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Accidental Criticality Estimation for Advanced CANDU Type Fuel Bundles containing Th-based Fuel Compositions

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Abstract: The paper aims to present the criticality (effective multiplication coefficient, k-eff) estimation for advanced CANDU type fuel bundles containing Thorium and LEU (low enriched uranium) oxides mixture, in some accidental scenarios. For this purpose, advanced CANDU fuel bundles with 37 and 43 fuel elements have been considered, both for fresh and spent Th-based fuel compositions. The scenarios take into account fuel bundles accidental immersion in light or heavy water, for several "dangerous" geometrical configurations (2, 3, 5 or 6 fuel bundles stacked close together at distances that could lead to critical conditions, but maintaining their structural integrity).

For spent fuel analysis, Th-based fuels irradiation in CANDU reactor conditions was simulated, by means of ORIGEN-S burn-up code, to reach 20 MWd/kg HE (heavy element) fuel burn-up, using specific powers for each considered fuel bundle. A number of 27 combinations of (Th, LEU) mixed oxides fuel have been investigated by considering different values for: fuel pellet density (8.5 g/cc, 9.0 g/cc and 9.5 g/cc), enrichment in U²³⁵ (9 wt%, 10 wt% and 11 wt%) and U/Th mass content ratio (20%, 25% and 30%). The criticality estimation has been performed using the Monte Carlo KENO-VI code, considering that the fuel bundle elements contain the same fuel composition.

The study results are comparatively presented for different fuel compositions in each fuel bundle case, and also for the same fuel composition considering different fuel bundle projects. Considered accidental scenarios led to subcritical k-eff values (less than 1), for both fresh and irradiated fuel compositions, assuming realistic neutrons leakage treatment. Estimated k-eff values are greater for fuel bundle with 43 fuel elements comparatively with the other fuel bundle project, but as U/Th content ratio increased the situation changed k-eff becoming greater for the fuel bundle with 37 elements.

Keywords: Accidental criticality, Advanced CANDU fuel bundle, Th-based fuel.

1 Introduction

Nuclear energy must find a reliable, affordable and public acceptable answer in dealing with an increased pressure due to availability of fissile resources, security of nuclear reactors operation and progressive spent fuel accumulation.

Actual requirements for higher nuclear fuel resources utilization put their mark on nuclear reactors design and construction, an increasing interest for advanced fuel cycles development being registered. Close and open options for nuclear fuel cycle are analyzed, with or without spent fuel reprocessing, in order to extend fuel resource utilization, to reduce radioactive waste amounts generated by reactors operation and to increase the proliferation resistance [1, 2].

The advanced fuel bundles selection should be based on criteria as follows: discharge fuel burnup, uniform radial distribution of bundle power, Uranium fraction in the spent fuel, minor actinides concentration in spent fuel etc. Fulfillment of such criteria for T37 and T43 fuel bundles has been investigated by RATEN ICN experts [3-12].

CANDU reactors are the illustration of a mature and wellproven technology, with high safety level and an evolutionary potential regarding the proliferation resistance, their versatility for different nuclear fuel cycles creating the premises for a better fuel resource utilization.

To initiate and sustain the fission reaction, Th-based fuels have to contain small amounts of fissile material (so called driver-fuel) such as low enriched uranium (LEU), Pu or U recovered from LWR spent fuel reprocessing; Pu or U^{233} obtained from fast breeder reactors operation could be also a potential solution in the future.

Present study proposes to estimate the criticality (effective multiplication coefficient, k-eff) associated to advanced



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CANDU type fuel bundles containing (Th, U)O₂ fuels, in potential accidental scenarios, for fresh and spent fuel. Advanced CANDU type fuel bundles with 37 and 43 fuel elements have been considered. Fuel bundles accidental immersion in H_2O or D_2O was assumed, for several "dangerous" geometrical configurations (fuel bundles stacked close together at distances that could lead to critical conditions, but maintaining their structural integrity).

2 Theoretical Basis for k-eff Calculations

Accidental criticality analyses for advanced CANDU fuel bundles have been performed by using KENO-VI [13], a 3D Monte Carlo code included as functional module in SCALE6 system, a worldwide implemented system of programs for criticality, shielding, thermal and fuel depletion analyses.

KENO-VI Monte Carlo code is used for criticality multigroup calculations considering a 3-dimensional system. It benefit of several characteristics: easy input for specific data; super-grouping of energy dependent data; scattering treated by means of Legendre polynoms; differential albedo coefficients used for particles tracking inside a reflector material. Several predefined geometrical shapes are available, but based on a set of quadrature equations, the user can model any volume according to the characteristics of the problem to be solved, [13].

The calculations start from Boltzmann transport equation, written as follows:

$$\frac{1}{v}\frac{\partial\Phi}{\partial t}(X,E,\Omega,t) + \Omega \cdot \nabla\Phi(X,E,\Omega,t) + \Sigma_t(X,E,\Omega,t) \cdot \Phi(X,E,\Omega,t) =$$
(1)
= $S(X,E,\Omega,t) + \iint_{E'\Omega'} \Sigma_s(X,E' \to E,\Omega' \to \Omega,t) \cdot \Phi(X,E',\Omega',t) \, d\Omega' \, dE'$

where: $\Phi(X, E, \Omega, t)$ = the neutron flux (neutrons/cm²/sec) per unit of energy, at energy E, per unit of solid angle in direction Ω , at position X and time t, moving with the speed v corresponding to energy E; $\Sigma_t(X, E, \Omega, t)$ = the total interaction macroscopic cross-section of the medium (cm⁻¹) at position X, energy E, in direction Ω and at time t; $\Sigma_s(X, E' \rightarrow E, \Omega' \rightarrow \Omega, t)$ = the differential interaction macroscopic cross-section of the medium (cm⁻¹) per unit of energy at energy E', per unit of solid angle in direction Ω' , at position X and time t, for scattering at energy E in direction Ω ; $S(X, E, \Omega, t)$ = the neutrons source (neutrons/ cm³/sec) born at position X and time t, per unit of energy at energy E, per unit of solid angle in direction Ω (the scattering source is excluded).

Defining $q(X, E, \Omega, t)$ as the neutrons total source obtained by summing on all the sources (external, scattering, fission and other contributions), following relation is written, [13]:

$$q(X, E, \Omega, t) =$$

$$= S(X, E, \Omega, t) + \iint_{E'\Omega'} \Sigma_s(X, E' \to E, \Omega' \to \Omega, t) \cdot \Phi(X, E', \Omega', t) \, d\Omega' \, dE'$$
(2)

Assuming the medium isotropy and ignoring the interaction cross-sections time dependence, the multigroup neutron transport equation is obtained:

$$\frac{1}{v_{g}}\frac{\partial\Phi_{g}}{\partial t}(X,\Omega,t) + \Omega \cdot \nabla\Phi_{g}(X,\Omega,t) + \Sigma_{tg}(X) \cdot \Phi_{g}(X,\Omega,t) = q_{g}(X,\Omega,t)$$
(3)

where: g = the energy group of interest; $v_g =$ average speed of neutrons from energy group g; $\Phi_g(X,\Omega,t) =$ angular flux of the neutrons having energies in energy group g, at position X and time t; $\Sigma_{rg}(X) =$ total interaction macroscopic cross-section of the medium at position X for energy group g; $q_g(X,\Omega,t) =$ the total source of neutrons contributing to energy group g, for position X and time t, in direction Ω .

Using the relation $X' = X - R\Omega$, to account for the time dependence, an integration factor is considered in both parts of Eq.(3), [13].

Defining
$$T(R) = \int_{0}^{K} \sum_{tg} (X - R'\Omega) dR'$$
, we obtain:

$$\Phi_{g}(X, \Omega) = \int_{0}^{\infty} q_{g}(X - R\Omega, \Omega) \cdot e^{-T(R)} dR \qquad (4)$$

If there is no external source of neutrons, the total source of neutrons can be defined as:

$$q_g(X,\Omega) = \sum_{g'} \int d\Omega' \Phi_{g'}(X,\Omega) \cdot \Sigma_s(X,g' \to g,\Omega' \to \Omega) + \frac{1}{k} Q_{g'}(X,\Omega)$$
(5)

where: k = the greatest eigenvalue of the integral equation; $Q_g'(X,\Omega)$ = the fission source in position X, for energy group g and in direction Ω (all the fissions in energy group g produced by all energy groups in the previous generation); $\Sigma_s(X, g' \rightarrow g, \Omega' \rightarrow \Omega)$ = the interaction crosssection for scattering in position X from energy group g' and direction Ω ' in energy group g and direction Ω .

The multiplication constant k can be defined as the ratio between the number of neutrons from (n+1)-th generation and the number of neutrons from n-th generation.

Considering neutrons fission as an isotropic process, after several mathematical operations, the equation to be solved in KENO-VI is obtained [13]:

$$\frac{v_{g}(X) \cdot \Sigma_{fg}(X)}{\Sigma_{tg}(X)} \Sigma_{tg}(X) \cdot \Phi_{g,n}(X,\Omega) = \frac{v_{g}(X) \cdot \Sigma_{fg}(X)}{\Sigma_{tg}(X)} \Sigma_{tg}(X) \int_{0}^{\infty} dR \ e^{-T(R)} \cdot \left\{ \frac{1}{k} \sum_{g',\Omega'} \int_{\Omega'}^{V_{g'}(X-R\Omega) \cdot \Sigma_{fg'}(X-R\Omega)} \chi(X-R\Omega, g' \to g) \cdot \Sigma_{tg'}(X-R\Omega) \cdot \Phi_{g',n-1}(X-R\Omega, \Omega') \cdot \frac{d\Omega'}{4\pi} + \sum_{g',\Omega'} \int_{\Omega'}^{\Sigma_{g}(X-R\Omega)} \frac{\Sigma_{g}(X-R\Omega)}{\Sigma_{tg'}(X-R\Omega)} \Sigma_{tg'}(X-R\Omega) \cdot \Phi_{g',n}(X-R\Omega, \Omega') \ d\Omega' \right\}$$
(6)

where n designates the n-th generation of particles, and (n-1) the (n-1)-th generation.

In KENO-VI, the above equation is solved by an iterative process; the fissions produced in position X in energy group g by the neutrons from the (n - 1)-th generation, normalized to the system neutron multiplication factor, are given as [13]:

$$\frac{1}{k} \sum_{g' \; \Omega'} \int_{\Omega'} \frac{\nu_{g'}(X) \cdot \Sigma_{fg'}(X)}{\Sigma_{tg'}(X)} \chi(X, g' \to g) \cdot \Sigma_{tg'}(X) \cdot \Phi_{g', n-1}(X, \Omega') \frac{d\Omega'}{4\pi}$$
(7)

The particles collision points used in KENO-VI are chosen by selecting the travelling paths in distribution $e^{-T(R)}$, representing the transport probability of a particle from any position (X – R Ω) in position X.

In order to reduce the statistical deviation of the effective neutron multiplication factor, KENO-VI uses the weighted tracking of the particles, taking into account for absorption by reducing of the neutrons weight, without ending the particle history. To avoid the tracking of neutrons with very low weights, when the neutron weight reaches a defined threshold value (*WTLOW*), the Russian roulette technique is applied. The neutrons passing from the Russian roulette get a new associated weight, *WTAVG*. *WTLOW* and *WTAVG* values are functions of position and energy, [13].

3 Initial Data for Calculations, Scenarios, Assumptions

The present study analyses have been performed for the advanced CANDU type fuel bundles T37 and T43 developed by the specialists from the Institute for Nuclear Research (RATEN ICN) Pitesti, Romania.

T37 fuel bundle (similar to CANDU standard fuel bundle) contains 37 identical fuel elements, disposed in annular geometry with one central element and 3 concentrical rings of 6, 12 and 18 fuel elements. T43 fuel bundle (similar to CANFLEX fuel bundle) is composed by 43 fuel elements (different in diameter - 8 "thick" elements and 35 "thin" elements, "thick" or "thin" referring here as comparison with CANDU standard fuel element diameter), disposed in annular geometry with one central element and 3 concentrical rings of 7, 14 and 21 fuel elements.

A number of 27 combinations of (Th, LEU) mixed oxides fuel have been investigated [7-11] by considering different values for: fuel pellet density (8.5 g/cc, 9.0 g/cc and 9.5 g/cc), enrichment in U^{235} (9 wt%, 10 wt% and 11 wt%) and U/Th mass content ratio (20%, 25% and 30%).

Fuel irradiation was simulated by means of ORIGEN-S burn-up code [14], using specific spectral factors from previous performed lattice cell calculations [7, 10].

Th-based fuels irradiation in CANDU reactors conditions was simulated to reach 20 MWd/kg HE (heavy element) fuel burn-up, using the specific powers of 42 kW/kg HE (T37 fuel bundle) and 50 kW/kg HE (T43 fuel bundle). Analyses have been carried out considering that Th-based fuel bundle elements contain the same fuel composition.

Fresh and irradiated fuel criticality estimation has been performed by using Monte Carlo KENO-VI code [13], whose calculations were based on 238GROUPNDF5 nuclear data library (general nuclear data library used for criticality analyses, known as LAW, Library to Analyze Radioactive Waste). It contains data for more than 300 nuclides, on 148 rapid and 90 thermal neutron energy groups, all nuclides using the same averaged spectra, consisting of:

- one Maxwell spectrum (with a peak at 300 K) in the energy range from 10^{-5} eV to 0.125 eV,
- one spectrum evolving as $1/\!E$ in the energy range from 0.125~eV to 67.4~keV,

- one fission spectrum (with effective temperature of 1.273 MeV) ranging from 67.4 keV to 10 MeV,

- one spectrum evolving as 1/E in the energy range 10 MeV to 20 MeV.

In the Monte Carlo simulation, 203 generations of 1,000 particles each have been considered.

The following accidental scenarios have been considered:

- SCE1: Accidental immersion of fresh fuel bundles in H₂O, during the transport to the reactor;
- SCE2: Accidental immersion of fresh fuel bundles in D₂O, during the transport to the reactor;
- SCE3: Discharge of irradiated fuel bundles from the reactor and storage into the cooling pool (filled with H₂O).

The accident assumes that 2, 3, 5 or 6 fuel bundles become stacked close together ("dangerous" configurations) at distances that could lead to critical conditions, but maintaining their structural integrity.

For each scenario, the standard composition of nuclides is specified together with the mixing in which the nuclides are considered; it follows modeling of the "geometrical units" (fuel element, fuel bundle, groups of fuel bundles), shown in Figures 1 and 2.



As regarding the neutrons leakage treatment, the following options were considered [13]:

(a) \pm X:REFL, \pm Y:REFL, \pm Z:REFL: It assumes a limited layer of water above the fuel bundles (2 cm thick); in X and Y directions the lattice cell has been extended so that the water covers completely the fuel bundles

(b) \pm X: REFL, \pm Y: REFL, \pm Z: VOID: It assumes restrictive conditions justified by the hypothesis of a single "row" of fuel bundles; neutrons "escaping" through lattice cell faces +Z and -Z are considered to leave definitively the system;

(c) \pm X:REFL, \pm Y:REFL, -Z:CONC4; +Z:VOID: It assumes a single "row" of fuel bundles covered by a layer of water, fuel bundles being placed on a concrete platform; face -Z (bottom) has a specific albedo for a 10 cm thick concrete layer (CONC4), and we kept the option VOID for face +Z.



Figure 1. 3-D modeling of the fuel bundles with KENO-VI



Figure 2. 3-D modeling of "dangerous" arrangements of fuel bundles with KENO-VI

4 Results on Investigated Advanced CANDU Fuel Bundles

Figures 3 to 6 present the estimated values for the effective multiplication constant (k-eff) corresponding to T37 and T43 fuel bundles in the fresh fuel criticality scenarios (SCE1 and SCE2), for selected "dangerous" geometrical configurations, (Th,U)O₂ fuel compositions with: pellet density = 9.0 g/cc, enrichment in U²³⁵ = 10%, U/Th content ratio = 20% and 30%.

The criticality scenario SCE2 conducted to k-eff larger than 1 for the infinite lattice of fuel bundles (option a for neutron leakage treatment), which is over-conservative. However, 3D calculations performed for realistic situations (options b and c for neutron leakage treatment) conducted to k-eff values less than 1. It must be mentioned here that for realistic situations, k-eff estimated values were significantly smaller than the ones obtained in SCE1, both for T37 and T43 bundles. For criticality scenario SCE1, the most "dangerous" arrangements seems to be the stacks of 3 fuel bundles (option a for neutron leakage treatment) and 6 fuel bundles (options b and c for neutron leakage treatment), respectively. As regarding SCE2, the most "dangerous" arrangements are the stacks of 2, 5 and 6 fuel bundles (option a for neutron leakage treatment) and 6 fuel bundles (options b and c for neutron leakage treatment), respectively.



Figure 3. Estimated k-eff values for T37(left) and T43 (right) fuel bundles in criticality scenario SCE1 for different neutron leakage treatment options and U/Th content = 20%



Figure 4. Estimated k-eff values for T37(left) and T43 (right) fuel bundles in criticality scenario SCE1 for different neutron leakage treatment options and U/Th content = 30%.



Figure 5. Estimated k-eff values for T37(left) and T43 (right) fuel bundles in criticality scenario SCE2 for different neutron leakage treatment options and U/Th content = 20%.



Figure 6. Estimated k-eff values for T37(left) and T43 (right) fuel bundles in criticality scenario SCE2 for different neutron leakage treatment options and U/Th content = 30%.



Figure 7. Estimated k-eff values for T37 fuel bundles in criticality scenarios SCE1 (left) and SCE2 (right) for different enrichment in U²³⁵ (fuel pellet density = 9.0 g/cc, U/Th content ratio = 30%).

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Figure 8. Estimated k-eff values for T37 fuel bundles in criticality scenarios SCE1 (left) and SCE2 (right) for different enrichment in U^{235} (fuel pellet density = 9.0 g/cc, U/Th content ratio = 30%).

Figures 7 and 8 show the variations of k-eff values estimated for T37 and T43 "dangerous" geometrical configurations in SCE1 and SCE2, assuming the realistic option (c) for neutron leakage (\pm X: REFL; \pm Y: REFL; -Z: CONC4; +Z: VOID) and considering (Th, U)O₂ fuels with different enrichment in U²³⁵.

As the fuel enrichment in U^{235} increases, for both advanced fuel bundles a slightly increasing of k-eff values is observed, the most "dangerous" arrangements being the stack of 5 and 6 fuel bundles (for SCE1) and 6 fuel bundles (for SCE2). Estimated k-eff values for criticality scenario SCE1 were significantly greater than the ones obtained for scenario SCE2, regardless of the fuel bundle type or fuel enrichment in U^{235} .

Figures 9 and 10 present the estimated values for the effective multiplication constant (k-eff) corresponding to T37 and T43 spent fuel bundles "dangerous" arrangements in criticality scenario SCE3 conditions, considering (Th,U)O₂ fuel compositions characterized by: fuel pellet density = 9.0 g/cc, enrichment in $U^{235} = 10\%$, U/Th content ratio = 20% and 30%. k-eff has been estimated considering different neutron leakage treatment options, as mentioned before for the criticality scenarios associated with Th-based fresh fuel.



Figure 9. Estimated k-eff values for T37 (left) and T43 (right) fuel bundles in criticality scenario SCE3 for different neutron leakage treatment options and (Th, U)O₂ fuel with U/Th content ratio = 20%.



Figure 10. Estimated k-eff values for T37 (left) and T43 (right) fuel bundles in criticality scenario SCE3 for different neutron leakage treatment options and (Th, U)O₂ fuel with U/Th content ratio = 30%.

For criticality scenario SCE3 associated to the spent fuel, taking into account the neutron leakage treatment options and assuming no fission products in the irradiated fuel composition (to avoid as much as possible the neutron absoptions leading to k-eff reduction), k-eff estimated values were less than 1 regardless the neutron leakage treatment option, fuel bundle type or Th-based fuel composition.

Similar with SCE1 case, the most "dangerous" arrangements seems to be the stacks of 3 fuel bundles (option a for neutron leakage treatment) and 6 fuel bundles (options b and c for neutron leakage treatment), respectively. However, k-eff values were smaller than those obtained for fresh fuel bundles arrangements in SCE1, but significantly greater than k-eff values corresponding to the advanced T37 and T43 fuel bundles in SCE2 conditions.

Comparative analyses have been performed by means of calculated relative differences corresponding to:

• comparison of k-eff estimated values, considering the same fuel bundle project (T37 or T43) with different (Th,U)O₂ fuel compositions (variations of the fuel pellet density, enrichment in U^{235} , and U/Th content ratio);

• comparison of k-eff estimated values, considering the same (Th, U)O₂ fuel composition, but different fuel bundle projects.

In first comparison, the modification of Th-based fuel composition parameters led to an increasing of k-eff values as follows: up to 3.5% for each step of fuel pellet density variation (8.5 g/cc to 9.0 g/cc and 9.0 g/cc to 9.5 g/cc); (3.0- 5.0)% for each step of enrichment in U^{235} variation (9 wt% to 10 wt% and 10 wt% to 11 wt%); (3.0- 6.0)% for each step of U/Th content ratio variation (20% to 25% and 25% to 30%).

Table 1. Relative differences for T43 vs. T37 comparison.

SCE1	SCE2	SCE3
(%)	(%)	(%)
2.28	0.68	0.89
1.62	0.61	1.27
1.76	1.16	1.14
1.47	0.67	0.75
2.90	-0.82	-0.85
2.03	-0.12	-0.26
1.62	0.78	-0.81
2.90	0.73	-1.25
	2.28 1.62 1.76 1.47 2.90 2.03 1.62	2.28 0.68 1.62 0.61 1.76 1.16 1.47 0.67 2.90 -0.82 2.03 -0.12 1.62 0.78

Fresh fuel criticality scenarios lead to relative differences greater than the the ones obtained for the spent fuel criticality scenario.

The second comparison results are shown in Table 1, illustrating the relative differences calculated for selected "dangerous" fuel bundles arrangements and $(Th,U)O_2$ fuel composition characterized by: fuel pellet density = 9 g/cc, enrichment in U^{235} =10%, and U/Th content ratio = 20% and 30%, respectively.

k-eff estimated values for T43 fuel bundle project were greater than those corresponding to T37 fuel bundle project, but increasing of the U/Th content ratio change the situation, k-eff values for T37 fuel bundle becoming greater than those obtained for T43 fuel bundle.

4 Conclusions

k-eff has been estimated for two advanced CANDU fuel bundles containing Th-based mixed oxides fuels, in several accidental scenarios for fresh and irradiated fuel.

T37 and T43 advanced fuel bundle projects, developed in INR Pitesti - cylindrical geometry with 37/43 fuel elements arranged on concentrical rings - were used in the analyses.

Various (Th, U)O₂ mixed oxide fuel compositions were considered, taking into account different values for fuel pellet density, enrichment in U^{235} and U/Th content ratio.

Three accidental scenarios have been defined: SCE1 and SCE2 for fresh fuel (accidental immersion of fuel bundles in H_2O or D_2O), and SCE3 for irradiated fuel (immersion of fuel bundles in cooling pool filled with H_2O). The accident assumes that 2, 3, 5 or 6 fuel bundles become stacked close together, fuel bundles integrity being preserved.

Fresh fuel accidental criticality scenarios led to subcritical k-eff (less than 1), except for SCE2 assuming an infinite lattice cell for neutrons leakage treatment. However, 3D calculations performed assuming realistic neutrons leakage treatment (options b and c) revealed subcritical k-eff values for considered "dangerous" configurations, fuel bundles and fuel compositions.

In the case of accidental criticality scenario SCE3 for irradiated fuel, estimated k-eff values were less than 1, regardless the considered neutron leakage treatment option, fuel bundle type or Th-based fuel composition.

Generally, the estimated k-eff values were greater for T43 fuel bundle comparatively with those corresponding to T37 fuel bundle. Increasing of the U/Th content ratio to 30% led in present study to a change for irradiated fuel, k-eff values being larger for T37 fuel bundle comparatively with the ones obtained for T43 fuel bundle.

Acknowledgement

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