Nuclear Chemistry Laboratory

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When the RIKEN Ring Cyclotron (RRC) was ready for utilization, I proposed two new research subjects for my nuclear chemistry group: the development of a multitracer technique and in-beam Mössbauer spectroscopy using ^{57}Mn.

The former involves the preparation of multielemental radioactive nuclides by the fragmentation of target nuclei and their application to trace the behavior of elements in various research fields. In 1990, we started a series of studies to develop radiochemical procedures for the preparation of multitracers, namely the removal of the target material from the solution of an irradiated target, leaving as much as possible of the radionuclides, and the application of the multitracers thus obtained to chemistry and other fields.[1)] More than fifty elements were studied using this technique, with twenty or more of them being traced simultaneously.

Significant features of the multitracers are as follows: (1) High efficiency: a single experiment usually yields information on twenty or more elements. (2) Accurate comparison of the behavior of a number of elements is realized under strictly identical experimental conditions. (3) Serendipity: since many elements are included in every multitracer, it is often possible to discover new important or interesting behavior of an element, which is not the main subject of the investigation. (4) All the tracers are carrier-free, that is, the multitracers contain slight amounts of stable isotopes of the tracers, which can disturb the experiment. (5) Some multitracers contain scarcely obtainable tracers such as ^{28}Mg and ^{47}Ca.

Typically, a beam of $^{12}C^{6+}$, $^{14}N^{7+}$, or $^{16}O^{8+}$ ions with an energy of 135 MeV/nucleon is used against Au, Ag, and Cu targets. The irradiated target is dissolved, and the target material is removed, leaving the radioactive nuclides in a solution. For example, in the simplest case of an Ag target, it is dissolved in nitric acid, and Ag is removed as AgCl precipitate. The measurement of the tracers is performed by means of conventional Ge detectors.

The multitracers were used for the investigation of behavior of various elements in chemistry, biochemistry, and biology in cooperation with more than 10 groups in universities and research institutes.[1,2)] The behavior of elements in rats and mice was most extensively studied by different groups. The uptake, excretion, and retention of various elements were studied in organs of Al-, Cd-, and Vitamin D-overloaded or Se-deficient rats in comparison with those of normal ones. The following studies were also made on various elements: the transportation through the blood-brain barrier and retention in different parts of the brain, labeling of immunoglobulin G, reactions with blood components and transport proteins, and in vivo and in vitro binding with liver DNA.

In the case of plants, the uptake and accumulation of elements in mushrooms, carrots, and marigolds as well as the subcellular distribution and translocation of radionuclides were studied. In chemistry, the adsorption of metal ions on α-Fe_2O_3, formation of metallofullerenes with higher-group elements, effects of model acid rain on the adsorption of trace elements upon soils, and stability constants of humate complexes with various metal ions were studied.

In this period, investigations using unstable nuclei as probes, namely Mössbauer spectroscopy and time-differential perturbed angular correlation (TDPAC) of γ rays, were continued with the use of cyclotron-produced radionuclides in cooperation with the magnetism group of RIKEN, and a project using short-lived ^{57}Mn produced by RRC was newly started.[3)] With the use of the source nuclide ^{61}Cu produced by the α-irradiation of a Ni-V alloy disk, ^{61}Ni-Mössbauer spectra of chromite spinels, $Cu_{1-x}Ni_xCr_2O_4$, were measured. Both H_{hf} and EFG were found to be well related to a lattice distortion. In $Cu_{0.9}Ni_{0.1}Cr_2O_4$, the H_{hf} was 790 kOe, which is the largest among the values for Ni^{2+} ever reported. Carrier-free ^{99}Rh produced by the proton irradiation of ^{99}Ru was chemically separated from the target and was doped in $YBa_2Cu_3O_{6.8}$ and $YBa_2Cu_3O_6$. Their Mössbauer emission spectra and TDPAC showed that ^{99}Ru ions arising from ^{99}Rh are tetravalent and occupy the Cu sites.

^{119}Sn-Mössbauer emission spectroscopy using cyclotron-produced ^{119}Sb as the source nuclide was applied to the in situ study of the chemical state of $^{119}Sb^{5+}$ adsorbed on α-Fe_2O_3. The state of $^{119}Sb^{5+}$ was shown to change depending on the pH value of the solution in contact with the powder samples on the basis of broadening of the spectra due to the magnetic interaction.

A setup for in-beam Mössbauer spectroscopy was constructed at RIPS of RRC in order to measure the spectra of ^{57}Fe with ^{57}Mn (half-life 87.2 s) produced as a beam by the projectile fragmentation of ^{59}Co. With the use of a parallel-plate avalanche counter for detection of the internal conversion electrons, we succeeded in the first observation of Mössbauer spectra of ^{57}Fe following the decay of ^{57}Mn nuclei. However, the quality of the spectra was not satisfactory because of β-rays from ^{57}Mn. Later, Dr. Y. Kobayashi and his coworkers overcame this difficulty by developing an anti-coincidence detection system and also realized time-dependent measurement with the system. Now, ^{57}Mn-Mössbauer spectroscopy is used as a unique method for atomic-scale study of reactions in solids.

References
1) F. Ambe, ed., RIKEN Rev. No. **13** (1996).
2) Y. Yano, S. Enomoto, eds., RIKEN Rev. No. **35** (2001).
3) F. Ambe, ed., RIKEN Rev. No. **16** (1997).

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