

Source Intensity Analysis of DUPIC Fuel

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DUPIC 핵연료의 조사선량률 분석

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Abstract - Source intensities in terms of the exposure rates at 1 m from the fresh and spent DUPIC fuels, made from standard and extended burnup PWR fuels, were analyzed. Two cases were studied based on the degree of elimination of removable elements. Homogeneous mixture model was applied to get the exposure rate. The exposure rate turned out to be very high and sensitive to Cs elimination during the dry process. About 90% of exposure can be reduced in the case of fresh DUPIC fuel made from 10-year cooled spent PWR fuels if Cs is fully removed during the dry process. The main radiation source in spent fuels is Cs-137. The dry storage of spent DUPIC fuel may need a longer wet storage period and require a further review.

Key words : Source intensity, DUPIC fuel, exposure rate

요약 - 사용전 그리고 사용후 DUPIC핵연료의 선원분석을 연료다발에서 1m 떨어진 지점의 조사선량률을 기준으로하여 분석하였다. DUPIC핵연료 제조에 사용된 PWR핵연료는 표준 연소도와 장주기 연소도를 갖는 것으로 설정하였고, 건식 가공에서 제거되는 핵분열생성물의 양을 고려하여 두 가지의 경우를 고려하였다. 조사선량률은 균일 혼합체 모형을 사용하여 구하였다. 조사선량률 값은 매우 크게 나왔으며, 건식가공 중의 Cs제거율에 민감하게 변화하는 것으로 나타났다. 10년 이상 냉각된 PWR핵연료를 사용한 DUPIC핵연료의 경우 핵연료 내 모든 Cs를 제거하면 약 90% 이상의 조사선량률을 감소시킬 수 있다. 조사선량률에 주된 영향을 미치는 주요 방사선원은 Cs-137이다. Cs 제거에 관련된 연구는 DUPIC핵연료의 조사선량 뿐만 아니라, 건식 처리시설의 방사성 물질 관리에도 중요하다.

중심단어: 사용후 DUPIC핵연료, 선원분석, 조사선량률

INTRODUCTION

DUPIC (Direct Use of Spent PWR Fuel in CANDU) fuel cycle, which maximizes the use of valuable uranium resources and minimizes the release of radioactive wastes per electricity-production with less concerns about nuclear proliferation has been actively studied as a promising fuel cycle option in the Republic of Korea(1). DUPIC fuels contain lots of highly radioactive fission products(FPs), and this radioactivity may be one reason for utilities to hesitate in taking the resource-saving and waste-reducing DUPIC as a fuel option.

An elementary question would be how high the radioactivities of fresh and irradiated DUPIC fuels are DUPIC fuels are made through the Dry Process, where spent PWR fuel pellets become powder through repeated oxidation and reduction, then the powder is repelletized to be used in CANDU. Volatile radioactive nuclides are eliminated from the fuel during the process. These volatile nuclides not only affect the radioactivity of DUPIC fuels, but also complicate the treatment of wastes produced during the Dry Process. All these processes need more in-depth study of the radiological effect of the DUPIC fuel cycle. As a first step, we studied in this paper the radiation source intensity of the DUPIC fuel in terms of exposure rate at 1 m from the fuel bundle and its dependency on elimination of radioactive nuclides during the Dry Process.

Since the Dry Process used for the production of DUPIC fuel is under development, the definite source intensity cannot be estimated at this time. In this study, we set two cases: partial elimination of radioactive elements during the Dry Process and no elimination at all. Source intensity analyses of fresh and irradiated DUPIC fuels for these cases are performed, providing interesting results of which the meaning is discussed.

SOURCE TERMS OF THE DUPIC FUEL

Radioactivities in the spent PWR fuel

The main radioactive sources of DUPIC fuel are FPs generated during the burnup of uranium fuels in PWR. The amount of each radioactive nuclide in the spent PWR fuel was obtained by ORIGEN2(2). Two types of spent PWR fuels are considered based on the extent of burnup: standard (35000 MWD/MTU) and extended burnup (50000 MWD/MTU) fuels. The total radioactivities(Ci) per one metric ton of initial heavy metal(MTHM) are calculated with respect to the cooling time and shown in Figure 1. The radioactivity decreases rapidly up to about 10 years, then decreases slowly ($T_{1/2} \sim 30$ y).

Gamma radiation in fresh DUPIC fuels is the main radiation type of source determines the radiation field around them. The spectra of gamma radiation in standard and extended burnup spent PWR fuels were also calculated using both ORIGEN2 and MICROSIELD(3). Figure 2 shows three γ -ray spectrums: two from the decay library of ORIGEN2 (GXUO2BRM and GXNOBREM) and one from that of MICROSIELD. These spectra are based on 1 kg of 20-year cooled

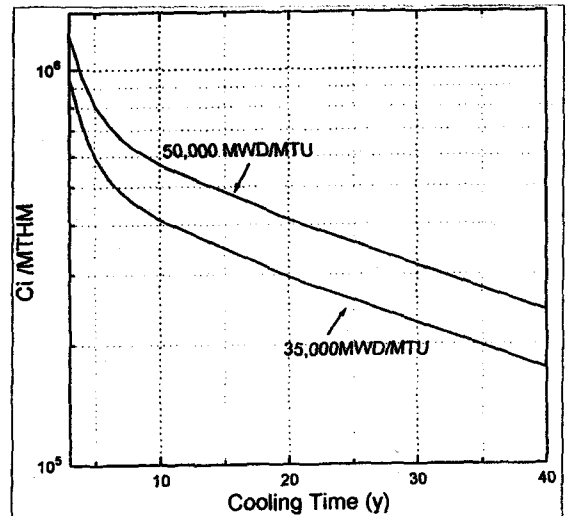


Fig. 1. Total radioactivity of spent PWR fuel.

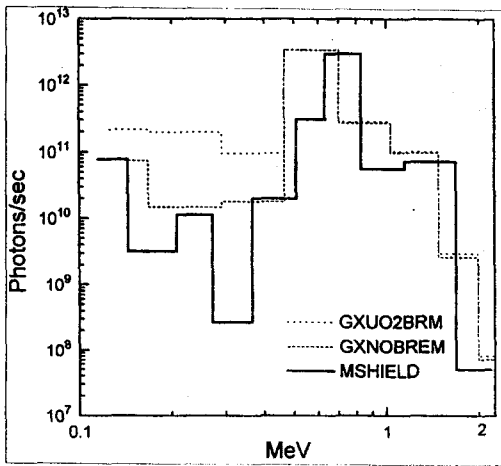


Fig. 2. The γ -ray spectrum of spent PWR fuel.

standard burnup PWR fuel. Gamma radiations below 2 MeV are dominant in intensity. In ORIGEN-2, γ -ray energies are divided into 18 fixed energy groups, while MICROSIELD sets the energy groups freely. In Fig. 2, only the important portion (0.1 MeV ~ 2 MeV) of the spectra is shown. All three spectra look similar in general and reasonably well represent the dominant source, Cs-137 (emit 0.622 MeV gamma via Ba-137m) in 20-year cooled spent fuels. The exposure calculation using the MICROSIELD program with self generated γ -ray spectrum has been checked in many benchmark problems(3) and is proved to be valid. Hence, we used the γ -spectrum from MICROSIELD for the calculation of the exposure rate from DUPIC fuels.

Table 1 shows important nuclides which contribute the main activity in each energy group. Cs-137, Sr-90 and Y-90 are β -ray sources, making low energy x-rays from Bremsstrahlung in the fuel materials. Y, Sr and Eu are soluble to fuel matrix(4) and not removable during the dry process.

Source intensity in Fresh DUPIC Fuel

Spent PWR fuels are decladded and the pellets become powder through repeated oxi-

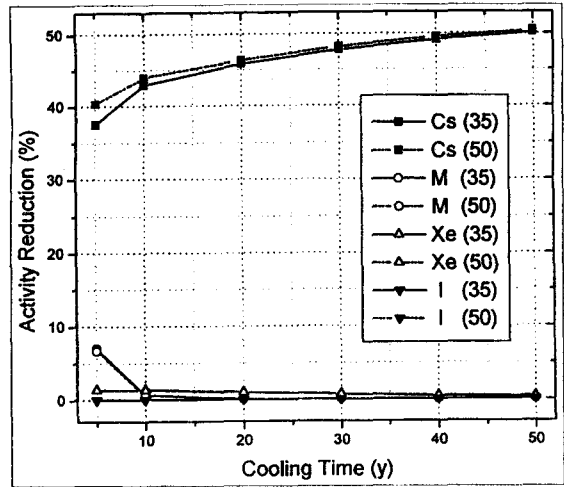


Fig. 3. The effects of the elimination of each group of elements on the radioactivity of fresh DUPIC fuel (35 = 35,000 MWD/MTU, 50 = 50,000 MWD/MTU).

dation and reduction. This powder is repelletized and then cladded to be used in CANDU. All these processes are called dry process (specifically, OREOX). While elements soluble to fuel matrix stay in the fuel during the dry process, volatile and noble elements are mostly eliminated. The reduction of total activity of fuel after the dry process is important from the viewpoint of exposure rate analysis and radioactive-waste production estimates.

We divided the elements of non-soluble FPs into 4 groups, inert gases (Xe), volatiles (I), volatile compounds (Cs), and metallics (M) depending on their chemical states in

Table 1. Important radioactive nuclei producing γ -radiation at each energy group (Bremsstrahlung is considered).

γ -ray energy (MeV)	Radioactive Nuclei*
0-0.2	Cs-137, Y-90, Sr-90, Eu-154
0.2-1.0	Ba-137m, Eu-154, Cs-134
1.0 or higher	Eu-154, Co-60, Tl-208

The order of radioactive nuclei follows approximately the amount of contribution. (* after 20 year cooling)

Table 2. Elemental grouping of FPs not soluble to fuel matrix.

Group Name	Elements
Xe	He, Ne, Ar, Kr, Xe, Rn
I	I, Br
Cs	Rb, Cs
M	Sb, Te, Cd, Sn, Ag, In Tc, Ru, Rh, Rd, Mo

the fuel. Table 2 shows the name and the elements belong to each group. The effect of the elimination of each group of elements on the radioactivity of fresh DUPIC fuel was also analyzed based on the grouping given in Table 2 (Fig. 3).

The values in Figure 3 indicate the degree of reduction in fuel activity after full elimination of each group of elements. The most activity-reducing group is Cs in which Cs-137 ($T_{1/2} = 30.17y$) is the dominant radioactive nuclide. Cs-137 is in a secular equilibrium with Ba-137m (daughter, $T_{1/2} = 2.252 m$). Total removal of Cs group reduces the fuel activity up to 50%. The rest of activity comes mainly from Sr-90, its daughter Y-90, and Pu-241.

The composition of fresh DUPIC fuel after the dry process is not exactly known, so far. Recently, Sullivan has analyzed the composition change of spent fuel after the dry process(10). His analysis indicates that the final composition is strongly dependent on the temperature during the dry process. Since the detail method of the dry process is not fully developed yet, we consider two extreme cases. One is the DUPIC fuel directly made from spent PWR fuel without

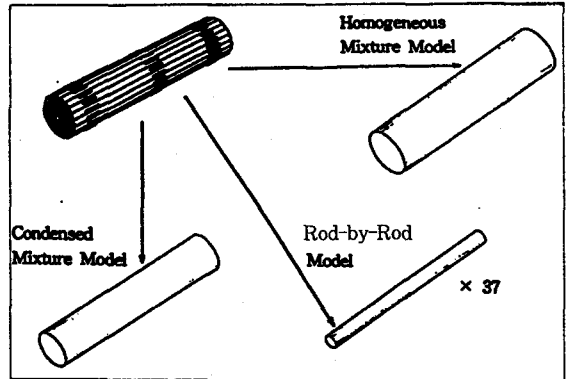


Fig. 4. The modelling of a fuel bundle assembly.

dry process (called 'direct DUPIC'). The other is the fuel made from the high temperature dry process (called 'processed DUPIC'). Based on the Sullivan's results and the vapor pressures of important elements and their compounds(5,11), the following elemental elimination model is set for the processed DUPIC fuel (table 3).

Source intensity in spent DUPIC fuel

The exposure-rate analysis of spent DUPIC fuels is also important from the viewpoint of the methodology development of spent DUPIC fuel management. Fresh DUPIC fuel from irradiated PWR fuels (35,000 MWD/MTU) contains about 0.8% U-235 and 0.6% Pu-239 on the average. The library, CANDUSEU in ORIGEN-2 is made for the calculation of nuclide composition in the slightly enriched uranium fuel in CANDU(9). Since the neutronic characteristics of DUPIC fuel during irradiation seem to be close to those of slightly enriched uranium fuel, we used CANDUSEU for the calculation of the elemental distribution of FPs in spent DUPIC fuels. The burnup of

Table 3. The assumed elemental elimination during Dry Process.

Elements				High TM Metal				Low TM Metal				
	Xe, Kr	I, Br	Cs, Rb	Ru	Rh	Mo	Tc	Te	Ag	Cd	In	Sb
Elimination(%)	100	100	98	100	100	50	50	80	80	100	80	80

(TM : Melting Temperature)

DUPIC fuel is set as 7,500 MWD/MTU.

EXPOSURE RATE ANALYSIS

Calculation model.

A CANDU fuel bundle consists of 37 fuel rods (see figure 4). Because of the complicated geometry of the assembly, modeling of the simplified geometry is necessary for the exposure rate calculation. DUPIC fuel is mainly composed of UO_2 and Zircaloy. UO_2 is an effective γ -radiation shielding material. So, the modeling of UO_2 portions in the assembly is crucial for the reasonable calculation.

The fuel bundle is assumed to be a homogeneous mixture of UO_2 , Zircaloy and void (i. e., empty spaces). The outer radius of the homogeneous mixture is set as that of the bundle. The average density of the mixture is calculated by considering the volume fraction of each material (see table 4). Another two geometry models for the fuel bundle were also considered: rod-by-rod and condensed models. Effectiveness of the homogeneous model can be checked by a high limit from the rod-by-rod model, where the exposure rate is simply obtained by multiplying the exposure rate of single fuel rod by the number of rods in a fuel bundle, and a low limit from the condensed mixture model, where a fuel assembly is assumed as a condensed mixture neglecting the empty space (see Fig. 4).

Results and Discussion

Exposure rate calculation is performed

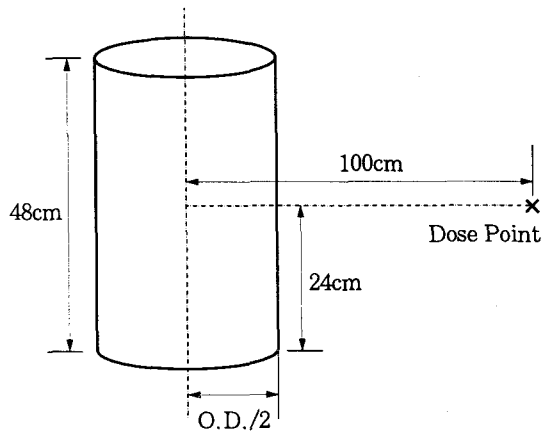


Fig. 5. The geometry setup for the exposure rate calculation.

using MICROSHIELD(3). MICROSHIELD is a code which is originated from ISOSHLD (7). MICROSHIELD conforms to ANSI 6.6.1 and is validated to be used in the documentation in accordance with 10 CFR 50 Appendix B. Several bench mark problems from ANSI/ANS-6.6.1 and ESIS were tested, and the results from MICROSHIELD were confirmed to be reasonably good(3). So, Microshield is accepted as a convincing tool for the calculation of exposure in this study.

Exposure rate in this study is set as the value of exposure rate at the position that is 1m apart from the center of DUPIC fuel (Fig. 5). Reflection or buildup of γ -radiation is only considered in the sample and surrounding air. Taylor buildup factor (3) is employed for the exposure rate calculation. The output of ORIGEN-2 is modified and

Table 4. The geometry of each model.

	CANDU Assembly	Homogeneous Mixture Model	Condensed Mixture Model	Multiplication Model
O.D. (cm)	10.24	10.24	7.90	1.22(one rod)
Density (g/cm ³)	UO ₂ : 10.5 Zry : 6.46	6.0	10.0	UO ₂ : 10.5 Zry : 6.46
Dose	--	Reasonable	Low Limit	High Limit

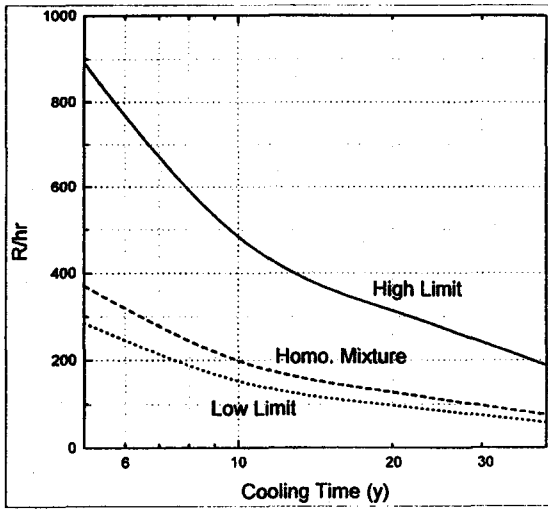


Fig. 6. The exposure rate of fresh direct DUPIC fuel made from 35,000 MWD/MTU PWR fuels

used as an input data for Microshield. First, the calculation is performed for fresh direct DUPIC fuels. Figure 7 shows the calculation results for direct DUPIC fuel from 35000 MWD/MTU spent PWR fuels

As shown in Fig. 6, the exposure rate of the homogeneous mixture is much lower than the high limit set by the rod-by-rod model, and higher than the low limit from the condensed mixture model. The exposure rate of a direct DUPIC fuel is very high (about 200 R/h in the case from 10 year cooling PWR fuels). The exposure rate decreases almost exponentially with cooling time after 10 years.

The contribution of each γ -ray source on the total exposure rate is also analyzed. In the calculation logic used in MICROSHIELD, each contribution of a γ -ray is additive, and the total sum becomes the exposure rate. Figure 7 shows the contributions of strong γ -ray sources on the exposure rate of fresh direct DUPIC fuels with respect to cooling time of spent PWR fuels. More than 95% of exposure rate is due to four nuclides - Ba-137m, Cs-134, Eu-154, and Co-60 - in more than 5 year cooled fuel. Elimination of Cs group reduces the exposure rate from 87%

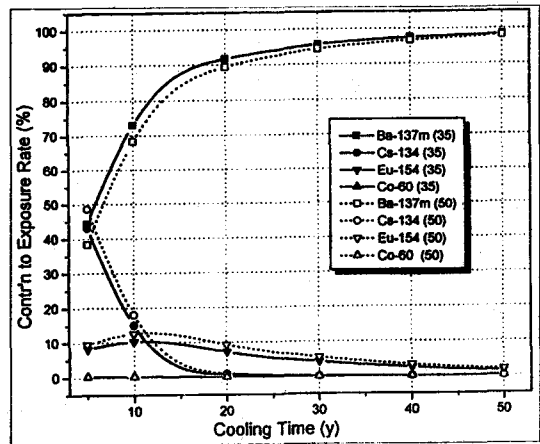


Fig. 7. Contribution of each strong γ -ray source to exposure rate of fresh direct DUPIC fuel.

(5 year cooled) to 98% (50 year cooled).

The exposure rates of processed fresh DUPIC fuels from 35000 MWD/MTU and 50,000 MWD/MTU burnup PWR fuels are also shown in Figure 8. The big difference of the exposure rates between processed and direct DUPIC fuels is mainly due to the elimination of Cs-137 during the dry process (see Table 3). The difference becomes bigger with the cooling time of spent PWR fuels because of increased contribution of Cs-137 on the exposure. Cs is known to be easily captured by the cold trap in the laboratory scale. However, the upscale of the Cs trap to the industrial use is not fully developed, and more intense study about the trap is needed for the prevention of radiation exposure to the workers in the dry process facility.

Exposure rates of spent DUPIC fuels - direct and processed from 20 year cooled PWR fuels - are calculated and compared with that of ordinary spent CANDU fuel. (fig. 9) Discharge burnup of CANDU fuel is set as 7500 MWD/MTU. The exposure rates of all three fuel types are close to each other when they are initially taken out from CANDU reactor after irradiation. Differences in the exposure rate look clear, as the cooling time of spent fuel increases. Direct DUPIC fuel has the highest exposure

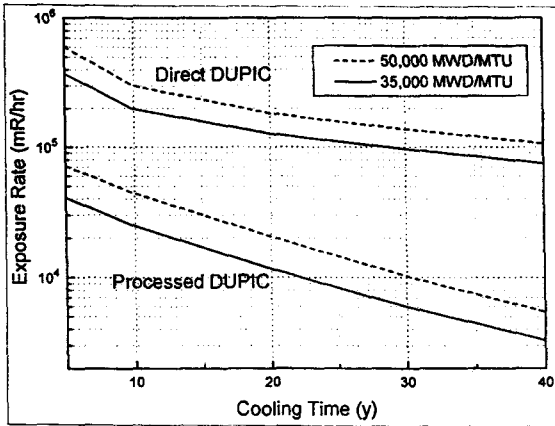


Fig. 8 Exposure rates of direct and processed DUPIC fuels with respect to spent PWR fuel cooling time.

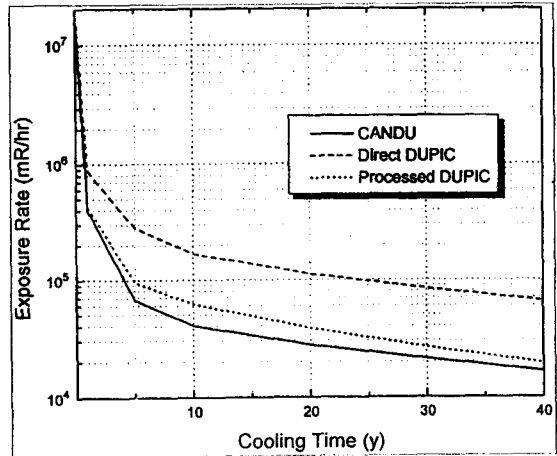


Fig. 9. Exposure rates of spent DUPIC (direct and processed) and spent CANDU fuels. Burnup is 7,500 MWD/MTU.

rate, while that of processed DUPIC fuel is quite reduced. But, the exposure rates of both direct and processed DUPIC fuels are higher than that of CANDU fuels. Due to the increased γ -ray intensity and decay heat of DUPIC fuels, spent DUPIC fuels may need longer wet-storage period before disposal to dry storage facilities, which needs further detail analysis.

CONCLUSION

Exposure rate analyses of the fresh and spent DUPIC fuels made from standard and extended burnup PWR fuels were performed using MICROSIELD. A bundle consists of 37 fuel rods was assumed as a homogeneous mixture for the calculation, and the exposure rate is set as the value at a point which is 1m apart from the DUPIC fuel bundle.

The exposure rate of DUPIC fuels strongly depends on the amount of Cs in the fuel. The main γ -radiation source is Cs-137. The exposure rate turned out to be quite high - e.g., the fresh direct DUPIC fuel made from 10-year-cooled standard-burnup PWR fuels gives the exposure rate of 200 R/hr.

The exposure rate decreases with the cooling time of spent PWR fuels and also with the amount of Cs elimination during the Dry Process. The exposure rate of spent DUPIC fuels is higher than that of the ordinary CANDU fuel, and also dependent on the amount of Cs elimination during the Dry Process. There is a possibility of longer period of wet storage of spent DUPIC fuels before dry storage. Further analysis of collected Cs in the Dry Process facilities is also required for the prevention of radiation exposure to the workers in the facility.

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