Paper

RELATIVE RADIOLOGICAL IMPACT FROM A REACTOR ACCIDENT IN THE CASE OF EMERGING NUCLEAR FUELS

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Fn1 *Abstract*—**An assessment has been carried out on the radiological impact on an area contaminated from an accident of a nuclear reactor loaded with different actinide fuels considered in transmutation and recycling schemes. The impact of these schemes is compared to reference cases of commercial UO_2 and MOX fuels. The effective dose equivalent delivered to permanent residents has been calculated using the RESRAD code and used as an index for the assessment purposes. The highest and lowest doses would be delivered from the self-generating recycling of actinides in fast and thermal reactors, respectively. External irradiation is the main contributor to the dose delivered to the target population in comparison to ingestion and inhalation. The external dose delivered would be attributed for the first few years to ^{134}Cs and for the following several tens of years to ^{137}Cs.**

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Key words: dose assessment; environmental impact; nuclear fuel cycle; radiation dose

INTRODUCTION

THE ISSUE of Partitioning and Transmutation (P & T) as a nuclear waste management option has brought forward several processes, relying on existing nuclear reactors, in order to reduce the radiotoxicity of the waste (Koch 1986; Tommasi et al. 1995; Walters et al. 2002; Nishihara et al. 2008). In this concept, the recycling of minor actinides and plutonium, either in a "once-through cycle" or in a "self-generated mode" has been proposed as emerging fuel cycles in P & T (Koch and Nicolaou 1993; Nicolaou and Tsagas 2006; Warin 2007). This has led to the proposal and development of possible candidate fuels and targets, with appropriate fresh compositions. These fuels should be able to withstand the extreme conditions of a transmutation process, rendering the processes safe from the viewpoint of fuel behavior and hence reactor safety (Salvatores 2002).

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In this simulation study, different fuels corresponding to the commercial and emerging cycles are compared with respect to the radiological impact on a population due to the fallout from a reactor accident fuelled with them. The comparison is based on the fission products ^{90}Sr, ^{103}Ru, ^{106}Ru, ^{131}I, ^{132}Te, ^{140}Ba, ^{134}Cs, and ^{137}Cs deposited on the ground and responsible for the external gamma irradiation and ingestion doses to the population (UNSCEAR 2000). These radionuclides can be readily monitored, depending on the time that has elapsed from their release to deposition, through mass and gamma spectroscopy, with the latter been carried out either in-situ or in the laboratory on soil samples collected from the affected area (Roca et al. 1989).

MATERIALS AND METHODS

Nuclear fuels considered

Emerging and commercial nuclear fuels in existing nuclear power stations were considered. The former include fuels in thermal (Pressurized Water Reactor, PWR) and fast (Liquid Metal Fast Breeder Reactor, LMFBR) nuclear power stations which, as a nuclear waste management option, have been considered for the transmutation (cases *T1, T2*) and self-generated recycling of actinides (*S1, S2, S3*). Existing commercial fuel cycles were considered in the same reactor types as reference cases for comparison purposes (cases *R1, R2, R3*):

- *PWR "once-through" UO_2 fuel cycle* (*reference case R1*). A PWR reactor with 1,150 MWe energy output and fuelled with 98 t of UO_2, i.e., 89 t of heavy metal (HM);
- *PWR MOX fuel cycle* (*reference case R2*). A PWR reactor fueled with $(UPu)O_2$ for the recycling of first generation plutonium in the same reactor and under the same operating conditions as the reference case *R1*. The isotopic vector of the plutonium for 238:239:240:241:242 was 1.4:55:25.3:13.3:5;
- *LMFBR MOX fuel cycle* (*reference case R3*). A fast reactor with 1,200 MWe energy output and fuelled with 32 t of $(UPu)O_2$, i.e., 28 t HM;
- *Transmutation in fast reactors*: regarding the transmutation of ^{237}Np and ^{241}Am in a fast reactor, fresh fuels

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with low concentrations of these nuclides have been considered, resembling the homogeneous recycling in the same reactor and under the same operating conditions as in the reference case *R3*. The nuclides of interest ^{237}Np and ^{241}Am are homogeneously mixed in a fast reactor fresh fuel (cases *T1* & *T2*);

- *PWR self-generated Pu recycle* (*S1*). Self-generated plutonium is recycled for 5 cycles in the same reactor and under the same operating conditions as in the reference case *R1*. The composition of the spent fuel refers to the "equilibrium" after 5 cycles and it is averaged over the total core;
- *PWR self-generated transuranium (TU) recycle* (*S2*). All self-generated transuranium nuclides are recycled for 5 cycles in the same reactor and under the same operating conditions as in the reference case *R1*. The composition of the spent fuel refers to the "equilibrium" after the completion of the 5 cycles and it is averaged over the total core; and
- *LMFBR self-generated transuranium (TU) recycle* (*S3*). All self-generated transuranium nuclides are recycled for 16 cycles in the same reactor and under the same operating conditions as in the reference case *R3*. The composition of the spent fuel refers to the "equilibrium" after the completion of the 16 cycles and it is averaged over the total core.

The fresh fuel compositions for the different fuel cases considered are summarized in Table 1. The loading of the core in each case and the burnup at end of irradiation (EOI) of the fuels are given in Table 2. It should be noted that the fresh fuel compositions for the three recycling cases are the ones of the 5th (cases *F6*, *F7*) and 16th (case *F8*) fuel cycles. These cycles in the recycling process correspond to the period necessary to achieve "equilibrium" in the inventory of the recycled nuclides (Koch and Nicolaou 1993). At this stage, the amounts of nuclides being formed and transmuted during irradiation are the same. The plutonium and minor actinides used in the recycling and transmutation schemes have been used 6 months after being retrieved from reprocessing which was carried out on spent fuel with a 1.5 y cooling time. The required radiation dose comparisons have been performed on the basis of a full loaded core for each of the fuel-reactor combinations, assuming that the accident has occurred with the fuel having attained its full burnup at the EOI (Table 2). Although current commercial burnup values have been considered in this study, values of up to 60 and 120 GWd/t for PWR and LMFBR fuel cycles, respectively, are envisaged in the future. Hence, absolute values of doses would increase, due to increase fission product content in the fuel and the source term. Nevertheless, the trend of the relative radiological impact from the different fuel cycles would remain the same.

Computational methodology

The relative radiological risk associated with the different fuel cycles has been assessed using the RESRAD (RESidual RADioactivity) computer code (Yu et al. 2001). The code is designed for dose and risk assessment based on given concentrations of radioactive materials in the environment. RESRAD includes the exposure pathways of inhalation, ingestion of soil and food products, and external exposure from the soil (Azlina et al. 2003).

Depletion calculations, required to simulate the reactor core inventory in actinides and fission products at the moment of the accident and hence the release of radioactivity to the atmosphere, were performed using the zero-dimensional isotope generation and depletion code ORIGEN-2 (Croff 1980). The calculations were carried out on the basis of 1 ton of the fresh fuel compositions and target burnup values given in Tables 1 and 2, respectively. In each fuel cycle, the total core inventory, at the moment of the accident, is the total composition of the spent fuel in the core with the burnup given in Table 2.

Scenario considered

It is assumed that the radioactivity released to the environment has occurred due to the core melt because of

Table 1. Fresh compositions of the different fuels considered.

Fuel case	Fuel type	Fuel charge composition*	Fissile content (wt%)
F1	UO2	UO2	3.5 (U)
F2	MOX	U0.96Pu 0.04	3 (Pu)
F3	MOX	U0.8Pu0.2	17.5 (Pu)
F4	MOX with 1%Np	U0.795Pu0.195Np0.01	17.2 (Pu)
F5	MOX with 1%Am	U0.795Pu0.195Am0.01	17.2 (Pu)
F6	Self-generated Pu recycle	U0.96Pu0.04	2 (Pu)
F7	Self-generated TU recycle	U0.95Pu0.04Am0.003Cm0.002Np0.005	1.8 (Pu)
F8	Self-generated TU recycle	U0.8Pu0.186Am0.007Cm0.006Np0.001	11.2 (Pu)

* Case F1: U enriched with 3.5% ^{235}U; cases F2–F8: depleted U (0.2% ^{235}U); cases F2–F5: 1st generation plutonium from PWR (1 y cooling, reprocessing, 3 y cooling); cases F6–F8: 'equilibrium' composition.

Table 2. Core loading of the fuel cycles considered.

Core case	Reactor	Fuel cycle	Core loading*	Burnup (GWd/t)
R1	PWR	U	100% F1	40
R2	PWR	MOX	70% F1 + 30% F2	40
R3	LMFBR	MOX	100% F3	100
T1	LMFBR	MOX with 1%Am	100% F4	100
T2	LMFBR	MOX with 1%Np	100% F5	100
S1	PWR	Self-generated Pu recycle	100% F6	40
S2	PWR	Self-generated TU recycle	100% F7	40
S3	LMFBR	Self-generated TU recycle	100% F8	100

* 100t HM for PWR, 30t HM for LMFBR.

AQ: A a LOCA and failure of the reactor containment in the PWR and LMFBR. The release fractions, as a result of cladding failure and core melt, of the radionuclides of interest are (Soffer et al. 1995): 2% for ^{90}Sr and ^{140}Ba, 0.25% for ^{103}Ru and ^{106}Ru, 40% for ^{131}I, 5% for ^{132}Te, and 40% for ^{134}Cs and ^{137}Cs. It is assumed that the extent of the accident in the different reactors considered would be the same, in order to assess their relative radiological impact. Hence, the same release fractions are employed in the computations for each reactor type and fuel load. The released radioactivity would eventually be deposited on the ground according to the meteorological conditions. In this study, it is assumed that the fraction of each of the released radionuclides that would eventually be deposited and contribute to the dose would be that of the radioactivity from the Chernobyl accident deposited in the far zone from the reactor (~100 km distance) (UNSCEAR 2000).

The case studied assumed a 100,000 m^2 area of a ground, at a distance of ~100 km from the reactor of interest, with 5 mm of its surface contaminated within 1 d after the accident of a reactor loaded with fuel from the recycling and transmutation of actinides. To be conservative, a resident farmer scenario was used with the permanent residents of a house located in the middle of the contaminated zone being the critical population group, raising crops and livestock for their diet. The target group can incur a radiation dose, over a 30-y period considered, by direct irradiation from radionuclides in the soil (external dose), inhalation of resuspended dust, radon, and decay products, and ingestion of soil, food, and water. The average fractions of time in a year, during which an individual from the target group stays indoors and outdoors on the contaminated T3 site, are 0.50 and 0.25, respectively (Table 3). Hence, an individual would spend on the site an average fraction of 0.75 over the period of 1 y, which corresponds to 273.75 d.

All calculations were based on the default values in RESRAD, except in the case of the contamination area and depth, and the erosion rate. The default values of key parameters used in the dose calculations using RESRAD are given in Table 3 (Yu et al. 1993). A contamination area of 100,000 m^2 was used as being a more realistic area contaminated following an accident, as it was the case of the far zone from Chernobyl considered. A 5 mm contamination depth was considered as a reasonable value for the fresh-fallout situation considered (Almgren and Isaksson 2006). An erosion rate of 0.0001 m y^{-1} was used, assuming a site with a 2% slope, in order to take into consideration the dependence of the 5 mm thickness on the erosion and hence allow the dose computations up to 30 y (Yu et al. 2001).

RESULTS AND DISCUSSION

The effective dose equivalent to the target population, over a period of 30 y following the ground contamination from the different core cases considered in Table 2, is shown in Fig. 1. The dose is due to all the F1 radionuclides and pathways considered. The cases are clustered together in two distinct well-separated families reflecting the two reactor types under consideration, namely the PWR (lower set of curves) and the LMFBR (upper set of curves). The clustering of the cases reflects the formation of the fission products during the irradiation of the fuel in the reactor. The formation of each

Table 3. Key parameters considered in different scenarios of exposed population (Yu et al. 2001).

Parameters	Value
Soil density (gr cm^{-3})	1.5
Hydraulic conductivity (m y^{-1})	
Contaminated zone	10
Unsaturated zone	100
Saturated zone	100
Total porosity	0.4
Effective porosity	0.2
Soil b parameter	5.3
Hydraulic gradient	0.02
Unsaturated zone thickness (m)	4
Inhalation rate (m^3 y^{-1})	8,400
Fraction of time indoors/outdoors	0.5/0.25
External gamma shielding factor	0.7

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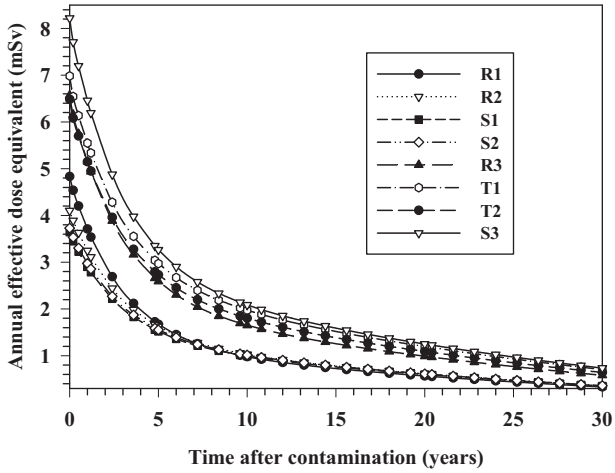

Fig. 1. Effective dose equivalent for the different cores on the basis of all nuclides and pathways considered.

fission product is a function of the neutron flux, the length of irradiation, the fission cross-section of the fissile radionuclides present, the fissile content in the fresh fuel and the fission yield for the fission product of interest.

Over the first year, an annual effective dose equivalent between 6.6–8.2 mSv would be delivered to the target population upon deposition of the fallout from the fast reactor fuel schemes (*R3*, *T1, T2, S3*). Within this family of cases, the highest dose would be delivered in the case of the fast reactor fuelled with all the self-generated transuranium nuclides for their recycling (case *S3*). This case would deliver a dose that would be higher by about 20% over the cases *R3, T1, T2* throughout the 30-y period of interest. The dose would reach the annual limit for the public of 1 mSv within 20 and 25 y after the contamination, for the cases (*R3, T1, T2*) and *S3,* respectively.

In the first year, the annual effective dose equivalent, delivered to the target population upon deposition of the fallout from the thermal reactor fuel schemes (*R1, R2*, *S1, S2*), would be between 3.6–4.9 mSv. Doses would diminish after that as shown in Fig. 1. Within the thermal reactor cluster, emerging fuel cases *S1* and *S2* are grouped together according to their similar plutonium isotopic composition in the fresh type loaded to the reactor. These recycling schemes present, up to 5 y after contamination, the lowest effective dose delivered within the cluster, while the highest corresponds to the reactor fuelled with UO_2 (*R1*). Beyond this time, all four cases would deliver the same dose reaching the limit of 1 mSv within 10 y after the contamination.

Hence, the comparison of all eight cases considered shows that, over the 30-y period after the contamination of the ground, the highest dose would be delivered by the fast reactor fuelled for the recycling of all the self-generated transuranium nuclides (*S3*). In contrast, the lowest would be delivered by the thermal reactor fuelled with all self-generated transuranium nuclides (*S1*). The dose would differ by a factor of almost 2 between the highest and lowest dose cases throughout the period of 30 y. In the case of *S3*, the effective dose equivalent, over the first year, 10 and 30 y after the contamination, would be 8.2, 35, and 50 mSv, respectively. In the case of the Chernobyl accident, the corresponding doses in the areas with a similar ^{137}Cs contamination level were lower by a factor of 2 (UNSCEAR 2000).

Case *S3* was further investigated in terms of the contribution of the different pathways and individual radionuclides to the dose delivered. The effective dose equivalent, delivered to the target population due to the pathways of food ingestion, inhalation, and external exposure to the radionuclides in the contaminated ground, are shown in Fig. 2. Throughout the 30-y period F2 after the contamination, 97.5% of the dose is due to the external irradiation of the population, while 2.5% is due to the ingestion pathway.

The influence of the key assumptions, considered in the calculations, on the dose output was examined for the parameters in Table 3. In accordance with the fact that the external component is the dominant contributor to the dose delivered (Fig. 2), the average time spent by an individual on the contaminated site and the erosion rate related to the site are the assumptions that influence the dose output. The contribution to the dose, from the indoor and outdoor time fractions considered in the study, is 56% to 44%, respectively. Therefore, in the case S3, where the highest annual dose of 8.2 mSv is given to the population, the indoor and outdoor time fractions would contribute 4.6 and 3.6 mSv, respectively. In the case

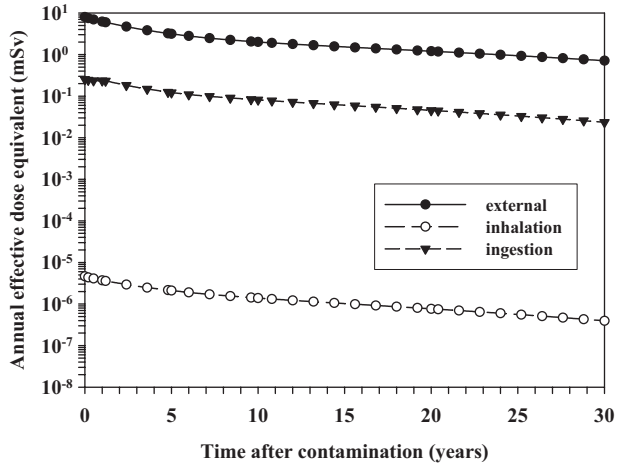

Fig. 2. Effective dose equivalent for the different pathways considered.

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when the average fraction of time indoors or outdoors is 1 (100%), the dose to the population would be 9.2 and 14.4 mSv, respectively. An erosion rate of 0.001 m y^{-1}, assuming a site with a slope greater than 2% (Yu et al. 2001), would decrease the dose below 1 mSv within 8 y after the contamination.

The relative contribution to the external dose delivered by the radionuclides of interest is shown in Fig. 3 for periods of up to 1 and 30 y following the contamination, respectively. Throughout the period of interest, 98% of the dose delivered is due to the ^{134}Cs ($\tau_{1/2}$ = 2 y) and ^{137}Cs ($\tau_{1/2}$ = 30 y), with the former being the main contributor over the first 3 y and the latter for the rest of the period. The other radionuclides of interest contribute the remaining 2%,—the short-lived ^{131}I ($\tau_{1/2}$ = 8 d), ^{132}Te ($\tau_{1/2}$ = 3 d), and ^{140}Ba ($\tau_{1/2}$ = 12 d) during the first month after contamination of the ground, and the ^{103}Ru ($\tau_{1/2}$ = 30 d) and ^{106}Ru ($\tau_{1/2}$ = 374 d) over the first few years (Fig. 3). ^{90}Sr, although long-lived in relation to the 30-y period ($\tau_{1/2}$ = 29 y), contributes less than 0.1% to the dose delivered. Thus, external gamma exposure would remain elevated for the first few years due to ^{134}Cs and for the following several tens of years due to ^{137}Cs.

It is seen in Fig. 3 that, although ^{137}Cs and ^{90}Sr have similar physical half-lives, over the 30-y period only the former is reduced by a factor of 2 according to its radioactive decay while the latter is reduced by three orders of magnitude. This is due to the different distribution coefficients K_d for the two radionuclides. The default values in the code of 1,000 and 30 cm^3 g^{-1} were employed for ^{137}Cs and ^{90}Sr, respectively (Yu et al. 2001). Hence, ^{90}Sr would be dispersed much faster than ^{137}Cs resulting in the response shown in Fig. 3.

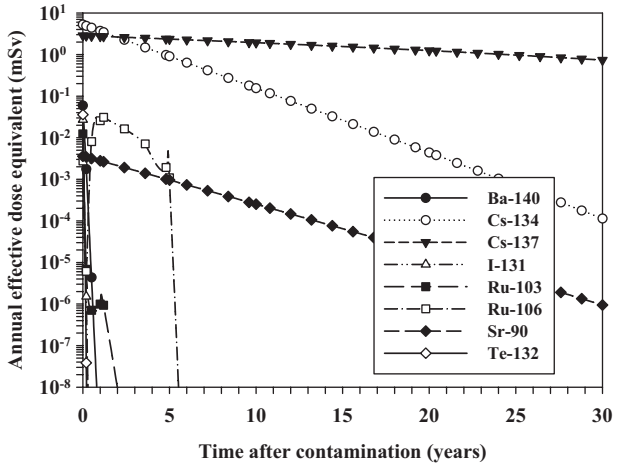

Fig. 3. Effective dose equivalent for the different radionuclides considered over the period of 30 y following the contamination.

CONCLUSION

The relative radiological impact of an area contaminated from an accident in a nuclear reactor that is loaded with emerging fuels has been assessed. Commercial thermal and fast reactors were considered for comparison purposes, while the emerging fuels represented those envisaged for the transmutation and recycling of plutonium and minor actinides in these reactors. The effective dose equivalent delivered to permanent residents has been used as the index for the assessment purposes.

The highest dose would be delivered in the case of the self-generated transuranium recycle in a LMFBR (*case S3*). The dose delivered would be higher by a factor of 1.3 and 1.9 than the dose due to the commercial fast (*R3*) and thermal (*R1*) reactor cases, respectively. The lowest dose would be delivered in the case of the self-generated plutonium recycle in PWR (*case S1*). The dose would be lower than the *R3* and *R1* cases by a factor of 1.8 and 1.4, respectively. The external irradiation contributes 98% to the dose delivered, in comparison to the pathways of ingestion and inhalation. The external gamma exposure would be attributed for the first few years to ^{134}Cs and for the rest of the 30-y period to ^{137}Cs.

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AUTHOR QUERIES

AUTHOR PLEASE ANSWER ALL QUERIES **1**

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