INTERNATIONAL CONFERENCE
ON NUCLEAR POWER AND ITS FUEL CYCLE
SALZBURG, AUSTRIA • 2–13 MAY 1977

INTERNATIONAL ATOMIC ENERGY AGENCY

IAEA-CN-36/402

EVALUATION OF PLUTONIUM, URANIUM, AND THORIUM USE IN POWER REACTOR FUEL CYCLES*

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ABSTRACT

The increased cost of uranium and separative work has increased the attractiveness of plutonium use in both uranium and thorium fuel cycles in thermal reactors. A technology, fuel utilization, and economic evaluation is given for uranium and thorium fuel cycles in various reactor types, along with the use of plutonium and ^{233}U. Reactors considered are LWRs, HWRs, HTGRs, and breeder reactors. Key technical factors are fuel irradiation performance and associated physical property values. Key economic factors are unit costs for fuel fabrication and reprocessing and for refabrication of recycle fuels; consistent cost estimates are used, given that each of the steps involved in such use can be effectively safeguarded.

In thermal reactors, the irradiation performance of ceramic fuels appears to be satisfactory, with attainable exposures corresponding to those economically desirable. The uranium cycle is the reference cycle in water-cooled reactors, with plutonium recycle planned. The unit costs of recovering plutonium appear less from LWRs than from natural uranium HWRs; further, use of LWR product permits plutonium-thorium fueling to compete with uranium and uranium-plutonium cycles. Converting uranium cycles to thorium cycles increases the energy that can be extracted from a given uranium resource. Thus, fuel utilization can be improved by fueling all thermal reactors with thorium, but this requires highly

*Research sponsored by the Energy Research and Development Administration under contract with Union Carbide Corporation.

enriched uranium; use of ^{235}U with thorium is most economic in HTGRs, with advantageous fuel utilization performance. Marked improvement in long-term fuel utilization can be obtained through high thorium loadings and short fuel irradiations, but with significant economic penalties.

In fast reactors, use of the plutonium-uranium cycle gives advantageous fuel resource utilization; the thorium cycle provides more negative core reactivity coefficients and more flexibility in use of recycle fuels containing uranium of less than 20% ^{233}U. Metallic fuels appear to be advantageous when based on thorium, but additional irradiation performance testing is needed.

1. INTRODUCTION

This paper examines the basic technology performance of urania, plutonia, and thoria fuels, and evaluates the power costs and fuel utilization performance of uranium and thorium fuel cycles in LWRs, HWRs, and HTGRs.

By far the most experience has been obtained with uranium oxide fuels, since both LWRs and HWRs use them. However, thoria along with urania also can be utilized as the fuel material; also, plutonium produced from the first uranium cycle can be recycled with either urania or thoria. While reactors initially fueled with uranium will, with time, also contain some plutonium, there is more flexibility in the plutonium content of recycle fuel. The basic regulatory decision on plutonium recycle has not yet been made in the U.S. and, as a matter of policy, such reprocessing, separation, and recycling of plutonium will not be permitted in the U.S. unless these activities are necessary and can be accomplished in a manner consistent with the U.S. national and international nonproliferation objectives. The technical discussions which follow should be understood in this context.

While the most prevalent commercial reactors are LWRs and HWRs, other thermal reactors being developed are the High-Temperature Gas-Cooled Reactor (HTGR) and the Light Water Breeder Reactor (LWBR). Fuels considered for HTGRs include oxides, carbides, and mixed oxide-carbide, while the fuel for the LWBR consists of thorium and uranium oxides. Pelleted fuels are of most interest to the water-cooled reactors, while pyrolytic-carbon-coated particles of oxides and carbides are of interest to HTGRs. Fuel enrichments and irradiation conditions associated with fuels in the different reactor types can differ significantly with reactor operating conditions (environment, pressure, temperature, and fuel exposure).

2. FUEL PERFORMANCE IN WATER-COOLED REACTORS (LWRs AND HWRs)

2.1 Urania Fuel

The performance of uranium oxide pellets in water-cooled reactors depends upon a number of factors, and extensive knowledge and technology have been developed to improve the irradiation behavior of such fuels. While problems have been encountered in fuel performance, they have presently been eliminated/alleviated to the extent needed. Factors, often interrelated, that have contributed to fuel failures include (1) densification and ratchetting of fuel, (2) stress-corrosion cracking [SCC] of Zircaloy, (3) influence of fission products, (4) excessive moisture in fuel and hydriding of cladding, (5) pellet-cladding interactions, (6) cladding embrittlement at high fluences, and (7) water corrosion of the Zircaloy. No distinction is made here between urania performance in LWRs and HWRs, since the general performance features are similar; however, it should be remembered that HWRs use

natural uranium fuel and have relatively low average fuel exposures relative to LWRs (7500 vs 30,000 MWd/t). Thus, those factors relating to fuel exposure should be considered when specifically comparing the technology performance of LWR and HWR fuels.

Fuel densification and fuel ratchetting can lead to cladding collapse and subsequent cladding failure. Densification occurs during initial reactor irradiation and was largest for the "early" oxide fuels having unstable microstructures. However, in-reactor densification can be reduced substantially by increasing the initial fuel density and/or by using improved fabrication methods leading to large pores and stable fuel microstructures [1,2,3]. Further, prepressurization of fuel rods or improving and strengthening the cladding can avoid cladding collapse over the fuel lifetime [4,5], so densification and/or fuel ratchetting caused by power cycling need not cause fuel rod failures.

Stress-corrosion cracking of Zircaloy takes place under the influence of stress and strain and in the presence of certain fission products such as iodine [6,7]. Since the concentration of fission products increases with time, SCC is more likely at the higher fuel burnups, although it can occur as low as 2000 MWd/t [7]. By its nature, SCC is influenced by power ramping, cladding embrittlement, and stress concentrations due to fuel chips. These effects can be decreased by using stronger, more ductile cladding. Moisture inhibits SCC [6] but is undesirable because it causes hydriding of the cladding.

Fission products are generated during reactor exposure and lead to fuel expansion, generation of fission gas pressures, and stresses in the cladding. Also, certain fission products along with excess oxygen generated during the fission process corrode and thin the cladding. Surface reactions are particularly linked to cesium and tend to be small for fuel exposures up to about 10,000 MWd/t [8]. Increasing fuel burnups and temperatures increases the chemical attack on the cladding. The influence of fission products on fuel rod failure can be controlled by proper attention to rod design and operating conditions and by proper control of materials and fabrication methods.

During the early production of water-reactor fuel, excessive moisture was present, leading to hydriding of the inner surface of the cladding and weakening it mechanically. As a result, hydriding caused failures early in fuel life (0–11,000 MWd/t exposure)[9,10]. Present fabrication processes have eliminated the problem of excessive moisture in the fuel [3,9–11].

Pellet-cladding interactions are caused by fuel expansion due to exposure and associated generation of solid and gaseous fission products, and by pellet cracking and the wedging of fuel chips between the main pellet and the cladding along with the imposition of thermal gradients. Pellet-cladding interactions are also influenced by fuel restructuring and by the peak heat ratings of the fuel pins; they are more pronounced with increasing fuel burnup and increasing density of the initial fuel. Pellet-cladding interactions occur later in fuel life (exposures of ~6–30,000 MWd/t) [9] and also can be a function of power level, power changes, fuel rod design characteristics, and manufacturing variables.

Damage by high-energy neutrons embrittles cladding and enhances susceptibility of Zircaloy to SCC by iodine [6] at fluences above 10^{20} neutrons/cm^2 (>1 MeV). Rod failures from this can be controlled by proper cladding production processes and methods, and by keeping cladding stresses low.

Corrosion and hydrogen pickup on the outer surface of the cladding weakens it; they can be maintained at low levels by careful water chemistry and coolant flow control [3].

Combination of the above factors, such as fuel densification and internal hydriding of the cladding, tends to accelerate rod failures. Further, other factors can have an influence, such as axial rod bowing due to irradiation conditions and weakening of the cladding due to fretting corrosion caused by high local coolant velocities [10]. In general, since cladding stresses and strains can eventually lead to fuel rod failure, steps taken to reduce them will tend to improve fuel rod performance. Cladding stresses can be reduced by restricting reactor operating conditions so as to avoid large and rapid power changes.

Overall, through prevention of fuel rod manufacturing defects and deficiences through careful quality control, by improving materials and fuel rod designs, and by careful control of reactor operating procedures and power distribution, satisfactory fuel rod performance in power reactors has been obtained [3,9,11–13]. Average fuel exposures in excess of 30,000 MWd/t are technically attainable.

2.2 Recycle Fuel

Recycle of plutonium in water reactors is considered to take place with use of UO_2-PuO_2 mixed-oxide (MOX) fuels. Irradiation of such fuels has confirmed that MOX fuels can have good irradiation performance [9]. By paying special attention to fuel rod fabrication methods, exposing MOX fuel to low power densities, operating the reactor in moderate load-following modes, and giving attention to water coolant chemistry control, excellent performance has been obtained with prototype MOX assemblies. There have been many tests of MOX fuel in a number of reactors at various fuel exposures and linear heat ratings, as well as under transient conditions; results indicate that the irradiation performance of MOX fuel in water reactors is satisfactory [9,14–18] such that MOX fuel should perform about the same as UO_2 from a licensing viewpoint of reactor operations. However, irradiation testing to high fuel exposures is limited, and the chemical attack of cladding tends to be higher with MOX fuels than with urania unless the oxygen-to-plutonium ratio of the fuel is initially low [19].

2.3 Vibratorily Compacted Fuel Particles

Another oxide fuel type of interest is vibratorily compacted (vi-pac) fuel; fuel microspheres vibrated to reasonably high fuel densities have given satisfactory irradiation performance, and in general, the use of small-particle fuels tends to reduce interactions between fuel and cladding. Also, the performance of vi-pac MOX fuel particles appears to be about the same as for pelleted MOX fuel [15], although vi-pac fuel tends to release higher amounts of fission products [16].

3. PERFORMANCE OF THORIUM FUELS IN THERMAL REACTORS

3.1 Water-Cooled Reactors

Thoria is of interest as a reactor fuel in water-cooled reactors, and its performance has been studied in a number of irradiation experiments [20,21]. Thoria-urania fuels (pelleted and vi-pac) perform well at burnups up to 80,000 MWd/t.

More recently, extensive irradiation experience with zircaloy-clad thoria and thoria-urania pellets has been obtained in the LWBR Program [22] with much effort on determining the performance of the cladding. For LWBRs and their prebreeders (average fuel exposures of about 10,000 and 30,000 MWd/t, respectively), the irradiation performance of the fuel should be satisfactory.

Limited irradiation experience [21] indicates that PuO_2-ThO_2 fuels should perform about as satisfactorily as UO_2-ThO_2. The higher thermal conductivity and melting temperature of thoria relative to urania should be beneficial to thoria use [23].

3.2 High-Temperature Gas-Cooled Reactors

High-Temperature Gas-Cooled Reactors use coated microspheres as fuels [24,25]; failure of these fuel particles is associated with failure of the coating layers. Several mechanisms are associated with coating failure; these include (1) transport of fission products through intact coatings, (2) mechanical failure of coatings, (3) fuel transport through the coatings, and (4) fission product attack of the coatings.

Fission products such as cesium and strontium can be transported by diffusion through the pyrolytic carbon (PyC) coatings; such transport can be reduced by limiting the fuel temperature, using oxide kernels, adding Al_2O_3 and SiO_2 to the kernels to form stable fission product compounds, forming PyC coatings under proper gaseous decomposition conditions, or adding a SiC layer.

Mechanical failure of coatings can arise from excessive pressure within the fuel kernel and from excessive stresses in coatings due to irradiation shrinkage of anisotropic coatings and of the matrix structure containing the fuel. These failures can be controlled by proper fuel design and by careful control of materials and fabrication processes.

Fuel transport through coatings occurs most severely in oxide fuels; it can be controlled by limiting temperature gradients across fuel particles, limiting maximum fuel temperatures, diluting fissile kernels with ThO_2, and incorporating UC_2 in UO_2 kernels.

Fission product attack of SiC coatings occurs in fissile particles of UC_2; it can be controlled by addition of UO_2 to UC_2.

Overall, the performance of HTGR fuel elements under reactor conditions appears to be satisfactory according to results from reactor experiments and from research and development programs [24–26], if proper attention and quality control are given to fabrication methods and processes. Fertile particle exposures in excess of 10% burnup and fissile particle exposures of 80% burnup appear technically attainable under reactor conditions, permitting average reactor fuel exposures in excess of 100,000 MWd/t. Also, limited irradition experience with coated plutonia microspheres shows that oxide fuels should perform satisfactorily to high exposures if the initial oxygen-to-metal ratio in the plutonia particles is low.

4. THORIUM FUELS IN FAST BREEDER REACTORS (FBRs)

In FBRs, use of thorium or thorium-uranium fuel cycles provides a more negative void coefficient of reactivity in the core than does use of the uranium fuel cycle; however, use of such cycles with oxide fuels in FBRs leads to lower breeding ratios than does use of the uranium cycle.

Relative to metallic fuels, thorium-base alloys appear more practical [27] than uranium alloys; use of thorium metals leads to breeding ratios comparable with those obtained with urania-plutonia fuels [28]. However, much more development and analysis are required before utilization of thorium alloys can become a reality.

A potentially significant feature of thorium fuel cycles in FBRs is related to safeguard aspects. Developing a mixed uranium-thorium fuel cycle permits the "denaturing" of recycle fissile fuels since ^{233}U can be diluted with ^{238}U.

5. ECONOMIC AND FUEL UTILIZATION PERFORMANCE OF VARIOUS FUELS IN THERMAL REACTORS

Thermal reactors of the LWR, HWR, and HTGR types are considered here, with operation on uranium, thorium, or mixed fuel cycles. Results are obtained with regard to the relative energy extraction from a given ore resource under a specified nuclear power growth scenario, along with associated power costs and related economic benefits or penalties, treating the prices of uranium ore and separative work as parameters [29].

The basic nuclear power growth scenario considered a power capacity growth starting in 1970 of 15 GW(e)/year up to a level of 450 GW(e) in the year 2000. After the power level reached 450 GW(e), it was maintained at that level until reduction was necessary because of limitations in U_3O_8 resources. The ore resource base was considered to be either 2.5 million or 3.5 million tons of U_3O_8.

In these studies [29], reference LWRs operating on the uranium cycle were used initially; these were termed LWR_1s. New reactors were built after the year 2000; the new reactors were either additional LWRs operating on the uranium cycle (termed LWR_2s), LWRs operating on the thorium cycle [LWR(Th)s], HWRs operating on the thorium cycle [HWR(Th)s], or HTGRs operating on the thorium cycle. After the year 2000, LWR_1s were withdrawn from use as their 30-year lifetime was attained and replaced with a second type reactor as indicated above. The power capacity was maintained at 450 GW(e) for a period of time t_e defined as the time of extension associated with maintaining the power capacity at 450 GW(e). After time t_e, no new reactors were built, and those in use operated until the end of their 30-year lifetime.

The above power growth scenario, along with the estimated lifetime U_3O_8 requirements of the various reactors, was used to calculate the energy that can be generated as a function of new reactor use. In doing this calculation, a given reactor was considered to operate for 21 full-power years over its lifetime, and the uranium tails from enrichment plants corresponded to 0.2% ^{235}U. Further, it was assumed that 1200 t of fissile plutonium generated by LWR_1s was stored for fast breeder reactor use; that amount of plutonium permits a significant breeder economy to develop.

As indicated above, LWR_2s represent a continued construction of LWR_1s, but are specifically identified to clarify results. The energy generated by LWR_1s plus LWR_2s is the reference energy generation based on the use of LWRs with uranium and plutonium recycle, in which the entire ore resource is utilized (except for the plutonium stored for FBRs). The energy generation when the new reactors introduced after the year 2000 were either LWR(Th)s, HWR(Th)s, or HTGRs was also calculated and compared with the reference energy generation. (The conversion ratios of the reactors were those estimated to give the most economic performance.) The results obtained [29] are given in Table I.

Table I. Relative Energy Generation and Extension Time for Assumed Power Growth Scenario as a Function of Reactor Use and U_3O_8 Resource

Reactor Use	Fuel Conversion Ratio for Second Reactor	Relative Energy Generation (REG) and Extension Time (t_e) for Two U_3O_8 Resource Levels			
		2.5×10^6 tons		3.5×10^6 tons	
		REG	t_e, years	REG	t_e, years
LWR_1 + LWR_2	0.60	1	8.6	1	25
LWR_1 + LWR(Th)	0.70	1.12	13.4	1.16	34
LWR_1 + HTGR	0.66	1.10	12.5	1.20	37
LWR_1 + HTGR	0.82	1.20	16.5	1.41	48
LWR_1 + HWR(Th)	0.82	1.15	14.7	1.31	43

The higher the value of t_e, the longer the time available for FBR development without a closeout of nuclear power generation. For the cases studied, use of the thorium fuel cycle rather than the uranium cycle permits a significant increase in energy generation, even though thorium reactors are not introduced until the year 2000.

The economic performance of the various reactor types is given below, where a consistent evaluation is given of the relative power costs in the various thermal reactor concepts as a function of U_3O_8 costs and separative work costs. The fuel recycle cost factors employed are listed in Table II. Effective fuel storage costs are \$25/kg for HWR(natural U)s, \$100/kg for LWRs and HWR(Th)s, and \$400/kg for HTGRs. The cost of thoria is \$30/kg. The cost for recovered fissile plutonium considers only that cost associated with reprocessing the material. On the above bases, and assuming that the first reactor cycle always pays the equivalent of fuel storage costs toward fuel reprocessing, the cost to recover fissile plutonium is about \$20/g for LWRs and about \$50/g for natural uranium HWRs. The above effectively assumes that the first reactor fuel cycle obtains no credit for any fuel value of the product plutonium.

Additional power cost factors [29] are such that the sums of capital costs and operating and maintenance costs are 21.5 mills/kWh(e) for LWRs and HTGRs, 23.8 mills/kWh(e) for HWR(U)s (including D_2O costs), and 23.0 mills/kWh(e) for HWR(Th)s (including D_2O costs).

The resulting calculated power costs are summarized in Table III. The costs of the Pu/U and Pu/Th fueled systems consider that the plutonium cost is only that for its recovery, and that fertile uranium and thorium do not change in cost. Under these conditions, recycle of plutonium in thermal reactors appears more attractive with thorium than with uranium. However, such plutonium fueling will not have a significant influence on fuel utilization since only a fraction of reactors can be fueled with plutonium as the fissile fuel; further, such use of plutonium impacts its use in FBRs.

Table II. Fuel Cycle Cost Factors [29]

Reactor	Fuel Cycle	Cost Factor, $/kg		
		Fresh Fuel Fabrication	Fuel Reprocessing[a]	Recycle Fuel Refabrication
LWR	$^{235}U/^{238}U$	114	221	
	Pu/U		221	500
	$^{235}U/Th$	152	250	
	$^{233}U/Th$		250	570
	Pu/Th		260	510
HWR	Natural U	50	150	
	Enriched U	80	160	
	Pu/U		160	310
	$^{235}U/Th$	100	210	
	$^{233}U/Th$		210	390
	Pu/Th		220	320
HTGR	^{235}U	400	750	
	$^{233}U/Th$		750	1030
	$^{235}U/^{238}U$	360	730	
	Pu/Th		750	1030

[a]Inclusion of waste disposal charges will increase fuel cycle costs, with reactors having low fuel exposures being affected more than those with high fuel exposures.

Table III. Calculated Power Costs in Thermal Reactors

Reactor Type	Conversion Ratio	Fuel Cycle	Fuel Recycle	Power Cost, mills/kWh(e), for each U_3O_8 Cost (Separative Work Cost) in $/kg			
				55 (75)	88 (100)	220 (150)	660 (200)
LWR	0.60	U	no	26.1	27.8	33.7	~50
LWR	0.60	U	yes	26.7	28.0	32.0	41.2
LWR	0.60	U/Pu	Pu makeup	28.5	28.5	28.5	28.5
LWR	0.70	Th	yes	27.6	28.6	32.2	41.3
LWR	0.70	Th/Pu	Pu makeup	27.0	27.0	27.0	27.0
HWR	0.75	U	no	26.7	28.5	31.0	43.1
HWR	0.82	U/Pu	Pu makeup	30.0	30.0	30.0	30.0
HWR	0.82	Th	yes	27.7	28.6	31.6	40.0
HWR	0.90	Th/Pu	Pu makeup	28.5	28.5	28.5	28.5
HTGR	0.66	Th	no	25.2	26.4	30.1	40.0
HTGR	0.66	Th	yes	25.2	26.2	28.9	36.8
HTGR	0.82	Th	yes	26.2	27.0	29.3	35.6
HTGR	0.66	Th/Pu	Pu makeup	25.2	25.2	25.2	25.2
HTGR	0.82	Th/Pu	Pu makeup	26.3	26.3	26.3	26.3

Based on the above economic bases, a 7.5%/year discount factor, the assumed power growth scenario, and the energy generation possible with the various reactors, the economic benefits of the various systems were calculated relative to the power cost of the LWR (U/Pu recycle) system. Table IV gives the results for U_3O_8 costs of \$220/kg and separative work costs of \$150/kg SWU.

Table IV. Discounted Benefits of Various Reactors Relative to LWR (U/Pu Recycle) Systems

Reactor System	Discounted Benefits, $\$10^9$ for each U_3O_8 Resource (tons)	
	2.5×10^6	3.5×10^6
LWR_1 + LWR_2	reference value = 0	
LWR_1 + LWR(Th)	negative benefit	
LWR_1 + HTGR (0.66)	5.1	8.0
LWR_1 + HTGR (0.82)	6.4	8.7
LWR_1 + HWR(Th) (0.82)	1.1	1.5

According to the above, use of the thorium fuel cycle provides better U_3O_8 utilization, improved potential for long-term economics, and additional flexibility with regard to fuel recycle alternatives. It also provides additional power generation capability in case of delayed introduction of commercial FBRs. If the uncertainties regarding commercial introduction of the HTGR can be resolved favorably, the HTGR appears to offer the best combination of economics and fuel utilization with the thorium fuel cycle.

6. BREAK-EVEN THERMAL BREEDERS

Use of the thorium fuel cycle permits operation of LWRs, HWRs, and HTGRs as break-even breeders. The advantage of such breeders is that they permit long-term generation of energy for a limited fuel resource, using thermal reactors; however, thermal reactors operate more economically as converters. This is due [29] to the high fuel recycle costs associated with low fuel exposures and the need for producing ^{233}U breeder inventories by operation of "prebreeders." To build large numbers of break-even breeders, prebreeders need to operate with high net production of ^{233}U, which makes the prebreeders uneconomical; if prebreeders are limited to low values of net ^{233}U production to improve their economic performance, the number of break-even breeders that can be constructed is very limited. Economic ground rules that would improve the economic performance of break-even breeders and their prebreeders are: low effective charge rates for fuel inventory; low costs for fissile fuel; and low unit costs for fuel recycle.

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