Annals of Nuclear Energy 37 (2010) 285–288

Contents lists available at ScienceDirect

Annals of Nuclear Energy

journal homepage: www.elsevier.com/locate/anucene

Technical Note

Criticality aspects of nuclear power reactor cores in the case of emerging nuclear fuels

G. Nicolaou *

Demokritus University of Thrace, School of Engineering, Department of Electrical and Computer Engineering, Laboratory of Nuclear Technology, Kimeria Campus, 67100 Xanthi, Greece

ARTICLE INFO

Article history:
Received 3 August 2009
Received in revised form 26 October 2009
Accepted 30 October 2009
Available online 25 November 2009

ABSTRACT

Reactor cores of PWR and LMFBR, loaded with different commercial and emerging nuclear fuels, have been simulated and compared at BOI with respect to criticality with and without chemical shim, control rods and sodium. The different cases considered, within each of the reactor types, are grouped together according to their fissile content, when compared on the basis of the neutron multiplication factor (k_{eff}). For both PWR and LMFBR reactor types, the reactivity worths of the control rods do not change significantly when replacing commercial fuels by emerging ones. In the case of the LMFBR, the Na void reactivity effects are small and comparable using either emerging or commercial fuels. Hence, operation and control of the core at beginning of irradiation are similar for emerging or commercial fuels.

1. Introduction

Nuclear waste, in the form of spent nuclear fuel or high level waste from the reprocessing of the fuel is associated with a radiotoxicity potential due to its actinide and fission product content. Partitioning and Transmutation (P and T), as a nuclear waste management option, has brought forward a number of processes, relying on existing nuclear power reactors, towards reducing the radiotoxicity of the waste (Koch, 1986; Tommasi et al., 1995; Mennerdahl and In de Betou, 2007; Nishihara et al., 2008).

Hence, recycling schemes of minor actinides and plutonium, either in a 'once-through cycle' or in a 'self-generated mode', have been proposed as emerging Partitioning and Transmutation fuel cycles (Koch et al., 1997; Salvatores, 2002; Nicolaou and Tsagas, 2006). Hence, possible candidate fuels and targets, based on an oxide, nitride, metallic or inert matrix, have been proposed to contain the actinides to be transmuted. These emerging fuels must withstand the extreme conditions of a transmutation process, rendering the processes safe from the viewpoint of fuel behaviour and safety. Furthermore, the use of emerging fuels, either in the homogeneous or heterogeneous recycling mode, should not prejudice the safe operation of the core.

In this study, simulated Pressurised Water Reactor (PWR) and Liquid Metal Fast Breeder Reactor (LMFBR) cores, loaded with either emerging or commercial oxide fuels, are compared. The aim of the study is to obtain information on the behaviour of cores loaded with emerging fuels in the homogeneous recycling mode, in terms of criticality and control. Hence, the deviation of these cores from the commercial PWR and LMFBR ones could be assessed. The comparisons have been performed with respect to the multiplication factor (k_{eff}) and reactivity worth ($\%\Delta k/k$) in the case of control rod and shim use, as well as sodium void. The simulations have been carried out for fuel compositions at the beginning of irradiation (BOI).

2. Materials and methods

The required criticality analysis was based on the determination of the neutron multiplication factor (k_{eff}) and was carried out for a range of nuclear fuels from commercial and transmutation fuel cycles on the basis of their composition at BOI.

2.1. Nuclear fuels considered

Emerging and commercial nuclear fuels in existing nuclear power stations have been considered. The former include fuels in PWR and LMFBR nuclear reactors which, as a nuclear waste management option, have been considered for the transmutation and self-generated recycling of actinides. Existing commercial fuels were considered in the same reactor types for the required comparison. The fresh compositions for the different fuels considered in this study are summarized in Table 1.

- *Commercial fuels:* UO_2 in PWR (case *R1*); MOX $(UPu)O_2$ in PWR (case *R2*); MOX $(UPu)O_2$ in LMFBR (case *R3*). First generation plutonium from the PWR-UO_2 case *R1*, with an isotopic vector for 238/239/240/241/242 of 3.42/48.58/24.06/14.19/7.98, was used in the PWR- and LMFBR-MOX fuels (cases *R2* and *R3*).

* Tel.: +30 2541079736.
E-mail address: nicolaou@ee.duth.gr

0306-4549/$ - see front matter © 2009 Elsevier Ltd. All rights reserved.
doi:10.1016/j.anucene.2009.10.018

Table 1
Fresh compositions of the different fuels considered.

Fuel case	Fuel type	Fuel charge composition*	BOI fissile content (wt.%)
R1	UO_2	UO_2	3.5, 2.8 and 2.25 (U)
R2	MOX	$U_{0.96}Pu_{0.04}$	2.5 (Pu)
R3	MOX	$U_{0.80}Pu_{0.20}$	12.55 (Pu)
T1	MOX with 1%Np	$U_{0.795}Pu_{0.195}Np_{0.01}$	12.24 (Pu)
T2	MOX with 1%Am	$U_{0.795}Pu_{0.195}Am_{0.01}$	12.24 (Pu)
S1	Self-generated Pu recycle	$U_{0.96}Pu_{0.04}$	2 (Pu)
S2	Self-generated TU recycle	$U_{0.95}Pu_{0.04}Am_{0.003}Cm_{0.002}Np_{0.005}$	1.8 (Pu)
S3	Self-generated TU recycle	$U_{0.8}Pu_{0.186}Am_{0.0067}Cm_{0.0058}Np_{0.0015}$	11.2 (Pu)

* Fuel case *R1* with a 3.5% enrichment in ^{235}U; all other cases with depleted U (0.2% ^{235}U); cases *R2*, *R3*, *T1*, *T2* with 1st generation plutonium from PWR; cases *S1*, *S2*, *S3* with 'equilibrium' composition.

- *Transmutation fuels:* fresh fuels with low concentrations of either ^{237}Np or ^{241}Am have been considered, regarding their homogeneous transmutation in the same reactor and under the same operating conditions as in the commercial LMFBR-MOX case *R3*. The nuclides of interest ^{237}Np or ^{241}Am are homogeneously mixed in the LMFBR-MOX fuel (cases *T1* and *T2,* respectively).
- Fuels with plutonium or transuranium (TRU) from their self-generated recycling in a PWR (*S1* and *S2*).
- Fuel with transuranium (TRU) from their self-generated recycling (*S3*) in an LMFBR.

In the three self-generated recycling schemes, the Pu and TRU composition of the fresh fuels are either the ones after the 5th recycling step in the cases *S1* and *S2*, or the 16th recycling step in the case *S3*. These steps correspond to the periods necessary in order to achieve 'equilibrium' in the content 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 'equilibrium' charge composition of the fuels, given in Table 1, has been averaged over the total core. 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 criticality comparisons have been carried out at BOI on the basis of the reactors loaded with the fuels given in Table 1. The characteristics of the reactor cores (*CR1,CR2,CR3,CT1,CT2,CS1,CS2,CS3*) are listed in Table 2. It should be noted that the PWR-UO_2 core (*CR1*) at BOI consists of zones with 2.25%, 2.8% and 3.5% enriched ^{235}U fuel assemblies. In the case of the PWR-MOX (*CR2*), the UO_2 fuel is that with an enrichment of 3.5% ^{235}U (*R1*).

2.2. Computational methodology

Depletion calculations, required to simulate the composition of the first generation Pu, as well as the 'equilibrium' plutonium and TRU after the 5th and 16th recycle steps, respectively, were performed using the zero-Dimensional isotope generation and depletion code ORIGEN-2 (Croff, 1980, 1883). The nuclear data used in the depletion calculations were from the libraries ENDF/B-V (ENDF/B-V, 1979). The calculations were carried out with target burnup values given in Table 2. Validation of the ORIGEN predictions have been performed through post-irradiation examinations on fuels of the type *R1*, *R2*, *T1* and *T2* (Glatz et al., 1994; Barrero Moreno et al., 1996, 1999; Sasahara et al., 1996, 1997). The agreement between calculations and measured compositions is within ~10% for the actinides and fission products.

The neutron multiplication factor for the PWR and LMFBR cores considered, was calculated using the general purpose 3-D Monte Carlo N-Particle code MCNP5 (X-5 Monte Carlo Team, 2003). The code was used coupled with the continuous energy neutron ENDF/B-VI data library (Rose, 1991; Hendricks et al., 1994). In particular, the neutron reactions cross-section table 66c, covering releases 0–3 and 5 of the B-VI data library, was used. The k_{eff} parameter was calculated, using the *kcode* card in the MCNP code, on the basis of the fresh fuel compositions in Table 1. The calculations were performed for arrays of assemblies in water and sodium (Na), with and without chemical shim and control rods as appropriate. Whole cores of PWR and LMFBR were simulated in a pin by pin model. In the modeling performed, only the fuel rod part with or without control rods was considered as a fuel assembly, while any structural parts were ignored. Hence, the height of the assembly would be considered as that of the fuel rod. The physical description of the fuel assemblies considered in the study is given in Table 3 (Knief, 1992; Todreas and Kazimi, 1993). In the case of the LMFBR core, sodium plenum regions were modeled above and below the fuel. The have a height of 40 and 10 cm, respectively. The presence of the Na plenum regions has been observed to minimize the sodium void reactivity effect in case of a completely dried core (Rachi et al., 1997; IAEA, 2007).

The core was described by defining once a fuel assembly cell which repeatedly appear in the geometry. Each calculation was carried out on the basis of 500 million neutron histories: 50,500 generations were simulated, with 10,000 neutron histories each, skipping the first 500 generations before averaging. The absolute

Table 2
Loading of the core for the fuel cycles considered.

Reactor	Core case	Fuel type	Core loading*	Burnup (GWd/t)
PWR	*CR1*	U	100% *R1*	55
PWR	*CR2*	MOX	70% *R1* + 30% *R2*	55
LMFBR	*CR3*	MOX	100% *R3*	110
LMFBR	*CT1*	MOX with 1%Am	100% *T1*	110
LMFBR	*CT2*	MOX with 1%Np	100% *T2*	110
PWR	*CS1*	Self-generated Pu recycle	100% *S1*	55
PWR	*CS2*	Self-generated TU recycle	100% *S2*	55
LMFBR	*CS3*	Self-generated TU recycle	100% *S3*	110

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

Table 3
Physical description and irradiation history of the fuel assemblies considered.

	PWR assemblies	LMFBR assemblies
Assembly		
Type	17 × 17 pins square	271 pins hexagonal
Number in core	193	364
Assembly pitch (mm)	207	179
Number of control rods	24	19
Number with control rods	12	16
Fuel pin		
Height	4	2.7
Active length (m)	3.81	1.6
Pellet diameter (mm)	9.5	8.5
Clad material	Zr	Stainless steel
Clad thickness (mm)	0.57	0.7
Pin pitch (mm)	12.6	9.7
Irradiation in reactor	37.5	123

standard deviation (σ), associated with the calculation of k_{eff}, ranged between 0.015% and 0.035% for k_{eff} values between 1.3 and 0.8 (Table 4).

3. Results

The neutron multiplication factor k_{eff}, for the PWR core (Table 2), has been calculated at BOI for three different core configurations: without chemical shim and all control rods withdrawn (reference configuration); with the presence of chemical shim and all control rods withdrawn; and, with the 12 control rod clusters fully inserted (total control element configuration). In the LMFBR core cases (Table 2), k_{eff} was calculated at BOI for three different core configurations: with all control rods withdrawn (reference configuration core); three control rod clusters fully inserted; and, sodium coolant removed (Na void) while control rods withdrawn. The k_{eff} with an associated standard deviation (σ), for the PWR and LMFBR core cases considered, are shown in Table 4. Furthermore, the corresponding reactivity worth values ($\%\Delta k/k$) have been included, describing the changes induced in the core reactivity by the chemical shim, the control rods, and the Na void. The different core cases in Table 4 have been compared at the 99% confidence level ($\pm 2.75\sigma$).

Within the LMFBR, the k_{eff} of the reference configuration cores *CT1*, *CT2* (Table 2), is close to that of the commercial reference configuration core *CR3*, on the basis of their comparable fissile content. The reference configuration core *CS3* shows a lower k_{eff} due to its lower plutonium content in relation to the other fast reactor cores. The expected negative reactivity worth induced in all four core cases, when the three control rod clusters are fully inserted and resulting to subcritical cores, is observed. The reactivity worth in the *CT1* and *CT2* core cases is close to the *CR3* commercial reference configuration core, indicating that similar changes are induced by the control rod insertion. Hence, the behaviour of the core fuelled with the emerging fuels *T1* or *T2* would be similar to the commercial LMFBR fuelled with the fuel *R3*. A slightly lower reactivity worth is observed in the core *CS3* fuelled with *S3*. Hence, with respect to the control rod worth which remains similar between the four core cases, the use of the emerging fuels would render the cores similar in operation and safety with the reference configuration core *CR3*.

In the case of Na void, with the three control rod clusters withdrawn, the LMFBR cores remain critical, while k_{eff} has increased in comparison to the reference configuration cores with the Na coolant present. This is due to the increase in the neutron population within the cores, since the absence of Na decreases the number of neutrons lost through absorption. Hence, Na void has induced a small positive reactivity worth in all four core cases, increasing marginally the already above criticality configuration cores.

Within the PWR group, the cores *CR1, CR2*, fuelled with fuels *R1* and *R2*, show a similar k_{eff}, on the basis of their fissile content. The cores *CS1* and *CS2* loaded with the recycling fuels *S1* and *S2* have also similar k_{eff} values, which are lower than for cores *CR1* and *CR2* because of the lower fissile content of fuels *S1* and *S2*. In the PWR cases, boric acid is diluted as chemical shim into water except in the control rod channels. The expected negative reactivity worth, induced in all four fuel cases, is observed. The chemical shim in sufficient amount brings down the reference configuration PWR core to just above criticality ($k_{eff} = 1.0008$). On the basis of the fissile content, the reactivity worth in the cores *CR1* and *CR2* is similar to core case *CS1* and much higher than the core case *CS2*. The MCNP calculations of the k_{eff}, for different boron contents in water have shown that just above criticality ($k_{eff} = 1.0008$) would be achieved with 1960 ppm natural boron used as chemical shim, for the PWR core cases *CR1, CR2* loaded with fuels *R1* or *R2*. The two self-generated recycling core cases *CS1* and *CS2*, having less fissile content than the cores *CR1* and *CR2*, would be just above criticality with 1540 ppm natural boron. Hence, the boron worth in reactors loaded with the self-generated fuels is drastically reduced compared to reactors loaded with commercial fuel.

The full insertion of the 12 control rod clusters would bring the core to subcriticality in all cases considered, as expected. In the core cases *CR1* and *CR2*, a reactivity worth of $\sim 18.6\%\Delta k/k$ would induce this change of k_{eff} to ~ 0.97, while in the *CS1* and *CS2* cases a worth between 17.4 and $20.3\%\Delta k/k$ would reduce k_{eff} to $\leqslant 0.85$. Noting that since the total control element worth does not change significantly between the four cases considered, the operation and safety of the emerging fuel cores would be similar to the reference configuration core *CR1*.

Table 4
k_{eff}, its standard deviation σ ($\times 0^{-2}$) and reactivity worth values for the fuel cycles considered.

PWR core configurations						LMFBR core configurations					
Core case	Reference configuration core	Chemical shim		Total control element		Core case	Reference configuration core	3 control rod clusters inserted		Na void	
	k_{eff} (1σ)	k_{eff} (σ)	$\%\Delta k/k$	k_{eff} (σ)	$\%\Delta k/k$		k_{eff} (σ)	k_{eff} (σ)	$\%\Delta k/k$	k_{eff} (σ)	$\%\Delta k/k$
CS1	1.03174 (0.023)	1.00080 (0.027)	2.99	0.85183 (0.035)	17.43	CT1	1.06181 (0.020)	0.99807 (0.025)	5.99	1.06684 (0.019)	−0.47
CS2	1.01842 (0.024)	1.00080 (0.027)	1.73	0.81201 (0.034)	20.26	CT2	1.05847 (0.020)	0.99805 (0.025)	5.71	1.06299 (0.020)	−0.42
CR1	1.19169 (0.016)	1.00080 (0.027)	16.01	0.96787 (0.026)	18.78	CS3	1.02690 (0.024)	0.98458 (0.025)	4.12	1.03218 (0.023)	−0.61
CR2	1.20725 (0.015)	1.00080 (0.027)	17.09	0.98464 (0.025)	18.43	CR3	1.06324 (0.021)	0.99811 (0.024)	6.13	1.07193 (0.019)	−0.78

4. Conclusions

Simulated reactor cores of PWR and LMFBR, loaded with different commercial and emerging fuels, have been compared with respect to their reactivity worth at BOI when criticality control is employed. On the basis of k_{eff} and, within each of the two reactors studied, cores are grouped together according to their fissile content. The reactivity worth does not change significantly when the commercial fuels are replaced with the emerging ones, in the case of the total control element (PWR) and three control rod (LMFBR) configurations. In the LMFBR four core cases, the reactivity worth values due to Na void are comparable between them and very small. Hence, operation and control of the reference core at beginning of irradiation are similar using emerging and commercial fuels (*CR1* and *CR3*). The boron worth in the PWR cores would be significantly reduced if commercial fuels were to be replaced by the self-generated fuels.

Further work is pursued to study core behaviour in the case of emerging fuels with respect to: the heterogeneous recycling of TRU in the fast reactor core; the sensitivity of the reactivity worth on different data libraries; the evolution of k_{eff} and $\%\Delta k/k$ during reactor operation and the change in fuel composition with burnup; and, fuel management aspects in the cores considered and implications on the economics of the whole fuel cycle and assessment of waste reduction.

References

Barrero Moreno, J., Garcia Alonso, J., Arbore, P., Nicolaou, G., Koch, L., 1996. Characterization of spent nuclear fuels by ion chromatography-inductively coupled plasma mass spectrometry. J. Anal. Atom. Spectrom. 11, 929–935.

Barrero Moreno, J.M., Betti, M., Nicolaou, G., 1999. Determination of caesium and its isotopic composition in nuclear samples using isotope dilution-ion chromatography-inductively coupled plasma mass spectrometry. J. Anal. Atom. Spectrom. 14, 875–879.

Croff, A.G., 1980. A User's Manual for the ORIGEN2 Computer Code, ORNL/TM-7175. Oak Ridge National Laboratory, Knoxville TN, USA.

Croff, A.G., 1983. ORIGEN2: a versatile computer code for calculating the nuclide compositions and characteristics of nuclear materials. Nucl. Technol. 62, 335–352.

ENDF/B-V Library, 1979. National Nuclear Data Centre, Brookhaven National Laboratory, Upton, NY.

Glatz, J.P., Babelot, J.F., Nicolaou, G., Koch, L., 1994. Theoretically and experimentally derived criteria for partitioning and transmutation of radionuclides in existing reactors. In: Proceedings 3rd International Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation, Cadarache, France, OECD/NEA, 12–14 December 1994.

Hendricks, J.S., Frankle, S.C., Court, J.D., 1994. ENDF/B-VI Data for MCNP. LANL Report LA-12891, (Loa Alamos National Laboratory, Loa Alamos, NM).

IAEA, 2007. Liquid metal cooled reactors: experience in design and operation TECDOC series-1569, Vienna, Austria.

Knief, R.E., 1992. Nuclear Engineering: Theory and Technology of Commercial Nuclear Power, second ed. Taylor & Francis, Washington.

Koch, L., 1986. Minor actinide transmutation – a waste management option. J. Less-Com. Met. 122, 371.

Koch, L., Nicolaou, G., 1993. Comparison of possible partitioning and transmutation schemes when added to the existing nuclear fuel cycles. In: Proceedings Meeting of the Technical Committee to IAEA on Safety and Environmental Aspects of Partitioning and Transmutation of Actinides and Fission Products, IAEA, Vienna, 29 November–2 December 1993, pp. 195–201, (IAEA-TECDOC-783).

Koch, L., Glatz, J.P., Nicolaou, G., 1997. Demonstration of a closed P&T cycle. In: Proc. Global 1997 Int. Conf. Future Nuclear Systems, Yokohama, Japan.

Mennerdahl, D., In de Betou, J., 2007. Guide for nuclear criticality safety analysis and review – accounting for neutron irradiation and radioactive decay, IAEA-TECDOC-1547, Vienna.

Nicolaou, G., Tsagas, N., 2006. Criticality safety of spent nuclear fuel assemblies from the transmutation of minor actinides in fast reactors. An. Nucl. Energy. 33, 305.

Nishihara, K., Nakayama, S., Morita, Y., Oigawa, H., Iwasaki, T., 2008. Impact of partitioning and transmutation on LWR high-level waste disposal. J. Nucl. Sci. Technol. 45, 84.

Rachi, M., Yamamoto, T., Jena, A.K., Takeda, T., 1997. Parametric study on fast reactors with low sodium void reactivity by the use of zirconium hydride layer in internal blanket. J. Nucl. Sci. Technol. 34, 193.

Rose, P.F., 1991 ENDF-201, ENDF/B-VI Summary documentation, fourth ed. Brookhaven National Lab, USA, BNL-NCS-17541.

Salvatores, M., 2002. Transmutation: issues, innovative options and perspectives. Progr. Nucl. Energy 40, 375.

Sasahara, A., Matsumura, T., Nicolaou, G., Glatz, JP., Toscano, E., Walker, C., 1996. Post-irradiation experiments and computational analyses of a high burnup UOX fuel and MOX fuel for spent fuel dry storage. In: Proceedings 10th Pacific Basin Nuclear Conference, Kobe, Japan, 20–25 October 1996.

Sasahara, A., Matsumura, T., Nicolaou, G., Bottomley, D., Rondinella, V., Walker, C.T., 1997. Post-irradiation examinations of MOX spent fuel for interim dry storage. In: Proceedings GLOBAL '97 International Conference on Future Nuclear Fuel Cycle Systems, OECD/NEA CEC IAEA, Yokohama, Japan, 5–10 October 1997.

Todreas, N.E., Kazimi, M.S., 1993. Nuclear Systems I: Thermal Hydraulic Fundamentals, second ed. Taylor & Francis, Washington.

Tommasi, J., Delpech, M., Grouiller, J.P., Zaetta, A., 1995. Long-lived waste transmutation in reactors. Nucl. Technol. 111, 133.

X-5 Monte Carlo Team, 2003. MCNP-A General Monte Carlo N-Particle Transport Code (Version 5), LA-UR-03-1987, Los-Alamos, USA.