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CANDU® - A Versatile Reactor for Plutonium Disposition or Actinide Burning

CANDU^MD - Un réacteur polyvalent pour l'élimination du plutonium ou pour la combustion des actinides

P.S.W. Chan, M.J.N. Gagnon, P.G. Boczar, R.J. Ellis,
R.A. Verrall

Presented at Global '97 International Conference on Future Nuclear Systems, held in Yokohama, Japan, 1997 October 5-10

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by

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Chalk River, Ontario KOJ 1J0
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CANDU[MD] - UN RÉACTEUR POLYVALENT POUR L'ÉLIMINATION DU PLUTONIUM OU POUR LA COMBUSTION DES ACTINIDES

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RÉSUMÉ

Une économie neutronique importante, un renouvellement du combustible en marche et un type de grappe de combustible simple signifient une grande polyvalence dans l'utilisation du réacteur CANDU[MD1] en ce qui a trait à l'élimination du plutonium d'origine militaire et à la combustion des actinides radioactifs à longue période, notamment les isotopes du plutonium, du neptunium et de l'américium, produits dans les réacteurs de puissance civils. Des caractéristiques de sûreté inhérentes sont intégrées à la conception des grappes qui contiennent les combustibles au plutonium et aux actinides. Cela permet aux réacteurs CANDU existants de fonctionner avec différents cycles du combustible à base de plutonium sans nécessiter des modifications importantes de la conception actuelle du réacteur.

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[1] CANDU[MD] est une marque déposée d'Énergie atomique du Canada limitée (EACL).

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ABSTRACT

High neutron economy, on-line refuelling, and a simple fuel-bundle design result in a high degree of versatility in the use of the CANDU® reactor for the disposition of weapons-derived plutonium and for the annihilation of long-lived radioactive actinides, such as plutonium, neptunium, and americium isotopes, created in civilian nuclear power reactors. Inherent safety features are incorporated into the design of the bundles carrying the plutonium and actinide fuels. This approach enables existing CANDU reactors to operate with various plutonium-based fuel cycles without requiring major changes to the current reactor design.

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1. INTRODUCTION

The fuel-cycle flexibility of CANDU reactors provides many options for the management of weapons-derived plutonium. These options include conventional mixed-oxide (MOX) fuel, optimized for plutonium dispositioning, as well as advanced options, optimized for plutonium annihilation.

One of the advanced options, which uses an inert matrix (SiC) as the carrier for the weapons-derived plutonium, can also be used to destroy actinides created in civilian power reactors.

Detailed fuel-management simulations demonstrate that an existing CANDU reactor can burn either conventional MOX fuel, or inert-matrix fuel containing plutonium or actinides without major modification. Fuelling rate, channel power, bundle power and element power ratings are all within current limits. The time-dependent fuel-management simulations were performed using RFSP [1], with lattice parameters from WIMS - AECL [2]. Spent fuel compositions were derived from a coupled WIMS / multi-region ORIGEN (Ian Gauld, AECL, unpublished report, 1997) calculation.

2. CONVENTIONAL MOX OPTIONS FOR DISPOSITION OF WEAPONS-DERIVED PLUTONIUM

The conventional MOX fuel is a near-term, technically achievable, safe and economic option for disposition of weapons-derived plutonium. The main objective is not to destroy the plutonium, but to convert it to a form that has a high degree of diversion resistance with the characteristics of spent fuel, while producing electricity. The flexibility of the CANDU reactor allows the choice of several variants of this option, depending on the requirements and priorities. The important considerations are the timeliness of the deployment option, the plutonium disposition rate, and the economics.

2.1 Conventional MOX Option 1

The first option, MOX(1), was chosen to provide the fastest start for the plutonium disposition mission. The reference fuel uses the standard 37-element geometry and is designed to perform within the operating and safety envelopes for natural-uranium fuel. Depleted uranium (0.2 wt % ^{235}U) is the matrix material throughout the bundle. In the central element, and the next ring of 6 elements, 5% dysprosium (a burnable poison) is mixed with the depleted uranium. Plutonium is confined to the outer 2 rings of fuel: 2.0% plutonium in the third ring of 12 elements, and 1.2% plutonium in the outer ring of 18 elements. The bundle average burnup of the reference MOX fuel is 9.7 MW·d/kg HE, compared with 8.3 MW·d/kg HE for natural-uranium fuel in Bruce A reactors.

Peak element burnup is about the same as for natural uranium. The fresh fuel contains 232 g plutonium per bundle, of which 94% is fissile. The plutonium disposition rate in a Bruce A reactor is 1.0 Mg Pu per year per reactor (assuming an 80% capacity factor). The MOX fuel fabrication capacity requirement is about 80 Mg per year per reactor.

2.2 Conventional MOX Option 2

In the second option, MOX(2), the plutonium disposition rate was increased by 50% by increasing the amount of plutonium in the 37-element bundle, without significantly increasing the fuel burnup. To compensate for the excess reactivity, the burnable poison content in the central elements was increased from 5% to 15%, and the purity of the coolant and moderator was downgraded from 99.75% to 97%.

The resultant 37-element fuel bundle has a plutonium loading of 336 g confined to the outer 2 rings (3.1% Pu in ring 3, 1.6% Pu in ring 4), 15% dysprosium in the central 7 elements, and depleted uranium as the base material throughout the bundle. The average discharge burnup is slightly greater than in the earlier study, 10 MW·d/kg HE. The plutonium disposition rate in a Bruce A reactor is 1.5 Mg Pu per year. The MOX fuel fabrication capacity requirement is 78 Mg per year per reactor. As with the earlier design, the reactor operates within the natural-uranium licence envelope.

2.3 Conventional MOX Option 3

The third option, MOX(3), was designed to increase the energy output that can be extracted from the weapons-derived plutonium, which could be important if the plutonium is viewed as a valuable resource. This option uses a large amount of plutonium in the CANFLEX bundle, which has 43 fuel elements arranged in rings of 1, 7, 14 and 21 elements.

The CANFLEX bundle has 20% lower peak element ratings than does the 37-element bundle operating at the same bundle power, and improved thermalhydraulic performance. The lower ratings facilitate achievement of the higher burnup with this option, which has a core-average burnup of 25 MW·d/kg HE, and a peak element burnup of 35 MW·d/kg HE. Nominal D_2O purity of 99.75% is used in the calculations.

The plutonium is confined to the outer 2 rings of fuel: 4.6% plutonium in ring 3, and 2.6% in ring 4, mixed with depleted uranium. The central 8 elements contain 7% dysprosium mixed with depleted uranium. This advanced MOX bundle contains 473 g plutonium in the fresh fuel. The plutonium disposition rate in a Bruce A reactor is 0.8 Mg Pu per year. The higher fuel burnup reduces the MOX fuel fabrication requirement to 28 Mg per year per reactor, which would result in a significant reduction in the capital cost of the MOX fuel-fabrication plant.

2.4 Fuel Composition of Conventional MOX Options

Table 1 shows the composition of the fresh and spent fuel for the 3 conventional MOX options. The fissile plutonium destruction efficiencies for the 3 options are 58%, 41% and 70% respectively. The total amount of plutonium is reduced by 34%, 23% and 46% respectively for the 3 options.

3. ADVANCED OPTIONS FOR DESTRUCTION OF WEAPONS-DERIVED PLUTONIUM

The CANDU versatility enables advanced options that can achieve near-complete destruction of the weapons-derived fissile plutonium. One such option is the use of an inert matrix (non-fertile material) as the carrier for weapons-derived plutonium. Another option, Pu-ThO_2 fuel, would also achieve a very high efficiency in plutonium destruction.

3.1 Plutonium Annihilation Using an Inert Matrix

Besides neutronic considerations, the selection of suitable inert-matrix materials must consider their mechanical, physical and chemical properties. A partial list of candidates under consideration in Europe, the United States and Canada includes SiC, $MgAl_2O_4$ (spinel), $ZrSiO_4$ (zircon), ZrO_2, CeO_2, $CePO_4$ and BeO.

Based on detailed assessments of the desirable properties of these inert materials, AECL is focusing its efforts on SiC as the most promising candidate for inert-matrix applications.

The very high thermal conductivity of SiC will result in very low temperatures, both in normal operation and in postulated accidents, with the expected benefit of low fission-gas release; its high melting temperature is also a benefit. There do not appear to be any long-lived activation products resulting from its irradiation, and it would appear to be a stable waste form. Simulated irradiation performance, and other tests, are positive to date. Further studies and experiments will be conducted to determine which material is the best candidate for use as an inert matrix for plutonium annihilation.

The reference Pu-SiC fuel contains 250 g of weapons-derived plutonium mixed uniformly with SiC in the outer 30 elements of a standard 37-element bundle. The central element contains 20 g of gadolinium. Another 40 g of gadolinium are distributed uniformly over the 6 elements in the next ring. The distribution of the gadolinium, which has a very high depletion rate, was optimized to suppress the excess reactivity of the fresh fuel and to minimize the power ripple that is due to refuelling.

Detailed fuel-management simulations using a uniform 2-bundle-shift refuelling scheme in a Bruce A reactor indicated that the maximum instantaneous channel power is below 7.1 MW and the maximum instantaneous bundle power is below 970 kW. Both are within the current licensing limits. The fuelling rate is about 15 bundles per full-power day of operation, which is within the current fuelling machine capability.

For this Pu-SiC fuel, as well as for all the conventional MOX options, void reactivity is negative. Hence there would be no power pulse in a postulated LOCA, and the safety and licensing analyses would be greatly simplified.

The fuel temperature coefficient is very slightly positive, about 12 $\mu k/^{\circ}C$; however, this is irrelevant in the safety analysis since any increase in heat in the fuel would immediately be transferred to the coolant because of the high thermal conductivity of the SiC, thus reducing the coolant density and producing a negative reactivity feedback because of the negative void-reactivity coefficient.

3.2 CANDU Pu-Thorium Fuel for Plutonium Destruction

The second CANDU plutonium-annihilation option employs the use of ThO_2 as a carrier for the plutonium. This is a forward-looking strategy that uses plutonium to convert ^{232}Th to ^{233}U, for possible use as a future energy resource. The ^{233}U would be safeguarded in the spent fuel, with all the proliferation-resistant features of spent UO_2 or MOX fuel. Moreover, the radiation fields caused by the presence of ^{232}U (which emits copious α particles) and its daughter products (particularly ^{208}Tl, which emits a 2.6 MeV γ ray), provide a high degree of self-protection and render ^{233}U unattractive as a weapons material. The ^{233}U could be recovered in the future using a proliferation-resistant technology.

To maximize the destruction of the plutonium, good neutron economy was desired. A reduction in void reactivity was also sought, to compensate for the faster dynamic behaviour of the fuel (shorter neutron lifetime and smaller delayed neutron fraction). To achieve these two objectives, the central elements in a CANFLEX bundle were replaced with a large central graphite displacer. Plutonium at 2.6% (354 g per bundle) was mixed with thorium in the remaining 35 elements in the outer 2 fuel rings of the CANFLEX bundle. Enrichment grading in the outer 2 fuel rings would result in peak element ratings that are comparable to those in a 37-element bundle with natural-uranium fuel. The resultant burnup was 30 MW·d/kg HE, a burnup for which there is CANDU experience with Pu-ThO_2 fuel. Void reactivity was 8.6 mk, which is judged to be acceptable with the current shutdown system.

3.3 Fuel Composition of the Advanced Options

Table 2 shows the composition of the fresh and spent fuel: 76% of the total plutonium is destroyed in the Pu-SiC option, 77% in the Pu-ThO_2 option, and 94% of the fissile plutonium is destroyed in both options. In the Pu-ThO_2 option, fissile ^{233}U (including its parent ^{233}Pa) is produced to the extent of 168 g per bundle, which can be recovered and recycled in the future using a proliferation-resistant process, when warranted by economic, resource-availability, and other considerations.

4. SUMMARY OF WEAPONS-DERIVED PLUTONIUM MANAGEMENT OPTIONS

Table 3 summarizes the relative merits of the conventional and advanced options for managing weapons-derived plutonium in CANDU reactors. Of the 3 conventional options, MOX(3) gives the best performance in terms of plutonium destruction efficiency, fuel economics and capital cost. However, its implementation would require more time for fuel qualification than the MOX(1) option does, which can be implemented in the shortest time. MOX(2) gives the highest plutonium disposition rate by sacrificing the plutonium destruction efficiency. Both advanced options have essentially the same efficiency in plutonium destruction. The Pu-SiC option does not create any new fissile material. The Pu-ThO_2 option creates fissile ^{233}U, which can be considered as a safeguardable future energy source.

5. REACTOR PHYSICS ASSESSMENTS FOR ACTINIDE BURNING

The results for annihilating weapons-derived plutonium are directly applicable to the case of burning actinides in CANDU reactors. An appropriate mixture of SiC and the actinide mix consisting of the ^{237}Np, ^{241}Am, ^{243}Am, and plutonium from spent PWR fuel was used in the outer 30 elements of the standard 37-element bundle. Gadolinium was used in the inner 7 elements to suppress the excess reactivity of the fresh fuel. Several combinations of actinide inventory and gadolinium loading were considered. The best result was achieved using a full-core loading of actinide-SiC fuel containing 400 g of actinides and 60 g of gadolinium per bundle in a CANDU 6-type reactor.

Table 4 gives the actinide composition for the fresh and spent actinide fuel. About 60% of the total original actinide inventory is destroyed, as is 90% of the initial fissile plutonium inventory. Detailed refuelling simulations were performed. The fuelling rate is 9.2 bundles per full-power day, resulting in the destruction of 0.68 Mg of actinides in a CANDU 6 reactor per year (assuming an 80% capacity factor). Maximum instantaneous channel power was less than 7.0 MW, and maximum instantaneous bundle power was less than 1000 kW. Fuel element power is expected to be below the current safety limit because of the high thermal conductivity of the SiC matrix. Fuel temperature coefficient is slightly positive, 12.2 μ k/°C, and the coolant-void reactivity is -4.2 mk. The power coefficient is expected to be significantly negative because of the negative coolant-void reactivity and the high heat conductivity of the SiC matrix.

6. SUMMARY

The CANDU system provides unsurpassed flexibility for plutonium management through high neutron economy, on-line refuelling, and a simple, economical fuel-bundle design. The CANDU MOX options offer timely and technically achievable ways of dispositioning weapons-derived plutonium. Advanced options, using an inert matrix or the thorium fuel cycle, can achieve near-complete annihilation of fissile plutonium and significant destruction of the other actinides in existing CANDU reactors.

REFERENCES

1. B. Rouben. 1996. Overview of Current RFSP-Code Capabilities for CANDU Core Analysis, Atomic Energy of Canada Limited Report, Atomic Energy of Canada Limited Report, AECL-11407.

2. J.V. Donnelly. 1986. WIMS-CRNL—A User's Manual for the Chalk River Version of WIMS, Atomic Energy of Canada Limited Report, AECL-8955.

TABLE 1. Fuel Composition: CANDU MOX Options (g/bundle)

	MOX (1)		MOX (2)		MOX (3)	
Nuclide	0 MW•d/kg	9.7 MW•d/kg	0 MW•d/kg	10.0 MW•d/kg	0 MW•d/kg	25.0 MW•d/kg
^{239}Pu	218.0	78.8	313.8	172.9	443.4	106.8
^{240}Pu	13.5	57.6	19.4	69.3	27.4	108.1
^{241}Pu	0.3	13.2	1.2	13.1	1.7	25.2
^{242}Pu	0.05	3.8	1.3	2.9	0.1	13.0
Total	*231.9*	*153.4*	*335.7*	*258.2*	*472.6*	*253.2*

TABLE 2. Fuel Composition: Advanced CANDU Options (g/bundle)

	Pu-SiC (Inert Matrix)		$Pu-ThO_2$ (Thorium MOX)	
Isotope	0 MW•d/kg	733 MW•d/kg	0 MW•d/kg	30 MW•d/kg
^{238}Pu	---	---	0.2	0.3
^{239}Pu	235.0	5.2	331.3	5.3
^{240}Pu	14.0	35.0	21.2	45.2
^{241}Pu	0.9	9.1	1.4	14.5
^{242}Pu	0.07	10.9	0.2	15.9
Total Pu	*250.0*	*60.2*	*354.3*	*81.2*
$^{233}U + ^{233}Pa$	---	---	---	167.7
^{237}Np	---	---	---	0.004
^{241}Am	---	0.10	---	0.59
^{243}Am	---	0.86	---	2.21
^{242}Cm	---	0.10	---	0.26
^{244}Cm	---	0.13	---	0.42

TABLE 3. Summary of CANDU Weapons-Derived Plutonium Management Options

	Pu-U MOX (1)	Pu-U MOX (2)	Pu-U MOX (3)	Pu-SiC (inert matrix)	Pu-Th (thorium)
Net Pu-Destruction Efficiency (%)	34	23	46	76	77
Net Fissile Pu-Destruction Efficiency (%)	58	41	70	94	94
Pu-Disposition Rate (Mg Pu/Gw_e a)	1.56	2.22	1.23	1.66	1.04
Energy Produced (GW_e•a/MgPu)	0.64	0.45	0.81	0.61	0.96

TABLE 4. Fuel Composition: Actinide Burning In An Inert Matrix (g/bundle)

Isotope	Actinide-SiC (Inert Matrix)		Net Destruction Efficiency (%)
	0 MW•d/kg	582 MW•d/kg	
^{238}Pu	5.9	5.4	8.5
^{239}Pu	205.2	8.3	96.0
^{240}Pu	95.8	56.7	40.8
^{241}Pu	31.0	15.2	51.0
^{242}Pu	18.5	44.4	---
Total Pu	*356.4*	*130*	*63.5*
^{237}Np	19.8	9.6	51.5
^{241}Am	20.3	2.4	88.2
^{243}Am	3.5	8.9	---
^{242}Cm	0	5.7	---
^{244}Cm	0	3.4	---
Total	*400.0*	*160.0*	*60.0*

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