

Preparation of Multitracer by Thermal Neutron Fission of  $^{235}\text{U}$ Koichi Takamiya,<sup>a</sup> Masaaki Akamine,<sup>a</sup> Seiichi Shibata,<sup>\*,a</sup> Atsushi Toyoshima,<sup>b</sup> Yoshitaka Kasamatsu,<sup>b</sup> and Atsushi Shinohara<sup>b</sup><sup>a</sup>Research Reactor Institute, Kyoto University, Kumatori-cho, Sennan-gun, Osaka 590-0494, Japan<sup>b</sup>Department of Chemistry, Graduate School of Science, Osaka University, Toyonaka, Osaka 560-0043, Japan

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Radiochemical procedures for preparing a multitracer using thermal neutron fission of  $^{235}\text{U}$  were established. The target material of  $^{nat}\text{UO}_2$  mixed with the catcher material for the fission products was irradiated at the research reactor (KUR) of the Research Reactor Institute of Kyoto University. After irradiation, the multitracer solution mainly consisting of fission products was prepared by quick removing of the target material. The multitracer enables simultaneous tracing of various elements, and comparison of their chemical behavior under strictly identical experimental conditions.

In the field of medical, pharmaceutical, biological and environmental science, radioactive isotopes are employed to trace the chemical behavior of various elements in a system. Although the trace analysis method using radioactive isotopes is highly sensitive, only one or two sort of tracers is usually applied in the experimental research. Recently, the research group at the Institute of Physical and Chemical Research (RIKEN) reported the preparing method of tracer solutions containing multiple radioactive isotopes (multitracer) without carrier materials.<sup>1-3</sup> They are preparing the multitracer solutions by heavy-ion reaction using the RIKEN Ring Cyclotron, and providing them to scientists in the various research fields. However, available elements are limited by their own half-lives and reaction yields. In the heavy-ion reaction, neutron-deficient isotopes are dominantly produced. Therefore, another method to prepare multitracers containing radioactive isotopes, which cannot be produced by heavy-ion reactions, is urgently required.

We tried to prepare a multitracer by thermal neutron induced fission of  $^{235}\text{U}$  using Kyoto University Reactor (KUR), because in nuclear fission neutron-rich isotopes are produced in relatively high yields in comparing with charged-particle induced reactions. A use of a multitracer prepared by nuclear fission of  $^{235}\text{U}$  was reported by Uchida et al.<sup>4</sup> They applied the multitracer solution without chemically separating the target material of uranium to the adsorption study of fission products onto ion exchangers. In this work, we newly developed the preparation method for the multitracer solution by quick removing of the target material of uranium.

A mixture of  $^{nat}\text{UO}_2$  and catcher material by which the fission products recoiled out from the  $\text{UO}_2$  were captured was irradiated by thermal neutrons in the reactor. After irradiation, the catcher material was dissolved in a dilute acid, and the  $\text{UO}_2$  undissolved was removed by filtering. The multitracer solutions prepared were provided for the purpose of medical research at Kanazawa University.

In this note, we report the radiochemical procedures established for preparing the multitracer and the results of investigating some salts as efficient catcher materials.

In order to prepare the multitracer solution efficiently, the following parameters for the sample preparation were examined; 1) catcher material, 2) mixing ratio of  $\text{UO}_2$  to catcher material, 3) particle size of the catcher material and 4) sample shape for irradiation. The catcher materials used in this work

were NaCl, LiF and  $\text{MgCl}_2$  by considering that these salts are soluble in water and easy for a quick chemical treatment. The mixing ratios ( $[\text{UO}_2]/[\text{catcher material}]$ ) examined were as follows; 1:1, 1:3, 1:10 and 1:30. The mixture was ground in an agate mortar to make it uniformed size. The particle sizes were controlled by the grinding time. In this work, we refer to the particle size as coarse and fine depending on the grinding time. For irradiation, powdered and pellet samples were prepared. The pellet samples were prepared using the fine ground powder. The content of samples is tabulated in Table 1.

The sample prepared was wrapped with an aluminum foil and sealed into an evacuated quartz tube. The tube was inserted into a polyethylene capsule and irradiated using the pneumatic transferring system (Pn-2) at KUR. The thermal neutron flux at Pn-2 is reported to be  $2.8 \times 10^{13}$  n/cm<sup>2</sup>/sec. The irradiation times were 1-10 min as indicated in Table 1.

TABLE 1: Contents of Irradiated Samples

Catcher Material	Form	Weight /mg $\text{UO}_2$ :Catcher	Irrad. Time
NaCl	Powder (coarse)	0.95 : 2.95	10min
		1.12 : 9.68	1min
		1.06 : 29.74	1min
	Powder (fine)	1.09 : 1.11	5min
		1.07 : 3.33	5min
		1.05 : 29.45	1min
		Pellet	5.12 : 5.18
	2.48 : 7.72		2min
	MgCl <sub>2</sub>	Powder (fine)	1.25 : 10.85
0.37 : 10.43			2min
1.00 : 29.61			5min
LiF	Powder (fine)	0.90 : 9.60	5min

After cooling for a few tens of minutes, the irradiated samples were picked up from the capsule. The fission products captured by the catcher material were dissolved in 0.1 N HCl, while the  $\text{UO}_2$  remained undissolved. The solution containing fission products was filtered with suction to separate the  $\text{UO}_2$ . Gamma rays from an aliquot of the solution and the  $\text{UO}_2$  on the filter paper were measured by a Ge detector. The peak areas of spectra were calculated using the BOB peak analysis code.<sup>5</sup> Assignment of the radioactive isotopes to each peak in the spectra was carried out on the basis of their energies and half-lives. Several measurements were usually

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performed for the sample at appropriate time intervals in order to determine the half-lives of the radioactive isotopes responsible for each peak. Because of the limited energy resolution of the Ge detector and the interference due to Compton scattering of the gamma rays from by-products induced by the catcher materials, the radioactive isotopes emitting low energy gamma rays and with low production yields were not detected precisely. Therefore, the separation method for the catcher material from the multitracer solutions is under investigation. An example of identified isotopes is as follows;  $^{91}\text{Sr}$ ,  $^{95,97}\text{Zr}$ ,  $^{105}\text{Ru}$ ,  $^{106}\text{Rh}$ ,  $^{131,132}\text{I}$ ,  $^{132}\text{Cs}$ ,  $^{140}\text{Ba}$ ,  $^{140}\text{La}$  and  $^{141,143}\text{Ce}$ .

Capturing efficiency of fission products by catcher materials was estimated by a ratio of  $Y_{\text{catcher}}/Y_{\text{total}}$  for each product nuclide, where  $Y_{\text{catcher}}$  indicates the yield of the nuclide captured in catcher materials and  $Y_{\text{total}}$  the total yield estimated by summing  $Y_{\text{catcher}}$  and the yield observed for the filter paper. There was no difference among the efficiency of each nuclide within the experimental error. The capturing efficiency of the whole sample is evaluated from the averaged value of the efficiencies for all identified nuclides. The obtained mean capturing efficiency for each sample is tabulated in Table 2.

Figure 1 shows the mean efficiency for each sample as a function of the weight ratio of  $W_{\text{catcher}}/W_{\text{total}}$ , where  $W_{\text{catcher}}$  and  $W_{\text{total}}$  are the weight of the catcher material and of the sample, respectively. Open and closed circles in Figure 1 show the efficiencies obtained for coarse and fine powder samples with the catcher material of NaCl, respectively. Square and diamond in the figure show those with LiF and MgCl<sub>2</sub>, respectively. The efficiencies obtained for pellet samples are shown as triangles. Solid and dashed lines indicate the results of the least square fitting for the mean efficiencies of powder and pellet samples with NaCl, respectively.

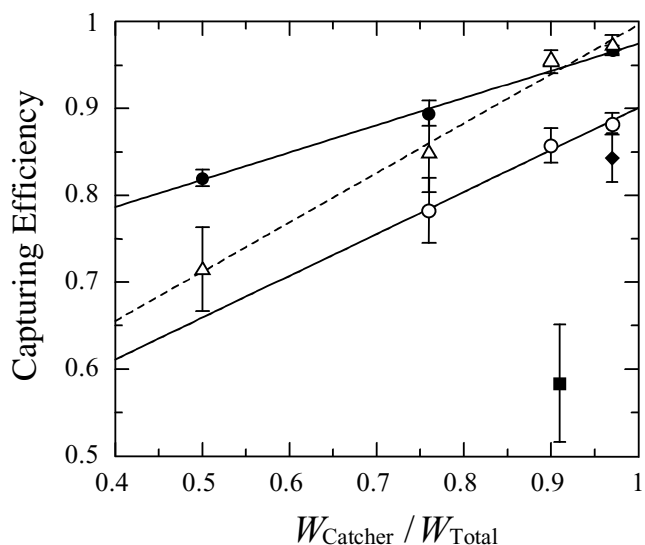
The capturing efficiency in the case of LiF is much lower than that of NaCl or MgCl<sub>2</sub>. This might be dependent on mean atomic size of catcher material, since that of LiF is smaller than that of NaCl or MgCl<sub>2</sub>. The sample preparation with MgCl<sub>2</sub> was much more difficult than that with NaCl, because of its high hygroscopicity. Comparing coarse and fine powder samples with NaCl, the capturing efficiencies for the fine powder sample are higher than those for the coarse one. This may be explained by that fission fragments produced in fine particles can recoil out easily than those in coarse particles. The capturing efficiencies for pellet samples are almost the same as those for fine powder samples except for the sample of low weight ratio. Since the UO<sub>2</sub> tends to contaminate more the multitracer solution from the sample of low weight ratio than that from the one of high ratio, an identification of fission products is relatively difficult in the case of the low weight ratio sample. The larger statistical fluctuation of the capturing efficiency for the sample of low weight ratio than that for the sample of high ratio is probably understood by the reason above.

As shown in Figure 1, the sample mixed with NaCl as a catcher material is most efficient for preparing multitracer solutions. Concerning the handling of samples, the pellet sample is more convenient than the powder sample. In case of pellet samples, it is possible to irradiate several samples simultaneously. The multitracer solutions prepared above are extensively used for the biological research to trace the element distribution in mice. In this research, the multitracer solutions obtained from the sample containing NaCl as a catcher material are especially useful, because a physiological salt solution can be easily prepared. The detail of this work will be reported elsewhere. Besides the above biological research, applications

to environmental, pharmaceutical and cytological sciences are now being planned.

**TABLE 2: Mean Capturing Efficiencies**

Catcher Material	Form	Weight /mg UO <sub>2</sub> :Catcher	Weight Ratio Catcher/Total	Capturing Efficiency
NaCl	Powder (coarse)	0.95 : 2.95	0.76	0.78 ± 0.04
		1.12 : 9.68	0.90	0.86 ± 0.02
		1.06 : 29.74	0.97	0.88 ± 0.01
	Powder (fine)	1.09 : 1.11	0.50	0.82 ± 0.01
		1.07 : 3.33	0.76	0.89 ± 0.01
		1.05 : 29.45	0.97	0.97 ± 0.00
		5.12 : 5.18	0.50	0.72 ± 0.05
	Pellet	2.48 : 7.72	0.76	0.85 ± 0.05
		1.25 : 10.85	0.90	0.95 ± 0.01
0.37 : 10.43		0.97	0.97 ± 0.01	
MgCl <sub>2</sub>	Powder (fine)	1.00 : 29.61	0.97	0.84 ± 0.03
LiF	Powder (fine)	0.90 : 9.60	0.91	0.58 ± 0.07



**Figure 1.** The mean capturing efficiency for each sample correlated with the weight ratio. Open and closed circles show the efficiencies obtained for coarse and fine powder samples with the catcher material of NaCl, respectively. Square and diamond in the figure show those with LiF and MgCl<sub>2</sub>, respectively. The efficiencies obtained for pellet samples are shown as triangles. Solid and dashed lines indicate the results of the least square fitting for the mean efficiencies of powder and pellet samples with NaCl, respectively.

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