# Development of the Multitracer Technique by Using Riken Ring Cyclotron

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**Abstract**

A versatile radioactive multitracer technique has been developed at Riken Ring Cyclotron. It enables efficient and simultaneous tracing of a number of elements under identical experimental conditions. Its principle and features are presented with example of recent application.

The Riken Ring Cyclotron can accelerate heavy ions up to 135 MeV/nucleon. Such energetic heavy ions produce a number of radioactive nuclides at once in a target by nuclear fragmentation. We have established the multitracer technique, which enables simultaneous tracing of a number of elements in various chemicals and biological systems. Here, its principle, features, and recent developments in preparation and biological applications of multitracers are presented.

The multitracer technique enables us not only to acquire efficiently data for various elements, but also to determine the characteristic behavior of different elements under identical experimental conditions, The principle is to irradiate a metal target by high-energy heavy ions, to dissolve it in acid, and to remove chemically the target material yielding a solution of a number of radioactive isotopes, namely, a multitracer solution. The solution can also be salt-free, if an appropriate separation procedure is employed.

A target stack of gold, silver and other metals are irradiated with a 135 MeV/beam of $^{12}C$, $^{14}N$, or $^{16}O$ ions accelerated by Riken Ring Cyclotron. The irradiated gold foil is dissolved in aqua regia. The solution is evaporated to dryness under a reduced pressure in a closed vessel with a trap. The residue is dissolved in 3 mol/dm$^3$ HCl and Au(III) is removed from the solution by extraction with ethyl acetate. Radioactive nuclides of Br, Sn, I, Re, and Os are recovered from the solution in the trap. The irradiated Ag foil is dissolved in (1:1) $HNO_3$ and silver is simply precipitated as AgCl with conc.HCl and filtered. The multitracers thus prepared are subjected to various chemical and biological experiments. The γ-rays of radioactive nuclides in the samples are measured by means of pure Ge detectors. The assignment of each peak of the γ-ray spectra to different nuclides is made on the basis of its energy and half-life.

A wide spectrum of radioisotopes is produced by irradiation of a target with high-energy heavy ions depending on the kind and energy of the heavy ions and the target materials. Both neutron-deficient and neutron-rich nuclides are produced, the former being dominant, with cross sections for isobars amounting roughly to 10 to 100mb. These nuclides are mainly the products of target fragmentation, covering elements with atomic numbers up to that of the target. Radioactive nuclides with an atomic number larger by one than that of the target are also produced as a result of proton transfer. Mass yield distribution, recoil properties, and angular distributions of the reaction products have been extensively studied for various combinations of heavy-ion projectiles and target elements. However, there had been no attempt before ours to utilize these mixtures of nuclides for chemical and other purposes.

The kind of elements traceable by the nuclides in the multitracer solution obtained from a target depends on time after the irradiation, radionuclides of different half-lives being produced. Because of the limited resolution of the detectors available at present, overlap of γ-ray peaks is inevitable in case of targets of heavy metals such as gold. Overlapped peaks can be resolved into two or more components by analyzing the decay curve. But, reliability of the data obtained by such analysis is not necessarily high and many of them must be discarded. Therefore, radiochemical procedures for separating the multitracer obtained from a gold target into two or three groups of elements have been developed. The elements traced so far by the multitracer technique are shown in Figure 1.

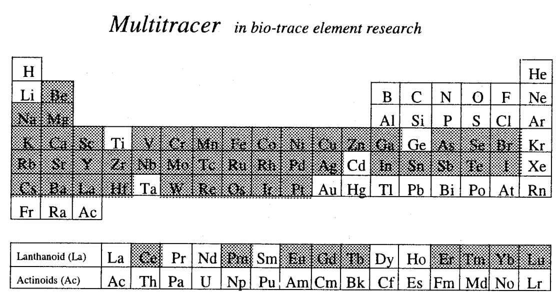


Fig. 1 Elements traced so far by the multitracer technique (shadowed ones).

The multitracer technique has been applied to various researches in the fields of basic chemistry, environmental chemistry, physiology and biochemistry of the bio trace elements in plants and animals in cooperation with many research groups of universities and research institutes. Among the subjects studied are:

(1) Distribution and excretion of various elements in normal, Zn-deficient, Ca-deficient, and Se-deficient rats,
(2) Metabolic physiological study on non-insulin dependent diabetes mellitus model, and hypercholesterolemic model mice
(3) Specific accumulation of vanadium in mice bone,
(4) Labeling of immunoglobulin G with a multitracer,
(5) Trace elements distribution in tumor-bearing mice,

(6) Radioimmunological and pharmaceutical studies on DOTA-coupled antibody,
(7) Biodistribution of gold complexes in mice,
(8) Biochemical properties of the platinum group elements,
(9) Metabolic physiology of the rare-earth elements,
(10) Metabolic physiology of various elements in alcoholic mice,
(11) Distribution of various elements in vitamin D overloaded and deficient rats,
(12) Biodistribution of various elements in Hepatopathic animals,
(13) Binding activities of various elements to blood components, metallothionein, and serum proteins,
(14) Comparative regional accumulation of a multitracer in brain of mice,
(15) Reduced uptake of Mn in Cd-resistant metallothionein null fibroblasts,
(16) Uptake of trace elements in Yeast *Saccharomyces cerevisiae*,
(17) Transfers of various trace elements from the placenta to the fetus of pregnant rats,
(18) Metabolic physiology on space field by using the multitracer,
(19) Transport of metal ions in rice, soybean, fern, turnip, carrots, marigold, and tobacco,
(20) Formation of metallofullerenes studied by the multitracer technique,
(21) Model study of acid rain effect on adsorption of trace elements on soil,
(22) Multitracer studies on the extraction and chemical separation.

Examples of more recent application will be presented in the lecture.

In conclusion, the results described above demonstrate the versatility of the multitracer technique in studying the behavior of a number of elements in various systems. The multitracer technique has opened a new way of application of heavy-ion physics to chemistry, biology and medicine.

Now, Riken accelerator research facility undertakes construction of a RI-beam Factory (RIBF) capable of providing the world's most intense RI beams at energies of several hundreds MeV/nucleon over the whole range of atomic masses. This factory is expected to realize a new era of not only nuclear physics, but also nuclear and radiochemistry, including development of new tracer technique.