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Neutronics analysis of Americium-based fuel for long-life core

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ABSTRACT

Feasibility study on the ultra long-life lithium (Li) cooled fast reactor loaded with conventional LWR-grade Am has been performed. Americium-241 has a potential to reduce the initial excess reactivity because it has relatively high capture cross section to be effectively converted to ^{238}Pu, a fissile nuclide in fast neutron spectrum. For the better neutron economy, ^{7}Li enrichment in coolant, nitride fuel with ^{15}N enrichment, and lead–bismuth (Pb–Bi) reflector were selected and parametrically analyzed to find the optimal condition of criticality achievement with Am-based fuel. In the case of single region homogeneous core with only Am nitride fuel, it was found the condition of criticality sustained more than 100 years operation though the core has a large gradient of flux level distribution. The flattening of geometrical neutron flux distribution were also studied by adjustment of the fuel composition of Am and fissile material in dual region core. With these mechanisms, the change of burn-up reactivity was within 3% and ultra long-life core with over 100-year-life and less than 1.5 radial peaking factor could be achieved simultaneously throughout the operation. Safety parameters such as Doppler and void coefficient are also improved by dual region core. This mechanism of ultra long-life core is expected to be applied to future nuclear reactor concepts such as a space nuclear reactor.

1. Introduction

In order to support the human activities in space, stable power resources for decades of long period will be required in the future. Because of the low power density and very severe mass restriction by the ability of rocket performance, there are difficulties to satisfy these demands by solar energy and chemical battery such as fuel cell. Space nuclear reactor is one of the answers to solve these problems. Space nuclear reactor is very attractive in its utilization as a long-life stable electric power resource for human activities in the space in, for example, Moon and Mars. Many concepts have been suggested to satisfy these contradicted conditions, and most of the concepts were based on the highly enriched uranium fuel. Highly enriched uranium, ^{235}U enriched over 20%, however, is defined as a "*direct use nuclear material*" for military purpose by IAEA (2001), there are many institutional restrictions to utilize such a fuel in non-weapon possessing countries.

Minor Actinides (MAs) has a very attractive potential to enables long life core both thermal and fast neutron reactor (Nikitin et al., 1999; Saito, 2002) because MAs of spent fuel in LWR have large neutron capture cross section. Mainly there are three elements in MAs, Neptunium (Np), Americium (Am) and Curium (Cm). Neptunium-237 and ^{241}Am have a comparable fission and capture cross section in fast neutron, the loading of these nuclides in the core does not affect the eigenvalue (*k-eff*) in unity, though Cm isotopes affects *k-eff* largely because of its large ν value compared to the values in Np and Am isotopes. In the transmutation chain of ^{237}Np and ^{241}Am by neutron capture shown in Fig. 1, these nuclides are mainly transmuted to ^{238}Pu with different paths, in the case of ^{237}Np, it takes the path of $^{237}\mathrm{Np}(n,g) \rightarrow {}^{238}\mathrm{Np}(\mathrm{beta}, 2.1\ \mathrm{d}) \rightarrow {}^{238}\mathrm{Pu}$, and in the case of ^{241}Am, it takes the path of $^{241}\mathrm{Am}(n,g) \rightarrow {}^{242}\mathrm{Am}(\mathrm{beta}, 16\ \mathrm{h}) \rightarrow {}^{242}\mathrm{Cm}(\mathrm{alpha}, 163\ \mathrm{d}) \rightarrow {}^{238}\mathrm{Pu}$. Because the half life of the compound nuclide in ^{237}Np neutron capture reaction, ^{238}Np, is 2.1 days shorter than that in ^{241}Am neutron capture reaction, ^{242}Am(beta, 16 h) and ^{242}Cm(alpha, 163 d), the production rate of ^{238}Pu, working as fissile nuclide in fast neutron, from ^{241}Am is more gradual than that from ^{237}Np. It is expected that the quantity of ^{241}Am would be increased very much in the future by short half-life of ^{241}Pu(beta, 14.3 y) included in Pu accumulating in the Pu fuel cycle, and ^{242m}Am is a very attractive fissile nuclide for designing a small space reactor because it has very large fission cross section (Ronen and Leibson, 1998). Furthermore, no usage of highly enriched uranium and Pu can make flexibility of technology development in non-weapon state countries. The present paper focuses on the possibility of long-life core loading Am.

As a coolant material, lithium (Li) is utilized in the present paper since it enables high efficiency of electricity conversion because of high boiling point and specific heat (Kambe and Uotani, 1997; Nikitin et al., 2003). Because gravity and air utilization in space is negligibly small, removal heat of convection and conductivity is not effective but radiation, so high operating temperature

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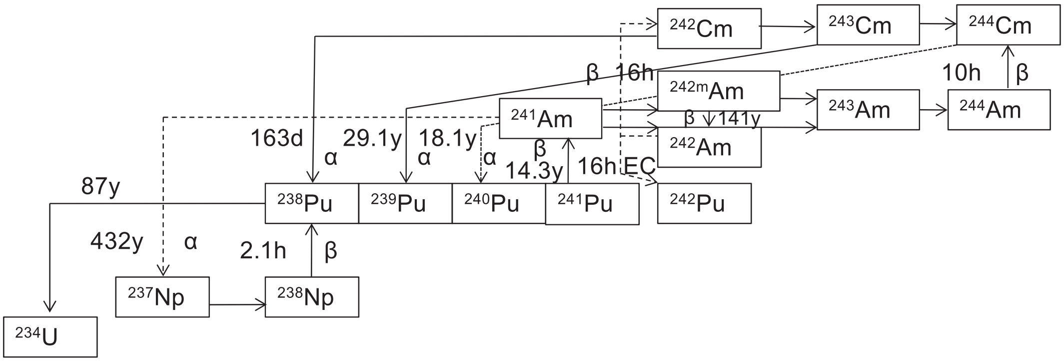

Fig. 1. Transmutation chain of trans-uranium.

is desirable from the viewpoint of much removal heat not converted to electricity. Though carbide and nitride fuel combine high thermal conductivity comparable to metallic fuel and high melting point comparable to oxide fuel, Am nitride is selected in the present paper because Am is very stable in nitride form chemically. Isomeric ratio of ^{242g}Am in total ^{241}Am capture cross section is used as 0.84 in fast spectrum based on recent experimental results (Sagara et al., 2010).

2. Goal of core specification

A small ultra long life core which enables launch by rocket and easily control in a remote area such as Moon and Mars is designed as a design goal. In the near future, core enables human activities in space has ultra long nuclear life time over 100 years as a first goal. It has a little burn-up reactivity change (under 10%) for reduction of load of equipment for control during burn-up as a second goal. It is decided to keep radial peaking factor (z = 35–40 cm) under 1.5 during burn-up using calculation result of JOYO MK-2 as a reference because it is desirable that radial neutron flux distribution is flat for control of burn-up reactivity by neutron absorber with control drum in outside of core is as a third goal. Finally, it is decided to keep negative Doppler coefficient to maintain minimum safety as a fourth goal. It is defined as Doppler coefficient that difference of reactivity between reference core temperature and increased up with 500 K and it is defines as void coefficient that difference between normal operating Li density and that changed to 0%. Goal of core specification is the followings:

1. Core life time is over 100 years.
2. Burn-up reactivity change is under 10%.
3. Radial peaking factor is under 1.5 during burn-up.
4. Negative Doppler coefficient.

3. Calculation model

Computer codes, SLAROM (Nakagawa and Tsuchibashi, 1984), JOINT and CITATION (Fowler et al., 1971) and cross section library, JFS-3-J-3.2R (Chiba et al., 2002), which was based on Japanese Evaluated Nuclear Data Library JENDL3.2 (Nakagawa et al., 1995), were used in the present calculation. The SLAROM input consisted only of the PREP block to obtain 70-group effective cross sections of each material region by homogeneous cell calculation. JOINT was used to convert 70-group effective cross section data sets from the SLAROM output to the CITATION input. The nuclear characteristics were investigated using a calculation of two-dimensional RZ diffusion theory with depletion chain by CITATION. In this calculation, each zone had uniform nuclide number densities, neutron flux, and neutron spectrum averaged effective cross-sections with 80 zones. Main transmutation chains shown in Fig. 1 were used in the present analysis. After each burn-up calculation, the average number densities were obtained in each zone. The geometry of the core was shown in Fig. 2 composed of a cylindrical core with 40 cm radius and 50 cm height and a reflector with 9 cm thickness radially and 15 cm thickness axially. Number densities of fuel and coolant were used with volume ratio obtained by MONJU. As Fig. 2 shows, nitride fuel was used as a fuel which has high melting point, thermal conductivity, heavy metal density, and smear density (85%), SS316 was used as a cladding in fast reactor and Li was used as a coolant which has high specific heat and melting point from the viewpoint of high conversion efficiency from heat to electricity. Furthermore, Beryllium (Be) reflector was used because it has a large scattering cross section and can be utilized to decrease neutron leakage in the small core. Calculation was done with mean temperature of inner core and outer core are 1273, 973 K, respectively. Cylinder for same mass has minimum S/V value (S and V are defined as surface and volume of core) as a function of core radius. S/V value is close to minimum, i.e. leakage of neutron is small compared to core volume, core radius = 40 cm, core height = 50 cm is chosen as a reference model. It is a two dimensional cylinder core of an equivalent size with JOYO MK-3.

4. Results

4.1. Parametric survey for criticality of single region core with Am nitride fuel

A Li-cooled small reactor core with only LWR-grade Am nitride fuel is studied parametrically. Atomic density ratio of Am was used as shown in Table 1. For the better neutron economy, ^{7}Li enrichment in coolant, ^{15}N enrichment in nitride fuel and a material for the reflector are considered as parameters shown in Table 2. Single region core means that fuel Am nitride fuel is loaded in inner core and outer core homogeneously. Thermal power of the whole reference core is designed as 5MW$_t$ and specific power is 4.3 MW$_t$/t. Mass loading, decay heat and radioactivity of initial Am nitride fuel in total core are estimated as about 1200 kg, 0.1 MW$_t$ and 4.4 MCi (3.7 Ci/g). In case A, natural Li as a coolant, Be as a reflector. Am nitride as a fuel with natural nitrogen isotope ratio is used. As a result of burn-up reactivity change in the case A, it takes subcritical during burn-up as shown in Fig. 3 with bold line. In the case B, pure ^{7}Li is used to reduce an excess neutron capture of ^{6}Li(n,t), and a result is shown in Fig. 3 with dashed line. In the case C, Be of reflector is replaced to Pb–Bi eutectic (Pb–Bi, Pb:Bi = 45:55) for hardening the neutron spectrum beside the reflector function and enhancing fission of Am and a result is shown in Fig. 3 with dotted line. Though the neutron reflection function of Be is much larger than that of Pb–Bi, burn-up reactivity