AECL-7697

ATOMIC ENERGY OF CANADA LIMITED

L'ÉNERGIE ATOMIQUE DU CANADA LIMITÉE

IRRADIATION PERFORMANCE OF $(Th,U)O_2$ FUEL DESIGNED FOR ADVANCED CYCLE APPLICATIONS

Performance sous irradiation du combustible $(Th,U)O_2$ conçu pour applications en cycle avancé

I.J. HASTINGS, A. CELLI, M. ONOFREI and M.L. SWANSON

Paper presented at the Third Annual Conference of the Canadian Nuclear Society,
Toronto, Ontario, 1982 June 8-9

Chalk River Nuclear Laboratories

Laboratoires nucléaires de Chalk River

Chalk River, Ontario

June 1982 juin

ATOMIC ENERGY OF CANADA LIMITED

IRRADIATION PERFORMANCE OF $(Th,U)O_2$ FUEL DESIGNED FOR ADVANCED CYCLE APPLICATIONS

by

I.J. Hastings, A. Celli, M. Onofrei and M.L. Swanson

Presented at the Third Annual Conference
of the Canadian Nuclear Society,
Toronto, Ontario 1982 June 8-9

Fuel Materials Branch
Chalk River Nuclear Laboratories
Chalk River, Ontario K0J 1J0
1982 June

AECL-7697

L'ENERGIE ATOMIQUE DU CANADA, LIMITEE

Performance sous irradiation du combustible $(Th, U)O_2$ conçu pour applications en cycle avancé*

par

I.J. Hastings, A. Celli, M. Onofrei et M.L. Swanson

Résumé

Notre processus de fabrication de référence pour le combustible de cycle avancé à base de thorine est classique en ce sens qu'il produit des pastilles frittées et pressées à froid. Cependant, nous évaluons également actuellement d'autres combustibles qui offrent la possibilité d'une fabrication plus simple dans une installation éloignée et dans certains cas avec une performance améliorée avec taux de combustion élevé. Ces autres combustibles à base de thorine sont imprégnés, "spherepac" et extrudés. Le combustible "spherepac" a été irradié à une puissance linéaire de 50-60 kW/m jusqu'à environ 180 MW.h/kg H.E. Il s'est produit des défauts inexpliqués dans le combustible avec une gaine droite et une gaine pliante. Le combustible imprégné a fonctionné jusqu'à 650 MW.h/kg H.E. à 50-60 kW/m. Une expérience permettant d'examiner le combustible provenant du procédé d'extrusion sol-gel a atteint 450 MW.h/kg H.E. à une puissance linéaire maximale de 60 kW/m. Les deux dernières expériences ont eu lieu sans défaut et avec un dégagement de gaz de fission inférieur à celui de l'UO_2 dans des conditions identiques. Le combustible extrudé avait une géométrie de pastille semblable à celle des combustibles classiques et c'est la première démonstration pratique de l'EACL d'un combustible à base de thorine dont le composant fissile est distribué de façon homogène sur une échelle atomique. Nous continuerons de contrôler le combustible extrudé jusqu'à un taux de combustion proche de 1000 MW.h/kg H.E. comme indicateur pour la performance attendue du combustible $(Th, U)O_2$ co-précipité ou du $(Th, U)O_2$ mécaniquement mélangé avec une bonne homogénéité fissile.

*Présenté au troisième congrès annuel de la Société nucléaire canadienne, Toronto, Ontario, 8-9 juin 1982.

Laboratoires nucléaires de Chalk River
Chalk River, Ontario KOJ 1JO

Juin 1982

AECL-7697

IRRADIATION PERFORMANCE OF $(Th,U)O_2$ FUEL DESIGNED FOR ADVANCED CYCLE APPLICATIONS

I.J. HASTINGS[+], A. CELLI, M. ONOFREI* AND M.L. SWANSON*

Atomic Energy of Canada Limited - Research Company
Chalk River Nuclear Laboratories
Chalk River, Ontario KOJ 1JO

*Whiteshell Nuclear Research Establishment, Pinawa, Manitoba ROE 1LO

ABSTRACT

Our reference fabrication route for Advanced Cycle thoria-based fuel is conventional in that it produces cold-pressed and sintered pellets. However we are also evaluating alternative fuels which offer the potential for simpler fabrication in a remote facility, and in some cases improved high burnup performance. These alternatives are impregnated, spherepac, and extruded thoria-based fuels. Spherepac fuel has been irradiated at a linear power of 50-60 kW/m to about 180 MW.h/kg H.E. There have been unexplained defects in fuel with both free-standing and collapsible cladding. Impregnated fuel has operated to 650 MW.h/kg H.E. at 50-60 kW/m. An experiment examining fuel from the sol-gel extrusion process has reached 450 MW.h/kg H.E. at a maximum linear power of 60 kW/m. The latter two experiments have operated without defects and with fission gas release less than that for UO_2 under identical conditions. The extruded fuel has a pellet geometry similar to that for conventional fuel and is AECL's first practical demonstration of thoria-based fuel with the fissile component distributed homogeneously on an atomic scale. We will continue monitoring the extruded fuel to a burnup approaching 1000 MW.h/kg H.E., as an indicator for the performance expected from co-precipitated $(Th,U)O_2$ or mechanically-mixed $(Th,U)O_2$ with good fissile homogeneity.

[+]Member, CNS

INTRODUCTION

The neutron economic CANDU (CANada Deuterium Uranium) reactor is readily adaptable to thoria-based Advanced Fuel Cycles without the development of a new reactor technology. A number of possible cycles exists. For example, in the Pu-topped cycle, average burnups approaching 1000 MW.h/kg H.E. are envisaged while in the uranium-conserving lightly-topped or self-sufficient $(Th,U\text{-}233)O_2$ cycles, only about 250 MW.h/kg H.E. is required [1].

At Chalk River Nuclear Laboratories, our reference Advanced Cycle fuel production process is the conventional blended powder, cold-pressed and sintered pellet route. We already have irradiation experience to show that conventionally fabricated $(Th,U)O_2$ fuel can comfortably reach the target burnup for the self-sufficient cycle [1]. However, the theoretical improvement in performance expected for $(Th,U)O_2$ fuels has not yet been observed. General behaviour has been about the same as that for UO_2 and work is underway to determine the reasons for this and achieve the theoretically predicted performance of the fuel at higher burnups.

Work has also been carried out to develop and evaluate alternatives to the reference fuel which offer the potential for simpler fabrication in a remote facility. They also have features which may improve performance at high burnup. In this paper we give interim irradiation performance results for the alternative thoria fuel types we have studied. These are:

(i) pellets impregnated with the fissile component by immersion,

(ii) extruded sol-gel-derived clays, and

(iii) vibropacked, high density microspheres (spherepac).

All experiments have been carried out in the 1L-2 pressurized water loop of the WR-1 experimental reactor at the Whiteshell Nuclear Research Establishment, Pinawa, Manitoba.

FUEL TYPES - Impregnated Pellets

Impregnated fuel is prepared by immersing pressed green pellets of pure thoria in a nitrate solution of the fissile heavy element component [2]. Uranyl nitrate has been used as the impregnating solution in laboratory experiments but plutonium in solution could also be present in a mature thorium fuel cycle. Following impregnation the pellets are rinsed, dried and then processed in a conventional manner. The primary advantages of impregnation are that the fissile component can be used in solution form, thus removing the powder conversion stage, and that the more dusty and complex operations associated with making pellets are confined to an unshielded facility where maintenance of equipment is relatively easy.

Uranium has an affinity for thoria surfaces, resulting in its being removed from solution and deposited on the outer regions of the pellet as the liquid is drawn through the pores. Figure 1 shows this feature in longitudinal cross-sections of green thoria pellets impregnated for different times. Analysis has shown that the first liquid reaching the pellet center (after 30-40 minutes of impregnation time) is essentially pure water.

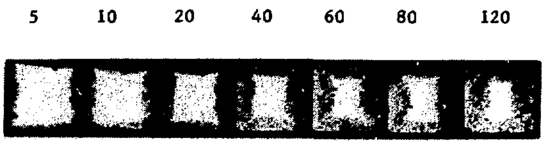

FIGURE 1: LONGITUDINAL CROSS SECTIONS OF GREEN THORIA PELLETS SHOWING URANIUM PENETRATION WITH INCREASING IMPREGNATION TIMES (min).

From an irradiation performance standpoint, the impregnated fuel concept is interesting because one expects the concentration of fissile material in the vicinity of the pellet outer surfaces should reduce fuel operating temperature in a manner similar to that for duplex pellets [3]. This comparison particularly applies to a newly-developed version of impregnated pellets in which the fissile component is introduced in the radial direction only.

Particle Fuel

There are several fuel forms generally referred to as particle fuel but our effort has been focused on the form commonly known as "spherepac". Spherepac fuel consists of almost fully dense mixed-oxide microspheres vibration-packed into fuel cladding. The sol-gel process of fabricating spheres has been detailed elsewhere [4]. A desirable feature is that the process involves dustless wet-chemistry operations. Enhancement of material transport, low sintering temperatures (1000-1200°C) and elimination of grinding are other advantages of the spherepac route.

Various microsphere packing configurations are possible; features of three fuels we have made are reviewed in Table 1. Figure 2 shows the view through a "glass sheath" of a packed two component (binary) and three component (ternary) mix. Binary fuel is the easiest to prepare but has the lowest smear density. Ternary fuel is the most difficult to fabricate but smear densities rival normal pellet fuel. The type referred to as "blended ternary" lies somewhere between the two in ease of fabrication and smear density.

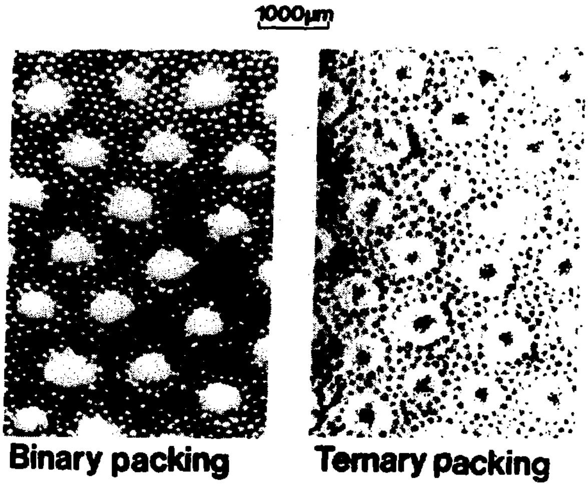

FIGURE 2 BINARY AND TERNARY SPHEREPAC FUEL AS SEEN THROUGH A TRANSPARENT SHEATH TO SHOW PACKING FORM. IN THE TERNARY PACKING THE 10 µm SPHERES ARE NOT RESOLVED INDIVIDUALLY, BUT OCCUPY THE SPACE BETWEEN THE 1000 AND 100 µm SPHERES.

TABLE 1 SPHEREPAC FUEL TYPES

Fuel Type	Nominal Microsphere Sizes (µm) Large	Medium	Fine	Typical Smear Density (%T.D.)
Binary	800	80	-	82
Blended Ternary	800	200	25	86
Ternary	1000	100	10	90

The irradiation of spherepac fuel in a CANDU reactor raises important irradiation performance questions:

(i) use of thin-walled collapsible cladding. All previous irradiations have been performed with free-standing cladding which reduces neutron economy,

(ii) waterlogging and defect behaviour. The envisaged low permeability of packed spheres to fluid flow could result in rupture if the element were defected and waterlogged and the power were raised quickly, as with on-power fuelling. Washout of fuel microspheres is also a concern,

(iii) ability of the sheath to resist ramp defects as imposed by on-power fuelling. One might expect higher resistance to such defects with spherepac because there are no fuel cracks near the sheath which could concentrate the sheath stress, and

(iv) general fuel performance, in particular gas release, at CANDU fuel operating conditions.

Extruded Fuel

Extruded fuel also offered the potential of reducing gamma-active dust during fabrication. A sol-gel process similar to that used for making microspheres but modifed to produce a clay was employed [5]. Figure 3 shows the clay being extruded into pellet-length slugs.

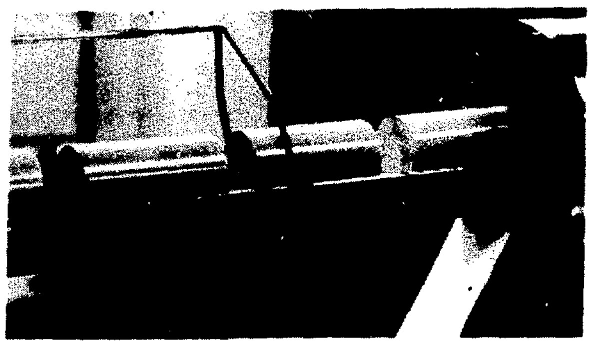

FIGURE 3 THE EXTRUSION PROCESS. EXTRUDED ROD IS SHOWN CUT INTO PELLET-LENGTH SLUGS.

A feature of extruded fuel is that it is homogeneous on an atomic scale. In contrast, normal pellet fuel is prepared by blending fissile and fertile powders and could result in inhomogeneity of the fissile component. This might have deleterious effects on fuel performance.

IRRADIATION PERFORMANCE - Impregnated Pellets

The W-223 experiment consists of twelve Bruce-sized elements of flat-ended impregnated pellets 12 mm in diameter. The UO_2 concentration is about 3 wt%, enriched 70 wt% U-235 in U. The 0.41 mm thick Zircaloy-4 sheaths are internally coated with siloxane. Element powers and burnups have ranged from 50-60 kW/m and 250-650 MW.h/kg H.E. respectively.

On puncturing two elements (WNB-4 and WNC-4) after burnups of approximately 250 MW.h/kg H.E. and average operating linear powers of 55 kW/m, the major portion of the gas collected was the initial helium filling gas. Only traces of fission gas were seen. A longitudinal section from WNC-4 is shown in Figure 4. A thin dish has formed between the originally flat ended pellets. Figure 5, a beta-gamma autoradiograph of the same section shows the fissioning events have occurred in the outer regions of the pellets. Bright spots on the autoradiographs indicate local high concentrations of fission products.

FIGURE 4: LONGITUDINAL CROSS SECTION OF NNC-4 IMPREGNATED FUEL ELEMENT AFTER A BURNUP OF 250 MW.h/kg H.E. NOTE SLIGHT DISHING OF ORIGINALLY FLAT-ENDED PELLETS.

Two additional elements (WNB-3 and WNC-3) were recently punctured after reaching burnups of about 650 MW.h/kg H.E. at average linear powers of about 50 kW/m. Gas release was about 6%. The ELESIM fuel performance code [6] indicated that release typical

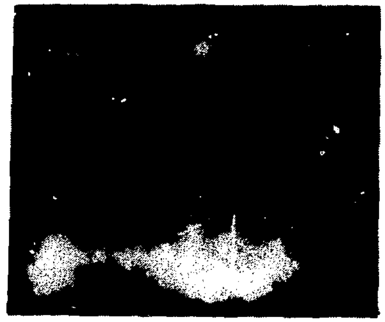

FIGURE 5: BETA-GAMMA RADIOGRAPH OF CROSS SECTION SHOWN IN FIGURE 4. FISSION PRODUCTS SHOW AS LIGHT SPOTS.

of normal UO_2 fuel operated with the same power history would be about 11%. A destructive examination of these elements is yet to be performed.

The gas release results of the lower burnup irradiation indicate that impregnated fuel operates at a lower temperature than conventional pellet fuel. This is supported by the slight post-irradiation dishing observed which suggests that low temperature sintering [7] rather than fuel swelling occurred in the central regions of the pellet. Operating the fuel to higher burnups appears to reduce the initial beneficial effect of impregnated fuel. One can expect gas release to increase because of increasing central temperatures due to the buildup of U-233 with burnup in the central regions of the pellet. The "hot spots" seen in the autoradiographs of the lower burnup fuels suggest initial local concentrations of uranium, probably introduced during impregnation due to large pores in the green pellets. The post-irradiation examination of the higher burnup fuels (WNB-3 and WNC-3), will provide additional information.

Spherepac Fuel

The irradiation program with spherepac fuel was started with related out-reactor waterlogging experiments. These were recently completed and our conclusion is that waterlogging failure due to any resistance to fluid flow out of the element by the fuel is unlikely. It would have to result from the flow resistance of the defect in the sheath, as would be the case with pellet fuel.

The average UO_2 composition in all spherepac irradiations was about 3 wt%, 70 wt% enriched. The W-227 experiment was our first irradiation test with spherepac fuel. It consisted of five binary Pickering-sized fuel elements (pellets 15 mm in diameter) in free-standing (0.63 mm thick) Zircaloy-4 sheathing. Peak linear powers were 60 kW/m. A defect signal was received on a reactor startup after a burnup of about 80 MW.h/kg H.E. but the fuel was permitted to continue operating for an additional five weeks to a burnup of about 115 MW.h/kg H.E. No further signals were received during reactor startups or shutdowns over this period. Following this period, the bottom end cap broke away from one element ((WML-7) while it was being removed from the loop for examination. Figure 6 shows the appearance of the fuel looking into the end with the missing end cap. The end cap itself was severly hydrided but no significant fuel loss from the element was detected. However, a small amount of fuel was lost from the end region when it was sharply rapped. Figure 7 shows a section exhibiting significant fuel restructuring in the pellet center. Although gross restructuring was not apparent in the outer regions, the photomicrograph in Figure 8 shows how the spheres have sintered together.

FIGURE 6: VIEW OF IRRADIATED BINARY ELEMENT WML-7 WITH END-CAP MISSING. IF THE FUEL HAD BEEN UNIRRADIATED THE SPHERES WOULD HAVE ROLLED OUT. ELEMENT OUTSIDE DIAMETER IS ABOUT 16.2 mm.

FIGURE 7: SECTIONED VIEW OF WML-7 SHOWING CENTRAL RESTRUCTURING.

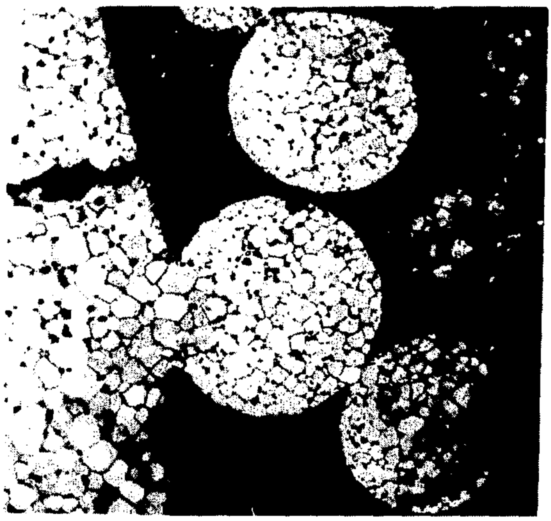

FIGURE 8: SINTERING TOGETHER OF SPHERES IN UNRESTRUCTURED AREAS. GRAIN SIZE IS ABOUT 10 μm, SMALL SPHERES ARE ABOUT 80 μm DIAMETER.

A further short irradiation of the W-227 experiment resulted in three more elements defecting. These were removed on receiving the defect signal but visual examination did not reveal the location of the defects. We suspect they were located in the end-cap weld region.

The W-229 experiment was designed to test spherepac fuel with collapsible Zircaloy-4 cladding (0.4 mm thick) in Bruce-sized elements. Binary, ternary and blended ternary fuels were used. Operating linear powers were about 50 kW/m. A defect occurred in a binary element (WMN-1) after a burnup of about 180 MW.h/kg H.E. but again it was too small to be visually identified. There was no evidence of sheath collapse in any of the elements examined.

W-230 contains fuel identical with that of W-229 but was operated at low linear power (35 kW/m) to be later ramped to 65 kW/m after a burnup of 180 MW.h/kg H.E. This type of ramp causes certain failure in UO_2 pellet fuel with uncoated sheaths [8]. Burnups have reached 37 MW.h/kg H.E. in this pre-ramp soak.

At the present time all three fuel strings are out of reactor awaiting more detailed post-irradiation examination to determine why the elements have defected.

The most significant positive result of the testing is the ability of the fuel to support a thin-walled sheath without collapse. This clearly demonstrates the difference between the stable porosity spherepac fuel, and low density pellet fuels with unstable porosity. Irradiation of the latter has resulted in longitudinal sheath ridges after a few days. Also significant is our realization that waterlogging-induced failures, which have been cited as a potential problem with spherepac, are not likely to be any more frequent than with pellet fuel. Only two such failures have been identified in the more then seven million UO_2 elements that have been irradiated in CANDU reactors. Given that a large hole does develop in the sheath, any washout problem will be reduced because of the irradiation-induced sintering and binding of the spheres.

Extruded Fuel

The W-228 test comprises Bruce-sized elements of (Th,2.9 wt% U)O_2 (enriched 70 wt% U-235 in U) fuel fabricated from a sol-gel-derived clay. The flat-ended fuel pellets are 12.14 mm in diameter, clad in 0.41 mm thick Zircaloy-4. The as-fabricated fuel density was 97% of theoretical, with an homogeneous initial uranium distribution. The internal surface of the Zircaloy- 4 sheathing is siloxane coated. The test has so far operated intermittently at a maximum linear power of about 60 kW/m to achieve a maximum cumulative burnup of about 450 MW.h/kg H.E. In an interim destructive examination, element WMM-5 released 0.7% fission gas after about 150 MW.h/kg H.E. at a maximum linear power of 52 kW/m; element WMM-9 released 3.5% fission gas after about 175 MW.h/kg H.E. at a maximum linear power of about 60 kW/m. For the power history of element WMM-9, ELESIM predicts release of about 16% for UO_2. Figure 9 shows a polished and etched cross section from this element exhibiting transverse and circumferential cracking, and equiaxed grain growth to a fractional fuel radius of 0.2. The appearance is identical with that for conventional pellet fuel.

SUMMARY

In our initial approach to the fabrication of thoria-based fuel we assumed that the fissile and fertile components would be separated in the reprocessing plant and then recombined in the correct proportions during the refabrication step. We also assumed that the irradiated thorium component would be stored for twenty years to permit its activity to decay to natural levels. Thus thorium handling for fabrication purposes could be performed on relatively inactive material. However, current thinking is that there are non-proliferation and economic benefits to co-processing and co-conversion, where the fissile and fertile components are not separated. This may have an important bearing

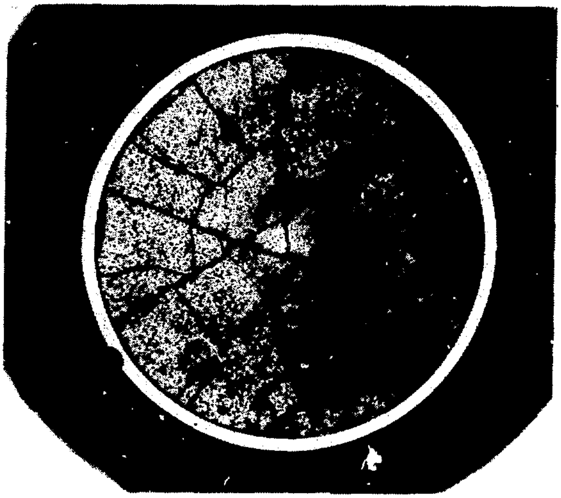

FIGURE 9: CROSS-SECTION OF IRRADIATED FUEL FROM ELEMENT WMM-9. OUTSIDE DIAMETER OF ELEMENT IS ABOUT 13 mm. SHEATH FEATURE AT BOTTOM LEFT IS SAW CUT.

on the viability of some alternate fuels. In this summary the future potential of alternative fuels is considered with co-conversion in mind.

Impregnated fuel has reached a burnup of 650 MW.h/kg H.E. at linear ratings of 50-60 kW/m. It has performed defect-free with gas release varying from zero at 250 MW.h/kg H.E., to about 6% at the maximum burnup. We have found the process as a fabrication route has simplifying aspects in comparison with the blended powder route, but fabrication development on impregnation has been discontinued because of its non-compatibility with co-conversion. The irradiation will continue, and fabrication work could re-start if difficulties arise with either co-processing or co-conversion.

Unlike impregnation, the extruded fuel process as a remote fuel fabrication route has been found to have no advantage over conventional pellet fuel under any fabrication scheme. No additional development work is planned. However the fuel is of interest at present from an irradiation performance standpoint because it represents AECL's first practical demonstration of fuel with the fissile component distributed homogeneously on an atomic scale. As such we will continue to monitor the performance of W-228 to a burnup approaching 1000 MW.h/kg H.E. as an indicator for the performance expected from co-precipitated $(Th,U)O_2$ or mechanically mixed $(Th,U)O_2$ with good fissile homogeneity.

Spherepac fuel of the binary and blended ternary form has demonstrated fabrication advantages and the process is compatible with co-conversion. Preparation of ternary fuel is too difficult to consider further. Irradiation performance tests of spherepac have shown it is compatible with thin-walled collapsible sheathing although unexplained defects have marred the performance of the fuel with both cladding types. Gas release and power ramp data still are not available. Waterlogging and washout problems are not expected to be as severe as originally expected but tests are still required. Development work will be continued, although most of the initial effort will be directed towards the pressing of microspheres into pellets, a process that takes benefits from both the spherepac and conventional pelleting route.

REFERENCES

[1] (a) A.D. LANE and A.T. JEFFS, "Advanced Fuel Cycles for CANDU: AECL's Fuel Development Program", Proc. CNS 2 (1981) 126.

(b) A.D. LANE, T.J. CARTER, A. CELLI, P.J. FEHRENBACH, I.J. HASTINGS, B.J.F. PALMER and A.M. ROSS, "The Development of Recycle Fuels for CANDU Reactors", Trans. ANS, 40 (1982) 261.

[2] A. CELLI, R.K. RONDEAU, and D.J. DREW, "Impregnation as an Alternate Fabrication Process for Recycle $(Th,U)O_2$ CANDU Fuel - A Status Review", 80th Annual Meeting of the American Ceramic Society (Nuclear Division), Detroit, Michigan, 1978 May 6-11.

[3] J.B. AINSCOUGH, D.N. COUCILL, D.A. HOWL, A. JENSEN and I. MISFELDT, "Duplex Fuel: British and Danish Experience and Evaluation", ANS Topical Meeting on LWR Extended Burnup - Fuel Performance and Utilization, Williamsburg, Va, 1982 April 4-8.

[4] R.B. MATTHEWS and M.L. SWANSON, "Fabrication of Large $(Th,U)O_2$ Microspheres", Bull. A. Cer. Soc., 58 [2], (1979) 223-227.

[5] M. ONOFREI and D.M. SOLBERG, "Extrusion Process for Thoria Recycle Fuel", 82nd Annual Meeting of the American Ceramic Society (Nuclear Division), Chicago, Ill., 1980 April 27-30.

[6] M.J.F. NOTLEY and I.J. HASTINGS, "A Microstructure Dependent Model for Fission Product Gas Release and Swelling in UO_2 Fuel". Nuc. Eng. and Design 56 (1980) 163.

[7] I.J. HASTINGS, and L.E. EVANS, "Densification Algorithm for Irradiated UO_2 Fuel", J. A. Cer. Soc., 62 (1979) 217.

[8] W.J. PENN, R.K. LO and J.C. WOOD, "CANDU Fuel-Power Ramp Performance Criteria", Nuc. Tech. 34 (1977) 249.

ISSN 0067 - 0367

To identify individual documents in the series we have assigned an AECL- number to each.

Please refer to the AECL- number when requesting additional copies of this document

from

Scientific Document Distribution Office
Atomic Energy of Canada Limited
Chalk River, Ontario, Canada
K0J 1J0

Price $1.00 per copy

ISSN 0067 - 0367

Pour identifier les rapports individuels faisant partie de cette série nous avons assigné un numéro AECL- à chacun.

Veuillez faire mention du numéro AECL- si vous demandez d'autres exemplaires de ce rapport

au

Service de Distribution des Documents Officiels
L'Energie Atomique du Canada Limitée
Chalk River, Ontario, Canada
K0J 1J0

Prix $1.00 par exemplaire

1755–82