b)

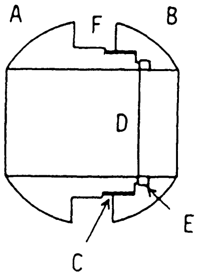

Fig. 1. (a) The whole layout of the falling ball irradiation system. Upstairs: Irradiation room (A: irradiation chamber, B: stand-by holder tube for the balls, C, D: vacuum systems, E: radiation monitor). Downstairs: Hot laboratory (F: hood, G: end station for the balls, H: control system). (b) The falling ball. A, B: Separable two parts of the ball, C: screw groove, D: target foil, E: spacer ring. F: groove for guide.

evacuated to $1\text{x}10^{-5}$ and $1\text{x}10^{-1}$ Pa, respectively. Targets are mounted in the falling balls as shown in Fig. 1(b). The balls (diameter 42 mm) are made of aluminum and have a piercing cylindrical hole for the heavy-ion beam. They are transferred one by one from the stand-by tube to the irradiation position of the chamber, the direction of the target against the beam being properly kept with a groove on the ball and rails in the chamber. After irradiation, the ball is let fall onto a radiation monitoring station under the chamber, and is further rolled down to the end station in the hood, where the target is ready for chemical treatments within one minute after the end of irradiation.

In the present work, a stack of 24 mm Φ x 50 μm thick gold foil was irradiated with a $^{14}\text{N}^{7+}$ beam of 1.89 GeV (135 MeV/nucleon). The beam current was about 100 nA and the beam profile was roughly 10 mm x 10 mm. The irradiation time was 150min.

In separation of the reaction products after the irradiation of gold with 2.6 GeV protons, Szweryn et al. extracted gold with diethyl ether from an aqua regia solution followed by precipitation as metal with sulfur dioxide from an aqueous solution.$^{3)}$ We employed only precipitation of gold metal with hydrogen peroxide in our separation procedure in order to minimize contamination of various elements and to make the procedure simple. Three pieces of the irradiated gold foil were dissolved in aqua regia. The solution was evaporated to dryness under a reduced pressure of about 4 kPa in a closed vessel with a trap. The residue was dissolved in conc. hydrochloric acid. Gold ions in the solution were reduced to metal with hydrogen peroxide and the precipitate was filtered. The same procedure was repeated several times to ensure complete removal of gold from the solution. The residue was finally dissolved in 6 mol dm^{-3} hydrochloric acid to yield a carrier- and salt-free multitracer solution containing a variety of radioactive nuclides produced in the gold target.

The γ-ray spectra of the multitracer solution and the precipitated gold metal were measured with a pure Ge-detector of 18% relative efficiency. Figures 2 and 3 show the spectra of the solution and the gold precipitate, respectively. The spectra were analyzed with the "BOB76" code$^{4)}$ on FACOM M780 computer of the institute. Only radionuclides of gold were detected in the fraction of precipitated gold, while no radioactive nuclides of gold were found in the multitracer solution. The present observations indicate the complete separation of the radioactive nuclides other than those of gold from the target. The kind of elements traceable by the nuclides in the solution depends on time after the irradiation, because radionuclides of different half-lives are produced. The dominant peaks observed in the spectrum of the tracer solution shown in Fig. 2 are those due to ^{67}Ga, ^{89}Zr, ^{87}Y, ^{99}Mo, ^{103}Ru, ^{111}In, ^{131}Ba, ^{145}Eu, ^{146}Eu, ^{146}Gd, ^{147}Gd, ^{149}Gd, ^{152}Tb, ^{153}Tb, ^{155}Tb, ^{166}Tm, ^{167}Tm, ^{169}Lu, ^{171}Lu, ^{170}Hf, ^{173}Hf, ^{181}Re, ^{183}Os, ^{195m}Hg, and so on. The solution collected in the trap of evaporator contained ^{181}Re and ^{195m}Hg.

It is noted that a multitracer solution containing neither carriers nor salts is obtained by the present radiochemical procedure. This is a great advantage in applying it to a variety of chemical researches. Now, application of it to a model experiment on marine chemistry is in progress. We also plan to prepare multitracer solutions covering the elements in different ranges of atomic number by combining different projectiles and targets as well as appropriate radiochemical procedures.

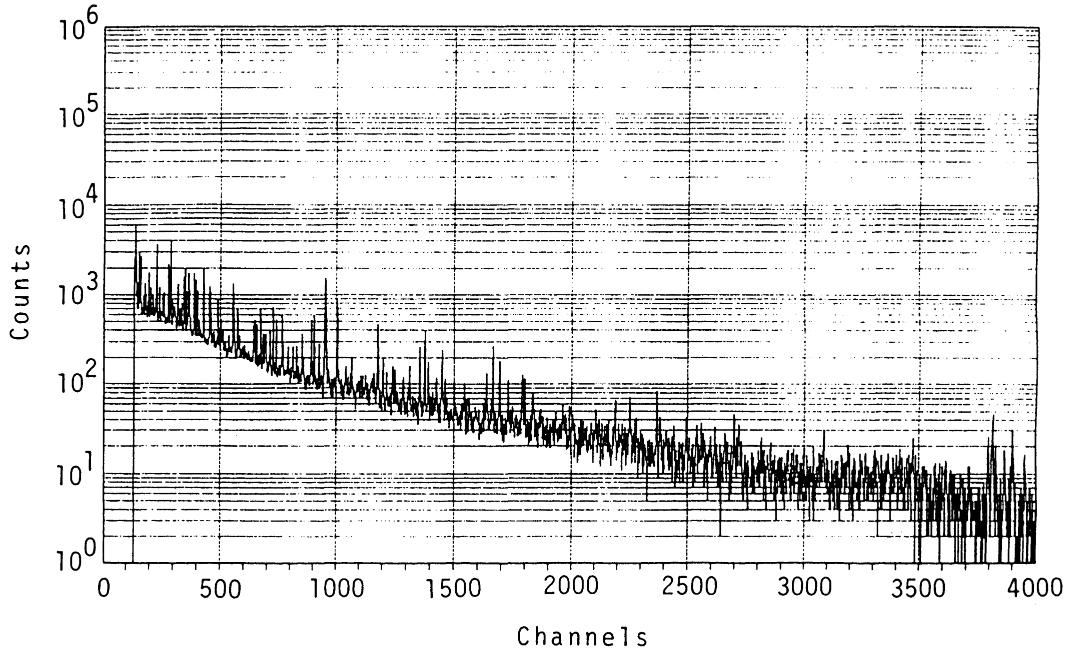

Fig. 2. The γ-ray spectrum of the multitracer solution measured 4 days after irradiation of the gold target with 1.89 GeV $^{14}N^{7+}$ beam. One channel corresponds to 0.538 keV.

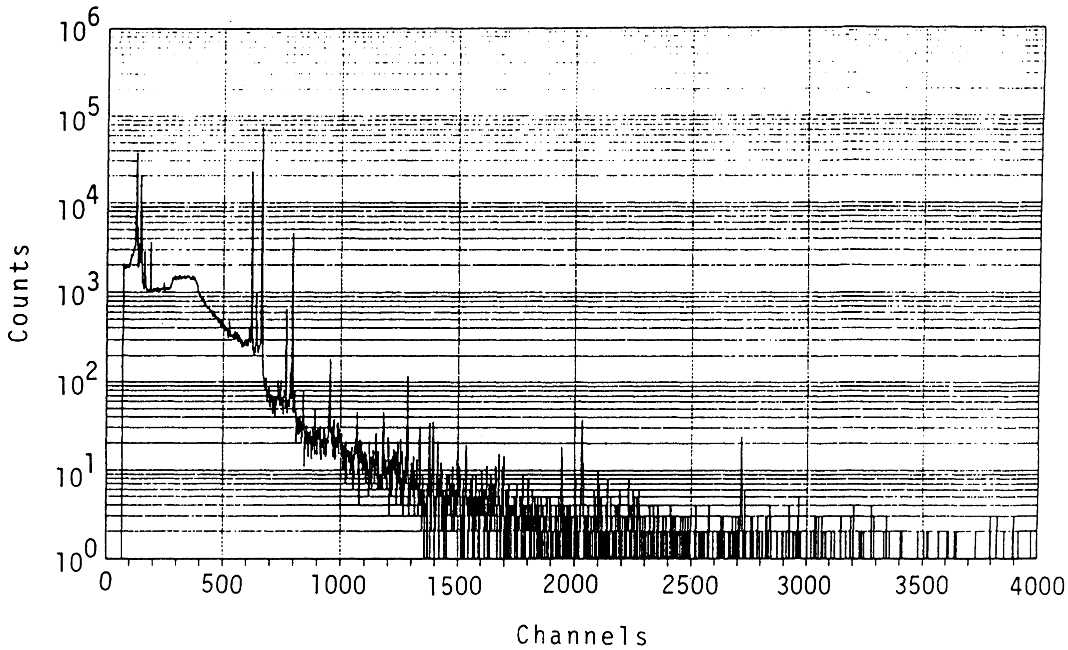

Fig. 3. The γ-ray spectrum of the gold precipitate measured 21 days after the irradiation. One channel corresponds to 0.537 keV.

We thank heartily Drs. Y. Yano, A. Goto, and K. Hatanaka for their helpful cooperation in the irradiation.

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(Received September 1, 1990)