2. Session B: Present State of the Technology Development in Japan

2.1 Research and Development of Nitride Fuel Cycle Technology in Japan

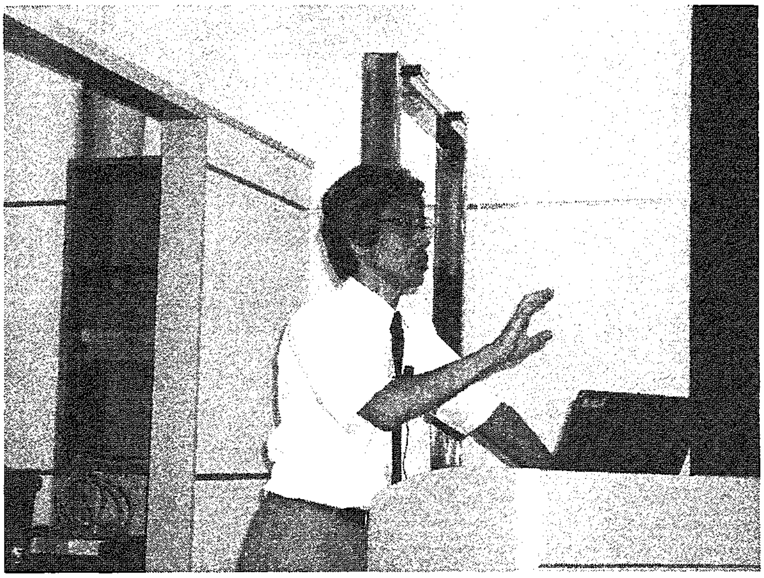
Kazuo MINATO [1], Yasuo ARAI [1], Mitsuo AKABORI[1], Masayoshi UNO [2], Yoshihisa TAMAKI [3], Kunihiro ITOH [4]

[1] Japan Atomic Energy Research Institute

[2] Graduate School of Engineering, Osaka University

[3] Mitsubishi Materials Corporation

[4] Nuclear Development Corporation,



Research and Development of Nitride Fuel Cycle Technology in Japan

Kazuo MINATO [1], Yasuo ARAI [1], Mitsuo AKABORI[1], Masayoshi UNO [2], Yoshihisa TAMAKI [3], Kunihiro ITOH [4]

[1] Japan Atomic Energy Research Institute, Tokai-mura, Ibaraki-ken, 319-1195 Japan
[2] Graduate School of Engineering, Osaka University, Suita-shi, Osaka, 565-0871 Japan
[3] Mitsubishi Materials Corporation, Naka-machi, Ibaraki-ken, 311-0102 Japan
[4] Nuclear Development Corporation, Tokai-mura, Ibaraki-ken, 319-1111 Japan
minato@popsvr.tokai.jaeri.go.jp

Abstract

The research on the nitride fuel was started for an advanced fuel, (U,Pu)N, for fast reactors, and the research activities have been expanded to minor actinide bearing nitride fuels. The fuel fabrication, property measurements, irradiation tests and pyrochemical process experiments have been made. In 2002 a five-year-program named *PROMINENT* was started for the development of nitride fuel cycle technology within the framework of the Development of Innovative Nuclear Technologies by the Ministry of Education, Culture, Sports, Science and Technology of Japan. In the research program *PROMINENT*, property measurements, pyrochemical process and irradiation experiments needed for nitride fuel cycle technology are being made.

1. Introduction

The research on the nitride fuel was started for an advanced fuel for fast reactors. Fuel fabrication, property measurements, irradiation tests and pyrochemical process experiments of (U,Pu)N fuel have been made, and these activities have been expanded to minor actinides (MA: Np, Am, Cm) bearing nitride fuels for transmutation of these elements. The main player in this field in Japan is the Japan Atomic Energy Research Institute (JAERI). The nitride fuel has good potentials. The melting point of the nitride fuel is higher than the metal fuel and comparable to the oxide fuel. The thermal conductivity of the nitride fuel is higher than the oxide fuel and comparable to the metal fuel. The nitride fuel supports a hard neutron spectrum needed for fissions of the minor actinides. The actinide mononitrides would form solid solution, which could accommodate a wide range of the combination and composition of actinides. Highly enriched N-15 would have to be used for the nitride fuel in order to prevent the formation of hazardous C-14. By applying pyrochemical process in the treatment of spent fuel, N-15 could be readily recovered and recycled.

For the development of nitride fuel cycle technology, a five-year-program named *PROMINENT* was started in 2002 within the framework of the Development of Innovative Nuclear Technologies by the Ministry of Education, Culture, Sports, Science and Technology of Japan. In the research program *PROMINENT*, property measurements, pyrochemical process and irradiation experiments needed for nitride fuel cycle technology are being made.

In the present paper, the research and development of nitride fuel cycle technology in Japan is reviewed and the research program *PROMINENT* is introduced.

2. Fast reactor and transmutation fuels

The research activities of the nitride fuel fabrication, property measurements, pyrochemical process experiments and irradiation tests for fast reactors and for transmutation before starting the research program *PROMINENT* are summarized briefly in this chapter.

2.1 Fuel fabrication

Actinide nitrides of UN, PuN and NpN with high-purity have been produced from respective oxides with carbothermic reduction method, and the high-density pellets of UN, PuN and NpN and their solid solutions (U,Pu)N, (U,Np)N and (Np,Pu)N were successfully fabricated [1-4]. AmN, (Pu,Cm)N, (Am,Y)N and (Am,Zr)N were also synthesized with carbothermic reduction method though the batch sizes of them were in the order of ten mg[5-7]. The U-free fuel pellets with inert matrix materials (Pu,Zr)N and PuN+TiN were fabricated for irradiation tests [8]. Figure 1 shows (Am,Zr)N and (Pu,Zr)N pellets.

Fabrication of mixed nitride fuel pellets with high-purity and high-density in the (U)-Pu-Np-Am-Cm system is needed for the next step for the nitride fuel development.

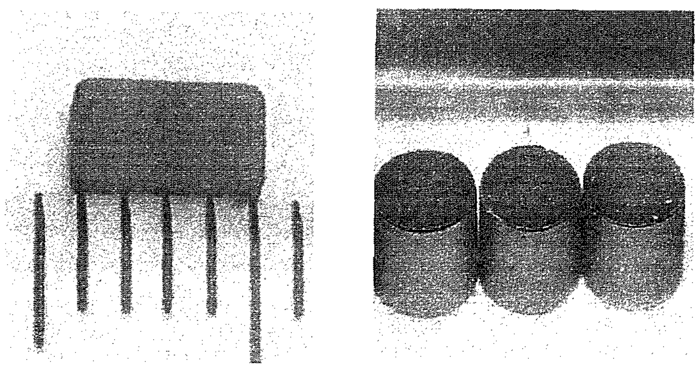


Fig. 1 (Am,Zr)N (left) and (Pu,Zr)N (right).

2.2 Property measurements

Vaporization behaviors of UN, NpN, PuN and the solid solutions of (U,Pu)N and (Np,Pu)N were studied with Knudsen-effusion high temperature mass-spectrometry [9-11]. The specific heat capacities of UN, NpN and PuN were measured with a differential scanning calorimeter [12]. Preliminary experiments of the thermal expansion measurements of UN by high-temperature X-ray diffraction were made. The thermal diffusivities of UN, NpN, PuN and the solid solutions of (U,Pu)N, (U,Np)N and (Np,Pu)N were measured by the laser-flash method and the thermal conductivities were determined as a function of temperature [13-15]. Figure 2 shows thermal conductivities for the UN-NpN-PuN system as a function of composition at 1273 K.

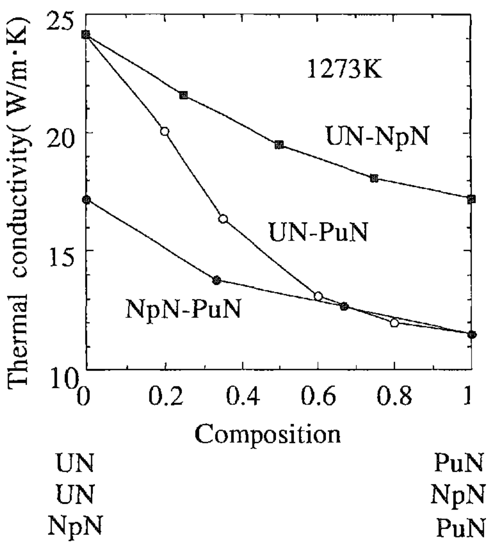


Fig. 2 Thermal conductivities of (U,Pu)N, (U,Np)N and (Np,Pu)N.

The properties for Am and/or Cm bearing nitrides were not available. These properties data are needed to design the MA-bearing fuels and to understand the irradiation behavior of them.

2.3 Pyrochemical process

The pyrochemical process developed by ANL for the metal fuel was applied to the nitride fuel. Electrolyses of UN, NpN and PuN in LiCl-KCl eutectic melt were made, and electro-depositions of U, Np and Pu metals on solid cathodes were carried out [16-18]. Electrode reactions of Np and Pu at liquid Cd and Bi electrodes were studied, and recovery of Pu into liquid Cd was made successfully [19]. As a feasibility test of N-15 recycling, dissolutions of DyN and NdN in LiCl-KCl by $CdCl_2$ were made, and nitrogen could be recovered as N_2, as shown in Fig.3 [20]. During the electrolysis of UN and PuN, nitrogen evolving as N_2 was observed and nitrogen could be recovered more than 90% as N_2.

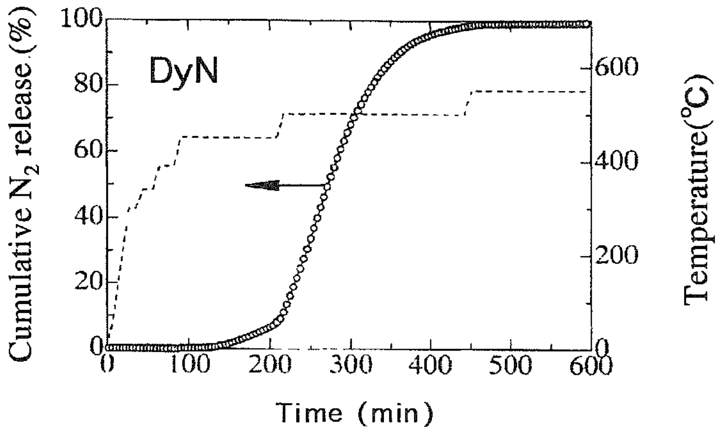


Fig. 3 Evolving behavior of N_2 gas.

For the next step, the electrolysis behavior and electrode reactions for Am and Cm need to be clarified, and re-nitridation of actinide elements recovered in the liquid metal cathode should be made.

2.4 Irradiation tests

Irradiation tests of He-bonded (U,Pu)N fuel pins have been carried out. Four fuel pins of (U,Pu)N were irradiated in the Japan Materials Testing Reactor (JMTR) [21], and two fuel pins of (U,Pu)N were irradiated in the fast test reactor JOYO under the joint research with the Japan Nuclear Cycle Development Institute (JNC). The irradiation behavior of the fuels was found to be good up to 5.5%FIMA. The irradiation tests of U-free fuels (Pu,Zr)N and PuN+TiN are under irradiation in JMTR. Irradiation tests of MA-bearing fuels are needed for the next step.

3. Research program *PROMINENT*

The research program *PROMINENT* was started in November 2002 and is to be valid till March 2007. The purpose of the program is to clarify the feasibility of an innovative nuclear fuel cycle including minor actinides recycling based on the nitride fuel and pyrochemical reprocessing. In the program, the fuel fabrication and pyrochemical process technologies are developed and the fuel properties and irradiation behavior are studied for the MA-bearing fuels, as shown in Fig. 4.

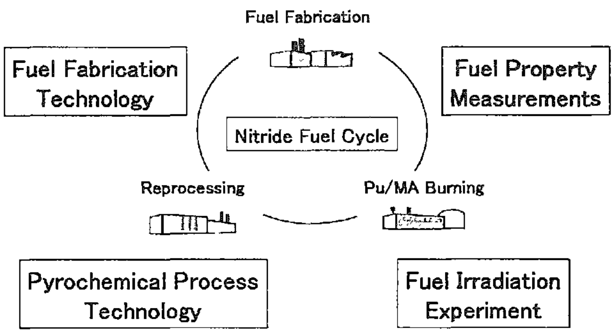


Fig. 4 Research areas in the program *PROMINENT*.

3.1 Fuel fabrication technology

Nitride is hard to be sintered and Am vaporizes easily during fabrication. One of the objectives is to fabricate high-density and high-purity nitride pellets. To survey a suitable fabrication condition, lanthanide elements are used as surrogates for actinide elements, which aims to accelerate the experiments. Mixed nitrides of the Np-Pu-Am-Cm-N system will be fabricated with the carbothermic reduction method. The other objective is to fabricate simulated burnup fuels, where non-radioactive fission product elements are added to uranium nitride. These fuels are supplied to the property measurements and pyrochemical reprocessing tests to support the understanding and modeling of the properties of irradiated nitride fuels and the development of pyrochemical process.

3.2 Fuel property measurements

The handling of minor actinides nitrides is not as easy as that of uranium nitride because of their radioisotopic nature. The size of the specimens containing minor actinides is small, and the number of specimens available is also small. The property measurements of minor actinides nitrides take a longer time since equipments for property measurements are in glove boxes and/or even in hot cells. One of the objectives is to measure thermal properties of Np-Pu-Am-Cm mixed nitrides. For this purpose, new equipments are installed in glove boxes. The second objective is to measure thermal and mechanical properties of the burnup simulated fuels. These measurements support the understanding and modeling of properties of MA-bearing nitride fuels as a function of burmup. The third objective of this area is to develop micro-indentation technique for mechanical property measurements of MA-bearing nitride fuels.

3.3 Pyrochemical process

To develop the pyrochemical process for the nitride fuel, experiments are carried out with MA-bearing nitride and burnup simulated fuels. One of the objectives is to understand the behavior of Am, Cm and fission products at anode and cathode during electrolysis. The other objective is to understand re-nitridation behavior of actinide elements recovered in liquid Cd and to fabricate fuel pellets with recovered actinide nitride. For the enrichment of N-15, the enrichment level of N-15 is evaluated taking account of C-14 production and the cost of enrichment. The process of N-15 recovery in the pyrochemical process and fuel fabrication process is discussed.

3.4 Irradiation experiment

MA-bearing nitride fuel has not been irradiated in fast neutron spectrum. We participate in the irradiation program FUTURIX, where MA-bearing nitride fuels are to be irradiated in Phenix. JAERI will make a contract with CEA on this subject.

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