7-4 Toward Clarification of Minor Actinides Behavior in Pyrochemical Processes
—Preparation of Minor Actinide Chlorides without the Use of Corrosive Gases—

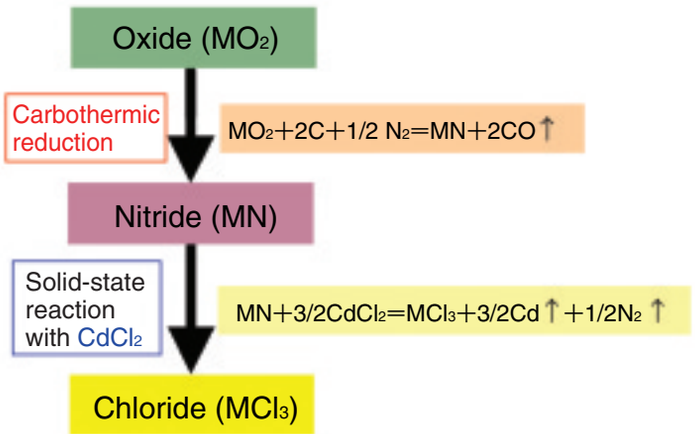

Fig.7-12 Outline of the method for the synthesis of MA chlorides
MA chlorides are synthesized from MA oxides without the use of corrosive gases such as chlorine. M stands for MA elements including Am.

Fig.7-13 Appearance of the synthesized Am chloride ($AmCl_3$)
$AmCl_3$ in fine powder form was obtained by heating the pellets of the mixture of AmN, which was synthesized by carbothermic reduction, and $CdCl_2$ at 600-723K in vacuum.

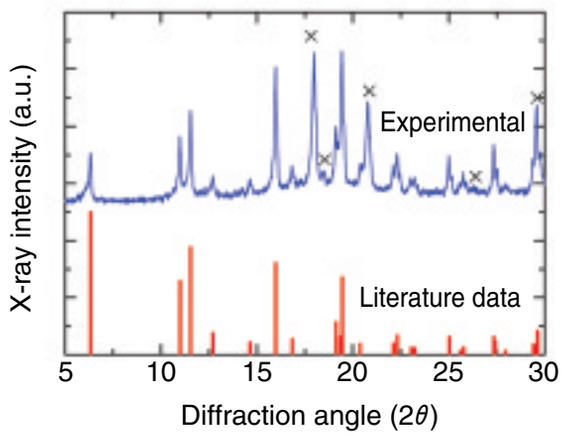

Fig.7-14 The X-ray diffraction pattern of the synthesized Am chloride ($AmCl_3$)
All the observed peaks correspond to the reported peaks of $AmCl_3$ synthesized by a conventional method. No peaks which can be assigned to impurities were found. The peaks assigned to the Pt sample holder are marked with ×.

In the advanced nuclear fuel cycles, minor actinides (MA: Np, Am, Cm), which are classified as high level wastes in the current nuclear fuel cycle, are to be recycled to reduce the burden of waste disposals. R&D of pyrochemical processes using molten salts as a solvent for treating MA-bearing fuels is underway. The pyrochemical processes are expected to be suitable for the treatment of spent nuclear fuels with high radioactivity and decay heat, and to have some advantages over hydrochemical processes in proliferation resistance, compactness, and economy. To develop the pyrochemical processes, understanding of MA behavior in molten salts is necessary, but few data are available. In order to obtain such data precisely, we installed a hot facility to handle MA chlorides in an inert gas atmosphere, because MA chlorides easily react with moisture and oxygen in air. A new method for the synthesis of MA chlorides on a gram scale without the use of HCl or Cl_2 gas, which are used conventionally, was needed because these gases can corrode the materials of the hot facility.

We developed a method for the synthesis of MA chlorides from MA oxides, which are relatively easy to obtain, without the use of corrosive gasses. This method consists of two steps. First, nitrides are synthesized from the oxides by carbothermic reduction. Then, chlorides are synthesized by the solid-state reaction of the nitrides with cadmium chloride ($CdCl_2$) (Fig.7-12). We succeeded in synthesizing high purity MA chlorides including americium trichloride ($AmCl_3$) (Fig.7-13, Fig.7-14) in the Module for TRU High Temperature Chemistry (TRU-HITEC) maintained with argon gas. The solid-state reaction of the nitrides with $CdCl_2$ is suited for synthesis not only of MA chlorides but also of other chlorides with high purity, because reagents containing oxygen are not used; they can easily react with chlorides to form stable by-products such as oxides.

Our experiments using the prepared MA chlorides are elucidating the behavior of MA in molten chlorides and the behavior of americium during the electrolysis of americium nitride in molten chlorides. We will continue to study on the behavior of MA in pyrochemical processes.

This study was carried out within the collaborative research program of TRU behavior in pyrochemical processes with Tohoku Electric Power Company, Tokyo Electric Power Company and The Japan Atomic Power Company.

Reference

Hayashi, H. et al., Synthesis of Americium Trichloride by the Reaction of Americium Nitride with Cadmium Chloride, Journal of Alloys and Compounds (2007), DOI:10.1016/j-jallcom.2007.02.011, in press.