

Besides, the XRD pattern after the complete oxidation and reduction is shown in Fig. 4. The result is in agreement with standard parameter of U_3O_8 and UO_2 respectively, which confirmed chemical conversion of uranium can be fully realized.

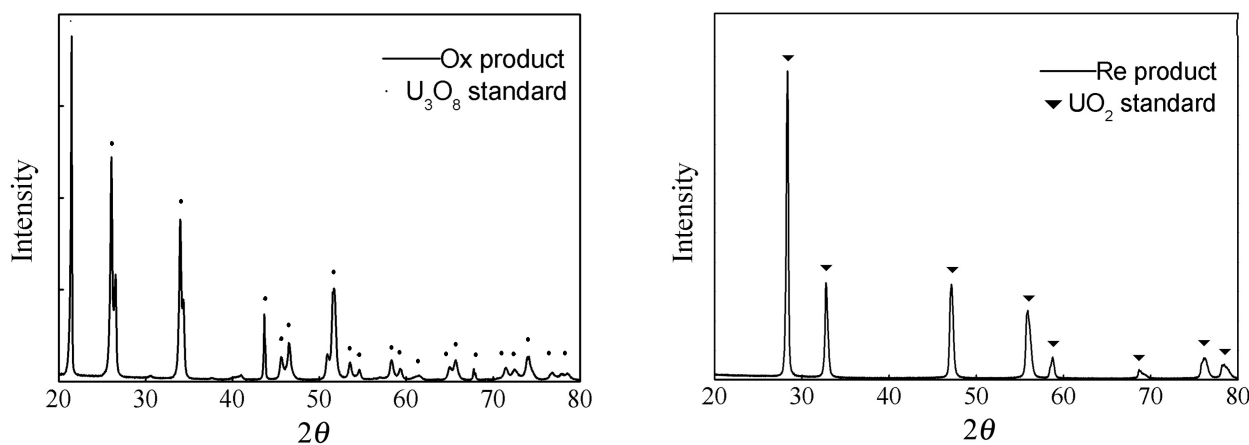


Fig. 4 XRD pattern of products after the oxidation and reduction.

Based on these, our group is doing more in-depth research for chemical composition and crystal structure of the UO_2 real fuel pellets in oxidation/reduction cycle process in order to lay a foundation for the advanced dry head-end processing of spent fuel reprocessing.

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2 - 23 Research of A New Triazoly-pyridine Ligand for Lanthanide/Actinide Extraction

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If high level radioactive waste (HLW) is directly disposed in stable deep geological formations for long periods of time (some hundred thousand years), any release of which might take place in the future will pose no significant health or environmental risk. Partitioning and transmutation (P&T) is the one of the most important strategies to reduce the long-term radiotoxicity of HLW^[1]. The basic idea is to separate the minor actinide elements (MA = Np, Am, and Cm) from the waste and convert them by neutron fission (transmutation) into shorter-lived or stable elements^[2]. The CHON type extractions as the first choice have been developed and tested for Lanthanide/Actinide separation properties, especially Kolarik showed that the BTPs (bis-triazinyl pyridine) are able to extract Am(III) with high selectivity over Eu(III) under acidic conditions^[3]. Unfortunately, these molecules extract actinide-(III) too efficiently, resulting in problems during stripping and has too slow extraction kinetics.

In our latest study, a new type of triazoly-pyridine ligand was synthesized through a series of steps (Fig. 1)^[4]. The ligand was obtained as brown solid (yield 60%) and its structure was determined by ¹H NMR (400 MHz, CDCl₃): δ ppm 8.61(s, 1 H), 7.70-7.66(m, 1 H), 7.60(s, 1 H), 7.30-7.27(m, 2 H), 7.15-7.13(m, 1 H), 5.66(s, 2 H), 3.77(s, 2 H), 2.42(s, 4 H), 1.47-1.41(m, 4 H), 1.31-1.24(m, 4 H), 0.93-0.85 (m, 6H).

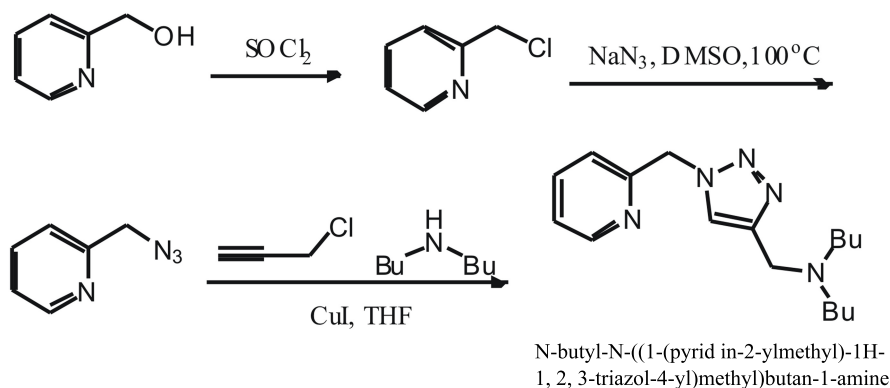


Fig. 1 Synthesis process of the new Triazolo-Pyridine ligand.

In preliminary liquid-liquid extraction experiments, radioactive actinide tracer isotope Am-241, lanthanide tracer isotope Eu-152, element Nd, and U was used. The triazolo-pyridine ligand, N-butyl-N-((1-(pyridin-2-ylmethyl)-1H-1,2,3-triazol-4-yl)methyl)butan-1-amine, is insoluble in nonpolar organic solvents, such as carbon tetrachloride, but

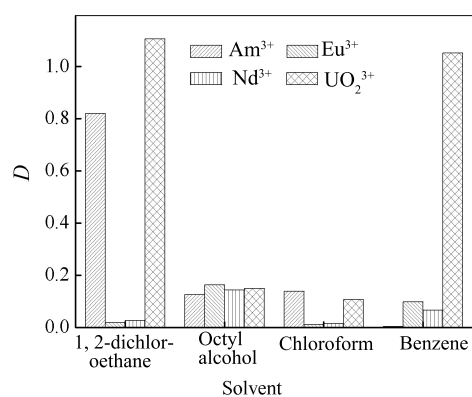


Fig. 2 (color online) Effect of solvents (pH= 4).

dissolve in most polar solvents such as chloroform and benzene. In this study, the distribution ratios are diverse as the diluents of different polarities. As shown in Fig. 2, we chose four different polar solvents to research the effect of distribution ratio, D .

In the case of low nitric acid concentration, the distribution ratio of Am³⁺ reached 0.8, not very high, in 1, 2-dichloroethane and that of UO₂²⁺ is 1.1, higher than $D(\text{UO}_2^{2+})$ in benzene. However, the distribution ratio of lanthanide was very low in the four kinds of diluents. As a result, the maximum separation factor, $SF_{\text{Am/Eu}}$, of about 30 was obtained in 1, 2-dichloroethane, which is the best one to be used as a diluent of organic phase. So the triazolo-pyridine agent can reach the requirements and purpose of the separation of lanthanide/actinide elements.

References

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