4–3 Toward High Recovery Yields of Pyrochemical Reprocessing for Minor Actinide Transmutation Nitride Fuels
— Chlorination of Neptunium in Platinum Group Elements Compounds Without Using Corrosive Gases —

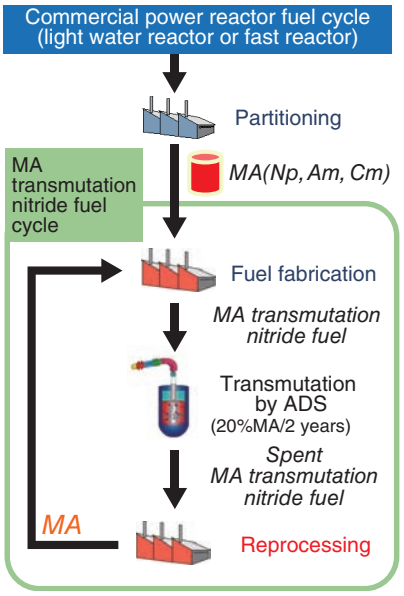

Fig.1 Outline of the proposed minor actinide (MA) transmutation nitride fuel cycle
MA remaining in the spent MA nitride fuels are selectively recovered and used in the fuel to be loaded in the core of the accelerator-driven system (ADS).

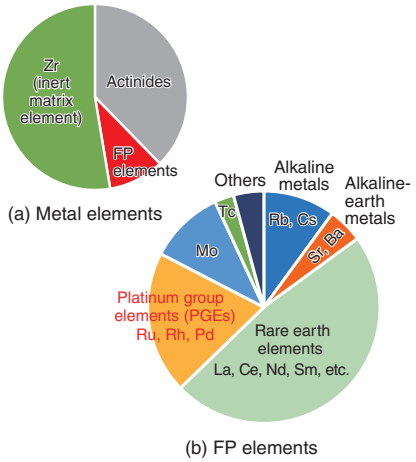

Fig.2 Composition of the spent MA transmutation nitride fuel: (a)metal elements and (b)FP elements
Calculation results for the solid components of the spent MA transmutation nitride fuel cooled for 630 days after the burnup in a typical ADS (800 MWt) for 600 days* are shown. Platinum group elements, which account for 2% of the metal elements, are expected to form stable compounds with MAs.

Fig.3 $NpPd_3$ sample obtained by the reaction of neptunium nitride with Pd

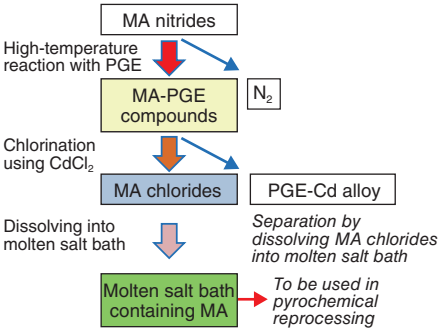

Fig.4 Formation of MA–PGE (platinum group elements) compounds in the MA nitride fuels and the proposed method for treating them

We have been conducting research and development activities on technology for partitioning elements in high-level radioactive wastes discharged from reprocessing plants and for transmuting highly radioactive long-lived nuclides to reduce the volume and radiotoxicity of radioactive wastes. For the transmutation of minor actinides (MAs) using accelerator-driven systems, nitride fuels containing high MA concentrations are chosen. Previous feasibility studies indicated that the transmutation ratio of MAs in the core is limited to 20% from the viewpoint of fuel safety. To transmute MAs more effectively, a dedicated fuel cycle, which includes the reprocessing of the spent MA fuels and reusing of the MAs recovered from the spent MA fuels, has been proposed (Fig.1).

Fig.2 shows the calculated elemental composition of the spent MA transmutation nitride fuel, in which the fission product (FP) elements are generated and MA–FP compounds are expected to be formed. To increase the recovery yields of reprocessing, treatment of stable MA–FP compounds, other than unburnt fuel materials, should be considered. We focused on the pyrochemical process in which MAs are dissolved into molten alkali chlorides and selectively recovered. With regard to the dissolving process, we aim to develop the technology without using corrosive gases such as chlorine, which can corrode the facility and equipment easily.

In this study, experiments on high-temperature reactions using neptunium, a MA, were performed. The reaction products were identified by powder X-ray diffraction analysis. We observed the formation of $NpPd_3$ by the reaction of neptunium nitride with palladium (Pd) at 1323 K (Fig.3). It was also observed that $NpPd_3$ reacts with cadmium chloride ($CdCl_2$) at 673 K to form neptunium chloride and Pd–Cd alloys.

These results suggest that intermetallic compounds with platinum group elements (PGEs) such as $NpPd_3$ are formed in spent MA transmutation nitride fuels, and the MAs in MA–PGE compounds can be chlorinated without using corrosive gases. MA chlorides easily dissolve to form a molten salt bath containing the MAs, which will be used in the pyrochemical process (Fig.4). We will continue to develop the pyrochemical process with high recovery yields by adding the process to treat stable MA–PGE compounds.

This study contains the results of "R&D on Nitride Fuel Cycle for MA Transmutation to Enhance Safety and Economy," supported by MEXT Innovative Nuclear Research and Development Program.

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*Tateno, H., Hayashi, H. et al., Material Balance Evaluation of Pyroprocessing for Minor Actinide Transmutation Nitride Fuel, Journal of Nuclear Science and Technology, vol. 57, issue 3, 2020, p.224–235.

Reference

Hayashi, H. et al., Formation of MPd_{3+x} (M = Gd, Np) by the Reaction of MN with Pd and Chlorination of MPd_{3+x} Using Cadmium Chloride, Journal of Radioanalytical and Nuclear Chemistry, vol.332, issue 2, 2023, p.503–510.