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CERAMIC FUELS
FOR HEAVY WATER REACTORS

June 1970

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FOR HEAVY WATER REACTORS

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CERAMIC FUELS FOR HEAVY WATER REACTORSI. INTRODUCTION

Ceramic oxide fuels have achieved a commanding lead for water cooled reactor fuels. The purpose of this report is to summarize fuel fabrication technology, fuel irradiation behavior and limitations for these materials, emphasizing pertinent work since 1964. The use, properties and behavior of ceramic fuels before 1964 are documented in "Uranium Dioxide - Properties and Nuclear Applications" (1961)⁽¹⁾ and the Geneva III conference papers (1964).

Most oxide fuel fabrication and irradiation experience are valid for all water reactor fuels, including heavy water reactors unless otherwise specified.

II. SUMMARY AND CONCLUSIONS

Heavy water moderated, ceramic fueled reactors which are being operated, built, planned or studied indicate that a PWR is favored over a BWR, 41 to 4, respectively.

Not all of the heavy water moderated reactors employ heavy water coolant. Of the above reactors, five employed boiling light water coolant, nine an organic coolant, and five a CO₂ coolant.

In nearly all the heavy water reactors operating or under construction, the fuel is natural or slightly enriched UO₂ with only two exceptions; the PRTR which used UO₂-PuO₂ and the ORGEL (organic cooled) which used UC. At present most HW reactors use natural UO₂ (four out of six), but the trend seems to be changing. Most U. S. studies indicate enriched UO₂ will be used in heavy water reactors of the future.

The majority of water reactors use fuel rods in cluster assemblies. Over 250,000 fuel rods have operated satisfactorily in water reactors and over 80,000 fuel rods have been irradiated in Canadian heavy water reactors. The present day rod cluster fuel element is well proven in heavy water reactors for exposures to 8,000 or 10,000 MWd/tonne. Work on UO₂-PuO₂ fuels for BWR and PHWR is moving at an accelerated pace.

A few tabular fuel elements are being and were used in heavy water reactors but there are no apparent plans to use tube-in-shell elements in any reactor.

Zircaloy cladding is used for most reactor fuels. Other clad materials are being considered but there is no concerted effort to find or develop new clad materials.

Higher linear heat ratings for oxide fuel elements are forecast. For a rod fuel geometry the $\int kd\theta$ ratings are expected to increase from 36 W/cm to 70 W/cm.

Burnups are also increasing. Present Canadian reactors attain an average burnup* of up to $\sim 8,800$ MWd/tonne and plan to increase to about 10,000 MWd/tonne average. Other heavy water reactors using rod geometries quote present goals at 11,000 to 49,500 MWd/tonne while tubular geometries quote present burnups in tests from 1,100 to 17,300 MWd/tonne and future goals of 19,000 in 1970 and 27,000 MWd/tonne in 1980.

Adequate industrial capability and competition exist in the U. S. today to process and fabricate depleted, normal and enriched uranium to the various fuel forms required in research, test and power reactors. There is, in addition, competition in some areas by foreign countries. There is little doubt that technology, industrial capability and sufficient in-reactor irradiation experience exist to provide present day heavy water ceramic fuels of the rod cluster type.

Basic research on ceramic nuclear fuels is being pursued by many countries, particularly in the areas of physical, chemical and mechanical properties.

However, the effects of irradiation, fission products and fuel stoichiometry need better definition. At present, the melting point of oxide fuels represents a physical limit. There is a need to show that an oxide fuel element can be designed and can be operated predictably above the melting point.

*Burnup is expressed as MWd/tonne U (or) MWd/tonne (U+Pu) unless otherwise specified.

Adequate technology exists for the production of oxide fuels. However, optimization of fuel fabrication processes are subject to further development. The importance of high, fissile density should encourage a penetrating investigation of ceramics other than oxides for use as reactor fuels.

Mechanical mixing of mixed oxide fuels is probably more economical than coprecipitation. There are only a few installations with coprecipitation capabilities and experience. However, chemical processes such as the latter when developed and operated on a large scale can usually be made very economical. In the future, unless oxide conversion charges and losses are decreased, coprecipitation may become the primary fuel preparation method for mixed oxides. For thermal reactor fuels there is no well defined technological or nuclear advantage for either fuel.

Nuclear fuels are usually specified that will operate satisfactorily to burnups now contemplated for heavy water reactors. However, present fuel specifications are probably much more rigid than necessary because of uncertainties in clad - impurities - fuel interactions.

Fuel element loading and assembly methods are well developed but there is no "standard" method. Both pellet and vibrational compaction offer advantages under certain conditions in a bundled-rod geometry. Tubular and tube-in-shell fuel elements have not been tested for use extensively, but there are applications believed pertinent to heavy water systems which make their continued consideration very important.

Ceramic fuels have performed excellent in-reactor; however, a critical study is necessary to delineate those properties most affecting the irradiation behavior and longevity of ceramic fuels in a HWR.

A few examples are listed:

- The elastic constants and plasticity are not known for UO_2 and UO_2 - PuO_2 at high temperatures nor the effect of irradiation on these properties.
- The behavior of molten UO_2 , and the influence of impurities thereon, are not well known.

- Crystal damage by irradiation is obvious at 6-7 a/o burnup; at nominal 1-3 a/o the effects on thermal conductivity, density or grain boundary impurity migration are not well known.
- Plutonium and fission product migration are not resolved, particularly at low temperatures.
- UO_2 thermal conductivity increases at high temperatures, and more in-reactor conductivity measurements are required.
- Substoichiometric UO_2 is believed to have better irradiation damage resistance, but more experimental data are required.

Slumping of fuel, vapor transport of fuel, fuel creep and swelling do not seem to be limiting or serious problems at present HWR power levels and burnups.

III. PROPERTIES OF CERAMIC FUELS

A multitude of ceramic fuels have been prepared, characterized and irradiated including oxides, carbides, nitrides, borides, sulfides, and combinations thereof. For operating water reactors, the oxides are now used exclusively. The physical and thermal properties of uranium and plutonium ceramics are well documented⁽¹⁻³⁾ and need not be repeated.

Mixed oxides of $\text{UO}_2\text{-PuO}_2$, $\text{ThO}_2\text{-UO}_2$, and $\text{ThO}_2\text{-PuO}_2$ are potential heavy water reactor fuels, particularly with respect to future recycle concepts. In most cases the second component is in low concentration and does not materially alter the properties of the matrix ceramic. The mixed oxides form a complete series of solid solutions, and additions of up to $\sim 30\%$ of the second component do not materially affect the fabricability or sinterability of the mixed oxides.

IV. FABRICATION TECHNOLOGY

A. Oxide Fuels

Natural UO_2 and low enriched UO_2 fuels are the leading candidates for HWR. However, mixed oxides of $\text{UO}_2\text{-PuO}_2$, $\text{UO}_2\text{-ThO}_2$, and $\text{ThO}_2\text{-PuO}_2$ have been irradiated in heavy water prototypic or test reactors.

1. Fuel Preparation

Commercial capability⁽⁴⁾ exists to convert ore concentrates of UF_6 since the AEC requires that all normal uranium feed furnished for toll enriching be in the form of UF_6 . Allied Chemical Corporation is the only commercial facility capable of converting ore concentrates to UF_6 in the U. S. This plant, which has a capacity to convert about 5,000 tons U per year from ore concentrations to UF_6 , has been shut down since mid-1964, and brought back to operation in 1963. In April 1967, Allied announced an expansion program which will increase the plant's capacity to about 7,000 tons U early in 1968 to about 10,000 tons U sometime in 1969.

Allied announced in August 1966, that its future charge for converting ore concentrations to UF_6 will be reduced from \$1.25 per pound U to \$1.04 per pound U. The reduced rate will apply to UF_6 to be delivered to AEC for toll enrichment, but the \$1.25 charge will continue to apply for conversion accomplished in conjunction with ad hoc barter arrangements between U. S. and foreign countries.

In July 1967, the Kerr-McGee Corporation announced plans to build a \$25 million UF_6 conversion plant, with a capacity of 5-10,000 tons U per year. The plant is expected to go on stream early in 1970. The charge for converting ore concentrates to UF_6 is estimated at \$2.52 per kg U.

No commercial capability exists today for converting enriched uranium to UF_6 . However, the quantities of enriched uranium recovered from unirradiated scrap and irradiated fuels, returned to AEC's gaseous diffusion plants for re-feeding, have been relatively small. Commercial facilities capable of converting slightly enriched uranium to UF_6 can probably be expected within the next 2-3 years, followed by UF_6 conversion facilities for highly enriched uranium. As soon as commercial capability is achieved to convert enriched materials to UF_6 , the AEC will probably require that these materials be returned to AEC gaseous diffusion plants as UF_6 . Presently, leased enriched uranium is returned to

AEC in the form of uranyl nitrate, upon payment of charges calculated to cover AEC's cost of converting to UF_6 . The conversion charge is \$5.60 per kilogram of contained uranium for low U-235 enrichments (< 5.0% U-235) and \$32 per kilogram of contained uranium for high U-235 enrichments (> 5.0% U-235), plus allowances for losses and conversion time.

Uranium Dioxide Preparation: Preparation of UO_2 powders is well documented.^(1,4,5) Today a reproducible, sinterable UO_2 powder can be prepared via the ammonium diuranate route for depleted, natural and enriched UO_2 .

Enriched Fuel Preparation: Enriched fuels may be handled by several different methods, each of which has its own special problems and characteristics. In each case the objective is to increase the concentration of the fissile component of the fuel which is accomplished by adding ^{235}U , ^{239}Pu - ^{241}Pu or ^{233}U . Enrichment techniques include mechanical mixing, coprecipitation, the sol-gel process and gaseous diffusion.

A continuous coprecipitation process is described by NUMEC⁽⁶⁾ for the preparation of a sinterable $(U-Pu)O_2$. The process involves blending of uranyl and plutonium nitrates and coprecipitating ammonium diuranate and plutonium hydroxide with ammonia. The precipitate is filtered, washed, dried, thermally decomposed and reduced to UO_2 - PuO_2 in N_2 - 6% H_2 atmosphere.

Powder characteristics required for pellet fabrication by cold-pressing and sintering to 93-95% density were easily controlled within acceptable limits.⁽⁶⁾ These included particle size, bulk density, surface area, and O/U ratio. Any desired surface area within the range 1-12 m^2/g can be obtained by controlling the reduction temperature. (The surface area/temperature coefficient is nominally $-0.02 m^2/g-^{\circ}C$.) Reduction time and temperature may both be used to control the final O/M ratio. An advantage of the basic precipitation process is the low aqueous Pu losses (0.005%), minimizing waste-disposal cost.

Mechanical mixing of PuO_2 and UO_2 may be accomplished in a number of ways. For best results the particle size and surface area of the two components should be similar. Mechanical mixing techniques usually include a twin shell preblending followed by hammer milling, mixmulling or ball milling. Wet milling produces uniform mixtures but has the disadvantage of utilizing liquids that must be removed by drying and of small batch sizes because of criticality control.

A novel method which provides high density fuel particles is high energy, high temperature, pneumatic impaction.⁽⁷⁾ PuO_2 ($< 44 \mu$ particle size) is blended with minus 210μ UO_2 and the mix is sealed in an evacuable, stainless steel can, heated to 1200°C while applying vacuum and hot impacted by a high energy-rate impact in a Dynapak machine at $175,000 \text{ lb/in}^2$ or greater. The billet is cooled, grit blasted to remove scale and carbonized die lubricant and opened. The resulting product ($> 10.80 \text{ g/cc}$) is crushed to desired particle sizes for vibrationally compacted or swaged fuel rods without fear of plutonium segregation.

Development of fused⁽⁸⁻¹⁰⁾ natural UO_2 was sponsored for several years since fusion provided high density particles for swaged or vibrationally compacted fuel elements. However, the pneumatic impaction method is particularly applicable to fissile-enriched materials wherein the quantity of material during fusion or impaction is limited by criticality considerations.

Recycled fuels may also be handled by several methods. Fuel recycle must be given a more serious consideration in R&D planning for future fuels because it is of particular interest to HWR's utilizing ^{233}U and plutonium enriched thorium fuels.

The sol-gel process^(11,12) has been developed in the U. S., Italy, and in other countries and is particularly adaptable to fuel recycle and can be used to provide homogeneous mixed oxide microspheres or particles. The process consists of evaporation or denitration of the particular hydrosol to a gel and formation

of the glassy high density sphere or particle by drying and sintering. The process has been proven on a pilot plant scale by ORNL and B&W, and is adaptable to shielded facilities for fuels recycle.

2. Fuel Specification

Most heavy-water-moderated reactors now in operation, under construction, or in the design stage rely on the use of natural or slightly enriched UO_2 contained in Zircaloy.⁽¹³⁾ Minimum specifications for reactor grade UO_2 are presented in an American Standards Association specification.⁽¹⁴⁾ Various specifications have been issued by a number of research organizations covering their requirements for UO_2 and for PuO_2 .⁽¹⁵⁻¹⁸⁾

It has generally been the policy of industry to set fuel specifications for a particular reactor loading. Such specifications have features in common but may also include items pertinent to the particular reactor design involved, and reflect the inevitable compromise by requiring that degree of purity which is economically obtainable and which experience has shown to give acceptable performance.

A representative specification is outlined below for nuclear fuels that have been successfully irradiated in water moderated reactors.⁽¹⁹⁻²⁵⁾

Physical Properties

- a. Fuel Density: Sintered pellet: $94 \pm 2\%$ T.D.
as determined from volume and weight measurements.

Dense particle: $\geq 98\%$ T.D.
as determined by Hg penetrometer on
-6 + 10 mesh granules.

- *b. Smeared Density of Fuel in the Rod: 84-90% T.D.
as determined from volume and weight measurements.
- c. Homogeneity of Enriched Fuel: Fissile oxide particles,
< 44 microns.

Chemical Properties

- a. Fissile Content
Isotopic Content: within $\pm 3\%$ of that specified.
- b. Total Impurity Content: ≤ 4 ppm E.B.C.
- c. Oxygen-to-Metal Atom Ratio: 2.00 ± 0.2 .
- d. Halides: < 10 ppm F, < 20 ppm Cl.
- e. Hydrogen: < 20 ppm (by wt.) total H_2 , < 100 ppm H_2O .
- f. Nitrogen: < 200 ppm N_2 .
- g. Carbon: < 200 ppm.
- h. Element Impurities, maximum permissible:

<u>Element</u>	<u>ppm (by wt.)</u>	<u>Element</u>	<u>ppm (by wt.)</u>
Ni	300	Mn	10
Mo	150	Co	6
Cu	50	Sn	5
W	50	In	3
Pb	20	Bi	2
Zn	20	V	1

3. Element Assembly and Testing

a. Pellet Fuels

The development and production of reactor fuel rods by the pellet-in-tube process are well advanced for UO_2 fuels. The process lends itself well to the stringent inspection requirements necessary for reactor loadings. With a few exceptions, commercial reactors utilize elements containing pellet fuel.

*The fuel bulk density is commonly quoted for pellets. It is convenient to define a "smeared density" as the percentage of actual fuel weight contained in the volume enclosed by a unit length of cladding relative to the weight which could be contained if the same volume were occupied by 100% dense fuel. This measure is equivalent to the bulk density of vibratory compacted particle fuel. For pellets, the smeared density takes into account radial gap, axial gaps between pellets, central void and porosity of the fuel.

Fuel shapes are usually fabricated⁽²⁶⁻³⁷⁾ by pressing a free-flowing powder containing an organic lubricant and/or binder. Sinterable grade powders have an average particle size of about one micron and are not free-flowing. Therefore, the powder is either dry pressed at 8,000 to 16,000 psi or mixed with an organic binder and then granulated to yield 14-180 mesh granules. Pellets of coprecipitated or mechanically blended powders of $\text{UO}_2\text{-PuO}_2$ are compacted to a green density of about 55% of the theoretical density.

Final densification of the compacted fuel shape is usually achieved by sintering in an inert atmosphere containing H_2 . Stoichiometry can be controlled in mixed oxide fuels by the use of a dry or wet atmosphere.

Sintered fuel pellets are centerless ground to within the specified diametral tolerance. These grinders usually require a recirculating water spray for cooling and lubrication during grinding. Critical mass control of the grinder sludge in the coolant system can be accomplished either by weighing pellets before and after grinding and limiting the plutonium accumulation within the safe limit by accountability or by limiting the total liquid volume to less than 3 liters. Recent work reveals that dry centerless grinding is feasible. However, fuel for most heavy water reactors is sintered natural UO_2 and so poses no criticality problems.

Thoria and $\text{ThO}_2\text{-}^{233}\text{UO}_2$ pellet fuel fabrication have also been investigated.⁽³⁸⁻⁴⁸⁾ NUMEC fabricated over 14,000 kg of ThO_2 powder having a surface area of $6 \pm 1 \text{ m}^2/\text{g}$ into high-density pellets ($95 \pm 2\%$ T.D.). The surface area and the carbon content of the high-purity ThO_2 powder were of major importance in the sinterability. A limit of 300 ppm of carbon in ThO_2 is suggested. Carbon present as CO_2 can easily be removed by pre-heating the green pellets but carbon present as oxalate inhibits sintering, causing porosity, low density and cracking.

Two operating power reactors, Indian Point and Elk River, employ $\text{ThO}_2\text{-UO}_2$ sintered pellets. For current and future thorium applications pellets are preferred because of past experience.

b. Vibrationally Compacted Fuels

A simple way to make a fuel element is to vibrate a ceramic fuel in a tube and to cap it. Such elements have been evaluated at a number of sites. ⁽⁴⁹⁻⁵⁷⁾ The elements have the disadvantage of lower density and higher fission gas release when compared with pelleted fuel.

Vibrational compaction has obvious advantages in handling fissile-fertile materials that require remote operation and shielded facilities, and is also well adapted to fuel geometries not amenable to conventional fabrication processes. ⁽⁵⁸⁾

Two complete core loadings for nuclear superheat critical experiments were fabricated by vibratory compaction of fused UO_2 . For double annular elements, vibratory compacted powder elements provided adequate performance at significantly less cost than the pelletized elements. Furthermore, the probability of fabricating a third core loading of different fuel geometry but requiring the same fuel enrichment offered the interesting possibility of reusing the original fuel, a condition not possible with a pelletized product. The first core loading consisted of 401 dual annular nested elements, containing a total of about 3000 kg of 2.55% enriched UO_2 clad in stainless steel. The second core loading consisted of 105 tube-in-shell elements, stainless steel clad, containing a total of about 2200 kg of UO_2 .

c. Cladding

The primary cladding material for water reactors is Zircaloy. In Canada and Sweden where heavy water reactors are favored, Zircaloy is employed for reasons of neutron economy.

Cladding thickness varies widely within the industry. The Canadians have emphasized and have considerable operational experience with thin wall collapsible Zircaloy tubing for their PHWR fuels.

In U. S. boiling water reactors, both Zircaloy and stainless steel cladding are used. Zircaloy is now preferred. In HW-BLW reactors now planned both cladding materials are being tested.

Aluminum, aluminum alloys, SAP (sintered aluminum powder), magnesium alloys, beryllium and some iron-aluminum alloys have been evaluated as alternatives to Zircaloy and stainless steel.

d. Nondestructive Testing

Nondestructive tests to insure fuel enclosure integrity include the eddy current test and the ultrasonic test. The eddy current test is capable of detecting axial and circumferential flaws, 0.001-inch deep by 0.001-inch wide by 0.030-inch long. Standardization and sensitivity for the ultrasonic test are not fully established. It is anticipated that a lack of weld penetration equal to 10% of the tube wall thickness and encompassing 60 mils along the surface will be detectable.

e. Quality Control

Fuel material and processing, cladding, end caps, devices internal to a fuel rod and the finished product are subjected to a series of quality assurance tests and inspections during manufacture to insure conformance to product specifications.

Materials specifications, quality control and quality assurance established for light water reactor fuel elements are applicable to heavy water reactor fuels.

B. Fuel Geometry

1. Rod Clusters

The rod-cluster fuel element utilizing ceramic fuels is the most

tested fuel element used for commercial water power reactors. Selection of the cluster geometry must consider the number of rods in the cluster, length, and diameter of the rods, configuration and all of the fuel-cladding-reactor environment and physics parameters. For the cluster geometry, most fuel elements employ either a circular, hexagonal or square array. For heavy water reactors a 7-, 19- or 37-rod cluster is employed although a much greater number of rods are used in LW reactors. Factors affecting the number of rods per cluster for the Canadian HWR are extensively discussed by Notley and Lane.⁽⁵⁹⁾ Rod clusters with ceramic fuels are being used or are planned for use in the following pressurized water reactors:

<u>HMWPWRS WITH ROD CLUSTERS AND CERAMIC FUEL</u>			
	<u>Reactor</u>	<u>Press. Tube</u> <u>Press. Vessel</u>	<u>Coolant</u>
(1)	<u>Operating Power Reactors</u>		
	NPD-2	X	HW
	CANDU	X	HW
	MZFR	X	HW
	BR-3		X
	SGHWR	X	LW & HW
	Agesta		BLW
	EL-4	X	HW
			CO ₂
(2)	<u>Power Reactors Being or to be Built</u>		
	Pickering	X	HW
	Gentilly	X	BLW
	RAPP-1	X	HW
	KANUPP	X	HW
	KKN(AKB)	X	CO ₂
(3)	<u>Power Reactor Studies</u>		
	CVNA	X	HW
	HWRS (ORNL)	X	BLW
	SSCR	?	LW or HW
	PHWR	X	HW
	HVCCR	X	Organic
	B&W, (Th-U)O ₂	X	Organic
	AI-CE(UC)	X	Organic
(4)	<u>Other Operating Reactors</u>		
	PRTR	X	HW
	CVTR	X	HW
	WR-1	X	Organic
(5)	<u>Other Reactors Being or to be Built</u>		
	ORGEL	X	Organic
	CIRENE	X	BLW
(6)	<u>Other Reactor Studies</u>		
	DON	X	Organic

The rod cluster is the only commercial boiling water reactor fuel configuration, yet there are very few experimental data available on its hydrodynamic stability. Pate⁽⁶⁰⁾ points out that the greater freedom of movement offered to void by the rod cluster geometry may be the cause of its diffuse threshold of instability. A comparison of core data for a typical 400 MWe BWR was made with a similar BWR by Margen and Haza.⁽⁶¹⁾ Many parameters affecting fuel geometry were practically identical; however, the BWR assemblies did have fewer rods per assembly. This is not a very sensitive factor in BWR core economics. Rod clusters with ceramic fuels are being used or are planned for use in the following BWR reactors:

<u>Reactor</u>	<u>Press. Vessel</u>	<u>Coolant</u>
(1) <u>Operating Power Reactors</u> None (note SGHWR under PWR listing)		
(2) <u>Power Reactors Being or to be Built</u> Marviken (also note Gentilly under PWR listing)	X	BW
(3) <u>Power Reactor Studies</u> Yugoslavia (also note ORNL's HWR's under PWR listings)	X	BW
(4) <u>Other Operating Reactors</u> Halden	X	BW
(5) <u>Other Reactors Being or to be Built</u> None (note: CIRENE in PWR listing)		
(6) <u>Other Reactor Studies</u> None		

A Canadian paper⁽⁵⁹⁾ summarizes in the following table the factors which affect the design of UO_2 fuel clusters for high power outputs. The results indicate that more efficient packing and better matching of channel sizes is achieved with 19- or 37-element bundles; the 22- and 28-element designs have the lowest fuel/coolant ratios, hence are less suitable for LW than HW coolant. The 19-rod cluster is undoubtedly the most thoroughly tested and documented circular cluster geometry.⁽⁶²⁾

COMPARISON OF BUNDLE DESIGN PROPERTIES*

<u>Bundle Design</u>	<u>Fuel/Coolant Ratio (By Volume)</u>	<u>Zircaloy Volume (Normalized)</u>	<u>UO₂ Fuel/Zircaloy** Ratio (By Volume)</u>	<u>Relative Surface Heat Flux</u>
19-Element Radial Pitch	1.82	1.17	6.05	1.16
22-Element (with 2 Element Sizes)	1.59	1.08	6.24	1.11
28-Element	1.42	1.00	6.54	1.00
37-Element Radial Pitch	1.62	1.06	6.47	0.85
37-Element Hexagonal Pitch (with 2 Element Sizes)	2.04	1.13	6.51	0.83

*In these designs the Zircaloy cladding thickness increased with rod diameter in the ratio of thickness/outside diameter = 0.0275. The designs are also based on a minimum inter element spacing of 1.0 mm and a bundle to pressure tube spacing of 1.6 mm.

**Includes Zircaloy volume in sheathing, end caps, end plates and appendages.

2. Tubular Fuel Elements

Nested tubular fuel (oxide) elements are economically attractive for HW reactors.⁽⁶³⁾ Irradiation of nested tubular elements containing UO₂ mechanically compacted into Zircaloy sheathing and irradiated in the HWCTR⁽⁶⁴⁾ revealed that the tubular element, as compared with the rod cluster, offered several attractive features, namely:

- a. A large mass of fuel can be incorporated easily in a single fuel element with concomitant low fabrication cost.
- b. High specific powers can be achieved without resorting to expensive small diameter fuel rods.
- c. No geometrical transition is necessary to fit nested tubes into the pressure tubes of D₂O reactors.

- d. Adverse heat transfer effects caused by the radial gradient in neutron flux in individual fuel assemblies can be largely overcome by adjustments in thickness of individual coolant annuli and fuel tubes.

Tubular oxide fuel elements have been or are planned for use in these reactors:

<u>Reactor</u>	<u>Status</u>	<u>Reactor</u>	<u>Status</u>
HWCTR	~ 100 irradiated	HWRS1	These reactor studies call for tubular elements.
PRTR	1 irradiated	-2	
SGHWR	8 (23-tube) superheat clusters in reactor.	-3	

3. Tube-in-Shell

Canadian studies⁽⁶⁵⁾ on the Gentilly reactor (BLW coolant) have included the tube-in-shell element and, in comparison to a fuel rod bundle, it would increase burnup 6%, increase steam outlet quality 100%, and decrease fueling and capital cost. However, later work⁽⁶⁶⁾ shows that the tube-in-shell geometry provides a higher fuel-to-coolant ratio, but the economic incentive now is less than that calculated previously.

One tube-in-shell, Zircaloy-clad UO_2 packed particle element was irradiated in the PRTR to a burnup of ~ 150 MWd/tonne.⁽⁶⁷⁾

Recent studies on second-generation BONUS (light water, direct cycle BWR) superheat fuel elements showed pessimistic results for rod-type elements but the tube-in-shell type looked attractive.⁽⁶⁸⁾ Elements have been developed for in-reactor proof tests.

C. Fuel Channels

1. Pressurized Water Reactors

The fuel element for a PWR can be a circular, square or hexagonal array of rods. The fuel element may or may not be enclosed in a can, depending on the manufacturer.⁽⁶⁹⁾ In a pressure tube reactor,

the fuel element assembly slides into a process tube or fuel channel. The channel is usually round.

2. Boiling Water Reactors

The rod cluster is currently the only commercial BWR fuel configuration,⁽⁶⁰⁾ and is enclosed in a channel that provides guidance for the cluster and a bearing surface for the control rod blades.⁽⁶⁹⁾ Conventional BWR's normally employ square fuel channels but one study⁽⁶¹⁾ proposes the use of hexagonal fuel channels to facilitate tandem refueling between a BWR with fixed hexagonal channels and a BHWB with fixed round channels. Fuel element dimensions for a BWR and an optimized BHWB are compared below:

FUEL ELEMENT DIMENSIONS OF A BWR AND AN OPTIMIZED BHWB

	<u>BWR</u>	<u>BHWB</u>
Number of Rods in Bundle	64	37
Coolant-to-Fuel Area Ratio	1.95	2.27
Fuel Pin Diameter, mm	10.5	12.8
Active Core Height, mm	3650	3430

V. FUEL PERFORMANCE

Surveys of the status of oxide fuel performance through 1963 are available in the papers of the 1964 Geneva Conference. Summaries from the papers presented at this meeting are given below; thereafter this report will be concerned with more recent work. No attempt is made in the summary to differentiate between reactor types.

The great breadth and depth of U. S. experience with UO_2 fuel was stressed. Over 130,000 Zircaloy and stainless steel-clad rods were irradiated in operating reactors to 28,000 MWd/ton at heat fluxes from 350,000 to 450,000 Btu/hr-ft² with good results. Over 4,000 experimental rods made by 37 fabrication processes were irradiated to 16,000 MWd/ton and at heat fluxes up to 1,435,000 Btu/hr-ft². Superheat elements were successfully operated at 510°C to 3,600 MWd/ton at heat fluxes of 445,000 Btu/hr-ft².

Operation with a molten core was possible when the UO_2 was not too dense. Swelling was reported to be independent of temperature in the range of 760-2000°C to exposures of 36×10^{20} fissions/cm³. Good radiation resistance was also shown for $\text{ZrO}_2\text{-UO}_2$, $\text{ThO}_2\text{-UO}_2$, and $\text{UO}_2\text{-PuO}_2$. Swaged and vibratory-compacted ($\text{UO}_2 - 2\% \text{PuO}_2$) fuel performed excellently to 1.7×10^{20} fissions/cm³ in the PRTR.

The Canadians irradiated Zircaloy-clad UO_2 rods to 10,000 MWd/ton, with most in the range of 4000-7000, with dimensional changes of a fraction of 1% and fission-gas release typically 12%. Irradiations in NPD achieved 3000 MWd/ton without failure. Netherlands workers irradiated stainless steel-clad UO_2 rods 10-15 mm diameter to 4000 MWd/ton at an $\int k d\theta$ of 60 W/cm. Japanese workers reported gas release of a fraction of 1% from swaged rods, 0.03% from sintered rods, and no failures in irradiations to 2700 MWd/ton. Swedish investigators found little change in Zircaloy-clad UO_2 rods in irradiations to 1200 MWd/ton. Russian investigators reported only cracking of the UO_2 rods used in the icebreaker LENIN after operation at an $\int k d\theta$ of 10 W/cm. Examination of irradiated, burnout stainless steel-clad UO_2 rods by German investigators revealed no reaction between stainless steel and UO_2 , and no significant stainless steel/water reaction.

British investigators measured the lattice-parameter changes in $\text{UO}_2 - 1\% \text{PuO}_2$ and $\text{UO}_2 - 10\% \text{PuO}_2$ irradiated to 5-6% burnup. With central temperature up to 2400°C, fission-gas release was as much as 56%. The British reported a cladding reaction when nitrogen was left inadvertently in the cans of stainless steel-clad AGR elements. Nitrides formed in the clad adjacent to cracks in the fuel.

A. Irradiation and Other Effects in Oxide Fuel

1. Plutonium and Fission Product Migration

Axial migration of several fission products from mixed oxide into an adjacent UO_2 blanket region was determined.⁽⁷⁰⁾ The transport of fission products beyond the isotherm at about 940°C (the calculated fuel-blanket interface temperature for 0.220-inch, fuel diameter at 20 kW/ft) was almost entirely accounted for by the

daughters from Xe and Kr decay. The isotopic ratios observed preclude significant transport of cesium by distillation.

Radial migration in mixed oxide and UO_2 fuels was observed for three isotopes. Cesium migrated towards the cooler portions of the fuel; Ce-144 and Zr-95 migrated towards the center of the fuel.⁽⁷¹⁾ Plutonium migration in the mixed oxide samples was within the estimated errors of measurement and calculation. For molten UO_2 fuel the data scattered by $\pm 30\%$, but there was definite indication of plutonium segregation at the center of the fuel.

The PFR* program revealed evidence⁽⁷²⁾ of very substantial plutonium migration, as much as 50% of the initial concentration. The information is not yet sufficiently systematic to incorporate as a design parameter. Such migration of plutonium may introduce hotspots but the main effect would be on reactivity. The migration of plutonium to affect the fuel stoichiometry could have marked effects on the irradiation behavior of fuel and on cladding/fuel compatibility. Radial, plutonium migration within a PFR pin is not considered to be of much consequence.

Although upward and downward movements of fuel were noted as a result of transient melting in irradiated mixed oxide tests,⁽⁷³⁾ no evidence of significant axial redistribution was found in mixed oxides operated below center melting. Autoradiographs revealed an alpha gradient in molten fuel. Almost all of the characteristic microstructure changes within the fuel correlated with changes in either alpha, beta or gamma radiography rings.

The PRTR high-power-density program⁽⁷⁴⁾ with packed particle (and pelleted) oxide fuel indicated axial relation of fuel when the

*UKAEA, Prototype Fast Reactor

fuel operated under molten conditions. Variations in size of central void and degree of melting were observed in packed particle rods operated at high power generation.

Convective mixing was noted within the molten oxide in long fuel rods. Recent evidence⁽⁷⁵⁾ in PRTR UO_2 - PuO_2 fuel rods indicated a flow of liquid fuel into radial cracks during operation -- a previously unobserved phenomenon.

Some of the latest diffusion and segregation data for oxide fuels appear in fast reactor articles but the data apply equally well to thermal reactor fuel phenomena in most cases.

Smits and Linder⁽⁷⁶⁾ measured the diffusion coefficients of plutonium in UO_2 at temperatures to 1500°C , and Davies and Novak⁽⁷⁷⁾ at 2400°C . The 2400°C measurement showed a diffusion rate about 40% greater for plutonium ($3.4 \times 10^{-10} \text{ cm}^3/\text{sec}$) than for uranium ($2.5 \times 10^{-10} \text{ cm}^2/\text{sec}$).

Freshley and Mattys⁽⁷⁸⁾ observed slight plutonium redistribution towards the center region of ThO_2 - PuO_2 fuels (5.5% to 18.5% PuO_2), leaving behind a depleted region at about 40% of the radius.

Plutonium distribution along the radius of the fuel also coincided with the distribution of "nonmobile" fission products. Bailey and Chikalla^(79,80) investigated the redistribution of plutonium and fission products in mixed oxide fuels containing up to 7.5% PuO_2 . They concluded that fission gas release rates are primarily controlled by rod power rather than by plutonium concentration. They observed extensive radial redistribution of Ce-137 and Ru-106, but only "insignificant" migration of Pu, Zr-Nb-95, Ce-Pr-144 and Sr-90. Specific locations within the columnar grain region exhibited a high concentration of Ru-106 along with "metallic" inclusions. The outer 10 mils of the fuel radius revealed sharp gradients in the concentration of fission products. A high concentration of fission products at the extreme outer sintering boundary seemed to be unique to mechanically

mixed UO_2 - PuO_2 fuels.⁽⁷⁵⁾ Cause of the effect has not yet been determined.

After a series of observations of fission product distribution in irradiated UO_2 and UO_2 - PuO_2 , Bates⁽⁸¹⁾ concluded that fission products were depleted in the dense, pore-free region of large columnar grains, a fission product was often concentrated, but not always, near the thermal center and in the small adjacent columnar grains. Cesium and ruthenium showed the greatest tendency to migrate and the concentration of cesium may vary a thousandfold across the fuel radius, and several fission products concentrate into metallic inclusions after high-temperature irradiation.

The distribution of plutonium departs significantly from that calculated on the basis of the expected self-shielding of the capture resonances of ^{238}U . Maximum concentration is observed near the center and a minimal in the columnar grain region. The ratio of maximal to minimal concentration is the same, 1.4, for Pu, Zr-Nb, and Ce-Pr, and the radius at which minimal concentration is observed is essentially the same for all three.

2. Redistribution Mechanisms for Oxide Fuels

To allow for differential thermal expansion of fuel and cladding and for fuel swelling at high burnups, the available volume of the cladding is normally filled with less than the theoretically possible amount of fuel. Additional void is often provided to accommodate the volume change due to center fuel melting to prevent cladding deformation.

Several redistribution mechanisms may be postulated:

- a. Solid-state creep by axial and radial densification by "self-hot-pressing" under differential thermal expansion forces, swelling by nucleation and growth of retained fission gas bubbles, and axial and radial bulk displacement relative to cladding under differential thermal expansion forces.

- b. Vapor transport by sublimation or distillation in central void and densification by pore migration.
- c. Fluid flow by slumping of molten fuel by gravity and boiling or gas evolution from molten fuel.
- d. Self-diffusion in a thermal gradient by bulk diffusion or grain boundary or surface diffusion.

Evidence of the first two mechanisms is apparent in some regions of nearly all fuel operating above 1600°C. Fluid transport probably occurs in all fuel tests which reach central melting temperatures, but evidence of melting is often obscured by further operation of the fuel below melting. The fourth mechanism occurs as a result of a thermal gradient only. No evidence of this mechanism is available, since specifically designed experiments are required to discriminate from the first two processes.

3. Slumping of Oxide Fuel

Although the following data are taken from a fast oxide fuel study the fuel behavior is believed applicable to rod fuel geometries. Axial displacement of oxide fuel has been observed in most fuel pins operated above the melting point. For 93 to 96% dense mixed oxide fuel, 0.220-inch O.D., melting is observed at linear powers above 22 to 25 kW/ft. Axial transport occurs even in transient tests in which the center of the fuel rises above the melting point for only 2 seconds.⁽⁸²⁾

A series of fuel capsules operating with 10 to 20% of the fuel radius at central melting temperature was irradiated to 60,000 MWd/t.⁽⁸³⁾ Axial redistribution of the fuel was observed but the cladding remained intact. A similar element operating with ~ 40% of the fuel radius molten experienced cladding elongation (~ 5%) and cladding rupture accompanied by a 25% circumferential elongation at the rupture.

4. Vapor Transport of Fuel

Aitken et al.⁽⁸⁴⁾ have shown that UO_2 volatilizes rapidly above 2200 to 2400°C. In flowing dry hydrogen, the O/U ratio of the specimen decreased to 1.88 by reaction with hydrogen to produce water and substoichiometric oxide or U metal. The congruent vaporization composition in hydrogen varied between 1.89 at 2200°C to 1.87 at 2600°C.

Axial redistribution of fuel was also observed without central melting.⁽⁸⁵⁾ Fuel appeared to be transferred via the vapor phase from a region of high local power to a region of low local power. In fuel pins irradiated with nearly flat axial power distribution, a uniform cylindrical void having hemispherical ends was observed. In fuel with a significant axial power gradient (peak-to-minimum ratio greater than about 1.2) a tapered, sometimes ellipsoidal, void was observed.

Large Seed Blanket Reactor studies also showed that gross redistribution of fuel in the seed annular elements can occur below the fuel melting temperature.⁽⁸⁶⁾

In steady-state irradiation with constant power distribution, the fuel-center void boundary is believed to be isothermal since a hot region will evaporate material and nearby cooler regions will condense material. The local temperature is brought to equilibrium quickly since both the local linear heat generation rate and the length of the radial heat-flow path are reduced by the loss of material where there is local peaking of neutron flux, and consequently peaking of local specific power; this is manifested by localized enlargement in the central void. In thermal reactor irradiations, such peaks are often at the end of a fuel element.

5. Fuel Creep

Creep rate measurements as a function of stoichiometry and temperature were made by Armstrong and Irvine⁽⁸⁷⁾ from 975 to 1400°C.

Their results for UO_{2+x} are represented by the expression:

$$E = 0.064 \sigma x \exp (-55,700/RT)$$

where E is the unit strain rate, σ the stress applied, constant in poise^{-1} and x ranges from zero to 0.25. It is reasonable to assume that these data may describe mixed oxide with low plutonium content, but additional measurements are needed for substoichiometric mixed oxides and UO_2 .

6. Irradiation Effects upon Thermal Conductivity

Reactor designs often assume that UO_2 conductivity data apply to mixed oxide. This assumption may not be valid. Variations in UO_2 and single phase mixed oxide conductivity with stoichiometry suggest an optimum stoichiometry vs. conductivity relationship.

The data for UO_2 show extensive scatter in the high temperature range of importance to high performance fuels. The reasons for the discrepancies between three groups of results on UO_2 are not yet resolved. One group of results shows a sharply increasing conductivity above 1500°C , another shows conductivity nearly constant with temperatures from 1200 to 1700°C , while a third shows an undulating relationship of conductivity versus temperature. Most in-reactor results are consistent with the slowly varying curve for conductivity versus temperature with values asymptotic to about $0.02 \text{ W/cm}^\circ\text{C}$ from 1300 to 1600°C .⁽⁸⁸⁾

Results, similar to the in-reactor values, were obtained in recent out-of-reactor measurements on stoichiometric UO_2 .⁽⁸⁹⁾

An anomalous increase in conductivity below 1400°C was found for hypostoichiometric UO_2 , the transition temperature depending on the free uranium content at room temperature. It is hypothesized that the increased conductivity may be due to free uranium metal, and that the effect disappears at temperatures above 1400°C when the excess uranium goes into solution. This also supports the view that some of the reported differences in apparent UO_2 conductivity are due to uncontrolled changes in stoichiometry during the measurements.

Such effects are believed to be less likely for mixed oxides since the substoichiometric compositions are single-phase. de Halas⁽⁹⁰⁾ has hypothesized that microstructural changes, such as growth of radially-oriented columnar grains, can sharply increase conductivity. The presence of a pattern of thermal stress microcracks having appreciable circumferential components may also influence conductivity.

The radial heat flow was recently measured on UO_2 - 20 wt% PuO_2 specimens having an O/M ratio from 1.97 to 2.04.⁽⁹¹⁾ The conductivity of 1.97 material was slightly below the curve for stoichiometric UO_2 , to 1200°C. The results for O/M of 2.00 followed closely the results for stoichiometric UO_2 in the same apparatus. The results for hyperstoichiometric mixed oxide were significantly lower, particularly at low temperatures, than either of the above two compositions. An in-reactor measurement of mixed oxide conductivity showed similar results.⁽⁹²⁾ A mixed oxide fuel pin with O/M of 1.97 was irradiated simultaneously and under substantially identical conditions to a UO_2 fuel pin. The conductivity of the substoichiometric oxide pin was significantly below the conductivity of UO_2 at low temperatures (800 to 1000°C), but approached the value for UO_2 more closely at temperatures up to 1800°C.

Additional measurements are scheduled to explore the matrix of parameters for fuel and cladding geometry, smeared density, and stoichiometry. However, a provisional value of $\int k d\theta = 85 \text{ W/cm}$ for central melting is consistent with both in-reactor and out-of-reactor measurements to date for stoichiometric mixed oxide containing 20% PuO_2 . This corresponds to a permissible linear power generation of about 22 kW/ft in a thermal spectrum, with a cladding surface temperature of 650°C.

Out-of-reactor measurements of conductivity of mixed oxide with low PuO_2 contents were reported by Belgonucléaire.⁽⁹³⁾ Mixed oxides containing 3.6 and 5.5% PuO_2 in UO_2 were compared with UO_2

over the temperature range of 600°C to about 2400°C. A minimum in the conductivity curves was observed at about 800°C, gradually rising towards 2000°C. The curve for the 5.5% mixed oxide fell slightly below the UO_2 curve below 1400°C and rose somewhat above the UO_2 curve at higher temperatures. The authors concluded that the thermal conductivities of the two materials are similar, within experimental error.

A similar measurement on vibrationally compacted fuel in which the plutonium was contained in the fine particle component of a three component particle mixture, also gave a slightly higher conductivity than UO_2 alone (both particle mixtures at 88% T.D.). For these measurements, the minimum conductivity occurred at about 1500°C, with a slight upward trend at about 2000°C. Analyses of the $\int k d\theta$ values derived from the Belgo-Nucléaire measurements showed an uncertainty range from 60 to 90 watts/cm in the integral from 0° to 2600°C. The results for UO_2 range from 65 to 96 watts/cm (Hanford) and 70 to 85 watts/cm (Chalk River).

7. Fuel-Cladding Interactions

Cladding may be deformed by differential thermal expansion if the initial smeared fuel density is greater than 95%, redistribution of the fuel axially raises the local smeared density to over 95%, local melting of fuel produces a volume change greater than can be accommodated by the available local void and relief by axial movement is prevented by adjacent solid fuel, and fission product accumulation causes local fuel swelling which together with thermal expansion exceeds the locally available volume.

Excepting very short-term tests, the experimental results for a quantitative relationship in fuel-clad interactions can at present be interpreted only within fairly wide limits because of the extreme difficulty of doing well-controlled in-reactor experiments. For thermal reactor fuels, operating limits have been established by large-scale statistical tests of whole or partial core-loads of

fuel. Assessment of fuel-cladding interactions at present must be restricted to cautious interpretation of limited in-reactor data, with primary reliance on basic measurements of fuel properties.

GE-APD data suggest that while the gap size is important at the start of irradiation, it tends to decrease as a function of time and that tight dimensional tolerances on pellets and cladding may not be necessary.⁽⁷⁴⁾ On the evaluation of gap tolerances (for fuel-clad mechanical interaction effects), many prior results are subject to large uncertainties because of limitations in ability to control experimental conditions. Recent studies indicate that a 4-10 mil gap decreases to about one-half its initial value.⁽⁹⁴⁾

Melting of UO_2 produces a reported decrease in density of 9.6%. A total volume increase of about 20% must be accommodated between room temperature and the molten state for material initially at theoretical density.⁽⁹⁵⁾ Melting tests with short segments of mixed oxide fuel (2.25" long) indicate that a limited volume fraction of molten fuel is accommodated without clad deformation by a combination of densification of the fuel (initially 90-94% dense including gaps) and axial expansion. As the fraction of radius undergoing melting increases (~ 35 kW/ft) permanent cladding expansion is observed in one cycle of melting.

On repeated power cycling or on repeated transient tests above center melting, yielding of the cladding is also observed.⁽⁹⁶⁾

The deformation observed on multiple melting transients was not radially symmetric; in one instance the rod tended to a triangular cross section. Fuel which was first operated above melting in steady-state, and then subjected to transient melting also ruptured the cladding. In the experiments in which yielding or rupture occurred, axial slumping of the fuel was observed after molten operation. A central core void tapering in diameter from top to bottom was formed by the shrinkage in volume accompanying freezing. A portion of the fuel below the central void attained an average

smear density approaching 98%. Upon being brought back up to power it is hypothesized that the fuel in the high density region reached melting temperature first, with the accompanying volume expansion. The fuel in the region adjacent to the central void remains below melting because of the lower local heat generation rate per unit length. Depending on the rate and extent of volume expansion due to melting, the bridge of unmelted fuel near the bottom of the central void may restrain the relief of the volume expansion on melting which could occur by axial movement.

The possibility of increased specific power without reducing fuel diameter by operation at central melting was explored by M. F. Lyons and co-workers.⁽⁹⁷⁾ They suggested the possibility of avoiding clad yielding on successive startups by a programmed slow approach to power. By this approach, excessive pressure due to central melting might be relieved by the slow plastic yielding of non-molten regions of the fuel.

8. Fuel Swelling from Fission Products

A void fraction within the clad is necessary to accommodate transients (melting), thermal expansion and fuel swelling at high exposures. The void is provided by the use of, or combination of, a radial gap, annular pellets, low density pellets, or packed particle fuel. Of the alternatives, GE-APD⁽⁷³⁾ has not made a choice; however, their reference design employs a radial gap. GE-APD found that gap size very definitely controlled the center temperature initially. An element with a fuel smear density of 90% of theoretical could accommodate 75% of the fuel in the molten state.

Fuel swelling due to the combined effect of thermal expansion and retained fission products in UO_2 fuel operated at relatively low temperatures was reported to range up to 0.7% increase in volume per 10^{20} fissions per cm^3 .⁽⁹⁸⁾ This swelling rate was not attained until the initial porosity of the fuel was occupied.

Irradiation of unrestrained single crystal stoichiometric UO_2 to 7% and to 11% burnup in sodium at low temperatures resulted in gross swelling.⁽⁹⁹⁾ The unit cell parameter at 3.5% and 9% burnup remained at 5.476 Å. A Mo-rich phase, with some Pu and Zr, was observed.

A volume expansion of 1.5% per atom percent burnup was reported for UO_2 irradiated at low temperatures in BR-5.⁽¹⁰⁰⁾ About 20% of the fuel sustained ruptures after 5% burnup. These were attributed to the expansion of the fuel and embrittlement of the cladding during irradiation. The remainder of the fuel has attained 6.5% burnup. The magnitude of fuel swelling is still not reliably predictable. Swelling data on mixed oxide however, have been reported lower than that for UO_2 . Since a large radial gap is not feasible for oxide fuels without excessive central fuel temperature, accommodation of swelling is provided by clad restraint and low density fuel.

9. Melting Point of Oxide Fuels

The melting point of oxide fuel is important in determining the maximum permissible specific power for a given fuel design. In designing for center temperature, t_m , just at melting, the specific power (watts/cm) of the fuel is proportional to $k(t_m - t_s)/r^2$ where k is the average conductivity and r is the fuel radius. The difference $(t_m - t_s)$ is of the order of 1500°C. A variation in melting of $\pm 150^\circ\text{C}$ corresponds to a range of 20% in specific power.

Melting point data on mixed oxide as a function of U-Pu ratio were measured by Chikalla⁽¹⁰¹⁾ who found a maximum at 10% plutonium content. Because of uncertainties in the O/M ratio and the possibility of a rapid change in composition due to loss of oxygen, measurements were repeated in high pressure argon using specimens with known O/M ratios. These measurements generally confirmed the Chikalla data except for a shift in the location of the peak which was found at 5 wt% PuO_2 and at $2900 \pm 30^\circ\text{C}$. A melting point of

2770 $\pm$ 30°C was determined for stoichiometric UO_2 in the same apparatus. The effect of stoichiometry on the melting point of UO_2 - 20 wt% PuO_2 was also determined.⁽¹⁰²⁾ The melting point increased as the O/M ratio decreased from 2.06 to 1.97. The most rapid change in melting point occurred between an O/M of 1.985 and 1.97. This paralleled a preliminary result by Duncan and Ferrari⁽¹⁰³⁾ for hypostoichiometric UO_2 which showed increasing melting point with decreasing oxygen below 2.00.

In a closed system typical of a fuel element, an effective partial pressure of oxygen is established which represents an equilibrium between different compositions at different temperatures along the radius and axis of the fuel. Measurements to determine O/M ratio as a function of radial position in the fuel, including melting point measurements are needed before the maximum permissible central temperatures can be established.

Accumulation of fission products decreased the apparent melting point of UO_2 at 5% burnup by about 150°C.^(104,105) One possible explanation for this effect is a change in stoichiometry with burnup. The effect on UO_2 - PuO_2 has not been determined. The effect of burnup on melting point will be unimportant for reactor designs in which the core conversion ratio is below 0.9. For example, the maximum kW/ft rating of the fuel for the core design described in a recent 1000 MW design study decreases about 30% with burnup⁽¹⁰⁶⁾ which would accommodate a 450°C decrease in melting temperature.

10. Failure Mechanisms

In selecting a fuel-clad combination for a reactor, two modes of failure must be considered. Either the fuel element will crack and release fission products or it will swell without cracking and block a coolant passage. Prediction of failure mechanisms remains difficult. For example, in one study of superheat fuel elements, the fission product release rate from a defective

element could not be predicted from measurements made on deliberately defected elements.⁽¹⁰⁷⁾

GE-APD studies indicate that it is necessary to define a local damage factor (the point of incipient damage) where cladding deformation begins. This factor can be described analytically in terms of fuel temperatures but must be experimentally verified.

TREAT tests with vibrationally compacted UO_2 indicated that simulated high burnup fuel failed by clad rupture and that simulated low burnup fuel failed by clad melting.⁽¹⁰⁸⁾

TREAT tests with high burnup (70,000 MWd/tonne) mixed oxide by GE-APD⁽⁸⁷⁾ indicated that the release of fission gas from irradiated fuel during a melting transient constituted a primary contribution to the failure mechanism.

Effects of fuel-cladding transient performance of fission product formation, fission gas release, and central void formation resulting from short- and long-term, steady-state irradiations need to be established.^(73,109)

Examination of irradiated fuel has shown that fuel melting during a transient resulted in agglomerates of extremely radioactive, metallic fission products.

Need exists to simulate and to evaluate effects of different types of transients, e.g., power excursion and loss-of-coolant. In some experiments, molten UO_2 ran out-of-clad in later but not in earlier tests.⁽¹¹⁰⁾

B. Coolant-Fuel Interactions

A Canadian paper⁽⁵⁹⁾ concludes that if an incentive exists to operate at thermal ratings in excess of $\int kd\theta$ of 60 W/cm, the defect behavior should be examined in more detail. The authors stated that with the few defected fuel elements that have operated at ratings ($\int kd\theta$) of over 50 W/cm, the following results were noted:

<u>No. of Fuel Elements</u>	<u>$\int kd\theta$, W/cm</u>	<u>Time In-Reactor</u>	<u>Remarks</u>	<u>Reference</u>
1	83	6 hours	Failed in too severe a manner to be contemplated for a power reactor.	59 111
1 3	68 60	8 hours to 35 days	No significant damage	59 112 112
--	> 60	< 90 min.		113

1. Coolant Entry in Defected Fuel Elements

PRTR⁽¹¹⁴⁾ showed behavioral differences between nondefected and defected mixed oxide fuel rods. It is felt that the proposed rupture mechanism is not unique with vibrationally compacted UO_2 - PuO_2 fuels and that a similar occurrence could take place with pellet-containing fuel. Studies conducted with intentionally defected fuel rods are discussed in detail in the following section on fuel experience.

Operation of experimental superheat reactors,⁽¹¹⁵⁾ although not yet far enough along to prove fuel-element lifetime, has demonstrated that although defects in oxide-fueled superheat elements cause operational inconvenience, they do not cause prohibitive radioactive contamination of the steam.

2. Coolant "Logging"

No evidence of water logging was noted in an irradiation test of a defected Zircaloy-clad oxide fuel element in Saxton reactor after operating at 13.5 kW/ft (peak) for 60 effective full-power days.⁽¹¹⁶⁾

No water logging occurred in in-service failed or intentionally defected fuel elements (one UO_2 , three Al-Pu, and thirty-four UO_2 - PuO_2) in the PRTR.⁽⁶⁷⁾

VI. FUEL DESIGN AND PERFORMANCE IN-REACTOR

The basic heavy water fuel element design is the rod cluster. Several fuel bundle types were used in CANDU-PHW reactors for the shorter elements. The first type used was wire-wrapped but the brazed split-spacer bundle, which was a cheaper bundle containing less Zr-2 and overcame the concern of sheath fretting by wire-wrap, was used in the later fuel concepts. Latest Canadian fuel elements use thin-wall tubing (0.38 mm wall) which collapses onto the UO_2 pellets under coolant pressure.

A. PWR Fuel Element Performance

Significant improvements were made in design of Zircaloy-2 - UO_2 elements for Canadian power reactors. A replacement 82.5 mm diameter, 19-element bundle ($\int k d\theta = 40 \text{ W/cm}$) for the Douglas Point reactor was developed using a brazed split-spacer design, instead of wire-wrap, thus preventing inter-rod fretting of the sheath. Both wire-wrapped and brazed split-spacer bundles successfully attained burnups in the range of 10,000 - 20,000 MWd/tonne at an $\int k d\theta$ of 15 to 35 W/cm in the NPD reactor.

An 103.4 mm diameter, 28-rod bundle ($\int k d\theta = 42 \text{ W/cm}$) was developed for the 500 MW(e) Pickering Station. Fission gas release of less than 5% at an $\int k d\theta$ of 35 W/cm and less than 20% at an $\int k d\theta$ of 50 W/cm was measured.

Fission gas release decreased with increased UO_2 density in the range 10.3 to 10.7 gm/cm^3 and was independent of burnup to 8000 MWd/tonne U.

Deuterium pickup in Zircaloy-2 cladding was insignificant after 600 days in-reactor.

During three years' operation of NPD only five bundles were defective. ^(117,118) Two of these defective bundles operated for three years with little or no deterioration. All failures were attributed to mal-operation of the fuel loading equipment.

Other operating experience has shown that defected rods can be tolerated in-reactor for relatively long periods. In the Shippingport LWR, a defected element survived more than 13,000 hours at full power without serious damage. At higher power outputs, failures of Zircaloy-clad UO_2 fuel occurred in Dresden-1 without requiring removal before the scheduled shutdown. Experience with particle fuel elements has also been satisfactory, with in-reactor sintering of the UO_2 apparently preventing any significant washout.

Under normal reactor operation the oxidation rate of Zircaloy-2 in boiling water has been greater than that observed in the laboratory. Hydriding, however, has been very low and should not limit performance. A more likely limitation to an increased surface heat flux would be the exponential increase in oxidation rate which could arise from the temperature drop across the oxide layer. Canadian experience in the PHWR-NPD reactor has indicated a higher deuterium absorption rate than would have been expected from the oxidation rates, which appeared normal. The increased rate soon leveled off, possibly indicating an initial absorption of excess deuterium early in the irradiation. Extrapolating to four years' exposure, a deuterium content of about 600 ppm (equivalent to 300 ppm hydrogen) might be expected. Experience with Shippingport PWR has shown that, if there were any irradiation enhancement of the corrosion rate, the factor was less than 1.2.

The Spanish Minister of Industry recommended that the JEN close down the DON Project, 30 MW(e) prototype heavy water moderated, organic cooled reactor and instead dedicate the effort to developing a fast reactor. The project started in 1962 in collaboration with Atomic International.

Plutonium-enriched fuel pins were inserted into 1 of the 32 fuel elements in the BR-3/Vulcain, 12 MW(e) spectral shift PWR prototype power reactor at Mol, Belgium. Half of the plutonium pins in the 37-pin assembly were manufactured by UKAEA using pelleted fuel and the other half using vibratory compacted fuel were made by Belgonucleaire. This fuel element will be in the core throughout the

present experiment which is aimed at verifying the operating characteristics of a Vulcain core throughout its life and to determine the fuel behavior at high burnups. Maximum burnup of 30,000 to 40,000 MWd/t is expected by the end of the test, yielding valuable data on the behavior of plutonium-enriched fuels in thermal reactors. A more economical use of plutonium is expected in this cycle than that employing ^{235}U . The European Community has established that about 90 tons of plutonium will have accumulated by the time breeder reactors enter utility service in 1980. BelgoyNucIèaire and CEN have been pursuing the plutonium evaluation program for several years, with Euratom backing.

The objective of the advanced HWR Vulcain core design is to develop a reactor suitable for land or marine applications over a range of powers from 20 to 100 MW(e) at low operational costs to facilitate commercial exploitation of small power reactors. Advanced concepts of variable moderation and extensive fuel life are features of the design. The in-reactor instrumentation of the BR-3/Vulcain is the most complete ever built to record operating parameters throughout the reactor's operating life. This represents over 600 days of full power operation without opening the intermediate vessel, to a burnup of 25,000 MWd/tonne. It is also the first time that D_2O has been used at pressures as high as 2000 lbs/in².

Refueling and radial shuffling of fuel can be done on load in the MARVIKEN reactor. The superheater channels are located in the inner half of the core volume, where one out of three channels is a superheater channel. The design of the reactor is such that it can operate also without superheater elements, i.e., with saturated steam, though at reduced power.

Probably the largest program in the world specifically directed towards defect characteristics and in-reactor behavior has been conducted at Richland, Washington, in the PRTR. Intentionally defected fuel rods were irradiated in the heavy water PRTR with supporting tests in the MIR and ETR at NRTS, Idaho.

The batch core experiment now in the PRTR is providing statistically significant irradiation behavior data on high power $\text{UO}_2\text{-PuO}_2$ fuel elements operated at power generations greater than those currently employed in commercial power reactors. In addition, irradiation of intentionally defected PRTR rods operating under power reactor conditions have provided information on fission product release rates from different defect types, water logging and fuel washout. The cladding of swage-compacted and of vibrationally compacted PRTR fuel rods was defected with holes and longitudinal slits up to 6 inches long. (119-121)

Release characteristics for the different types of defects were determined by analyzing the ratios of nuclides released to the coolant. Generally, the defect behavior of ceramic packed-particle elements has been excellent. (122) No appreciable fuel washout, fuel rod deformation, water logging effects or hydriding of the Zircaloy cladding was observed in any of the packed-particle fuel element defect tests. The only exception was the rupture of a deliberately defected $\text{UO}_2\text{-PuO}_2$ rod operating with considerable fuel melting at the plane of the defect. (123) Defect tests conducted in the Fuel Element Rupture Test Facility of the PRTR, are listed in the table on the following page. (67)

Since startup of the PRTR in July 1961, nearly 5300 fuel rods were irradiated prior to the present High Power Density Core which started in 1967. Of the 5300 fuel elements, most of which were 19-rod cluster bundles, only 38 failed in service (1 UO_2 , 3 Al-Pu , and 34 $\text{UO}_2\text{-PuO}_2$). Most of these failures occurred early in the history of the new fuel element developmental technology, particularly that of the packed-particle $\text{UO}_2\text{-PuO}_2$ elements. All of the $\text{UO}_2\text{-PuO}_2$ failures, with one exception were attributed to impurities in the fuel: (1) high fluoride in the plutonium, (2) sorbed moisture, and (3) traces of hydrocarbons introduced by failure of mechanical processing equipment. These failures characteristically occurred at low burnups as a result of severe internal hydriding and embrittlement of cladding caused by a gas phase hydriding mechanism. Only one failure has occurred since

COOLANT ACTIVITIES FROM DEFECTED PRTR ELEMENTS IRRADIATED IN THE FERTF

Test No.	Element Type	Cladding Defect	Maximum Rod Power, (kW/ft)	Loop Activity, (cpm)			Type of Release
				Steady-State GM ($\beta\gamma$)	DN (Delayed Neutron)	Burst Activity GM ($\beta\gamma$)	
A	A1-Pu FE-5008	6" cracks	~ 7.0	15,000	13,000	---	Recoil ^(a)
B	Swaged UO ₂ FE-1069	0.06" diam. hole	7.3	8,000	4,000	220,000	Equilibrium ^(b)
C	Swaged UO ₂ FE-1039	0.62" long Longitudinal Slit	7.0	~ 23,000 ^(c)	~ 13,000 ^(c)	700,000	---
D	Swaged UO ₂ FE-1039	3.2" long Longitudinal Slit	7.0	23,000	13,000	100,000	Diffusion ^(d)
E	Swaged UO ₂ FE-1030	6.5" long Longitudinal Slit	6.1	68,000	38,000	150,000	Diffusion
F	Vibrationally Compacted UO ₂ FE-1067	3.0" long Longitudinal Slit	7.3	47,000	26,000	170,000	Diffusion
G	Vibrationally Compacted UO ₂ FE-1067	3.9" long Longitudinal Slit	7.3	60,000	31,000	---	Diffusion
H	Vibrationally Compacted UO ₂ 2 wt% PuO ₂ FE-6004	0.06" diam. hole	24	80,000	4,000	660,000	Equilibrium
I	Vibrationally Compacted UO ₂ 4 wt% PuO ₂ FE-6504	0.06" diam. hole	27	7.7×10^6 ^(c)	1.6×10^5 ^(c)	---	(Ruptured)

- (a) Good communication between fuel and coolant - zero delay time.
 (b) Poor communication between fuel and coolant - long delay time.
 (c) Test was discontinued before stable form of release was established.
 (d) Good communication between fuel and coolant - short delay time.

these impurities were identified and their acceptable limits imposed in fuel specifications.

Even though severe localized embrittlement and loss of cladding fragments occurred in some instances, with one exception, little or no fuel loss into coolant resulted and no severe reactor operating difficulties were experienced.

Fuel washout resistance was improved as a result of in-reactor sintering which occurred during the early stages of irradiation. No water logging occurred in in-service failed and intentionally defected elements. In many instances, the release of fission products was first detected during post-shutdown depressurization of the reactor system.

An intentionally defected Zircaloy-clad, oxide fuel element, irradiated in the Saxton reactor (a LW-moderated PWR) for 60 effective full-power days at a peak linear heat rating of 13.5 kW/ft, showed no evidence of water logging, gross internal hydriding of the clad, fuel washout or detectable increase in coolant activity during operation. (116)

A Zircaloy-clad UO_2 fuel rod, having an 0.015-inch hole defect leaving only one mil of cladding was irradiated in Saxton reactor for 60 effective full-power days and did not fail during operation. (116)

Stainless steel-clad fuel behaved well in the Yankee LW reactor. No failures occurred in the annealed, free-standing clad for burnups as high as 27,500 MWd/tonne U in VBWR and Dresden; however, cold-worked collapsible sheaths suffered intergranular stress-induced corrosion cracking by 5000 MWd/tonne U. However, some annealed stainless steel sheaths cracked while cold-worked free-standing sheaths did not fail in the Saxton LWR after 15,000 MWd/te U.

The initial fuel cladding for NPD had a high deuterium concentration after irradiation. This was due to the poor quality control of the Zircaloy-2, which was not quenched during its fabrication,

and the high deuterium concentration (greater than 1.5 ml D₂/kg D₂O) in the coolant. The Douglas Point cladding, in the same coolant chemistry, showed less D₂ pickup because the forged billets were quenched before extrusion. The Douglas Point cladding exposed to low deuterium concentration (less than 1.0 ml D₂/kg D₂O) in the coolant has the lowest amount and the lowest pickup rate. Ni-free Zircaloy-2 irradiated in the same coolant with less than 1 ml D₂/kg D₂O contained a lower average D₂ concentration than Zircaloy-2, but by only a few ppm. The Canadians are using Zr-4 in their latest fuel bundle design. This choice was made on the better performance of the material with respect to hydrogen pickup.

1. Tubular Elements

Tubular elements for various reactors were designed for:

<u>Reactor</u>	<u>Burnup MWd/tonne</u>	<u>Maximum Fuel Temperature (°F)</u>	<u>On-Line Refueling</u>
HACTR	30,000 ⁽¹³³⁾	--	No
SGHR	19,800	3450	Yes
PRTR	5,000 - 7,000	--	No
HWR-1	14,000	--	Yes
HWR-2	15,000	--	--
HWR-3	8,500	--	Yes

Tubular UO₂ fuel elements⁽⁶⁴⁾ were operated in the HACTR at $\int k d\theta$ of:

<u>Long (~ 10 ft) Tubes</u>	<u>Short (~ 1 ft) Tubes</u>
Average 14-20 W/cm	19-66 W/cm
Maximum 14-33 W/cm	25-68 W/cm

The maximum burnup attained ranged from 1,100 to 17,300 MWd/tonne U for short tubes and from 1,700 to 12,400 MWd/tonne with long tubes. HACTR studies⁽⁶³⁾ with tubular UO₂ fuel elements showed that a columnar grain growth began and fission gas release

increased sharply at an $\int kd\theta$ of 25 to 30 W/cm. Core melting was observed at about 60 W/cm and core sintering began at ~ 5 W/cm.

One tube-in-shell, Zircaloy-clad UO_2 packed-particle fuel element was irradiated at low power in the PRTR to a burnup of about 150 MWd/tonne.⁽⁶⁷⁾ Canadian studies⁽⁶⁵⁾ included a tube-in-shell element and some ex-reactor tests on a 16-foot tubes. Tube-in-shell superheater elements for BONUS (second generation) were developed to the point of in-reactor proof tests.⁽⁶⁸⁾

HWCTR studies^(63,64) indicated that tubular fuel elements failed from dimensional instability at about 3500 MWd/tonne at an $\int kd\theta$ about 66 W/cm. Failure of two long tubular elements was attributed to vibrational damage. Out of a total of about 100 tubular element irradiations, 6 failed (2 were ~ 10 -foot elements and 4 were ~ 1 -foot elements).

HWCTR studies indicated that large tubes of fused UO_2 mechanically compacted in Zircaloy-2 sheathing appeared to be practical elements for HW-moderated power reactors. The authors concluded that resistance of the tubes to sheath collapse and wrinkling by coolant pressure was good at an $\int kd\theta$ of 40-50 W/cm if fuel bulk density was $\geq 90\%$ T.D.

One vibrationally compacted UO_2 tubular element was irradiated in the PRTR to a maximum burnup of 1500 MWd/tonne. The element (2.95-inches O.D. by 88-inches long) operated successfully at low power for eight months.^(124,125) Some in-reactor sintering of the UO_2 occurred.

The only tubular oxide fuel elements currently operating in a HW-moderated power reactor are the superheat elements in the SGHWR. The reactor went critical in September 1967, and on power in 1968.

No tube-in-shell elements are in currently operating HW-moderated power reactors.

One Zircaloy-clad UO_2 tube-in-shell was irradiated for 500 hours at low power in the PRTR to a maximum burnup of 150 MWd/tonne. The element (3.23-inches O.D. and 97-inches long) had seven internal cooling channels and operated at a tube power of 607 kW.^(126,127) Maximum fuel temperatures were 1500-1600°C. Cooling channels were 0.505-inch I.D. and fuel bulk density was 81% T.D.

The excellent fuel element behavior in the Canadian NPD was reported in the previous section, wherein only five elements failed because of fueling incidents during more than three years of operation.

2. Rod Cluster Elements

The Plutonium Recycle Test Reactor (PRTR), in support of the Plutonium Utilization Program, has produced over 43,000 MWd of heat energy. Approximately 360 experimental fuel elements were irradiated in the PRTR. Utilizing a basic 19-rod cluster fuel element design, such fuel materials as UO_2 , Al-Pu alloy, and UO_2 - PuO_2 were evaluated. Packed-particle oxide fuels fabricated by swaging⁽¹¹⁹⁾ or by vibrational compaction^(120,121) have been emphasized. Over 10 tonnes of UO_2 - PuO_2 packed-particle fuel have been irradiated in the PRTR. Fuel material was prepared by mechanically mixing UO_2 and PuO_2 powder and high energy-rate impacted to yield high density particles. Plutonium compositions ranged from 0.5 to 4.0 wt% PuO_2 in UO_2 . Maximum burnups of over 11,000 MWd/tonne and maximum central fuel temperatures above melting were attained with prototypic UO_2 - PuO_2 power reactor fuel assemblies.

The Batch Core Experiment in progress in the PRTR is providing statistically significant irradiation behavior data on High Power Density UO_2 - PuO_2 fuel elements operated at power generations greater than those currently employed in commercial power reactors. Maximum linear rod power generations are 20 kW/ft with maximum fuel temperatures near melting. Goal burnup is over 13,000 MWd/tonne.

The comparative performance of particle and pellet fuels is also being evaluated. Two vibrationally compacted $\text{UO}_2\text{-PuO}_2$ elements instrumented to measure plenum gas temperature and pressure are included in the Batch Core loading. Results of the program to date indicated that for the low plutonium concentrations of interest in thermal reactor fuels the performance characteristics were not significantly different for UO_2 and $\text{UO}_2\text{-PuO}_2$ fuels. Experience with in-service failed and deliberately defected packed-particle fuels has been very encouraging. Deliberately defected irradiation tests with $\text{UO}_2\text{-PuO}_2$ fuels, some operating under molten core conditions, were conducted in the Fuel Element Rupture Test Facility (FERTF), a pressurized water loop in the PRTR.

Concurrent with the Batch Core Experiment, irradiation tests of $\text{UO}_2\text{-PuO}_2$ fuel rods utilizing an 8-rod test element are being conducted at higher power generations in the FERTF. The effects of fuel form (powder, pellet), power generation (20 to 28 kW/ft), and defect behavior on fuel performance will be evaluated.

A summary of the fuel elements irradiated in the PRTR is given in Table I, and a **summary** of the present core loading status as of August 1967 is presented in **Table II**.

An important part of the Plutonium Utilization Program fuel development studies is to procure and test plutonium fuel rods designed and fabricated by commercial fuel vendors. A 19-rod cluster of commercially designed and fabricated fuel rods containing hot-pressed $\text{UO}_2 - 1.94 \text{ wt\% PuO}_2$ pellets was irradiated in the FERTF at a linear rod power of $21.2 \pm 1.0 \text{ kW/ft}$ to an average burnup of 1900 Mwd/tonne.

Periodic examination of the element in the basin revealed that the element performed satisfactorily and no adverse irradiation effects were observed. Post-irradiation examination of a rod

TABLE I
PRTR FUEL ELEMENT IRRADIATION SUMMARY
(as of August 1967)

Element Type	Number of Elements (subtotals)	Maximum Linear Rod Power, kW/ft	Maximum Element Burnup, MWd/tonne
Al-Pu	75	~ 15	(~ 80% of Pu)
UO ₂	68		
Vipac	(1)	~ 10	2,200
Swaged (1 Hot-Swaged)	(65)	~ 14	9,400
Vipac Tubular	(1)	--	1,500
Vipac Inverted Cluster	(1)	--	150
UO ₂ -PuO ₂	216		
UO ₂ - 0.5 wt% PuO ₂	(81)		
Vipac	(20)*	~ 16	11,000'
Swaged	(61)	~ 13	10,200
UO ₂ - 1.0 wt% PuO ₂	(49)		
Vipac	(16)	~ 15	6,300
Swaged	(33)	~ 17	8,500
UO ₂ - 2.0 wt% PuO ₂	(84)		
Vipac	(79)	~ 20	5,400
Swaged	(2)	~ 20	3,200
Pellet (Hot-Pressed)	(2)	~ 21.5	1,900
Vipac Salt Cycle	(1)	~ 17	1,600
UO ₂ - 4.0 wt% PuO ₂	(1)	~ 27	1,100
UO ₂ - 1.5 wt% PuO ₂	(1)	~ 6	3,000
MgO - 2.1 wt% PuO ₂ (Swaged)	1	~ 12	--
ThO ₂ - 5 wt% PuO ₂	1	~ 10	--

*A rod power of 20.5 ± 0.5 kW/ft, which produces onset of center melting, equivalent to an $\int k d\theta$ of about 68 W/cm.

TABLE II
PRTR CORE LOADING STATUS
(as of August 1967)

<u>Element Type</u> ⁽¹⁾	<u>Elements</u>	<u>Maximum Rod Power Generation (kW/ft)</u>	<u>Burnup (MWd/tonne)</u>
<u>Batch Core Experiment:</u>			
Vipac PI ⁽²⁾ UO ₂ - 2 wt% PuO ₂ (Batch Core Experiment Elements)	66	~ 20	2,100
Hot-Pressed Pellet UO ₂ - 1.94 wt% PuO ₂	1	~ 21.5	1,900
<u>Fringe Position Tests:</u>			
Vipac PI UO ₂ - 2 wt% PuO ₂ (Pilot HPD ² Elements)	4	~ 20	5,400
Swaged PI UO ₂ - 2 wt% PuO ₂	1	~ 20	3,200
Swaged PI UO ₂ - 1 wt% PuO ₂	1	~ 17	8,500
Vipac PI UO ₂ - 1 wt% PuO ₂	2	~ 15	6,300
Vipac PI UO ₂ - 1.5 wt% PuO ₂	1	~ 6	3,000
Vipac MM ⁽³⁾ UO ₂ - 0.5 wt% PuO ₂	1	~ 16	11,000
Swaged MM UO ₂ - 0.5 wt% PuO ₂	2	~ 13	10,200
Swaged UO ₂	1	~ 14	9,400

(1) PRTR elements are 19-rod clusters of 0.565 O.D. Zircaloy-clad rods with active fuel lengths of 58.5 and 88.5 inches.

(2) PI = High energy-rate pneumatically impacted fuel.

(3) MM = Mechanically mixed fuel.

indicated that incipient fuel melting occurred. Irradiation of this pellet rod cluster will be continued in the Batch Core. A second pellet rod cluster will also be irradiated in the Batch Core.

The UO_2 pellet fuel elements in CVTR were clad in 10% cold-worked Zircaloy-4, operated at a peak linear power rating of 17 kW/ft and were scheduled to operate until late 1967 to a peak burnup of 44,000 MWd/tonne.⁽¹¹⁶⁾ The burnup as of December 1966, was $\sim 19,800$ MWd/tonne. The CVTR program was cancelled prior to goal burnup.

B. BWR Fuel Element Performance

The only operating BWR that uses heavy water for both moderator and coolant is the Halden reactor in Norway. This reactor operated satisfactorily for five years but was shut down most of 1967. The first core was uranium metal clad in finned aluminum but the second core was UO_2 clad in Zircaloy. The MARVIKEN reactor in Sweden, also HW moderated and cooled, was commissioned in 1968.

Reactors which use heavy water moderator and light boiling water coolant include the UK-SCHWR, which went critical in 1967, and the Canadian Gentilly reactor scheduled for operation in 1971. The ND 19.3 MW(e) PHWR at Rolphton, Ontario, will be converted to a BHW under current plans by AECL. It is thought that the operating costs of large commercial plants such as Pickering can be reduced by as much as 0.1 to 0.2 mils per kWh by this treatment. The conversion of NPD will help to determine such factors as reactor stability and control, startup and cycling.

The design of the Gentilly boiling light water fuel bundle is based on the 19-rod geometry, containing 18 fuel elements arranged in two rings about an unfueled central tubular tie rod, with the fuel rod O.D. increased to 19.74 mm to fill the larger diameter (103.4 mm) pressure tube. The nominal inter-rod clearance was reduced to 1.03 mm from the 1.27 mm used in the PHW design. The larger diameter

and smaller spacing were necessary to minimize the light water holdup in the core and to improve core burnup. Elements operating at these higher heat ratings were designed to allow a total diametral strain of 0.8%. This strain may be produced by both the thermal expansion of the UO_2 and the internal fission gas pressure being greater than that of the coolant at fuel burnups greater than 10,000 MWd/tonne U. Axial strain produced by thermal expansion of the UO_2 was limited by dishing one end of each pellet. Creep strain due to fission gas pressure was limited by providing a cold void in the elements with hollow end caps.

The UKSGWR is a 97 MW(e) reactor using a 36-rod Zr-2 clad UO_2 fuel cluster in the boiling region and a 23-tube cluster in the superheat region. The reactor was commissioned in September 1967, and went on power operation in 1968.

The Gentilly type fuel elements were irradiation tested as a part of the Canadian BLW cooled fuel program. No in-reactor fretting greater than 0.05 mm was observed on inter-rod spacers of moderate exposure to 10 wt% steam quality at a mass velocity of $440 \text{ g/cm}^2\text{-sec}$.

Although BWR elements usually contain sintered UO_2 pellets, a promising new process, vibratory compaction, offers potentially lower fabrication costs. Several hundred Zircaloy-2, compacted- UO_2 particle fuel rods are under irradiation in the Dresden, Big Rock Point and JPDR reactors. So far, pellet and particle fuel have performed equally well.

GE-San Jose reported⁽¹²⁸⁾ that stainless steel cladding in thermal reactors was susceptible to cracking above 7500 MWd/tonne. Experience with stainless steel clad UO_2 fuel rods (pellet and packed-particle) in the LW-moderated VBWR showed that Type-304 stainless steel was susceptible to stress and subsequent intergranular corrosion attack.⁽¹²⁹⁾ Failure of over 40 fuel rods clad with 304 stainless steel occurred during irradiation of 950 fuel rods of several types.

VII. FABRICATION COSTS AND STATUS FOR CERAMIC FUELS FABRICATION

A. Fabrication Costs

Fuel fabrication costs show a downward trend over the past years and, with a recent Canadian announcement of only \$44/kg U as the contracted price for Pickering natural UO_2 fuel, the trend appears to be continuing down. Most studies project this trend into the future although there is concern about increased uranium costs and the necessity of plutonium recycle.

Since the Geneva II Conference, the fuel cycle cost for power reactors has been reduced by an approximate factor of two. Longer fuel life, higher fuel power density and lower fabrication costs have been contributing factors. A comparison of fuel fabrication costs today is most difficult because there is no "standard" fuel element.

1. Uranium Dioxide Fuels

Fuel fabrication charges are assumed to cover all costs from UF_6 withdrawal to delivery of the fuel elements to the reactor site unless otherwise specified.

Page⁽¹¹⁷⁾ recently stated that a firm price order by Ontario Hydro was awarded to Canadian General Electric, Ltd., for the first four charges of the Pickering 500 MW(e) reactors involving approximately 19,000 bundles or 375,000 kg of uranium. The fuel cost was \$44/kg U (1967 Canadian dollars) which will produce a fueling cost of 0.68 mils/kW at an average core burnup of 9,300 MWd/tonne and a station efficiency of 29.1%. The low cost was reported possible due to the use of natural UO_2 , the large production throughput of the same design which will be 85,000 kg U per year for at least four years, large bulk UO_2 orders, decreased Zircaloy clad costs and improvements in the fabrication process. The first Pickering reactor, now under construction, is scheduled to go critical in 1970 with a first fuel loading of natural UO_2 sintered pellets clad in Zircaloy-4. The fuel rods will be

15.25 mm O.D. by 49.5 cm long with an 0.38 mm clad; 28 rods per fuel bundle and 12 bundles per channel.

Two recent fuel cost quotations were published in the U. S. for enriched UO_2 fuels procurement. Westinghouse was reported as bidding \$100/kg U for the Sacramento Metropolitan Utilities Department (SMUD) 800 MW(e) PWR first core scheduled for 1973. Three different vendor prices were quoted for the Bolsa Island 900 MW(e) core.

	<u>\$ per kg U</u>				
	<u>GE</u>	<u>B&W</u>	<u>Westinghouse</u>		
Initial Core	78.28	79.49	100		
Core I	72.43	78.08	87.50	82.50	77.50
Core II	68.02	78.08	74.50	71.50	69.50

The exact **burnup** or enrichment is not known for this fuel but the ~~GE-BWR~~ price is reported based on something similar to the Niagara Mohawk which is 2.1% enriched. The Westinghouse **three-zone PWR** probably varies between 2 and 4% enrichment.

Nearly all **commercial** water reactor fuels are fabricated by the pellet process. The above contracted and bid vendor prices indicate the pellet fuel prices for the immediate future. There is really no way to compare other fuel process prices to these. Other fuel processes are usually for much smaller cores and usually of more specialized design; while fuel cycle studies of pelleted fuels assume relatively large throughputs. Although Big Rock Point is now operating with a 2/3 packed-particle fuel core, no **commercial** fuel costs for vibratory packed fuels or for swage compacted fuels have been **publicly** quoted for **commercial** power reactors.

ORNL⁽¹³⁰⁾ estimated \$62/kg, in 1963, for a stainless steel clad 2.46% enriched UO_2 tubular element for use in the EGCR, but the reactor study was terminated.

2. UO₂-PuO₂ and Other Oxide Fuels

Other fuel cost studies are summarized for special fuel fabrication operations.

A series of UO₂-PuO₂ mixed oxide fuel fabrication cost analyses were made by C. H. Bloomster⁽¹³¹⁾ for small test or prototype reactor fuels. The fuels were compacted by the high energy pneumatic impaction process and, after sizing, were vibrationally compacted into tubes.

A comparative cost study by PNL⁽¹³²⁾ of oxide fuel fabrication attempts to standardize and relate the effect of various fuel fabrication parameters to costs for UO₂-PuO₂ and enriched UO₂ combinations. The study indicated a questionable difference between pellet and vibrational compaction loading techniques, and indicated a 20% difference between ²³⁵UO₂ and PuO₂ enrichment. Nearly one half of the differential was due to the high cost for conversion of plutonium nitrate to the oxide. A minimum differential of 4% was shown between high and low exposure plutonium for enrichment, depending upon the time that the high exposure plutonium was used after chemical separation.

Estimated base fabrication costs are shown in the following table:

<u>FABRICATION COSTS</u>			
<u>Fuel</u>	<u>VIPAC</u>	<u>\$/kg (U+Pu)</u>	<u>Pellet & Sinter - \$/kg/U+Pu</u>
Enriched UO ₂		41.69	43.36
Low Exposure PuO ₂	(< 10% ²⁴⁰ Pu)	50.79	51.90
High Exposure PuO ₂	(> 10% ²⁴⁰ Pu)	52.40	43.47

However, U. S. industry has not yet achieved the throughput, standard design, confidence and experience to achieve these base prices.

3. Other Ceramic Fuels

The Indian Point and Elk River reactors used $\text{ThO}_2\text{-UO}_2$ fuel and Peach Bottom used ThC-UC, but there is little fuel fabrication cost information available in the literature.

Magnus⁽¹³³⁾ quoted a UC fuel fabrication cost of \$74.81/kg U in a study projecting a 20-element pilot plant operating at a 500 kg/week throughput basis. Costs were \$107/kg U for the 20 elements fabricated in 1963.

Magnus⁽¹³⁴⁾ reported a fuel fabrication cost of \$66.97/kg U for the Hallam reactor second core. Stainless steel clad, 8-rod elements were produced containing Na bonded, 4.9% enriched UC pellets, 0.852-inch diameter. Throughput was slightly over 2000 kg of fuel per month.

Washburn and Lotts⁽¹³⁵⁾ quoted a fuel fabrication cost for a low temperature desalination reactor study at \$77/kg (U+Pu) based on a 660 kg/day (heavy metal) throughput.

Washburn, Lotts and Harrington⁽¹³⁶⁾ estimated fuel fabrication costs for $(\text{U}_{0.82}\text{Pu}_{0.18})\text{O}_2$ fuel elements for fast reactors. Four processes (550 kg per day) were compared: (1) low energy packing of microspheres, \$331/kg; (2) vibratory compaction, \$337/kg; (3) pelletizing, \$342/kg; and (4) sol-gel extrusion, \$331/kg.

B. Status on Fabrication⁽¹³⁷⁾

1. United States

a. Uranium Processing and Fabrication

Adequate industrial capability and competition exist today to process and fabricate depleted, normal and enriched uranium to the various fuel forms required in research, test, and power reactors.

The principal producers of depleted, normal and enriched uranium metal, oxides and compounds are:

- General Electric Company, San Jose, California
Kerr-McGee Oil Industries, Inc., Oklahoma City, Oklahoma
National Lead Company, Albany, New York
- Nuclear Fuel Services, Inc., Erwin, Tennessee
- Nuclear Materials and Equipment Corporation, Apollo, Pennsylvania
- United Nuclear Corporation, Hematite, Missouri

UO₂ Fuels

Uranium dioxide is used for all the light water reactors being built or ordered. Fuel manufacturers are adding to their fabrication capabilities, as follows:

- Combustion Engineering is building a fuel fabrication facility addition to their Windsor, Connecticut, plant.
- General Electric is constructing a plant near Wilmington, North Carolina, scheduled for operation in 1969, which will produce fuel for nuclear power plants as well as equipment components.
- Babcock & Wilcox Company is building a commercial reactor fuel fabrication plant in Campbell County, Virginia, at a first-phase cost in excess of \$2 million. The plant is scheduled for operation by 1970.
- Westinghouse is building a \$20 million manufacturing plant near Columbia, South Carolina, to fabricate nuclear fuel elements for commercial nuclear power plants. Initial operation is scheduled for the third quarter of 1969.

The principal fabricators of UO₂ fuels are:

- Atomics International, Canoga Park, California
- Babcock & Wilcox Company, Lynchburg, Virginia
- Combustion Engineering, Inc., Windsor, Connecticut
- Gulf General Atomics, San Diego, California

- General Electric Company, San Jose, California
- National Lead Company, Albany, New York
- Nuclear Fuel Services, Inc. , Erwin, Tennessee
- Nuclear Materials and Equipment Corporation, Apollo, Pennsylvania
- United Nuclear Corporation, New Haven, Connecticut
- Westinghouse Electric Corporation, Pittsburgh, Pennsylvania

b. Plutonium Fuels

A great amount of interest has been expressed by nuclear utilities and fuel cycle suppliers in the value of plutonium as a fuel to recycle in thermal reactors and its impact on the fuel cycle and power cost estimates for reactor systems. In addition to its effect on thermal reactor system economics, plutonium used in a reactor as a fuel in place of U-235 provides greater flexibility of independence from an enriched uranium supply and also permits extending the available supply of fissionable material through breeding.

Private firms having various degrees of capability in plutonium processing are:

	<u>Processing</u>	<u>Fuel Fabri- cation</u>	<u>Scrap Recovery</u>	<u>Source Fabri- cation</u>	<u>R&D</u>
Atomics International	X	X			X
Babcock & Wilcox					X
Battelle Memorial Institute (Columbus, Ohio)					X
Carboryndum Co.					X
General Electric Co.	X	X			X
Monsanto Research Co.				X	X
Nuclear Fuel Services	X	X			X
Nuclear Materials & Equipment Corp.	X	X	X	X	X
United Nuclear Corp.					X

c. Thorium

The present need for thorium as a fertile fuel continues to be limited primarily to research and development. The development of certain advanced converter and breeder reactors for commercial applications could eventually expand the use of thorium as a source of nuclear fuel.

The principal firms producing thorium are:

- American Potash & Chemical Corporation, Chicago, Illinois
- Babcock & Wilcox Company, Lynchburg, Virginia
- The Dow Chemical Company, Midland, Michigan
- General Dynamics Corporation, San Diego, California
- Kerr-McGee Oil Industries, Inc., Cusning, Oklahoma
- Metal Hydrides, Inc., Beverly, Massachusetts
- National Lead Company, Albany, New York
- Nuclear Fuel Services, Inc., Erwin, Tennessee
- Nuclear Materials and Equipment Corporation, Apollo, Pennsylvania
- United Nuclear Corporation, Hematite, Missouri

d. Uranium-233

The present need for uranium-233 continues to be limited and is essentially of a developmental nature. Due to the interest in the thorium - uranium-233 fuel cycle, development work in this area will no doubt increase in the future.

The principal companies having U-233 processing and fabrication capabilities are Nuclear Fuel Services, Nuclear Materials and Equipment Corporation, and Babcock & Wilcox Company.

e. Fuel Cycle Arrangements

Nuclear utilities can select from a multitude of fuel cycle arrangements being offered by both the reactor manufacturer, as well as independent fuel cycle suppliers. The range varies from that of the reactor manufacturer's full fuel cycle service

to that of the utility controlling many of the major steps in the fuel cycle.

f. Fuel Core Manufacture

United Nuclear Corporation is the only independent producer so far that makes UO_2 powder and pellets, fabricates the complete reactor core for commercial reactors and that has bid and received orders for replacement cores. Commonwealth Edison's award of a replacement core for the GE Dresden I reactor to United Nuclear during 1966 was the first breakthrough of the independent supplier in the replacement core business.

The potential market in the nuclear fuel cycle is sufficiently attractive that several companies are making plans to enter all phases of the fuel cycle even though the magnitude of business may not be adequate to assure a reasonable return on an investment until the mid-1970's.

At the present time, Kerr-McGee, Nuclear Fuel Services, and Atlantic Richfield (NUMCO) produce UO_2 powder and pellets for sale to the reactor manufacturer. The Gulf Oil Corporation has indicated plans to enter the total fuel cycle business, and has acquired General Atomics. The National Lead Company has indicated plans to enter the replacement core business and other steps in the fuel cycle such as fuel reprocessing. Aerojet-General states it could have an automated fuel production facility in operation during 1972-1973. It would convert UF_6 to UO_2 , as well as fabricate fuel elements. Atomics International is involved in reactor development with a major effort in fast breeder reactors and plans for replacement fuel business, including plutonium recycle.

The reprocessing of fuel elements and handling and storage of the residual radioactive material is presently being performed by one commercial firm, Nuclear Fuel Services. General Electric Company has applied for a construction permit for a

chemical reprocessing plant and Allied Chemical Corporation has indicated intentions to reactivate their reprocessing plant. National Lead has also announced studies for a chemical reprocessing plant. Atlantic Richfield, who acquired NUMEC and is operating the chemical reprocessing facilities for the AEC at Richland, has indicated that they would eventually consider building a chemical reprocessing plant for commercial purposes at Richland, Washington. At present, the market is insufficient to operate the NRS plant at capacity, and a market to support a second plant probably will not be available until about 1973.

g. Enriched Uranium Scrap Recovery

The major source of enriched uranium scrap results from the fuel fabrication process. Since the nuclear material involved in this scrap is quite valuable, its recovery is economically necessary. Normally a large part of the scrap generated during fabrication is recycled into the process stream by the fabricator. Only a small percentage, estimated at 1.0 to 1.5, of the scrap cannot be recycled directly into the fabrication line and must be purified by an anion exchange or solvent extraction process.

Adequate private capability exists to recover enriched uranium from all types and forms of scrap generated within both private and AEC programs. These firms are:

- Gulf General Atomics, San Diego, California
- Kerr-McGee Oil Industries, Inc., Oklahoma City, Oklahoma
- Nuclear Fuel Services, Inc., Erwin, Tennessee
- Nuclear Materials and Equipment Corporation, Apollo, Pennsylvania
- United Nuclear Corporation, Westerly, Rhode Island

2. Canada

Canadian General Electric Company, Ltd. , Peterborough, Ontario, has fabricated several reactor fuel cores consisting of sintered UO_2 pellets, natural and enriched, clad in Zircaloy. Reactors include CANDU-PWR, Pickering , and the CANDU-type under construction overseas.

Canadian Westinghouse Company, Ltd. , Port Hope, Ontario, fabricated uranium metal shapes and UO_2 pellets and partial or full initial cores for the RAPPI (India) and Gentilly.

Eldorado Nuclear Company, Ltd. , Port Hope, Ontario, has complete facilities for conversion of yellow cake to natural UO_2 by calcination and reduction of uranyl nitrate or by precipitation and reduction of ammonium diuranate. The latter route yields a reproducibly sinterable UO_2 powder.

3. Europe*

a. Germany

NUKEM - Fabrication of sintered natural and enriched UO_2 shapes, including fuel element assembly. Research and development work on vibrational compaction, swaging and fusion of UO_2 . Fabrication of UC and UC_2 fuels by sintering and by skull casting. Conversion facilities for UF_6 to UO_2 . Ceramic fuel elements were supplied for the KRB, KNK FR-2 reactors (Germany) and the SENA and ESSOR reactors (Italy).

ALKEM - Fabrication of plutonium-bearing ceramic fuel elements. Plate elements were fabricated for the SNEAK.

AEG - Fabrication of UO_2 pellets.

b. France

CERCA - Conversion of UF_6 to UO_2 . Fabrication of UO_2 pellets.

*Fuel fabrication capabilities in Europe and Asia are briefly tabulated and do not represent complete fuels capability.

CEA - Conversion of UF_6 and UHN to UO_2 via ammonium diuranate route. Fabrication of UO_2 and $\text{UO}_2\text{-PuO}_2$ pellets and fuel assembly. Nearly all fuels fabrication is performed within the French AEC complex.

CICAF - Fabrication of UO_2 and UC shapes.

c. Sweden

A. B. Atomenergi - This government owned facility is engaged in mining, milling, yellow cake conversion to ceramic grade UO_2 and fuels fabrication.

ASEA - Fabrication of uranium and UO_2 fuels.

d. Italy

FIAT-Breda established a new facility, "Fuels for Nuclear Reactors, Inc.," near Saluggia to fabricate UO_2 fuels.

General Electric and Ansaldo are building a new facility, "Nuclear Fabrication, Inc.," near Genoa.

e. England

UK-AEA - This facility is engaged in nearly all areas of fuels fabrication--uranium, plutonium, oxides, carbides and cermets. A new, fully automated UF_6 to UO_2 conversion plant employing a new process was scheduled for operation in 1968.

f. Belguim

Metalurgie et Mechanique Nucléaires does fuel fabrication work with uranium, uranium alloys, and UO_2 . BelgoNucléaire is engaged in processing and fabricating U and Pu ceramic fuels.

g. Netherlands

Neratoom, N.V. - Fabrication of UO_2 fuels.

4. Asia

Japan

Mitsubishi Atomic Power Industries, Inc. - Preparation and fabrication of uranium, alloys, UO_2 powder and pellets, uranium carbides and ThO_2 .

Sumitomo Atomic Energy Industries, Inc., is engaged in fabrication of uranium, alloys, dispersions and carbides.

Japan Nuclear Fuel Company was organized for fabrication of UO_2 fuels in the near future. The company is owned by General Electric, Hitachi and a Japanese utility.

VIII. CAPABILITIES FOR CERAMIC FUELS

A. Linear Power Densities

1. Present Designs

The physical factors limiting the power output of fuel rods, and hence the center fuel temperature, were briefly described at Geneva Conference III.

Several authors stated their wish to avoid central melting, but this was not yet seriously restrictive as there was general agreement that an $\int_{k_{eff}}^{T_m}$ of about 90 W/cm would be required. The wish to avoid excessive sheath strain due either to fuel expansion or fission gas pressure was also noted, but no arguments were advanced to show that these factors limited present ratings. In several designs the real limitation may have been on surface heat flux to avoid burnout in the coolant. Alternatively, the present power ratings may reflect the current lack of knowledge concerning fuel behavior. Experimental results reported for both fuel expansion and gas release gave confidence in increased power output. Although experimental UO_2 fuel elements have operated above the melting point without failure, it was postulated⁽¹³⁸⁾ that power transients, unpredictability of fuel movement and the effects of volume expansion on cladding integrity were sufficiently great that melting represents an operational limit for present fuel element designs. In typical PWR and BWR plants, the present transient for melting with Zircaloy-clad fuel elements is 118 - 130% of maximum steady-state power.

2. Proposed Designs and Limitations

The trend in BWR⁽¹³⁹⁾ fuel design is to higher core power density (38% increase since Dresden-2) and longer fuel life (19,000 Mwd/tonne first core, 25,000 Mwd/tonne equilibrium core). Burnable poisons are used to permit the higher exposures without an increase in reactivity worth of control rods. Both high power density and the burnable poison concept were demonstrated in boiling water power reactors. Europium and gadolinium are being used as burnable poisons in Dresden, and gadolinium is being used in Big Rock Point. The Big Rock Point plant has achieved a power density of 60 kW/liter.

Among the first fuel irradiations in the PRTR⁽⁶⁷⁾ Batch Core Experiment were tests to determine the maximum allowable reactor power level based on a maximum fuel temperature limit. The limiting PRTR power level was established by the linear rod power that produced fuel temperatures just below melting. Ceramographic examination of thirteen irradiated fuel rods revealed that melting in vibrationally compacted $\text{UO}_2 - 2 \text{ wt\% PuO}_2$ fuel rods occurred at $20.5 \pm 0.5 \text{ kW/ft}$, or at an $\int k d\theta$ of about 68 W/cm. These examinations also revealed that: columnar grain growth began at approximately 2000°C for irradiation times up to 80 hours; a typical once-molten fuel structure, formed during irradiation, was erased by a time-temperature dependent diffusion phenomena in less than 72 hours irradiation under non-molten, but high temperature conditions; fission product distribution patterns typical of molten fuel operation, as indicated by β - γ autoradiographs were not completely eradicated during 72 hours of subsequent non-molten irradiation; fuel structures formed in different rods irradiated under the same non-molten conditions were comparable; and structural equilibrium was reached more rapidly at the higher power generations.

A review⁽⁶⁹⁾ of fuel performance in light water reactors revealed that perhaps 10-15% more heat could be attained from current cores with only modest fuel element design changes.

Canadian tests with 19-, 22-, and 28-rod bundles for the BLW-coolant version of CANDU indicated a burnout heat flux of 10 W/cm^2 .⁽⁶⁵⁾

B. Burnup

From the utilities viewpoint the structural integrity of a fuel element to its designed burnup is of primary importance.⁽¹⁴⁰⁾ Limited irradiation experience indicates that UO_2 should be capable of achieving better than 50,000 MWd/tonne of uranium. The cladding must also withstand the effects of prolonged irradiation in a high temperature water environment. The general accepted criteria⁽¹³⁸⁾ are that no fuel damage shall occur during steady-state operation, operational transients and accidental transients.

To preclude damage to fuel elements, including cladding perforation, fuel rod bowing, cladding strain and fretting damage to cladding, a knowledge of the cause-effect relationship is essential to establish design limits. Significant core operating parameters must be measured and stored for later use.

Swelling of UO_2 during irradiation is little understood but its importance as an economic factor is great.⁽¹³⁸⁾ Assuming that swelling rates in UO_2 wafers are applicable to UO_2 rod elements, present fuel rod designs would be expected to allow burnups to 45,000 to 55,000 MWd/tonne; however, the validity of this assumption has not been extensively tested and is subject to more tests.

C. Clad Capabilities

1. Production

U. S. industry has considerable capability for producing Zircaloy cladding at the present time and that capability is expanding rapidly in the U. S. A brief resumé is presented of current production capability, irradiation experience and alternative cladding alloys.

Wah Chang Corporation and AMAX Specialty Metals, Inc., produce zirconium tubing. The Sandvik Steel Works Company has constructed

a zirconium tube plant, in partnership with United Nuclear Corporation, at Richland, Washington. The company is known as the Sandvik Special Metals Corporation. General Electric and Westinghouse have announced plans to construct zirconium tubing fabrication plants. The total existing zirconium tubing fabrication capacity in the U. S. is 417 million-feet per year. Plans for expansion will increase this capacity to 9.8 million-feet by 1969. Details on the Zircaloy fabrication capabilities of the leading manufacturers are listed in the following table.

ZIRCONIUM AND ZIRCALOY PRODUCERS AND FABRICATORS

<u>Company</u>	<u>Sponge & Chunklets</u>	<u>Ingots</u>	<u>Plates & Strips</u>	<u>Tubing</u>
Carborundum Metals Climax	X	X	X	X
Chase Brass & Copper	--	--	--	L
General Electric Co.	--	--	--	o
Harvey Aluminum Co.	--	X	--	X
Jessop Steel Co.	--	--	X	--
Latrobe Steel Co.	--	--	X	--
Reactive Metals, Inc.	**	**	X	X
Sandvik Special Metals	--	--	--	o
Sandvik Steel Inc.	--	--	R	R
Superior Tube Co.	--	--	--	X
Wh Chang	X	X	X	oo
Wolverine Tube Division				
Calumet & Hecla, Inc.	--	--	--	X
Westinghouse Electric	--	o	o	o
Total Present Capability*				
Millions of lbs/yr	2.2	4.2	7.5	
Millions of linear ft/yr				3.2
Total Expected Capability				
As of January 1, 1969*				
Millions of lbs/yr	3.2	5.2	7.5	
Millions of linear ft/yr				9.8

Notes :

- * - Total capability numbers represent only the plants marked X, above
- X - Current capacity
- ** - Standby capacity
- o - Planning or developing capacity
- L - Large sizes only
- R - Representing Swedish parent company

2. General Remarks - Water Reactors

Selection of stainless steel over Zircaloy for the first and second cores of the Yankee Reactor and the first core of the Indian Point Reactor was made primarily on the basis of greater strength, cheapness, ease of fabrication and availability. Stainless steel was first preferred because of failures of Zircaloy-clad fuel in VBWR; subsequently shown to be due to fluoride impurities in the UO_2 . Zircaloy is now used because of stress-corrosion failure of stainless-steel cladding.

For zirconium alloys, the design limitation is based on embrittlement due to hydriding. A limit of about 250 ppm hydrogen is generally accepted. A USSR paper on the stress orientation of hydrides emphasized that a strong alloy that is less susceptible to orientation for a given stress should be selected. The Russians were concerned about hydriding as a limitation but also reported that sheaths prehydrided to 600 ppm survived 4000 hours in water at 300°C and 1400 lb/in^2 while the fuel operated at 520 W/cm .

Canada also considered hydride orientation and reported results showing that the embrittling effect of the hydrides was confined to temperatures under 105°C . The U. K. presented results showing that at a given temperature crack propagation occurred for a lower hydrogen content in Zircaloy-2 than in ZR - 2.5 wt% Nb.

For stainless steel the limitations appeared to be set by residual stress in the cladding and its cyclic stressing by the fuel. A U. S. presentation reported that the coolant chemistry should avoid conditions favoring stress corrosion and that surface oxide deposits concentrated chlorides onto the cladding.

Canada stated that the present lower limit for collapsible sheath thickness was set to avoid longitudinal ridges and consequent sheath failure on power cycling. Circumferential ridges at pellet interfaces were commonly seen, but did not cause trouble and were probably unaltered after initial start-up. Norway, the U. S., and

the U. K. presented semi-empirical data relating to the collapse of thin sheaths but the detailed mechanisms were not yet properly understood.

3. Zirconium

In the Geneva III Conference, corrosion of zirconium-base alloys in aqueous systems was reviewed in American and Russian papers. Zircaloy cladding has seen major service in power reactors in the United States without any problems, with the Shippingport and Dresden experience cited as examples. A 50% reduction in cost of Zircaloy-2 tubing in the past five years has added to its attractiveness as a cladding material.

Although accelerated in-reactor corrosion for Zircaloy-2 was reported, this does not provide a limit for in-reactor operation. Lower hydrogen pickup has been demonstrated for Zircaloy-4 than for Zircaloy-2; however, since hydriding has not been a problem with Zircaloy-2 in power reactors in the United States, the incentive for changing to Zircaloy-4 is not clear. The Swedish observed that the corrosion rate of Zircaloy-2 and Zircaloy-4 was dependent upon preferred hydride orientation.

Canadian interest in Zircaloy-4 stemmed from an attempt to replace the cold-worked Zircaloy-2 used for pressure tubes in NPD and CANDU with an improved alloy, although adequate performance of Zircaloy-2 in these reactors has been demonstrated. The German Federal Republic reported that Zr-Nb alloys pick up less hydrogen than Zircaloy-2 at 350°C. For creep and corrosion resistance, a Zr - 3%Nb - 1% Sn alloy was preferred. A Zr - 1.15% Cr alloy has also been selected in the United States for comparison with Zircaloy-2 in boiling water reactors.

The Russians concluded that operational experience with zirconium-base alloys as cladding has been good. They revealed that at low heat fluxes hydride tended to orient circumferentially in tubes, whereas at high heat fluxes and high radial stresses orientation

occurred. Work in the United States indicated that severe losses in ductility may take place in Zircaloy with oriented hydrides at a hydrogen level of only 37 ppm. Hydriding has not been a problem in any of the United States power reactors, and hydrogen levels to 250 ppm have little effect on the tensile properties of irradiated materials.

A Canadian paper reported a ductile to brittle transition occurred between 150°C - 200°C in hydrided Zircaloy. This observation led to the conclusion that on-line refueling was safer than other methods since the zirconium alloy cladding always operated above its brittle-ductile transition temperature. The existence of a brittle-ductile transition was also indicated by the British in the temperature range 100°C - 200°C for Zr - 2.15% Nb containing 200 to 400 ppm hydrogen. The effect of higher hydrogen concentrations was not reported.

Irradiation to 5×10^{21} nvt at 280°C was reported to have a greater effect on the room temperature tensile properties of Zircaloy-2 than similar exposure at 60°C. A marked increase in yield strength was observed, which varied according to the direction of testing with respect to rolling direction.

Creep rates of cold-worked Zircaloy-2 measured during irradiation were reported by the U. S. and by Canada. Near 300°C at a stress of 20,000 lb/in², higher rates were observed by the Canadians, compared to out-of-reactor rates. A series of U. S. tests showed that rates below 360°C were lower in-reactor while above this temperature rates were higher.

Latest reports from Canada attributed D₂ and H₂ pickup in Zircaloy-2 to the poor quality of the early Zircaloy-2 which was not quenched. There is little concern now about the effect of D₂ pickup on Zircaloy-2 sheathing because of proper heat treatment of the sheathing.

4. Stainless Steel

Corrosion of stainless steels in aqueous systems and in liquid metals was reviewed in two United States papers in the Geneva III Conference. In one of these papers the problem of stress corrosion of austenitic stainless steels was discussed together with a comparison of performance of these steels and nickel-base alloys in superheat environments. Both the U. S. and Sweden concluded that Incoloy-800 was a leading contender for superheat cladding. In the Swedish work, a dependence of corrosion rate on surface treatment was established and lowest weight gains were found for as-ground surfaces in static corrosion tests.

There was almost universal agreement among the United States, Great Britain and Russia that the high temperature ($> 500^{\circ}\text{C}$) ductility minimum observed in irradiated austenitic stainless steels was irreversible. The American paper showed that this high temperature ductility minimum was strain-rate dependent, with greater reductions in ductility occurring at the lower strain rates. There was almost universal agreement that this elevated temperature ductility effect was due to a transmutation, probably $\text{B}^{10}(\text{n},\alpha)$. The British investigated the possibility that this ductility effect was a result of precipitation observed within the grains of these irradiated steels. These precipitates went into solution at 1150°C with no recovery of ductility. However, gas bubbles were observed in the grain boundary. Since this high temperature ductility effect is accompanied by grain boundary failure, it is probable that helium is a major factor. Adjustments in B^{10} and B^{11} in these steels by the British established the thermal-neutron dependency of this ductility minimum. The Russians also indicated the probable source of hydrogen through the $\text{Ni-58}(\text{n},\text{p})\text{CO-58}$ transmutation.

The British and the Americans agreed that a low temperature ductility minimum, at about 250°C , existed for austenitic steels after irradiation at temperatures below 300°C . This low temperature minimum was associated with a large decrease in uniform strain

resulting from defects produced by fast neutrons. Fracture was primarily transgranular as opposed to the intergranular fracture noted for the high temperature ductility minimum, which was thermal-flux dependent.

5. SAP and Aluminum-Base Alloys

The Norwegians claim that aluminum-base cladding alloys reduced fuel cycle costs in their Boiling Heavy Water Reactor when compared with Zircaloy-2. Testing of an Al - 1% Ni - 12% Si - 1% Mg - 0.3% Fe alloy indicated better elevated temperature corrosion resistance and mechanical properties than either X8001 or M388. Because of creep strength limitations, a free-standing cladding could not be designed. However, results of strain fatigue tests of the new alloy were satisfactory for the intended application. Preliminary irradiation tests to 4×10^{19} nvt at room temperature indicated only a 10% reduction in elongation at fracture.

SAP materials for use in WR-1, a D₂O moderated, organic-cooled reactor, were evaluated by the Canadians. It was reaffirmed that SAP maintained its strength at elevated temperatures and that the major design limit was its low plastic deformation at elevated temperatures. SAP, as opposed to Zircaloy-2, was selected as the cladding for WR-1 because of uncertainty concerning hydriding of Zircaloy-2. Irradiation testing of SAP-clad UO₂ elements at $\int k d\theta$ of about 36 W/cm at clad temperatures of 450°C - 500°C resulted in two significant clad failures. Failures occurred only in 244-cm long rods, all of which were clad with SAP-930 which has a lower hoop-stress rupture strength than M257. No failures were attributed to the latter alloy.

The Danes utilized SAP for cladding, pressure tubes and structural members in their 250 MWe organic-cooled, heavy water plant. They indicated that SAP-895 will be used for the cladding and pressure tubes and that the major design problem was associated with fuel-clad mechanical interaction. A reaction was reported between SAP

and UO_2 after 8000 hours at 600°C under high contact pressures. No reaction was found after 9200 hours at 525°C . Similar reactions were reported by the Canadians at 430°C . The Danes reported a SAP/UC reaction after 7300 hours at 450°C and found that it could be eliminated through use of an internally anodized SAP sheathing. A similar conclusion was made by the Canadians for inhibiting the SAP/ UO_2 reaction. The Italians also planned to use SAP as cladding in ORGEL, a heavy water-moderated, organic cooled reactor.

6. Iron-Aluminum Alloys

The French discussed the properties of an Fe - 25 wt% Al alloy. This alloy was suggested as a desirable substitute for the austenitic stainless steel in cladding and structural applications because of its lower cost and more desirable thermal-neutron capture cross section.

D. Fuel Fabrication for Special Purpose Heavy Water Reactors

1. Desalination and Power-Desalination Reactors⁽¹⁴¹⁾

Desalination and dual-purpose power-desalination reactor studies have centered about the economics of the two types. There are no operating desalination plants using nuclear power so the data are the results of studies made by various organizations and countries. Economic desalination of seawater is predicated on larger central nuclear power stations than those now operating or scheduled for operation in the immediate future.

ORNL⁽¹⁴²⁾ has stated the lowest fuel cycle costs for converter reactors can be achieved by use of natural uranium fuels in heavy water reactors of very large size. Heavy water reactors are among the most popular candidates for desalination energy sources.^(143,144) Single-purpose, desalination-only plants, were found by most studies to be less economical than dual-purpose plant concepts.^(145,146) Heavy water reactors most frequently connected with desalination studies employ the CANDU type fuel elements^(147,148) or the tube-in-tube fuel element concept described by SRL.

The most important aspect of fuel element technology with respect to desalination and dual-purpose power-desalination reactors is that the studies to date have been concerned primarily with economics and feasibility or site selection problems. No real attempt has been made toward optimizing fuels or fuel geometries for these types of reactor. This is particularly important in the case of heavy water reactors because all heavy water reactors at present are quite small, 250-500 MW as compared to the 1000 MW(e) of the near future.

Large HWR plants are generally accepted as most economical for desalination; probably 10 to 25 times as large as those now being considered by the power industry.

2. Thoria Reactors

Thoria-fueled reactor cores have been used throughout the world in test and power reactors of almost every type including D₂O-cooled, PWR, BWR, gas-cooled, aqueous homogenous, seed-blanket, fast breeder, aqueous suspension and the HWOCR. At least three cores have been tested and operated satisfactorily in heavy water-cooled and moderated reactors. In most cases conventional fuel fabrication⁽¹⁴⁹⁾ methods were used and thoria fuels, per se, have been proven in water-cooled reactors. (150-152)

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