Nuclear Data for Innovative Reactors Having Persistent Resistance to Nuclear Proliferation

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The Protected Plutonium Production (P^3) project has been initiated in order to realize the proliferation-resistant plutonium. The first stage of this project is based on the idea to protect plutonium by ^{238}Pu generated from ^{237}Np doped in the fresh uranium fuel. Along with the burnup going on, the doped ^{237}Np is transmuted via ^{238}Np to ^{238}Pu, which is the intense neutron source of spontaneous fission ($2.6x10^3$ n/g/s). These neutrons will deteriorate the quality of the plutonium as the nuclear explosive material. In addition, high alpha-decay heat of ^{238}Pu (560W/kg) makes the processes of the explosive fabrication and its maintenance technologically difficult. In this background we surveyed the status of nuclear data relevant to the P^3-concept reactor design. The reliability of the capture cross section data for ^{237}Np and ^{238}Pu are far from satisfactory and more accurate experiments and evaluations are required. As fissions in these nuclides do not always play any decisive role in the P^3 concept LWR, we can be fairly tolerable about the uncertainty of the fission cross section data, but further study is needed.

1. Introduction

Converted use of reactor-fuel material to explosives is one of the most critical issues which must be resolved for the peaceful and sustainable use of the nuclear power. In this context the "Protected Plutonium Production (P^3)" concept has been proposed by M. Saito et. al [1,2] for the purpose of realizing the proliferation-resistant plutonium. In this P^3 concept, the plutonium produced in the nuclear reactors from ^{238}U is made highly proliferation-resistant by introducing a certain amount of the ^{238}Pu isotope and this is accomplished by doping some appropriate minor actinide (MA) in the fresh uranium fuel for the LWRs. The first stage of this project (P^3-A project [3]) focus on the transmutation of ^{237}Np, which has a large neutron capture cross-section, into ^{238}Pu without deteriorating, or even with improving, of the reactor-core performances. The isotope ^{238}Pu, which is the intense neutron source of spontaneous fission ($2.6x10^3$ n/g/s), will deteriorate the quality of the plutonium as the nuclear explosive material. In addition, high alpha-decay heat of ^{238}Pu (560W/kg) makes the processes of the explosive fabrication and its maintenance technologically very difficult.

Based on the preparatory studies for several years, P^3-A project has been initiated and carried out since 2003 by the financial support of the Ministry of Education, Culture, Sports, Science and Technology as an international cooperative work among the following organizations. They are Tokyo Institute of Technology, Idaho National Engineering and Environmental Laboratory, Musashi Institute of Technology, Japan Nuclear Cycle Development Institute, Japan Atomic Energy Research Institute, Engineering Development Co. Ltd., Mitsubishi Heavy Industries, Ltd., Nuclear Development Corporation, Advanced Reactor Technology Co., Ltd., Toshiba Corporation and Fuji Electric System Co., Ltd. The present paper describes the neutronic basis of the P^3-A project and then review the status and the needs of the nuclear data pertinent to this concept.

2. Generation and Depletion of ^{238}Pu

In the P^3-A concept[3], the potential of ^{237}Np is fully pursued as a novel fertile material which is transmutes into ^{238}Pu through a successive neutron capture and beta-decay. If we start LWR operation with the fresh uranium fuel doped with an appropriate amount of ^{237}Np, the plutonium generated mainly from ^{238}U in the reactor is persistently accompanied by a sizable amount of ^{238}Pu isotope coming from ^{237}Np at any stage of fuel burnup, reprocessing and storage as indicated below.

$$^{237}\mathrm{Np} + \mathrm{n} \longrightarrow {}^{238}\mathrm{Pu} / {}^{239}\mathrm{Pu} \longleftarrow {}^{238}\mathrm{U} + \mathrm{n}$$

(Fuel Protection) (Fuel Production).

The isotopic presence of ^{238}Pu makes it difficult to use the produced plutonium as the nuclear explosive material without some sophisticated isotope-separation technology, the atomic vapor laser method for example. In this sense the generated ^{238}Pu plays the role of the fuel protector against the nuclear proliferation.

Another merit of doping ^{237}Np in the fuel lies in the fact that it exerts a favorable effect of extending the fuel burnup. The capture cross section in ^{237}Np is much larger than the fission cross section in the LWR environment and, therefore, it reduces the reactivity at the beginning of the burnup. Then, along with the burnup going on, it is transmuted to ^{238}Pu, which has larger fission cross section than ^{237}Np, and further into

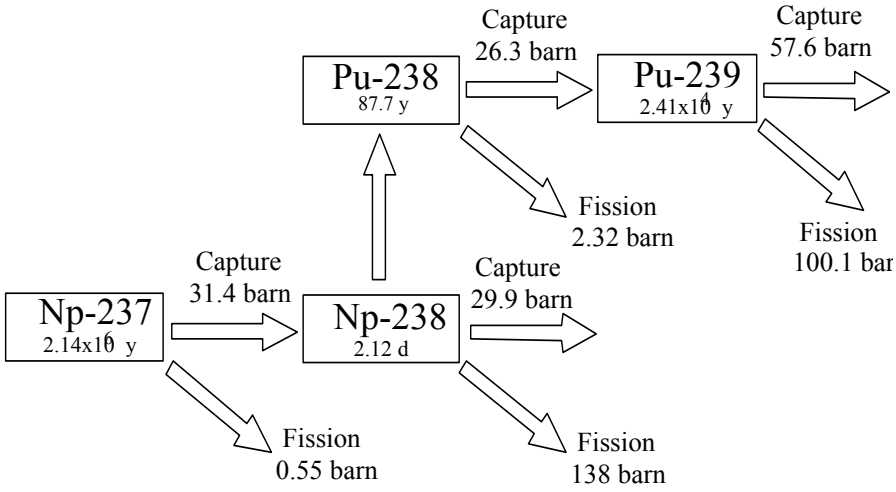

Fig. 1 Generation path of ^{238}Pu and ^{239}Pu from ^{237}Np

^{239}Pu through neutron capture. This means the negative reactivity of ^{237}Np is gradually replaced by the positive reactivity of plutonium. In this sense the doped ^{237}Np plays a role of an *active* burnable poison. Figure 1 shows the generation paths of Pu isotopes from ^{237}Np. The cross sections indicated in this figure are taken from ORIGEN-2 Library PWR47J33.LIB based on JENDL-3.3 for reference, which gives the effective one-group cross sections in PWRs with 4.7% enriched uranium fuel. As we can see here the capture reaction dominates in ^{237}Np and the ^{238}Np generated as the result will soon decay into ^{238}Pu. Note that, however, the fission and capture cross sections of this short-lived isotope are very large. In reality we can not ignore the neutron reaction in ^{238}Np under very high neutron flux though the life of this isotope is as short as 2.12 days. The quite recent measurement of the capture cross sectio of ^{238}Np by Harada et al.[4] indicates even larger value. The produced ^{238}Pu mainly undergoes capture reaction because the capture cross section is roughly one order larger the fission cross section.

Figure 2 indicate the fission rate in a P^3-A concept PWR along with the capture reaction rate in ^{238}Pu for reference. In ^{238}Pu the capture reaction (broken line) largely exceeds the fission reaction (triangle) at any satge of the burnup. This figure also indicates that the fission rate in ^{238}Pu isotope, which plays the dominant role in the P^3 concept, is one-order maller than that of ^{239}Pu. Even in the P^3 concept LWR, the reactivity is hold by ^{235}U fission in the low burup satge and then ^{239}Pu follows it in the later stage.

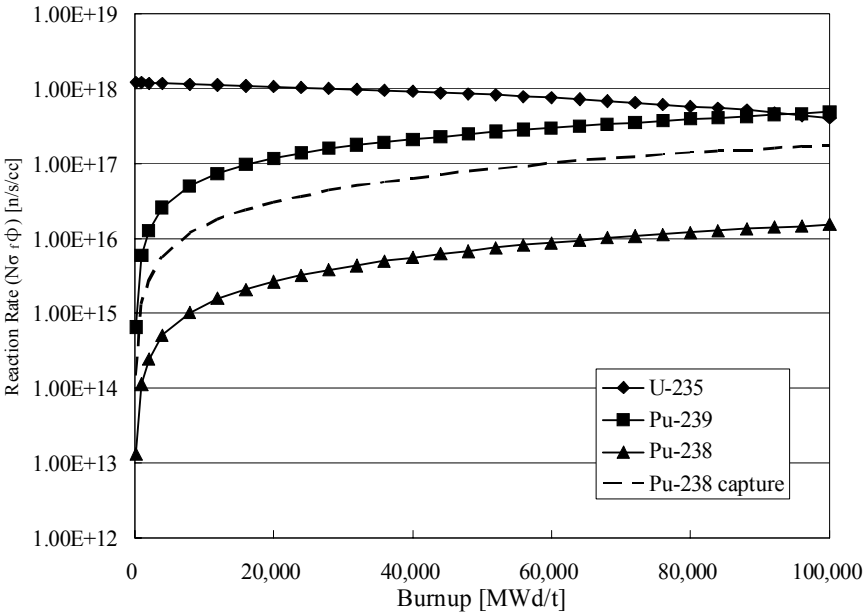

Fig. 2 Fission rate in a P^3-A concept PWR along with the capture reaction rate in ^{238}Pu. The uranium enrichment and the ^{237}Np doping fraction is 10% and 2%, respectively.

3. Nuclear Data

In the P^3 concept reactors, the isotopic fraction of ^{238}Pu is assumed to be fairy large. The percentage becomes from ten to several tens in total plutonium[3]. It is seen also from Fig.2, where the neutron capture rate in ^{238}Pu is comparable to ^{239}Pu fission rate at any stage of the burnup. The capture cross sections of ^{237}Np and ^{238}Pu are very important because these govern the production and depletion of ^{238}Pu, the protector of plutonium.

Figure 3 shows the capture cross section of ^{237}Np in the thermal energy region. Two libraries, ENDF/B-VI and JEFF-3 give 181.02 barns at 0.0235 eV after the rather old experiment by Weston and Todd[5]. For this particular value, JENDL-3.3 gives 161.71 barn on the basis of the measurement by Kobayashi et al.[6]. In addition Katoh et al.[7] reported 141.7 barns in their very recent experiment. This new value is 13% lower than JENDL-3.3, which gives the 12% lower value than ENDF/B-VI and JEFF-3. Considering this situation we have to assume an uncertainty more than ±10%. Being more conservative it might be ±20%. From a preliminary sensitivity study this value of uncertainty in the capture cross section give rise to the same order of ambiguity in the ^{238}Pu concentration generated from ^{237}Np. As for the energy range from thermal to keV we can, however, expect a new experimental data from a new Japanese project, in which Igashira et al.[8] plan to develop a novel neutron detector and obtain the neutron data for important MAs including ^{237}Np by applying it. Uncertainty mentioned above is expected to be remarkably reduced in a few years by their data.

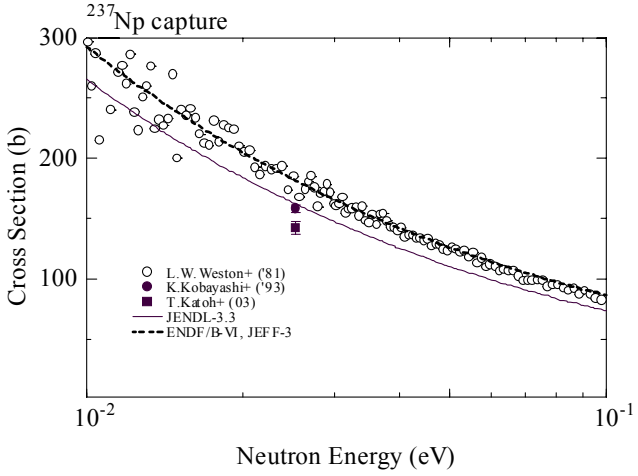

Fig.3 Capture cross section of 237Np

Another important reaction, which dominates the concentration of ^{238}Pu, is the capture in ^{238}Pu itself, because it is depleted mostly through this reaction. Further this reaction dominates the production of ^{239}Pu. Remember it is also generated from ^{238}U which is the main stream of the ^{239}Pu production. The present status of the capture cross section data is shown in Fig. 4. Both JENDL-3.3 and JEFF-3 adopt the same value of 540.3 barns after the rather old recommendation by Mughabghab[9], which is 540±7 barns. ENDF-B/VI takes 561.08 barns, that is 4% higher than the other two libraries.

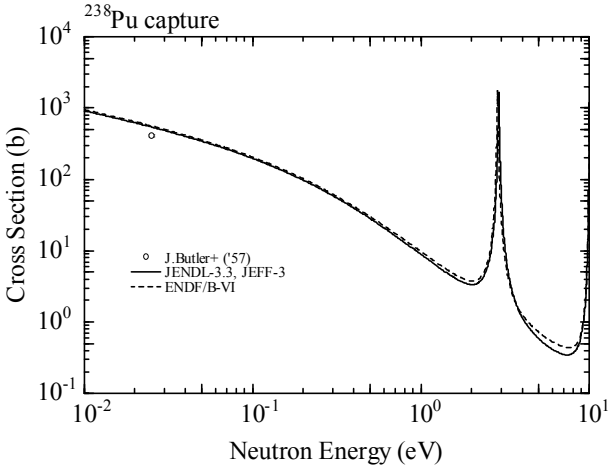

Fig. 4 Capture cross section of 238Pu

In this sense, the three libraries are rather consistent with each other because the 4% difference is not sizable. The experimental value, however, is very scarce and, then, we should expect large uncertainty in this cross section and need new reliable experimental data. Anyway, this will be one of the key cross section data in the P^3-concept reactor design.

As we saw in Fig. 2 the fission in ^{238}Pu is more than one order smaller than those in ^{235}U and ^{239}Pu. This means that this reaction play only a small role in the neutronic performance of the P^3 concept LWR core. In this sense fairy large uncertainty is tolerable for the ^{238}Pu fission cross section data. This is also the case for ^{237}Np fission because the effective fission cross section in LWR is even four times smaller than ^{238}Pu as we can see in Fig. 1. The ^{237}Np and ^{238}Pu are, however, plays dominant roles in the P^3 concept reactor. Therefore we have to try to evaluate and validate all the nuclear data more strictly for the P^3 Project than ever. For validation of the nuclear data, irradiation experiments are planned at Advanced Test Reactor in INEEL and also in the Japanese fast test reactor JOYO in the framework of the P^3-A project[10]. Some sample reactivity measurements at critical facilities are also in the scope. We can expect that the nuclear data needed for design of P^3-concept reactors will be tested and improved through the analysis of these experimental data.

4. Concluding Remarks

Peaceful use of nuclear energy will not go forward without solving the problem of the nuclear proliferation. A new concept called P^3 (Protected Plutonium Production) was proposed by M Saito et al. to resolve this problem. The concept is based on the fuel protection by ^{238}Pu isotope generated from ^{237}Np doped in the fresh fuel. We surveyed the status of nuclear data relevant to P^3-concept reactor design. The reliability of the capture cross section data for ^{237}Np and ^{238}Pu is far from satisfactory and more accurate experiments and evaluations are required. As fissions in these nuclides do not always play any remarkable role in the P^3 concept LWR, we can be fairly tolerable about the uncertainty of the fission cross section data.

Acknoledgement

This work has been supported by the Ministry of Education, Culture, Sport, Science and Technology.

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