

Nuclear and radiochemistry in China: present status and future perspectives

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Summary. Nuclear and radiochemistry is one of the frontier areas of chemistry with high impact on national security, energy supply, scientific advances, social and economic development. Nuclear and radiochemistry in China is now experiencing a renaissance, which is being strongly motivated by China's huge demand for nuclear energy. With this in review, the progress in nuclear and radiochemistry of China is selectively addressed. Some hot topics have been summarized and the main research results achieved by Chinese scientists in this field are highlighted, with emphasis on the basic nuclear chemistry, actinide and trans-actinide chemistry, chemistry of spent nuclear fuel reprocessing, radioanalytical chemistry, environmental radiochemistry and radiopharmaceutical chemistry, *etc.* Some measures about how to promote the radiochemical education and research in China are suggested, and future perspectives are briefly outlined as well.

1. Introduction

Radiochemistry was born in the early twentieth century with the discovery of radioactivity and radionuclides. It played a positive role in the extension of human knowledge, for example, it contributed to the discoveries of almost one-third of the elements in the Periodic Table. After the discovery of man-made radioactivity and the nuclear fission, the era of nuclear science and technology was consequently created. For nearly one century, radiochemistry played significant roles in establishing China's important position in the international arena. It also contributed a lot to the peaceful use of nuclear energy and nuclear technologies in human health and environmental protection, as well as to sustainable development of social economy.

Chinese radiochemistry was founded by Tcheng Da-Tchang, a proud student of Madame Marie Curie, who got his doctoral degree at Institute of Radium in Paris at the

end of 1933 and then came back to China in 1934 and established the Radium Research Institute in Beijing. His research interest was in protoactinium and uranium chemistry. Yang Cheng-Zong is another founder of radiochemistry in China. He commenced his study of radiochemistry with Tcheng Da-Tchang in 1934 and later worked in the Curie Laboratory under the guidance of Joliot-Curie from early 1947 onwards. There he was awarded the doctor degree in 1951. Immediately thereafter he returned from France without any hesitation. Yang was the pioneer in developing basic radiochemistry and its applications in China. He made outstanding contributions to the development of China's nuclear energy program, especially in the field of nuclear fuel production and radioisotope applications.

Historically, China's golden age of radiochemistry was in the 1950s and the 1960s. At that time, almost all leading universities in China offered radiochemistry training, achieving indelible contributions for the national security projects. However, from the 1980s, the global nuclear and radiochemistry confronted a downward trend. In particular, the US Three Miles Island accident and the former Soviet Union's Chernobyl nuclear disaster exacerbated this trend. In addition, because of the inappropriate exaggeration of hazardousness of radioactivity by some media and literary works, most of the public and young students got afraid to conduct research in all areas related to radioactivity. By the end of the last century, China's radiochemistry dropped to a valley; similar situation occurred in the rest of the world, although accelerator related radiochemistry for medical application has seen a tremendous upward trend in the western world. As the radiochemical research and education had been declining for many years, the overall level of China's current nuclear chemistry and radiochemistry can not meet the needs of the national security, construction of reliable nuclear power system and nuclear medicine, *etc.*

The non-rational and non-scientific downward trend of radiochemistry has gained strong attention from Chinese topmost leadership and leading scientists. The voice for promoting the teaching and research of radiochemistry is gradually rising. Since the new century, with the drastic development of Chinese nuclear power industry, there are huge basic research needs for the future advanced nuclear energy system. In response to this trend, the authorities have

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Table 1. The main radiochemical research institutes and universities and related research areas.

Institute or University	Main research areas
China Institute of Atomic Energy (CIAE)	Nuclear chemistry, radiopharmaceutical chemistry, actinide chemistry and spent nuclear fuel reprocessing
Tsinghua University	Actinide chemistry, nuclear energy chemistry
Chinese Academy of Engineering Physics (CAEP)	Actinide chemistry, nuclear fuel chemistry and nuclear waste disposal
Institute of High Energy Physics, Chinese Academy of Sciences (CAS)	Radioanalytical chemistry, nuclear energy chemistry and radiopharmaceutical chemistry
Institute of Modern Physics, CAS	Basic nuclear chemistry, radiochemistry of super-heavy elements
Shanghai Institute of Applied Physics, CAS	Radiochemistry related to thorium-based molten-salt reactors; radiopharmaceutical chemistry
Institute of Plasma Physics, CAS	Environmental radiochemistry, nuclear waste disposal
Beijing Research Institute of Uranium Geology, China National Nuclear Corporation (CNNC)	Uranium mining, nuclear waste disposal
Beijing Research Institute of Chemical Engineering and Metallurgy, CNNC	Hydrometallurgy of uranium mining, uranium chemistry
China Institute of Radiation Protection, CNNC	Environmental radiochemistry, nuclear waste disposal
Nuclear Power Institute of China, CNNC	Actinide chemistry and nuclear fuel chemistry
Peking University	Radiation chemistry, radiopharmaceutical chemistry and environmental radiochemistry
Lanzhou University	Environmental radiochemistry, nuclear waste disposal
Beijing Normal University	Radiopharmaceutical chemistry
Sichuan University	Environmental radiochemistry, nuclear energy chemistry
Shanghai Jiaotong University	Spent nuclear fuel reprocessing, separation chemistry
Zhejiang University	Fission products chemistry, separation chemistry
University of South China	Uranium mining and environmental radiochemistry

been taking positive measures to enhance China's radiochemistry recovery. After years of efforts, nowadays China's radiochemistry is at the recovery stage and keeps the upward trend, whose main signs are: (1) Under the financial support of the National Natural Science Foundation of China, the Ministry of Education and National Defence Science and Industry Bureau, some key radiochemistry laboratories are being established. (2) The Ministry of Education has formally included the nuclear chemistry and radiochemistry in the curriculum catalog for higher education, ranking it with the same status as inorganic, organic and physical chemistry. The enrollment of undergraduate and graduate students majoring in radiochemistry is beginning to continuously improve. Correspondingly, the radiochemical research in China has been expanding fast in recent years, and now almost all the important areas of radiochemistry are being covered in Chinese main research institutes and universities. The related information is shown in Table 1.

2. Important progresses in the past three decades

Despite many years of inadequate financial support from Chinese government, China's workers in the radiochemical areas have still made significant contributions and achieved a number of high level peer-recognized research results in the synthesis of super-heavy elements and nuclides, basic actinide chemistry, spent nuclear fuel reprocessing, radioanalytical chemistry, environmental radiochemistry and radiopharmaceutical chemistry related to tumor imaging and diagnosis, *etc.*

2.1 Basic nuclear chemistry

Basic nuclear chemistry refers to study of nuclear properties, nuclear structure, nuclear decay and transformation, and other nuclear parameters through radiochemical methods. With the development of particle accelerators, nuclear reactors, high efficiency detectors, and computer technologies,

research category of nuclear chemistry becomes larger and larger. Chinese nuclear chemists have been conducting fruitful studies and accumulating a number of experimental data in this field.

With respect to fission chemistry, scientists in CIAE have developed radiochemical separation procedures for many fission products and lanthanide elements, and improved various measurement techniques, including absolute measurement of fission product yields. Product yields in fission of ^{235}U induced by thermal neutrons, fission-spectrum neutrons and monoenergetic fast neutrons were systematically determined as well as of those in ^{238}U fission induced by neutrons at 6 different energies from 3 to 24.4 MeV [1]. Fission yields of some key products such as ^{93}Zr , ^{140}Ba , and ^{147}Nd were measured and the dependence of mean mass of light and heavy fission products upon the mass of fissionable nuclides was also addressed. Automatic rapid chemical separation apparatuses have been developed and successfully applied to the measurement of nuclear data of nuclides with half-lives of a few minutes such as ^{95}Y , ^{138}Cs , ^{91}Sr , ^{132}I , and ^{142}La . Through radiochemical methods and Inductively Coupled Plasma Mass Spectroscopy (ICP-MS), Chinese radiochemists successfully determined half-lives of long-lived ^{79}Se , ^{126}Sn , ^{93}Zr and obtained the thermal neutron cross sections of the reactions $^{126}\text{Sn}(n, \gamma)^{127\text{m}, 127\text{g}}\text{Sn}$ for the first time in the world [2, 3].

Mass and charge distributions in the fission reactions of ^{238}U , ^{232}Th , ^{237}Np induced by 14.7 MeV neutrons and the involved fission mechanisms were investigated by the Heavy Ion Research Facility (HIRFL) at the Institute of Modern Physics (IMP), CAS. In the fission of ^{232}Th , the fission yields of 57 fission products from $^{77\text{g}}\text{Ge}$ to ^{151}Pm were directly determined and 44 mass yields were successfully recorded. ^{197}Au , ^{209}Bi , and ^{238}U were bombarded by a 72 MeV ^{12}C beam and fission product yields were determined by radiochemical separations and γ -ray spectrometry. On the basis of these data, the Gaussian curves of complete fusion mass distribution were obtained and the dependence of the mass distribution width on the excitation energy of target nuclei was found. After heavy ions were successfully accelerated to an intermediate-energy level at HIRFL, a series of intermediate-energy reaction studies were carried out to examine the mass yield distribution of the target residues and linear momentum transfer through a thick target and thick catcher foil technique and γ -ray spectrometry. The experimental results were compared with fusion fragmentation calculations, sequential evaporation model and molecular dynamics method to shed some light on nuclear reaction mechanism [4].

In order to synthesize new isotopes and investigate their decay properties, various rapid chemical separation apparatuses such as on-line thermal chromatography and solvent extraction devices were developed at IMP. More than 20 new isotopes of neutron-deficient lanthanide elements and neutron-abundant heavy elements with $A > 170$ have been produced and identified. Decay chains of some isotopes were proposed as well [5]. A new isotope of ^{235}Am and the isotopes of the elements 105 and 107 with the mass weight 259 and 265, respectively, were synthesized for the first time [6]. The existence of ^{259}Db has been confirmed by Lawrence Berkeley National Laboratory. On-line isothermal

chromatographic apparatus coupled with a gas-jet system and a ^{252}Cf fission source was used in similar studies on the gas-phase chromatographic behavior of the short-lived technetium isotopes on quartz surface as model experiments [7]. Recently, the volatility of oxybromide complexes of the group 5 elements including Db has been addressed using this apparatus.

Additionally, in order to develop thorium-based reactors, basic nuclear chemistry related to thorium-uranium fuel cycle has been studied all along at Shanghai Institute of Applied Physics [8]. It includes: (1) nuclear transformation process of thorium irradiated by thermal neutrons and growth of ^{233}U ; (2) the influence of different flux ratio of fast and thermal neutrons on the relative amount of produced ^{232}U and ^{233}U ; (3) developing radiochemical separation technologies to obtain ^{233}U on a large scale.

2.2 Actinide chemistry

Actinide elements have some unique properties and require special treatment. For example, Pu has 6 phases in solid state, and the phase transition is accompanied by the dramatic change in volume, electrical conductivity, thermal conductivity and mechanic properties, which is unique in metallic elements. Since the 1950s, scientists in China have invested enormous endeavors towards the understanding of the actinide chemistry, and the contributions come from both theoretical and experimental sides.

The computational actinide chemistry has been a challenging field where the relativistic effect and strong electronic correlation effect require reasonable treatment. Over the years, significant advancement has been seen in the relativistic quantum mechanics, and now it is feasible to study the properties of actinide and trans-actinide elements. Due to the complexity of the actinide chemistry, the current theoretical study of actinide chemistry in China mainly concerns the structure and properties of small model compounds containing actinide atoms, and these include:

- (1) The ionization energy and excitation energy of actinide; geometry, electronic structure, spectroscopy and dissociation energy of U_2 and Pu_2 ; the bonding analysis of $\text{U}_2@\text{C60}$;
- (2) The geometry, dissociation energy and vibrational spectroscopy of tri-atomic systems, such as Pu_3 ;
- (3) The geometry, electronic structure, dissociation energy and thermochemistry of UH , PuH_2 , UC_2 , PuC , PuC_2 , PuO , UO_2 and OUH ;
- (4) Quasi-classic trajectory Monte-Carlo simulation of reactions between actinide, such as Pu, and small molecules, such as CO and H_2 ;
- (5) The electronic structure, spectroscopy and thermochemistry of UF_x compounds;
- (6) Relativistic quantum chemistry study of the multiple bonds between actinides, *e.g.* Th and U, and main group elements.

The concept of multiple bonding interactions in the molecules containing heavy atom complements our knowledge of chemical bonding interactions, and it is crucial to understand the properties of the actinide molecular systems. In this field, Li's group in Tsinghua University has contributed

to the characterization of a series of small molecular systems containing U and Th by means of relativistic quantum chemistry methods, and these results provide valuable information on the An=C double bond and An≡C triple bond (An = Th, U) [9].

The research in Liu's group of Peking University focuses on the development and application of relativistic methods to study the molecular systems containing heavy atoms. They have developed a composite relativistic DFT package (Beijing Density Functional, BDF) which features the complete, highly accurate and efficient application of relativistic DFT methods, and pioneered the investigation on the excited states and electronic fine structures of heavy atoms by means of relativistic TDDFT method [10]. They have studied the electronic structures of PuH₂ and PuH₃ with Discrete Variational method (DVM), the structures and properties of U₂ and Pu₂ using B3LYP method combined with the relativistic ECP (RECP), and the reaction dynamics of Pu and H₂, the property of δ-Pu (001) using full-potential linearized augmented plane wave method (FLAPW) with GGA functional employed; the structures of U and its compounds using plane wave method, *etc.* Because of Liu's distinguished work in the field of relativistic effect, he was awarded the "Pople Medal" in 2006 by Asia-Pacific Association of Theoretical & Computational Chemists (AP-ATCC), and the "International Academy of Quantum Molecular Award" in 2006, which is for the theoreticians below 40 years old. He is the first awardee of the "International Academy of Quantum Molecular Award" in China.

The interactions between actinides and nanomaterials have also been explored by the Chinese scientists. Among them is the Lu's group in Xiamen University. They studied the multiple bond in a U₂ dimer which is encapsulated in a fullerene cage (U₂@C₆₀) using PBE function with relativistic effect included [11]. It is shown that the 5*f* electrons of U do participate in the bond formation, and the interaction between the two U atoms has the feature of a one-electron-two-center multiple bond with a configuration characterized as $(5f\pi_u)^2(5f\sigma_g)^1 - (5f\delta_g)^1(5f\phi_u)^2$. This work shows that in a confined space it is possible for U₂ dimer to form a metal-metal multiple bond.

2.3 Radiochemistry of nuclear fuel reprocessing

Because of the high burn-up, the spent fuel of power reactors normally possesses strong specific activity and high content of fission products. In its reprocessing, high decontamination coefficients of uranium and plutonium products and high separation factor ($> 10^6$) of uranium to plutonium are required. Furthermore, the recovery yields of uranium and plutonium should be over 99.8%. Therefore, the reprocessing is extremely challenging and demands a well-equipped laboratory with high level of radiation protection. Up to now, this technology was only mastered by a few developed nuclear power countries in the world. The R&D activities of spent nuclear fuel reprocessing in China have been predominantly conducted in CIAE and Tsinghua University, concentrating on improving Chinese commercial reprocessing technologies of spent fuel and separation technologies of high level liquid waste (HLLW).

In late 2010, China succeeded in hot commissioning of pilot reprocessing plant, and qualified uranium and plutonium products were obtained, which marked that China has preliminarily mastered the spent fuel reprocessing technologies of power reactors. It was a significant achievement as a result of close cooperation in different fields, such as the basic research on the spent fuel reprocessing technologies, engineering design and construction, equipments installation, plant operation and management, *etc.* Among them the radiochemistry based spent fuel reprocessing is the core technology because it is the basis of the design and construction of the reprocessing pilot plant. In the past 30 years, cooperating with other related Chinese research institutes, CIAE carried out more than 40 research projects on process technologies and over 20 research projects on analytical techniques, which were results of test tube-scale, bench-scale and the whole process-linked-scale experiments. Consequently, the laboratory-scale advanced two-cycle PUREX process was established in CIAE and some innovative results have been successfully applied in the pilot plant project. Specifically, the basic researches associated with spent fuel reprocessing in CIAE include: (1) Solution chemistry of actinides: solution chemistry of uranium, neptunium and plutonium, valence change and control of actinides in the process; redox thermodynamics, chemical kinetics and extraction chemistry of actinides. (2) Chemistry of fission product elements: the chemical species of fission product elements and their extraction, stripping and hydrolysis behaviors. (3) Dissolution of spent fuel and off-gas treatment, especially optimization of dissolution conditions. (4) Reductants in PUREX process: the preparation of U(IV) and its related reduction behaviors, applications of salt-free organic reductants and complexants, aiming at excellent purification of uranium and clean separation of uranium/plutonium. (5) Two-cycle based processing technologies: Uranium-plutonium separation, uranium-line process, plutonium-line process, the end processing technologies of uranium-line and plutonium-line. (6) Solvent chemistry: Purification and recycle of solvents. (7) Remote analysis and control techniques.

The radiochemical research on spent fuel reprocessing at Tsinghua University is mainly concentrated on the treatment of HLLW, dealing with the raffinate from PUREX process which contains more than 95% radioactivity of spent fuel. The purpose of HLLW treatment was to remove the long-lived radionuclides and reduce the long-term risk of nuclear waste towards the environment. In the late 1970s, when the radiochemical research was still in recession in China, Zhu and his co-workers began their study on the extraction of transuranium elements (Np, Pu, Am, Cm) from HLLW [12]. They selected the mixed trialkyl phosphine oxide (TRPO) as the extractant, instead of the bifunctional extractants which were widely used in other laboratories at that time. TRPO with good physico-chemical properties and radiation stability has excellent extraction selectivity to tri-, tetra- and hexavalent actinides. After over 10 years of hard work, Zhu's group successfully developed the innovative TRPO process to remove transuranium elements from HLLW in 1992. During the past 30 years, they systematically studied the extraction behaviors and mechanisms of U, Pu, minor actinides and fission products by TRPO. The flow sheet of TRPO

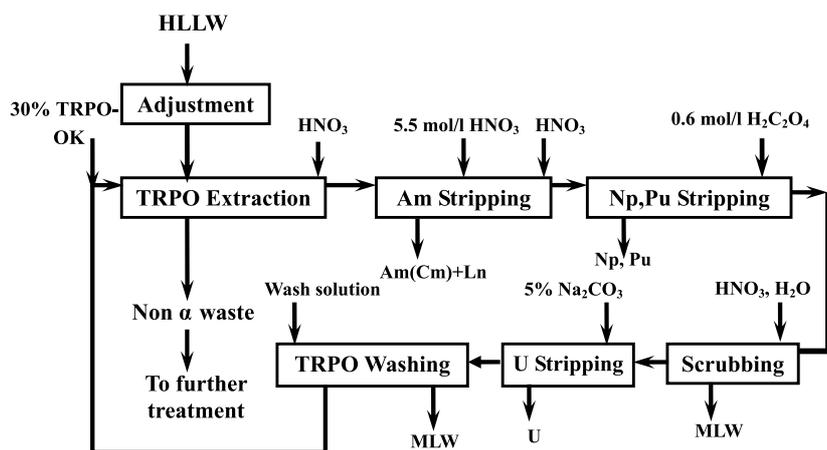


Fig. 1. The flowsheet of TRPO process.

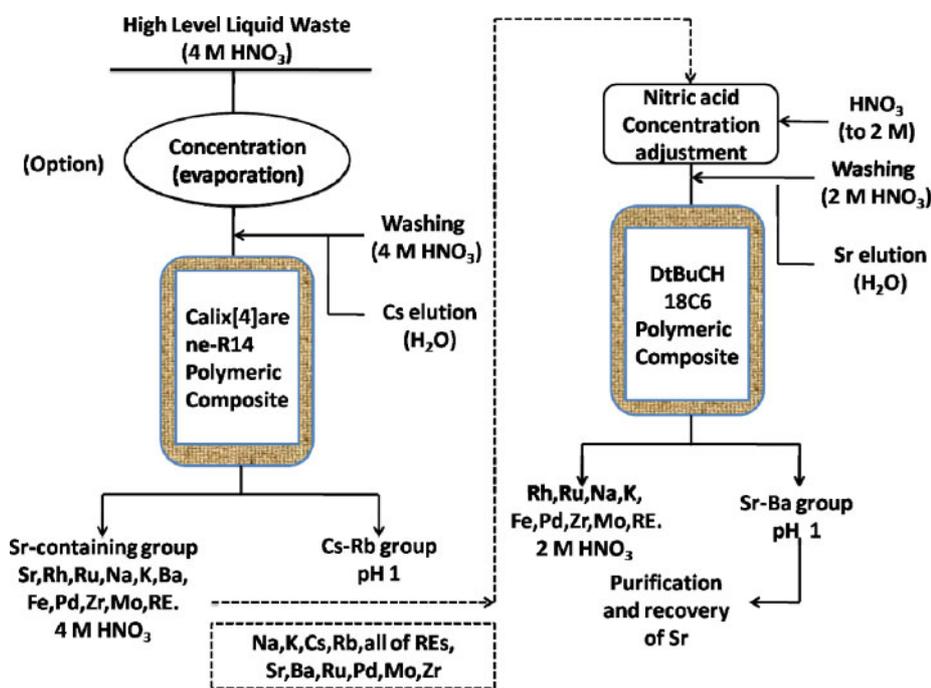


Fig. 2. SPEC process for Cs/Sr partitioning from an acidic HAW by extraction chromatography.

process is shown in Fig. 1. Now the Institute of Nuclear Energy Technology of Tsinghua University has carried out several hot tests and pilot tests of total partitioning with genuine HLLW, which used TRPO process for separating TRU and ⁹⁹Tc. The results demonstrated that the HLLW could be conditioned into non- α waste. Now the TRPO process is widely considered as one of the most promising processes for the treatment of HLLW in the world.

After the 1990s, Zhu's group made another breakthrough in the partitioning of lanthanides and actinides. They successfully isolated and purified the bis-(2,4,4-trimethylpentyl) dithiophosphonic acid (HBTMPDTP) from commercially available Cyanex 301, which was then used to extract trivalent Am and Cm from lanthanides with the separation factor of 5000 of Am over light lanthanides [13]. This work is also of strategic significance for establishing a future advanced nuclear fuel cycle system.

Zhang's group at Zhejiang University developed a series of novel macroporous silica-based supramolecular recognition materials with calixarene crown ethers as the functionally recognitional groups. These materials can be

readily synthesized by immobilization techniques and exhibit remarkable recognition properties and high selectivities towards heat-emitting elements such as Sr(II) and Cs(I) in HLLW. Based on the above silicon-based materials, Zhang also established a strontium/cesium partitioning from HLLW by extraction chromatographic (SPEC) process, which uses recognition material (calix[4]arene-R14+M)/SiO₂-P for Cs(I) extraction and (DtBuCH-18C6+M)/SiO₂-P for Sr(II) extraction, respectively [14–16]. The flow sheet of SPEC process is shown in Fig. 2. Besides high adsorption capacities, these supramolecular recognition materials also possess several other advantages such as acid and alkali resistance, high temperature enduring and radiation resistance. The SPEC process is quite prospective for the separation of Cs(I) and Sr(II) from HLLW.

2.4 Radioanalytical chemistry

Radioanalytical chemistry is the field in which the analysis is based on nuclear reaction, nuclear effect, nuclear spectroscopy, nuclear parameters and nuclear property. It

is crucial to multidisciplinary research, the national security and the social and economical development. In the past years, with the increasing demands from the rapid development of nuclear energy and the application of new materials and technology, the radioanalytical chemistry in China has achieved significant progress.

The burn-up analysis for the spent nuclear fuel is crucial for the design and safe operation of nuclear reactors. So far, the radioanalytical methods have been recognized as a good choice to fulfil this task. Scientists in CIAE have developed effective burn-up analytical methods based on heavy isotope ratios and nuclear fuel monitor, which were successfully applied to the burn-up determination of nuclear fuel components from the Qinshan nuclear power plant. The analytical uncertainty was 1.7%. In addition, they have also made important progress in direct determination of the quantity of ^{90}Sr in soil and in waste water with no need of chemical separation, and the rapid analysis of the short-lived fission products, which made it possible to study the properties of short-lived isotopes with the newly developed rapid separation process and apparatus towards the analysis of ^{91}Sr , ^{95}Y , ^{101}Tc , ^{114}Pd , ^{138}Cs , ^{142}La on the time-scale of minute or hour.

To tackle the complexity due to the special features of the solid samples such as complex gradient, low radioactivity, and many factors that may interfere with the measurement, scientists in Chinese Academy of Engineering Physics (CAEP) have developed the techniques of acid dissolution, alkali fusion and microwave radiation to digest samples, with high decontamination radiochemical separation processes of more than 40 isotopes and the preparation techniques of electroplating source, powder source, and mass spectrometry source [17]. In addition, to analyze the gas products in nuclear reactions efficiently, they have established the ^1H and He isotopic gas mass spectrometry and noble gas mass spectrometry for the rapid analysis. These techniques have been used for the CTBTO (the Comprehensive Test-Ban Treaty Organization) on-site monitoring. They have also invented the mobile ^{37}Ar rapid measurement system which has been demonstrated by CTBTO in Kazakhstan, and established xenon air/ground mobile monitoring system [18].

The most significant advance in nuclear activation analysis is the establishment of the so-called molecular activation analysis (MAA) method to realize the study of the chemical species of elements of interest in various biological, environmental and geological samples. This involves a combination of special separation techniques of element speciation, for example, the chemical separation or biochemical separation, with neutron activation analysis of high sensitivity. With this modern nuclear analytical technique, it is possible to carry out the chemical analysis of elements at the molecular or cellular levels.

The technique of molecular activation analysis has been widely used in the chemical speciation analysis of the platinum group elements (PGE), the rare earth elements, Hg, Se, I, and Cr [19]. Based on the successful experience, Chinese scientists pioneered the application of the nuclear analysis methods in study of metallomics, and published the first book in the world on this topic entitled "Nuclear analytical techniques in metallomics and metalloproteomics" [20]. In

addition, Chinese scientists are also leaders in using nuclear analysis to study nanotoxicology [21].

The development of science and technology brings new opportunities to radioanalytical chemistry which covers a broad field, including:

1. New nuclear analytical methods, nuclear instruments and radiation detectors enable the nuclear analysis with high sensitivity, accuracy, excellent time and spatial resolution, thus the possibility of real-time analysis on the molecular and atomic levels.
2. The heavy ion irradiation techniques to prepare nanopores with the controllable pore size on the scale of nanometer, which is also used in the sequence determination of nucleic acid, single molecule detection, simulation of the ion channel in the membrane of cell, and protein sensor, *etc.*
3. Nuclear analytical methods for the multidisciplinary field, such as molecular toxicology, nanomaterials, molecular environmental science, metallomics, space chemistry, *etc.*; meanwhile, it may develop into new nuclear analytical methods towards food safety, health, national security (nuclear proliferation, explosives, and drugs, *etc.*) and new energy resources.
4. The nuclear analytical methods based on large scientific facilities such as synchrotron radiation, spallation neutron source, and other advanced facilities, will have bright future to solve scientific frontier issues.

2.5 Environmental radiochemistry

The environmental radiochemistry investigations in China began in the 1980s. One of the most important achievements was the survey of radioactive level of the Yangtze River system at that time. The targets of the survey included water body, sludge, fish and soil along the river. For each sample, the quantities of total α , total β , U, Th, ^{226}Ra , ^{210}Po , ^{40}K , ^3H , ^{90}Sr , ^{137}Cs and ^{239}Pu were tested [22]. By the 1990s, Chinese researchers began to study the diffusion and migration of radioactive elements in the environmental media. During that time, one of the influential investigations involved the development of safety evaluation method for shallow ground disposal of low and intermediate level solid radioactive waste. This project was a five-year collaboration between China Institute for Radiation Protection (CIRP) and Japan Atomic Energy Agency (JAEA). In this project, the diffusion and migration of $^{85,89}\text{Sr}$, ^{134}Cs and ^{60}Co in the aeration zone of loess were studied systematically. In addition, this project basically established the safety evaluation method for shallow ground disposal of low and intermediate level solid radioactive waste in China. During the period of 1995–2001, the second five-year collaboration between CIRP and JAEA was launched, which focused on migration and diffusion of transuranium elements, like ^{237}Np , ^{238}Pu and ^{241}Am in the aeration zone of loess and shallow aquifer, and made a good start for Chinese safety evaluation of nuclear waste disposal [23, 24].

From the beginning of the new century, a lot of experimental and theoretical work has been performed in environmental radiochemistry, owing to the great concerns about nuclear waste treatment and disposal in China. Chinese re-

searchers began to study the adsorption, diffusion and migration of transuranium elements in granite at Bei-Shan, Gansu province, based on the significant national demand of high level solid waste disposal and decommissioning of old nuclear facilities. These investigations included many aspects, for example, the diffusion of weakly adsorbed radioactive elements in Bei-Shan granite, the possible influencing factors on nuclide migration, involving temperature, oxygen concentration, ionic strength, radiation field, *etc.*, the design of CHEMSPEC software for nuclide speciation analysis and subsequent calculations of speciation distribution of ^{125}I and uranium [25], the molecular dynamics simulations of uranyl ion adsorption in kaolin [26], and the adsorption of Eu on attapulgite [27].

Wang and his colleagues performed a series of experimental investigations on nuclear waste treatment and disposal [27–29]. One of their research areas is to explore the potential applications of multi-wall carbon nanotubes (MWCNTs) to treat nuclear waste. Carbon nanotubes have attracted great interest in multidisciplinary study since their discovery. Wang *et al.* studied the $^{243}\text{Am}(\text{III})$ sorption to uncapped MWCNTs in NaClO_4 solutions, and found that MWCNTs can adsorb $^{243}\text{Am}(\text{III})$ with extraordinarily high efficiency by forming very stable complexes, where chemisorption or chemicomplexation is the main mechanism [28]. Their investigations indicated that MWCNTs might be a promising candidate for the preconcentration and solidification of $^{243}\text{Am}(\text{III})$ or its analogue lanthanides and actinides from large volumes of aqueous solution. In addition, they also prepared a variety of carbon nanotube composites with experimental techniques such as chemical modifications and plasma-induced grafting. The macroscopic adsorption experiments showed that these carbon nanotube composites displayed better adsorption abilities compared with native carbon nanotubes, and were promising for the future processing of nuclear waste. They also found that the adsorption of Eu(III) and humic substances (HA) on MWCNTs was strongly dependent on pH and ionic strength, and the presence of HA would enhance Eu(III) adsorption to HA-MWCNT hybrids [30]. Liu *et al.* also studied the migration of radioactive elements like ^{85}Sr , ^{99}Tc , ^{237}Np , ^{238}Pu and ^{241}Am in a series of media including Chinese loess [31], loess aquifer [23], and granite [32, 33], *etc.* For example, they examined the diffusion of ^{125}I in deep granite taken from Bei-shan, Gansu Province, and obtained an effective diffusion coefficient of $(2.44\text{--}2.72) \times 10^{-12} \text{ m}^2 \text{ s}^{-1}$ [33]. Recently, Wu *et al.* studied the sorption of uranyl ions onto a decarbonated calcareous soil (DCS), and found that the sorption of U(VI) on DCS was significantly influenced by pH values of the aqueous phase, indicating the formation of inner-sphere complexes at solid-liquid interface [34].

In the future 20 years, the migration of geologically disposed high level nuclear waste will become one of the principal issues in environmental radiochemistry. The key research issues include: (1) the performance of solidification product of high level solid waste; (2) the long-term chemical stability of packing materials; (3) the aqueous solution chemistry of key elements; (4) colloid and interfacial chemistry; (5) the effect of gas release, germ and organic substance in the disposal environment; (6) the physical chemistry mechanism of nuclides migration. Among the

above aspects, the chemical species and migration behaviors of actinides in geological disposal media will attract special attention. Besides traditional coordination chemistry, the applicable methods nowadays have been expanded to the extended X-ray absorption fine structure, X-ray absorption near-edge structure and other advanced techniques. To calculate the speciation distribution of radioactive nuclides in ground water, a high quality thermodynamic database is highly desirable.

2.6 Radiopharmaceutical chemistry

Since the first application of radioisotopes in medical research in 1956, radiopharmaceutical chemistry has already made enormous contributions to diagnosis and therapy of various diseases in China. Currently, nuclear techniques are widely used in medical diagnosis, treatment and research activities in all provinces and large cities of China.

Generally, classified by their functions, radiopharmaceuticals consist of two categories, *i.e.* diagnostic radiopharmaceuticals and therapeutic radiopharmaceuticals. As for diagnostic radiopharmaceuticals, single-photon and positron emitting pharmaceuticals are generally involved. In the 1980s, the basic research on radiopharmaceuticals was predominantly focused on technetium (Tc-99m) labeled single-photon emitting pharmaceuticals. Since the beginning of this century, the research interest started to shift to the positron emitting pharmaceuticals. To date, both single-photon and positron emitting pharmaceuticals are regarded as equally important for clinical applications and research, particularly for the diseases of brain, tumor, heart, *etc.*

As for brain imaging, traditional research highly relied on the Tc-99m complexes. Zhang *et al.* reported the bis($\text{N-cyclohexyl-dithiocarbamate}$) nitrido Tc-99m complex [$^{99\text{m}}\text{TcN}(\text{-CHDTC})_2$] ($\text{CHDTC:N-cyclohexyl dithiocarbamate}$) as a potential brain perfusion imaging agent [35]. The complex was stable for over 6 h at room temperature. The partition coefficient also suggested that it was a good lipophilic complex, which accumulated in brain with high uptake and good retention. The ratio of brain/blood in mice was measured as high as 2.10 at 1 h post-injection. The results designated the complex as a potential brain perfusion imaging agent. Recent interest for brain imaging in China has been concentrated on the early diagnosis of the senile dementia and parkinsonism, and plenty of exciting results have been obtained, especially in the β -amyloid ($A\beta$) plaque imaging. Liu *et al.*, for example, synthesized a series of novel 4,5-dianilinophthalimide derivatives (DAPHs) as potential $A\beta$ plaque probes [36]. It was found that DAPHs bound efficiently to $A\beta$ plaques and showed high *in vitro* binding affinity with K_i in concentration of nM level. In particular, [^{125}I] N-methyl-4-(4-iodoanilino) phthalimide, because of its specific *in vitro* labeling of amyloid plaques, combined with its excellent BBB permeability and fast clear out, is considered as a promising candidate probe for SPECT imaging of the brain.

Regarding the heart imaging radiopharmaceuticals, also many interesting studies have been done by Chinese scientists. Liu *et al.* prepared a new promising myocardial perfusion-imaging agent $^{99\text{m}}\text{Tc-CO-MIBI}$ by a two-step procedure involving a convenient preparation of the

$[^{99m}\text{Tc}(\text{CO})_3(\text{OH}_2)_3]^+$ precursor followed by a substitution of the water molecules by the MIBI (2-methoxyisobutylisonitrile) ligands [37]. ^{99m}Tc -CO-MIBI was confirmed to be a mixture of two complexes: complex A and complex B, and the heart/liver ratios of complex A, the mixture, and complex B were found to be 1.57, 1.93, and 2.33, respectively, for 30 min post-injection, much better than the widely used ^{99m}Tc -MIBI. Subsequently, the authors prepared a series of novel $^{99m}\text{Tc}(\text{CO})_3$ -MIBI analogs, and evaluated lipophilicity and biodistribution of these complexes to determine whether different lipophilicity and structure of isonitrile ligands would improve their imaging properties for the heart [38]. The results showed that these new complexes exhibited low liver, lungs and blood uptake compared with $[^{99m}\text{Tc}(\text{CO})_3(\text{MIBI})_3]^+$, though their heart uptake was not so high. In a word, the discovery of chemical and biological properties of ^{99m}Tc -CO-MIBI series would certainly promote the research on a new promising myocardial perfusion-imaging agent.

As for other tissue imaging, Liu *et al.* prepared *N,N'*-bis(2-aminoethyl) propanediamine hexaacetic acid (BPHA) as a tetrachloride salt for kidneys imaging. The synthesized complex is easily labeled with ^{99m}Tc in the pH range of 2–5 using $\text{SnCl}_2 \cdot 2\text{H}_2\text{O}$ as a reductant [39]. The complex is accumulated in both kidney and liver and is fast excreted. Wang *et al.* investigated the therapeutic effects of both single and combination treatment with ^{99m}Tc -MDP (^{99m}Tc -conjugated with methylene diphosphonate, commercially named Yunke) and colloidal chromic phosphate ^{32}P (phosphonium-32) in rats with adjuvant arthritis (AA) [40]. It was found that a combination of ^{99m}Tc -MDP and ^{32}P colloid is more effective in the treatment of AA in rats compared with ^{99m}Tc -MDP or ^{32}P colloid alone. Zhang *et al.* synthesized ^{99m}Tc N complex of ciprofloxacin dithiocarbamate (CPFxDTC) as a potential agent for infection imaging. The synthesized complex was found to be a good lipophilic complex [41], and was stable at room temperature over a period of 6 h. The bacterial binding assay studies showed that ^{99m}Tc N-CPFxDTC had a better binding affinity compared to ^{99m}Tc -ciprofloxacin. Furthermore, the authors also performed the synthesis and biological evaluation of a novel $^{99m}\text{Tc}(\text{CO})_3$ complex of CPFxDTC as a potential agent to target infection [42]. The bacterial binding efficiency of $^{99m}\text{Tc}(\text{CO})_3$ -CPFxDTC was almost the same as that of ^{99m}Tc N-CPFxDTC, and was higher than that of ^{99m}Tc -ciprofloxacin. And $^{99m}\text{Tc}(\text{CO})_3$ -CPFxDTC had higher uptake at the sites of infection and better abscess/blood and abscess/muscle ratios than those of ^{99m}Tc -ciprofloxacin and ^{99m}Tc N-CPFxDTC, suggesting $^{99m}\text{Tc}(\text{CO})_3$ -CPFxDTC would be a promising candidate for further evaluation as infection imaging agent. Additionally, other radiopharmaceuticals for myocardial imaging [43, 44], tumor imaging [45], bone imaging [46, 47], liver imaging [48], kidney imaging [49], cerebral imaging [50] and blood pool imaging [51] were also studied by Chinese investigators, not illustrated here in detail for the sake of brevity.

Nowadays, the investigations of human genomics, proteomics, metabonomics, systematic biology and etiopathogenesis of human diseases provide vast opportunities for the development of radiopharmaceutical chemistry. And some advanced technologies and methods, such as computer aided

design (CAD) and “click chemistry”, are being widely used in synthesizing various radiotracers, radiopharmaceuticals, labeled compounds, and PET imaging agents, *etc.* Consequently, radiopharmaceutical chemistry in China will be fully developed in the next decades.

2.7 Education and training in radiochemistry

In China, higher education in radiochemistry, including undergraduate and graduate programs, was initially established in the 1950s. At that time, most leading universities in China provided training in radiochemistry, meeting the research demands from nuclear weapons development and other nuclear technologies. For example, from 1956 to 1957, over two hundred undergraduate students had intensive radiochemistry or nuclear physics training at Peking University. Many among this generation became the basic technical backbone of the Chinese nuclear science and technology. In the 1960s, a relatively comprehensive higher education system of radiochemistry was established in China. However, as the international nuclear chemistry and radiochemistry confronted a downward trend since the 1980s, radiochemical education in China also encountered a difficult time. With the shrinking demand for the radiochemistry specialists from nuclear industry and national defence, most universities which once provided higher education in radiochemistry started to reduce or even stop the enrollment plan related to radiochemistry. Many young students who were afraid to conduct research work related to radioactivity moved to other disciplines. Because of the adjustment of college admission professionals, undergraduate recruiting program of radiochemistry became more and more unpopular and was marginalized as a minor direction of applied chemistry in 1995. Many distinguished scholars had to give up radiochemical research and education and to turn to other chemical sciences. Correspondingly, the human resource situation of Chinese nuclear industry became serious. According to the human resource survey of China National Nuclear Corporation in 2003, the personnel constitutional status of China's nuclear industry is hard to be considered optimistic. It is estimated that only 3% of the employees of this corporation once received higher education, which obviously indicated the extremely limited innovative ability of this corporation. This situation would be seriously harmful to the national security and construction of reliable nuclear power system.

With the coming of “renaissance” of radiochemistry in the new century, the higher education in radiochemistry in China is experiencing the “Second Spring”. The Ministry of Education has formally included the nuclear chemistry and radiochemistry in the curriculum catalog for higher education, ranking it at the same status as inorganic, organic and physical chemistry. On the other hand, with the rapid growth of Chinese nuclear power industry, the demand for radiochemistry specialists is drastically increasing. According to the authorized estimates, the number of nuclear professionals needed in China would be 13,000 by 2020. In contrast, according to the statistics in 2005, the number of China's registered university students in this field was less than 500 annually. However, after over two decades of decline, the

Table 2. Five main Chinese universities and degree programs related to radiochemistry.

University or Institute	Undergraduate program	Graduate program related to radiochemistry (MS and Ph.D)
Tsinghua University	None	Nuclear fuel cycle and materials; radioactive waste treatment and disposal technologies
Peking University	Applied chemistry (radiochemistry)	Applied chemistry
Lanzhou University	Applied chemistry (radiochemistry)	Inorganic chemistry; applied chemistry; chemical engineering and technology
Beijing Normal University	None	Radiopharmaceutical chemistry
Sichuan University	Applied chemistry (radiochemistry)	Inorganic chemistry; Nuclear technology and applications

present state of higher education in radiochemistry is still far short of national needs. Fortunately, the enrollment of undergraduate and graduate students majoring in radiochemistry has begun to recover.

Around 2005, still only Peking University, Lanzhou University and Sichuan University had undergraduate training. For post-graduate training, in addition to the above three universities, Tsinghua University and Beijing Normal University can also offer graduate programs, slightly different in research directions. The five main universities and their degree programs related to radiochemistry are shown in Table 2. Furthermore, some research institutes such as CIAE, CAEP, China Institute for Radiation Protection, Northwest Institute of Nuclear Technology, Institute of High Energy Physics, Institute of Modern Physics, Shanghai Institute of Applied Physics and Institute of Plasma Physics, also provide high quality PhD and post-graduate training in radiochemistry.

With the increasing financial support from the National Natural Science Foundation of China, the Ministry of Education and National Defence Science and Industry Bureau, the infrastructure for radiochemical research in key universities has been significantly improved. Some key radiochemical laboratories have been established in recent years. For example, the Key Laboratory of Radiopharmaceuticals (Beijing Normal University) was established in 2003 and the Radiation and Radiochemistry Key Laboratory in Peking University was setup in 2007.

At present, higher education in radiochemistry is experiencing an excellent opportunity. Since 2006, it is reported that, besides the key universities such as Lanzhou University, Sichuan University, Shanghai Jiaotong University, Xi'an Jiaotong University and Harbin Engineering University, colleges of nuclear science and technology (or engineering) are being continuously established in over 25 universities; some of the endangered degree programs involving nuclear science and technology have been restored. For instance, 10 universities re-opened the undergraduate or graduate program for radiochemistry or radiochemical engineering; annually recruiting over 200 undergraduate students and 100 graduate students. This number will be hopefully doubled in a few years.

In all, higher education in radiochemistry is currently connected with the nuclear "renaissance" in China, and will definitely play pivotal roles in promoting the basic research

level of China's radiochemistry and improving the innovative potentials of China's nuclear industry.

3. Conclusion and perspectives

Entering the new century, radiochemistry is widely penetrating into other disciplines. One very representative example is the cross-fusion of radiochemistry with nano-sciences, life sciences and material sciences, which currently constitutes an important opportunity for the development of radiochemistry.

To make Chinese radiochemical researches comparable to the world's advanced level and to meet the demand of national security and nuclear power development, China still needs to strengthen the education in radiochemistry and attract more talented young scientists to this field. On one hand, we need to enhance the basic research in radiochemistry, especially in nuclear chemistry, evaluating physical and chemical properties and behaviors of nuclear fuels in strong radiation field, the radiochemistry under extreme conditions and plutonium chemistry, *etc.* On the other hand, we should promote the cross-integration of radiochemistry with other disciplines. It is necessary to establish a multi-disciplinary research center based on the large scientific facilities such as advanced synchrotron radiation facility, the advanced high neutron flux reactor and China neutron spallation source, aiming at future scientific frontiers. No doubt, it is foreseeable that radiochemistry in China will have a bright future.

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