Nuclear Chemistry in Japan

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Nuclear chemistry studies in Japan are briefly reviewed. Main topics are concerned with the works related to nuclear fission and discoveries of new radioisotopes. The other works, such as photonuclear reaction, high energy light ion-induced reaction, intermediate energy heavy ion-induced reaction, and neutron-induced reaction, are just outlined. The future plan for transactinide elements is also briefly introduced.

1. Introduction

In recent years, the field of nuclear chemistry in Japan has greatly progressed in accordance with the development of accelerators and their experimental facilities.

Researches on nuclear chemistry are mainly presented at the Symposium on Radiochemistry in Japan. In order to see a general view of the activities of nuclear chemistry in Japan, the papers presented at the Symposium on Radiochemistry for the last ten years are classified into several groups and displayed in Figure 1. Here the researches purely related to nuclei, such as nuclear reactions and decay properties, are classified as nuclear chemistry work. On the average, about 25 papers have been presented in a year, and almost half of them are related to nuclear fission.

In this paper, nuclear chemistry studies in Japan, including the future plan for transactinide elements, are briefly summarized.

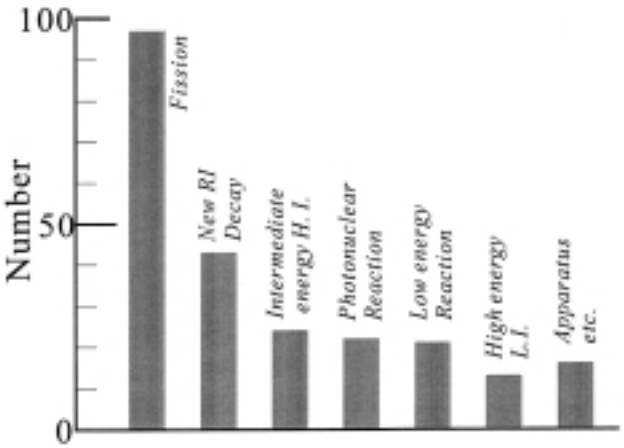

Figure 1. Number of papers related to nuclear chemistry presented at the Symposium on Radiochemistry for the last ten years from 1989 to 1998.

2. Brief Summary of Present Research

2.1. Nuclear Fission. Nuclear fission has been traditionally studied for a long time in Japan. Many precise and systematic measurements have been performed in the recent studies by the use of various experimental facilities.

The group of Osaka University has measured many short-lived fission products in proton-induced fission of ^{238}U by the use of a gas-jet transport system together with an conventional stacked foil method and discussed the fission mechanism from the charge and mass distributions of fission products.[1,2] They also carried out the double-velocity and double-energy measurement (2V2E) for thermal-neutron-induced fission of ^{233,235}U and ^{239}Pu in order to obtain the information of scission point configurations.[3] From the double-velocity data, masses of the primary fragments can be deduced with an assumption of isotropic emission of neutrons, while the double-energy data yield the secondary fragment masses. Thus, the number of neutrons $\bar{\nu}$ from each fragment can be obtained indirectly. They estimated the excitation energy of each fragment from $\bar{\nu}$ and neutron separation energies, and deduced the degree of deformation at scission assuming that the excitation energy only comes from the deformation energy of the nascent fragment of a spheroidal shape. They concluded that there exists two configurations at scission point, one corresponding to a deformed heavy fragment and the other to a spherical one.

Direct measurement of neutrons in thermal-neutron-induced fission of ^{235}U was carried out by the group of Kyoto University and Kyoto University Research Reactor Institute.[4,5] They measured energy and multiplicity of neutrons coincident with each fission fragment. The obtained energy specta of neutrons exhibit Maxwellian type distributions from which nuclear temperatures can be deduced. Combined with the excitation energy estimated from multiplicity data, one can deduce level density parameters of fission fragments.

The group of Niigata University measured short-lived fission products by the use of an ion guide isotope separator on-line (IGISOL) which was installed at the Cyclotron and Radioisotope Center of Tohoku University. IGISOL has no ordinary ion source. Reaction products are stopped in a helium buffer gas and remaining recoil ions of +1 charge state are guided by an electric field and extracted to the separator by a skimmer system. By the modification suitable for the measurement of fission products,[6] isomeric yield ratios for many fission products were obtained in proton-induced fission of light actinides.[7,8] The obtained isomeric yield ratios were converted to angular momenta of fission fragments and examined from various points of view. They found a good correlation between angular momenta and excitation energies of fission fragments. And the incident energy dependences of angular momenta were found to be different between symmetrically divided products and asymmetrically divided ones. They also measured the charge distributions of fission products in proton-induced fission of ^{238}U by the use of IGISOL and determined the most probable charges Z_p for 40 mass chains.[9] The charge polarization in fission process was examined with respect to the ground-state Q values. It was reported that the trend of Z_p is well explained by the ground-state Q values and the existence of deformed shell closure at neutron number of

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86–88 was suggested experimentally.

Among many studies related to fission phenomena, a series of researches performed by the group of Tokyo Metropolitan University and JAERI is worth to be noticed. They measured the velocities of complementary fission fragments by a double velocity time-of-flight (TOF) system with a high resolution in mass ($\Delta M \leq 2.0$ u) and kinetic energy ($\Delta TKE \leq 2.5$ MeV). In the proton-induced fission of ^{232}Th, there are two kinds of kinetic energies at the mass region near A=130.[10] The lower energy component is almost the same with those of symmetrically divided fragments and the higher one is corresponding to more asymmetrically divided fragments. This means that there are two kinds of elongation at scission, the one corresponding to an elongated shape and the other to a compact shape. On the other hand, the incident energy dependence of the yields corresponding to symmetrically divided products are quite different with those of asymmetrically divided ones. This fact means that there are two kinds of fission thresholds (fission barriers). The angular distributions also exhibit two kinds of thresholds. Nagame et al. performed the experiments which verify the connection between the two thresholds to the two scission configurations.[11] They measured the incident energy dependences of the relative yields of the components of two different kinetic energies and found that the higher threshold component which leads to symmetric mass division corresponds to the lower kinetic energy component which has elongated shapes at scission, and the lower threshold component corresponds to the rather compact shape at scission. This means that two different paths are present in fission process. The same kind of experiments were performed for the system of protons on ^{238}U and the existence of two deformation paths in the mass division process of actinides are experimentally verified.[12,13] They have systematically studied the low energy fission of actinides both by radiochemical method[14–17] and by TOF method[18,19] and deduced quite interesting results. From the radiochemically obtained excitation functions of fission products, systematic trends of fission barriers were deduced both for symmetric mass division and for asymmetric mass division.[16] Zhao et al. reported that the degree of deformation of asymmetrically divided fragments is almost the same ($\beta \simeq 1.53$) at scission regardless of fissioning nuclei and that of symmetrically divided fragments is also the same ($\beta \simeq 1.65$) among them except for the fragments of shell influenced symmetric deformation ($\beta \simeq 1.43$).[19] These results indicate a kind of universality of deformation in fission process.

2.2. New Isotopes. In the past ten years, many new isotopes were discovered by the use of various types of on-line mass separators which were constructed at various institutes in Japan. The separators used are roughly classified into two groups. One is the so-called isotope separator on-line (ISOL) which has an ion source, and the other is a recoil separator which has no ion source and is used for in-flight separation. In general, the time required for separation by the latter is shorter than that by the former, whereas the former has a better mass resolving power than the latter. In this connection, IGISOL is placed between these two groups. IGISOL was effectively used for the investigation of mirror nuclei, such as ^{45}V, ^{47}Cr, ^{49}Mn, and ^{51}Fe, at the Cyclotron and Radioisotope Center of Tohoku University.[20]

Many new isotopes of lanthanoides, such as ^{121}La (Ref. 21), ^{161}Sm (Ref. 22), ^{165}Gd (Ref. 22), ^{166}Tb (Ref. 23), and ^{167,168}Tb (Ref. 24), were discovered by the use of a gas-jet coupled ISOL connected to the tandem accelerator at Japan Atomic Energy Research Institute (JAERI), Tokai. Mono-oxide ions are separated for the investigation of neutron-rich isotopes of lanthanoid elements produced in proton-induced fission of ^{238}U, which is very effective because the cross sections of heavier elements by 16 mass number are quite small compared with the elements of interest.[22–24] Recently they have also succeeded in the mass separation of americium isotope, ^{236}Am (Ref. 25).

At Kyoto University Research Reactor Institute, the gas-jet coupled ISOL was also used for the discoveries of neutron-rich lanthanoid isotopes such as ^{150}La (Ref. 26), ^{152}Ce (Ref. 27), ^{154}Pr (Ref. 28), and ^{156}Pm (Ref. 29), which were produced in thermal neutron-induced fission of ^{235}U.

The ISOLDE type ISOL was constructed at JAERI, Takasaki and connected to the AVF cyclotron in the TIARA facility. The separator itself is the same with that at JAERI, Tokai and neutron deficient isotopes of praseodymium ^{125}Pr and ^{127}Pr were discovered in the reaction of ^{92}Mo + ^{36}Ar system.[30] In this ISOL system, a laser ion source in addition to ordinary ones can be combined and operated in an element selective mode.[31]

In the second group of separators, reaction products are separated in flight from the intense primary beam and practically no mass separation is made. The identification of new isotopes are usually performed by a time-of-flight system and a position sensitive detector on the basis of time- and position-correlated decay chains. Two kinds of recoil separators of such a type are used for the search for new isotopes of heavy elements. One is the gas-filled recoil ion separator (GARIS) at the Institute of Chemical and Physical Research (RIKEN). Recoil ions just ejected from the target material have great variety of charge states. Accordingly, the orbits of the ions with different charge states will differ under the applied magnetic field for separation. But if a suitable gas is filled in the separator, recoil ions behave as if singly-charged ions as a results of many charge exchanging collisions with the buffer gas. Therefore, the collection efficiency becomes larger. This is the main advantage of a gas-filled type separator. The high collection efficiency is essential in the case of new isotopes produced with extremely small cross sections. By the use of GARIS at RIKEN, new isotopes of radon ^{197}Rn (Ref. 32), ^{196}Rn (Ref. 33), and francium ^{200}Fr (Ref. 32) were discovered.

The other is the recoil mass separator installed at the tandem-booster facility of JAERI (JAERI-RMS). This separator consists of a quadrupole doublet, the first electric dipole, a magnetic dipole, the second electric dipole, a quadrupole doublet, and an octupole magnet. The characteristic feature of this separator is that the anode of the first electric dipole was vertically split into two parts separated in 1 cm, and the primary beam passed through without hitting the anode surface.[34] By this way, unfavorable backgrounds originating from the scattered beam were greatly suppressed. The new isotopes of thorium ^{209}Th (Ref. 35) and protoactinium ^{212}Pa (Ref. 36) were discovered by the use of JAERI-RMS.

Extremely neutron-rich light nuclei are being investigated by the RIKEN projectile-fragment separator (RIPS) for understanding the nuclear stability near the neutron drip line. Up to now, such nuclei as ^{10}He (Ref. 37), ^{31}F (Ref. 38), ^{31}Ne (Ref. 39), ^{37}Mg (Ref. 39), ^{38}Mg (Ref. 40), ^{40}Al (Ref. 40), and ^{41}Al (Ref. 40) were discovered.

2.3. Others. The rest of works related to nuclear chemistry in Japan were just outlined here.

2.3.1. Photonuclear Reaction. Tremendous yield data have been accumulated in 30 – 1200 MeV bremsstrahlung induced reactions with various targets (^{7}Li to ^{209}Bi) by Sakamoto of Kanazawa University and collaborators. Almost all of these data were obtained radiochemically except for a long-lived beryllium isotope, ^{10}Be, which was measured by accelerator mass spectrometry. Because of the continuous nature of the bremsstrahlung energy-spectrum, they performed the measurements at small energy steps less than 50 MeV and the obtained yields were unfolded into cross sections per photon of a specific energy. They discussed the reaction mechanisms of photonuclear reaction from various aspects, such as π emission channel,[41–44] spallation,[45–47] fragmentation,[48] and recoil proper-

ties of products.[49,50]

2.3.2. High Energy Light Ion Induced Reaction. Extensive investigations have been performed at the National Laboratory for High Energy Physics (KEK) to obtain production cross sections in spallation reactions with high energy light ions. Target materials are ranging from Al to Pb.[51] Some of radioactive nuclides of which decay mode are pure beta emission or electron capture have large production cross sections. It is practically impossible to estimate their activities by direct measurements. One of such nuclides is ^{3}H. Noguchi et al. measured production cross sections of ^{3}H with 12 GeV protons for various targets and deduced a simple relation between production cross sections and target mass numbers.[52] These cross section data are useful for radiation protection at high energy hadron accelerator facilities.

2.3.3. Intermediate Energy Heavy Ion Induced Reaction. Reaction mechanisms with heavy ions are radiochemically investigated at the energy region around 100 MeV.[53–56] In addition to the limiting behavior of product yields,[55] such as recoil properties of products[53] and angular momentum transfer[54] are examined, and target fragmentation mechanism is studied.

2.3.4. Neutron Induced Reaction. From the view point of radiation protection and astrophysical interests, production cross sections of long-lived light nuclides have been measured with semi-monoenergetic neutrons of energies up to 38 MeV.[57–59] The measurements of ^{14}C and ^{26}Al were performed by an accelerator mass spectroscopy method.[57,58]

2.4. Future plan. Syntheses of new isotopes of heavy elements and study of chemical properties of transactinide elements are intended at JAERI and RIKEN in near future by a large collaboration group in Japan. The scientific goals are (1) systematic understanding of nuclear properties of heavy nuclei, such as mass, deformation, beta-delayed fission, etc., and (2) clarification of the chemical properties of transactinide elements which are expected to strongly deviate from their light homologs because of the relativistic effect of orbital electrons.

The characteristic decay mode in actinide and transactinide elements is α-decay and the detection efficiency of α particles is quite large. In the case of even-even nuclei, it is easily possible to obtain masses of parent nuclei from the principal α energies and known masses of daughter nuclei. In general, the resolution of the α spectra is not so good as that of γ spectra and the probabilities of branching decays are small, but for some nuclei, it is possible to deduce Nilson levels from branching α decays. The other unique decay mode of heavy nuclei is β (β^-, EC)-delayed fission which is considered to be important in the process of nuclear syntheses in nature. It is necessary for β-delayed fission that β decay energy Q_β is larger than the fission barrier height B_f of the nucleus of interest. The more apart from the β stability line, the larger the Q_β is. Accordingly, the probability of β-delayed fission is larger for nuclei far from the β stability line. Therefore, it is very important to synthesize new isotopes of heavy nuclei far from the β stability line and to investigate their decay properties.

To investigate the chemical properties of the transactinide elements is one of the most important and challenging subjects in nuclear chemistry. Due to the strong Coulomb field of the highly charged atomic nucleus, the motions of orbital electrons near the nucleus are relativistic, and consequently the spherical *s* and $p_{1/2}$ orbitals are stabilized. While the diffused *d* and *f* orbitals are de-stabilized as a result of shielding of the nuclear charge by the inner shell electrons. Thus, the electron configurations of the heaviest elements may be different from those of lighter homologs, and which makes the strong deviation of the chemical properties of transactinide elements.

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