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ATOMIC ENERGY
OF CANADA LIMITED

ÉNERGIE ATOMIQUE
DU CANADA LIMITÉE

THE RELATIONSHIP BETWEEN NATURAL URANIUM AND ADVANCED FUEL CYCLES IN CANDU REACTORS

RELATION ENTRE L'URANIUM NATUREL ET LES CYCLES DES COMBUSTIBLES AVANCÉS DANS LES RÉACTEURS CANDU

A.D. LANE, F.N. McDONNELL and J. GRIFFITHS

International Symposium on Uranium and Electricity
(The Complete Nuclear Fuel Cycle)
Saskatoon, Saskatchewan 1988 September 18-21

Chalk River Nuclear Laboratories

Laboratoires nucléaires de Chalk River

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RÉSUMÉ

CANDU est le type de réacteur de puissance thermique le plus économique quant à l'uranium et est le seul utilisé au Canada. Les réacteurs CANDU consomment à peu près 15% de la production d'uranium du Canada et soutiennent une industrie d'exploitation du combustible évaluée à environ 250 millions $/an. En plus d'avoir un cycle du combustible d'uranium naturel à passage unique, les réacteurs CANDU peuvent fonctionner avec des cycles d'uranium légèrement enrichi (ULE), d'uranium-plutonium et de thorium, plus efficacement que les autres réacteurs. Actuellement, seul le cycle d'ULE est intéressant du point de vue écomique au Canada mais les autres cycles sont intéressants pour les pays sans ressources de combustible indigènes. Un programme est en cours pour établir les techniques de combustible nécessaires à l'utilisation du cycle d'ULE et des autres cycles du combustible dans les réacteurs CANDU.

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ABSTRACT

CANDU is the most uranium-economic type of thermal power reactor, and is the only type used in Canada. CANDU reactors consume approximately 15% of Canadian uranium production and support a fuel service industry valued at ~$250 M/a. In addition to their once-through, natural-uranium fuel cycle, CANDU reactors are capable of operating with slightly-enriched uranium (SEU), uranium-plutonium and thorium cycles, more efficiently than other reactors. Only SEU is economically attractive in Canada now, but the other cycles are of interest to countries without indigenous fuel resources. A program is underway to establish the fuel technologies necessary for the use of SEU and the other fuel cycles in CANDU reactors.

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BACKGROUND

All currently operating or planned nuclear-electric generating plants in Canada are of the CANDU type, and so the choice of fuel cycle for these plants will determine the use of fuel cycles in the domestic Canadian fuel industry for the foreseeable future, and consequently the impact they will have on the Canadian uranium industry. In Canada there are currently 18 CANDU reactors operating with a capacity of 11.8 GW(e) (1,2). Of these, 16 are operated by Ontario Hydro (capacity 10.5 GW(e)) and the remaining two are operated by Hydro Quebec and the New Brunswick Power Commission. There are four more reactors under construction by Ontario Hydro, that will provide an additional 3.5 GW(e) by 1992 (3), bringing the net generating capacity of CANDU reactors in Canada to 15.3 GW(e) by that date. All current CANDU reactors were designed for, and are still operating with, natural uranium fuel. Canadian requirements are estimated to be approximately 1700 tonnes of uranium in 1988, or approximately 15% of the Canadian production (4). A mature fuel industry has grown to service these reactors, with an annual product value of approximately $250 M per year focused on the supply of the uranium, its processing to UO_2 and manufacture into fuel bundles, plus the generation and application of the fuel technologies necessary for their optimal use, such as irradiation testing, behavioural modelling, fuel management and fuel handling.

There are six CANDU-type reactors currently operating outside of Canada (3 in India, and one each in Argentina, Korea and Pakistan), and eleven are under construction (6 in India and 5 in Romania) (1,2). Perhaps as importantly from the viewpoint of fuel cycles, there are a number of energy-resource-poor countries actively considering CANDU reactors for use with fuel cycles based on the spent fuel from existing light-water reactors (LWR), in order to maximize the energy they can extract from the uranium which must be imported. At least two of these countries (Japan and Korea) import significant quantities of uranium from Canada.

RATIONALE FOR FUEL CYCLES

The reason for using various resource-conserving fuel cycles in any reactor is to reduce the amount of uranium consumed by that reactor for each kW of electricity generated. This ability to reduce the amount of uranium required per unit of electricity produced is seen as an important hedge against potential uranium shortage and escalating price by utilities and planners in countries who must import their uranium. Having been vulnerable to external forces during the 1974 oil crisis, such countries now want technologies which will allow them to be more energy self-sufficient, even if they are more expensive. Even countries such as Canada, with large oil, coal and uranium resources, are not free from the price instability effects of events such as the 1974 oil crisis, since domestic prices are also forced up. It is worth noting that the price of uranium increased by a factor of approximately 5 during the late 1960's and early 1970's due to the pressure of rapid nuclear expansion during that period.

Uranium conserving fuel cycles have been extensively documented for both Light Water Reactors (LWR) (5-8), and CANDU reactors (8-15). Their importance to both the health and growth of the nuclear industry can be seen in Figure 1, where the projected uranium requirements (for both high and low growth scenarios) are compared with the projected uranium-plutonium capability from known uranium resources.

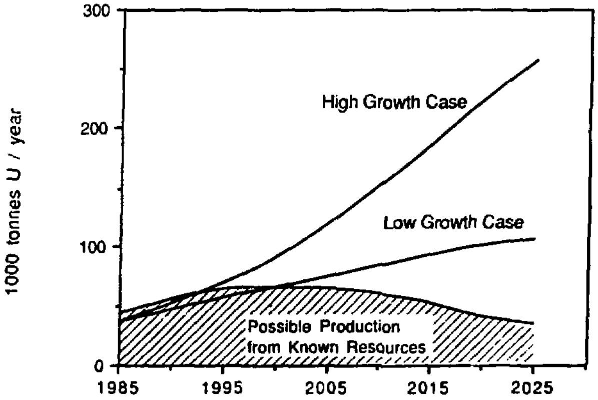

FIGURE 1: PROJECTED REACTOR REQUIREMENTS AND URANIUM PRODUCTION (CURRENT LWR ONCE-THROUGH STRATEGY).

The figure is based on 1986 OECD data for the world outside of centrally-planned economies (18), and uses information received from uranium-producing countries, plus countries with either programs or plans for producing electricity from nuclear reactors. The uranium consumption is based on the use of once-through cycles in LWR's (thus omitting

CANDU's, gas-cooled reactors and fast-breeder reactors), since LWR's represent ~80% of the operating power reactors (1). The uranium production capability is based on currently-known resources in existing, committed, planned and prospective production centers, which includes uranium recoverable at up to $130 US/kg in the "reasonably assured" and "estimated additional" resources categories in the NEA/IAEA resource classification system (18).

The CANDU reactor, because of its neutron-economic design and flexible, on-line fuelling capability, is ideally suited to operate with a wide range of fuel cycles. Not only can it operate on cycles ranging from natural uranium through enriched uranium, recycled plutonium, to various thorium cycles, but can do so with greater efficiency in fuel use than any other type of thermal reactor (i.e. excluding the complex and expensive fast-breeder reactors).

R&D programs to investigate the economics and technical feasibility of using advanced-fuel cycles in CANDU reactors, as well as to develop the key supporting technologies for them, have been pursued over the past 20 years (9,13,17). While the burnup and fuel-conserving advantages of advanced cycles have been clear in these studies, the economics have been uncertain for all cycles except enriched uranium. This is because the extra processing and complex fabrication required for each of the advanced fuels introduces significant extra costs, and these costs have been rising for key steps such as reprocessing. The economics are dependent on the difference between the extra cost for advanced fuels, and the cost for uranium. As the cost of uranium goes up, the advanced cycles become more attractive, and since the market price for uranium has moved both up and down significantly during this period, meaningful projection of when advanced fuel cycles will become economic has been difficult, other than that it will probably require uranium prices several times more expensive than current levels, possibly as high as $500/kg U (10).

However, while the use of advanced fuel cycles beyond enriched uranium does not appear to be economic for the foreseeable future in CANDU reactors in Canada, it is worth noting that uranium-plutonium fuel cycles are being introduced into existing PWR reactors in France on a commercial basis, and the use of such fuels are being actively studied in Japan for use in CANDU reactors. Recently, it has also become clear that the cost of transporting and disposing of spent fuel and other high-level wastes is increasing because of environmental concerns, and has thus become another important factor in the economic attraction of fuel cycles which increase burnup and reduce spent-fuel volume, particularly in highly-populated countries where such disposal is both difficult and expensive.

DESCRIPTION OF FUEL CYCLES

The various fuel cycles applicable to the CANDU reactor are shown in Figures 2 through 4 and 6. Figure 2 depicts the once-through, natural-uranium fuel cycle which is the basis for the design of the CANDU reactors, and has been used exclusively in these reactors up until now. The fuel fabrication step shown in Figure 2 includes the conversion of the uranium concentrate (or yellowcake) into nuclear-grade sinterable UO_2 powder, its fabrication into high density UO_2 pellets, the incorporation of these pellets into 50-cm-long Zircaloy-clad fuel elements, and the assembly of the fuel elements into CANDU fuel bundles. These bundles can have differing numbers of elements, depending on the reactor into which they are loaded, typically 28 or 37.

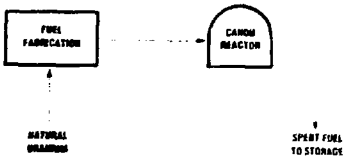

FIGURE 2: ONCE-THROUGH NATURAL-URANIUM FUEL CYCLE.

Figure 3 shows the once-through, enriched-uranium fuel cycle. This is the cycle commonly used in most power reactors other than the CANDU type, and involves an enrichment step prior to fabrication of the fuel assemblies or bundles. In this case the uranium concentrate is converted into the gaseous compound uranium hexaflouride (or HEX) to enable enrichment in the concentration of the isotope U-235 by means of either the gaseous diffusion or centrifuge process. Following enrichment to the desired level, the HEX is converted to ceramic-grade UO_2 powder for the fabrication process.

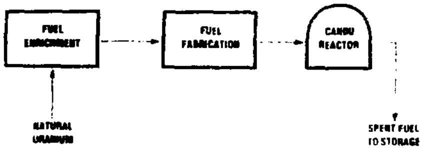

FIGURE 3: ONCE-THROUGH, SLIGHTLY-ENRICHED URANIUM (SEU) FUEL CYCLE.

Figure 4 shows the major steps in the uranium-plutonium fuel as it would be used in either the LWR or CANDU reactors. The fuel fabricated for this fuel cycle is normally referred to as mixed-oxide (or MOX) fuel because it is a mixture of uranium and plutonium oxides in the form of UO_2 and PuO_2. Plutonium is created in all power reactor fuel during its irradiation from neutron capture in the U-238 that it contains, and so spent power reactor fuel can be reprocessed to chemically separate this plutonium. The fabrication of fuel for the uranium-

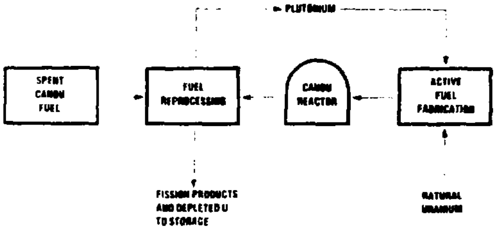

FIGURE 4: URANIUM-PLUTONIUM FUEL CYCLE.

plutonium cycle thus starts with the reprocessing of previously-irradiated spent fuel to separate the plutonium. This Pu can then be blended with natural or depleted U, either in the form of liquid nitrate solutions, or in the form of oxide powders. If liquid nitrate solutions are blended, homogeneous $(U,Pu)O_2$ can be obtained, whereas when dry powders are blended the degree of homogeneity is dependent on the particle size of the powders involved. The resultant $(U,Pu)O_2$ powder is then fabricated into fuel assemblies as described previously, except that because Pu is a very α-toxic carcinogen, all of the fabrication operations must be undertaken inside sealed, sub-atmospheric glove boxes (similar to the one shown in Figure 5), to the stage where the Pu-containing pellets are sealed within fuel elements. With MOX fuel it is the blending step which establishes the fissile loading or "enrichment" of the resultant fuel.

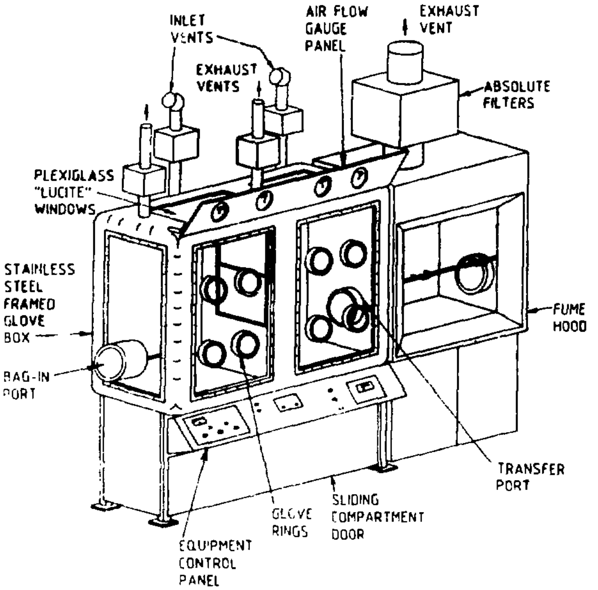

FIGURE 5: GLOVE BOX AND FUME HOOD USED FOR FABRICATION OF ALPHA TOXIC MOX FUEL.

The thorium fuel cycle is by far the most complex of the fuel cycles, and its main elements are shown in Figure 6. Unlike uranium, there is no naturally-occuring fissile isotope of thorium, and so all of the necessary fissile material must be added. Plutonium, as separated from spent uranium fuel, is probably the fissile material that would be used, although uranium which has been highly enriched in U-235 could also be used. The fabrication of fuel for the thorium cycle thus normally starts with spent fuel which is reprocessed to separate Pu, which is then blended with thorium to fabricate $(Th,Pu)O_2$ fuel as described above. Because of the Pu, this fuel must again be fabricated in a glove-box facility. Following irradiation, the spent fuel will contain U-233 from neutron capture in the Th-232, according to the reaction described in Equation 1.

$$\text{Th-232} + n \longrightarrow \text{Pa-233} \xrightarrow{\beta} \text{U-233} \qquad \text{(Equation 1)}$$

The major rationale for this fuel cycle is the U-233 which it produces. U-233 has by far the greatest yield of neutrons of any of the fissile isotopes when undergoing fission by well-thermalized neutrons - as is the case in a CANDU reactor. Although some of this U-233 is fissioned in the original fuel as it is irradiated, there is a considerable quantity of U-233 left in the fuel when it is discharged. Therefore, to optimize the cycle, the U-233 in the spent fuel must be recycled with fresh thorium and some Pu "topping". This requires a special facility for reprocessing thorium fuel when it has had the U-233 added to it, because of the high γ fields that are associated with the U-233. These additional special facilities, for reprocessing and fabricating the recycled U-233 containing thorium fuel, tend to make the reprocessing and fabrication portions of this cycle very expensive, and thus make the cycle less economic. However, the cycle does offer the opportunity for near breeding or uranium self-sufficiency.

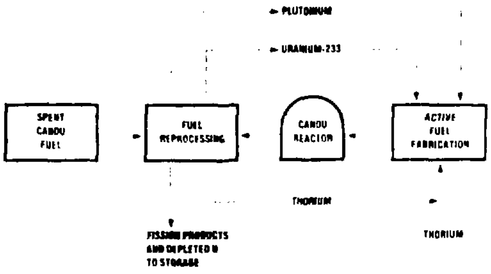

FIGURE 6: THORIUM FUEL CYCLE

FUEL CYCLES IN CANDU

As can be seen in Table 1, the uranium savings possible range from 30%, by the addition of slight enrichment to the present once-through cycle, to approaching 100% (self-sufficiency) within a low burnup, plutonium-topped thorium cycle. In Canada, the choice of what cycle is used at what time and under what circumstances, will depend upon a number of factors, such as rate of load growth, rate of uranium export and new discoveries, uranium price, and the cost and availability of the fuel reprocessing and fabrication services, but could be quite different in some other countries. The order in which the cycles are listed in Table 1 corresponds to the degree of technical difficulty involved in

TABLE 1: COMPARATIVE URANIUM USE FOR POTENTIAL CANDU FUEL CYCLES (AT EQUILIBRIUM)

	URANIUM SAVINGS (%)
NATURAL URANIUM, ONCE-THROUGH	0
SLIGHTLY-ENRICHED URANIUM (1.2%) ONCE-THROUGH	30
URANIUM-PLUTONIUM, RECYCLE	50-60
URANIUM-THORIUM, RECYCLE WITH PLUTONIUM TOPPING	
- HIGH BURNUP	70-75
- LOW BURNUP	75-100

implementing these cycles, their probable cost, and thus the probable sequence for introducing them in Canada (17). This is the inverse of the order of resource conservation. The salient features of these cycles in CANDU are outlined from a fuel viewpoint in the following paragraphs.

Natural Uranium

Since this is the standard fuel cycle in CANDU reactors, and is described in most publications on CANDU, it will not be covered here.

Slightly-Enriched Uranium

The use of slightly-enriched uranium (SEU), with enrichments ranging from 0.9 to ~1.5 wt% U-235 in total U, is the first fuel cycle that will be employed in CANDU reactors, and probably the only one in Canadian CANDU's within a 20-year planning horizon. This is because it is clearly economic at current price levels, and offers the incentive to become more so as enrichment price levels fall due to an industry over capacity, and the introduction of new, potentially lower-cost enrichment technologies such as the laser based AVLIS (Atomic Vapour Laser Isotope Separation) process (15,19,20).

For example, recent estimates of the potential savings that could be achieved through the use of SEU fuel in existing CANDU reactors range from about $5 M/a per reactor of approximately 750 MWe size, using 1.2 wt% U-235, to a present worth of about $800 M from all Ontario Hydro nuclear stations over the next 30 years using 0.9% SEU (21). These cost savings come from both the fuel-supply cost and the "back end" storage, transportation and disposal cost. It is worth noting that the "back end" component of the cost saving is due to the reduction in the volume of the spent fuel by a factor of two-three, and that if the cost of disposal rises above current estimates (particularly in countries with high population densities where the identification of areas for nuclear-waste disposal is a difficult political problem) then the advantage of SEU use is increased even further.

There is currently an excess capacity in the uranium-enrichment industry, resulting from the sharp decrease in the anticipated growth rate in the nuclear power business, principally in the USA. This excess capacity will be increased with the introduction of the new lower cost enrichment technologies - currently the improvements to the gas centrifuge, and in the future the introduction of the AVLIS or other laser-based technologies. The American AVLIS process is expected to be in commercial operation during the 1990's. As a result of the present and future over-capacity, the price of enrichment (or more correctly, separative work units) is currently depressed, and is expected to stay depressed, or possibly fall lower, between now and the end of the century. This situation provides a competitive benefit to those reactors that use enriched uranium, and it is desirable to have CANDU reactors similarly benefit from such low prices.

The SEU cycle will also establish most of the key reactor operating technologies, in-core fuel management and fuel design and behaviour-prediction technologies that will be necessary for the introduction of any other fuel cycles into CANDU which involve larger fissile loadings, and significantly higher fuel-burnups than in the current natural-uranium-fuelled reactors.

The use of SEU fuel, with the extra fuel reactivity and burnup that are available from its use, provides an extra degree of flexibility in the design of new CANDU reactors, by allowing the very strict neutron economy in CANDU reactors to be traded off against other economic benefits such as lower capital cost, or extended pressure-tube life. One particularly attractive economic benefit is the use of more than one enrichment level to flatten the power profile across the core of the reactor, thus allowing up to 19% more power to be generated from the same size of reactor core, without increasing maximum channel powers or fuel ratings, thus lowering the specific capital cost of a reactor using this feature (22).

In order to demonstrate that the fuel defect rate is maintained at less than 1% (the reference level for natural U CANDU fuel) for the higher fuel burnups available with SEU, and with the changes in fuel-power level likely to be associated with both fuel management and load following, significant irradiation testing will be required. The objective of such testing will be to establish an appropriate fuel-behaviour data base at the higher burnups during both steady operation and with a range of power ramps, plus a large-scale demonstration of operation within a power reactor. This testing program could be done using the current 37-element bundle common to most recent CANDU reactors. However, increasing the burnup by a factor of three, plus a significant degree of power manoeuvring associated with load following and fuel management, could probably increase the defect rate of the fuel, and extensive testing would be required to confirm the fuel's performance. Another option is to reduce the fuel rating by increasing the number of pins in the bundle, which would remove the fuel's performance further from the area of uncertain performance and reduce the amount of radiation testing required. This latter concept has given rise to the CANFLEX 43-element bundle, which is described elsewhere, along with its development program (15,20).

A variation on SEU which is economically attractive, and could find its way into CANDU reactors once SEU technology has become firmly established, is a group of cycles that would be based on utilizing the spent fuel from light-water reactors. The spent fuel from these reactors contains significant fissile material in the form of both U-235 and plutonium. Significant quantities of this fuel are being reprocessed now for various strategic and political reasons. The recovered U-235 from this fuel has an enrichment range of 0.7 - .95%, which makes this recovered enriched uranium (REU) a potentially cheaper alternative to SEU in CANDU reactors where strategic considerations are favourable. However, it is the technical problems associated with the other uranium isotopes, plus the traces of fission products which contaminate the REU, which cause most concern regarding the feasibility of its use.

To put this into perspective, the uranium isotopic composition in REU is compared with normal, natural and enriched uranium in Table 2 (8,11). Analysis has shown that the radiological problem with the enhanced concentration of U-232 is small and will not have a significant impact on fabrication or handling. Although the effects of the enhanced concentrations of U-236 are significant in LWR reactors, necessitating the use of additional U-235, the softer spectrum in CANDU reactors reduces this effect by a factor of approximately seven.

TABLE 2: TYPICAL URANIUM COMPOSITIONS

ISOTOPE	% COMPOSITIONS		
	NATURAL	ENRICHED	REU
U-232	0	0	1.1×10^{-7}
U-234	0.0055	0.03	0.02
U-235	0.72	3.25	0.91
U-236	0	0	0.42
U-238	99.275	96.72	98.65

Uranium-Plutonium

Once the SEU fuel cycle has been established in CANDU reactors, along with all of the operating technologies for higher fuel-burnups and the fuel-management technologies for higher fissile loadings, the next logical step in resource conservation involves the reprocessing of spent fuel, and the recycling of the recovered plutonium back in the form of a uranium-plutonium (MOX) fuel. The fuel contribution to the unit energy cost when MOX fuels are used is currently higher than the cost with natural uranium, and significantly higher than with SEU because the cost of plutonium is higher than that of U-235 in the form of SEU, and the fabrication cost of fuel containing Pu is significantly higher than that for natural U or SEU. Therefore, MOX cycles are unlikely to be economic in Canada until the price of uranium increases to well above $300 CDN/kg U. Given the fact that there are neither reprocessing facilities, nor a program to develop them in Canada, it is most likely that the first use of the U-Pu cycle in CANDU reactors would be outside of Canada, using plutonium from a source other than CANDU fuel. Because of the neutron efficiency of CANDU reactors the residual fissile plutonium in discharged CANDU fuel is about 2.8 g/kg, or a factor of two and one half less than that in the discharged fuel from LWR reactors operating on a normal non-extended-burnup cycle. Since the cost of reprocessing is dependent on the mass of fuel that must be reprocessed, the cost of Pu from spent LWR fuel is clearly much less. Although the fissile Pu contained in spent SEU fuel would be about 25% higher than in natural fuel, the extended-burnup cycles being introduced into LWR's will also increase the content of the Pu in their discharged fuel, so that LWR plutonium will always be cheaper.

The lower cost of LWR Pu gives rise to various tandem cycles with LWR's where CANDU reactors can burn reprocessed LWR fuel down to very low levels of residual fissile material, thus not requiring multiple reprocessing. This feature is attractive to countries that already have LWR reactors and a reprocessing capability, such as Japan.

Thorium

Thorium fuel cycles are uniquely advantageous for CANDU reactors, offering the potential for near breeding of fissile material, or self-sufficiency in uranium. However, their technical difficulty and high cost mean that it is unlikely that such cycles would be introduced before plutonium recycle. In fact, a plutonium fuel capability is required for the startup fissile inventory for thorium cycles unless more valuable U-235 were used. Even though Canada has large quantities of thorium sitting in tailings ponds from the production of uranium from ores in the Elliot Lake area, it is unlikely that the use of thorium cycles could become economic in Canada until well into the next century. Although these cycles do not have any significant near-term effect on the present natural-uranium fuel industry, their long-term impact on the viability of the CANDU reactor concept is of particular importance, because of the drastic reduction in uranium requirements which they allow.

CONCLUSION

Although natural uranium is currently used in all CANDU reactors, slight enrichment has already become significantly more economic, and will establish the operating technologies necessary for the use of other cycles which are expected to become economic early in the next century. For the CANDU reactor concept to survive in both Canada and abroad, it must remain competitive through the use of advanced fuel cycles where and when they are economically attractive. This ability is already a requirement for the use of CANDU in many countries that depend upon imported uranium. The ultimate incentive is that if nuclear fission is to remain a major energy source through the next century, it can only do so with the efficient recycle of fissile material in advanced fuel cycles and breeding.

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