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FUEL ELEMENT DESIGN HANDBOOK (U)

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Edited by

K. R. Merckx

Fuels Design

Fuels Development Operation

September 1, 1958

NOTICE!

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FUEL ELEMENT DESIGN HANDBOOKPREFACE

In writing a Handbook for engineers or scientists working in a particular field, the scope of the field must be determined. Since "Fuel Element Design" for nuclear reactors is not an established field, some arbitrary restrictions have to be made as to what is fuel element design. This book tries to separate the detailed fuel element design problem from the general reactor design problems. Thus, the broad criteria needed to successfully operate a reactor are assumed to be prescribed by the reactor designer. The fuel element designer determines by application of physical principles, material data, and experimental techniques what combinations of fuel, clad, and inter-face materials will perform adequately under the prescribed reactor conditions. Fuel element design has also been separated from the fuel element production; hence, the problems concerned with fabrication and testing of large quantities of fuel elements are not discussed. Since reactor design, fuel design, and economic fuel fabrication criteria must all be satisfied for an acceptable final design, the limitations of the material discussed in this Handbook does not eliminate the need for the fuel element designer appreciating the various factors used to select the final fuel element for a reactor.

The main purpose of this Handbook is to collect the information needed for designing fuel elements. This handbook is not intended to be a source book for general material properties useful in other phases of reactor design. Since the purpose of the Handbook is restricted, the bulk data used to obtain the material data is interpreted as pertains to its applications in fuel element design. By restricting the data and comments only to the area of fuel element design, this Design Handbook tells new fuel element designers what information he should be expected to understand. Also, a restricted field of interest does enable the many authors of this Handbook to emphasize those areas where data is vitally needed for future fuel design applications.

The organization of this book is such as to allow for easy revision and increased coverage of the field. The numbering system can be described by the following example:

"203.1.2-4" would be in the second chapter, (2) the third main subdivision (topic), (03), the first subdivision of this topic (section), (.1.), second subdivision of the section (.2), and page four of this subdivision, (-4). Hence, if section 203.1.2 is altered, the numbering of other subdivisions is not altered. The subdivision used for the numbering of various subjects will be selected for the convenience of revision by the authors.

If a subject is well established and understood, it can be described in a concise manner. Since so much of the fuel element design field is still an art, the design concepts are often not too well organized. At first extraneous material may be given and needed data omitted; thus, continual alterations, deletions, and additions will be needed if this Handbook is going to be of continuing use to you. Your comments to the author and the editor are needed in order to develop a useful Fuel Element Design Handbook. If you are going to use this book, will you please give us your comments on what is needed to cover the field of fuel element design and only this field.

FUEL ELEMENT DESIGN HANDBOOK100 INTRODUCTION - K. R. Merckx

The economic development of nuclear reactors depends upon the integrated progress in the fields of reactor design, fuel element design, reactor operation, and fuel production and separation. Broad criteria, which restrict the fuel element design, are determined by the mutual consideration of the problems encountered in all the above fields. Hence, no stage of reactor design or operation is independent of the fuel element problem, nor can the fuel element designer disregard the interest of any one field. As an introduction to the fuel element design problem, this chapter describes how the general criteria for a fuel element are determined.

101 Reactor Criteria

Since the operation of a reactor depends upon nuclear reactions, certain nuclear characteristics are required of the fuel element which are based on the purpose of the reactor. If a reactor produces materials such as plutonium or tritium (production reactors), it must be able to provide neutrons in excess of those required for the nuclear chain reaction. Since the amount of product directly depends upon the effective use of neutrons, the fuel element designer is restricted in the amount and type of materials that he can use as fuel clad and fuel alloying elements. Neutron economy is less important in power reactors because the cost of producing power is not directly related to the excess neutrons. Effective use must be made of the fissionable materials in creating heat, but factors such as increased operating temperatures or longer fuel life can increase power output per fissionable atom even if some neutrons are sacrificed. Neutron economy is less important in material test reactors, package power plants, or military applications, because size, control, length of operation, and

ease of operation under widely varying power conditions are stressed more than economic operation. Not only the use of the reactor, but also the energy range of neutrons used to maintain the chain reaction determines the extent of neutron capture in the coolant, structural and clad materials. Thermal energy reactors must have fuel and moderator arrangements which provide sufficient neutron moderation. Fast reactors (those using high energy neutrons) are more compact since moderation is not required. The compact fuel arrangements and the high power density of fast reactors require fuel elements with high ratios of surface area to volume. The variation of parasitic capture of neutrons with neutron energy allows cladding and alloying materials in a fast reactor which would be intolerable in a thermal reactor. Therefore, neutron energy range also determines the type of fuel element problems to be faced by the fuel element designer.

The reactor designer also has a choice of coolant. For high temperature low pressure operation, gas, liquid metal, or organics may be used instead of water. Each of these coolants create special problems for the fuel element designer. The low heat transfer film coefficient of gas coolants require fuel elements having extended surfaces for adequate removal of the heat generated. Gas cooled reactors operating gas turbines require high coolant temperature. The high coolant temperatures of liquid metal and gas cooled reactors can cause fuel designs to be limited by diffusion or melting. High temperature water or organic coolants create corrosion problems for both the fuel material and the clad material. Gases dissolved in or produced by radiation induced molecular changes in the coolant can create intolerable mechanical changes in the fuel material or its clad. Hence, the choice of a reactor coolant helps decide the type of fuel element design problems.

Once the use, energy range of neutrons, moderator, and coolant of a reactor are selected, the reactor physicist, the thermal hydraulic engineer, the reactor designer, and the fuel element designer must decide on the size and shape of fuel and coolant channels and the fuel power generation. Since the rate of heat removed determines the production rate in a production reactor and the heat generation and the outlet temperature of coolant determine the power and thermal efficiency in a power reactor, thermal-hydraulic considerations become directly involved in the design of fuel elements. Factors such as minimum coolant annuli, specific power generation, coolant pressures and flow affect fuel size, spacing, cladding, supports, and temperature limits.

Since most reactors are designed for economic nuclear conversion, power, or testing, economic factors determine the relative importance of fuel element integrity, power output, fabrication cost, etc. An important factor in determining the cost of a fuel element design is the integrated exposure which it will withstand before failure. Not only do fabrication costs increase as a result of a short fuel element exposure, but also reactor downtime is increased because of the increased rate of the discharging and charging. Thus, life expectancy must be considered in determining the economic merit of a fuel element.

102 Consequences of Failure

A guarantee against fuel element failure is impractical; thus, fuel element design must be made with both operational and failure characteristics in mind. Factors which make a fuel element inoperable include penetration of the clad by corrosion, excessive dimensional changes caused by swelling, contact between fuel segments, contact between process tubes and fuel elements, cladding failures induced by mechanical failure of the fuel material, and cladding failures caused by

geometrical strain concentrations or embrittlement of the cladding. The various types of fuel element jacket failures have different rates of corrosion of the fuel material which depend upon the extent of exposed fuel surface, temperature of the exposed fuel surface, and relative corrosion rates of the fuel and coolant. Contact of fuel segments with other fuel segments or process tubing can cause excessive temperatures, rapid diffusion, and high corrosion rates.

The damage caused to a fuel element because of its failure is not as serious as the possible damage to the reactor components. Jacket failure causes fission and corrosion products to be released into the coolant stream. Swelling, excessive corrosion products, or fuel element contact with coolant channels can cause the blocking of the coolant flow with subsequent failure of the process tube and damage to the moderator. Also, the spread of fission and corrosion products can restrict access to exterior reactor components. Hence, the effects of damage to the reactor outweigh the direct loss of a fuel element. Should the failure of a fuel element endanger the integrity of the reactor or spread contamination outside a reactor, then the possible economic consequences of fuel element failure are even larger.

Since the failure of a fuel element can affect the reactor and the surrounding areas, the failure characteristics of the fuel element determine the time required for fuel element removal. The time a fuel element must operate after failure is also determined by the type and purpose of the reactor. The economic consequences of failure, risk to personnel, property damage, possible loss of military objective (military reactors), and lost operational time must all be included in the design of a fuel element's failure characteristics.

103 Fabrication Limitations

Fabrication costs as well as the economic advantages for operational characteristics enter into the final choice of a reactor's fuel element. Availability of production equipment and techniques, materials, or testing methods are major considerations in scaling up the fabrication of a fuel element design concept into a production fuel element. In the case of plutonium and uranium 233 fuel elements, special remote methods of fabrication must be developed which cause additional expenses and delays. Since fabrication methods affect the mechanical state of the fuel and clad materials, it often becomes difficult to separate the fabrication process from the description of the fuel element's material characteristics. Many of the materials with desirable nuclear and mechanical properties for alloying or cladding have not been needed previously; hence, are not commercially available in sufficient quantities. If a fabrication process leaves certain properties, such as bond integrity, etc., in doubt, then testing methods have to be developed. Thus, the satisfaction of certain criteria by a fuel element produced by laboratory techniques does not assure that it can be produced in sufficient quantities for economic use in a reactor.

104 Separation

Since the full fuel cycle must be considered in determining costs, one cannot disregard the separation and recovery costs of the unreacted fuel material. Variations of fuel enrichment can cause criticality problems of the separation vessels. Alloying additions to increase the corrosion resistance of the fuel material can create separation problems in the initial dissolving of the fuel. Hence, along with fabrication costs and developments, one must also consider the problems encountered in the post-irradiation processing and recovery

of fuel material. The recovery process must also reduce γ -reactivity to a sufficiently low level or further fabrication costs will be greatly increased. Should one devise a fuel design for power, test, or military reactors effective enough to consider disposing of the fuel elements after the initial irradiation, the costs of disposal still has to be considered as part of the fuel costs.

This introduction points out the various factors which are used in defining an acceptable fuel element design. Since so many factors are involved in establishing fuel element design criteria, no one technological discipline is responsible for establishing "the fuel element criteria". The various developments made in reactor design, fuel element design, fabrication, and separations affect the work in each of the other fields. The fuel element design principles and data presented in the following chapters are not intended for economic evaluation of a fuel design, but for a possible fuel element candidate design which will satisfy the criteria needed for successful reactor operation.

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201 URANIUM
201.1 CONDENSED TABLE OF PROPERTIES

201.1 Composition and Reactivity
Typical analysis of production natural uranium

Element & ppm by weight										
C	Cr	Fe	H ₂	Mg	Mn	Ni	N ₂	Si		
150-750	35	30-140	1-4-6	10	15	50	25	<20-100		

Above analysis applied to metal recast into ingots after bomb reduction.
Ingots (Dingote) cast directly from the reduction bomb contain fewer impurities except for the hydrogen content.
Nuclear Properties (13)

Reaction Cross Sections at 2200 m/s (Barns)			
Total	Absorption	Activation	Fission per fission
Neutrons	Emitted/neutron	Absorbed	Neutrons

Isotope			
Natural U	238	235	233
Half life	4.5 x 10 ⁹ y	7.1 x 10 ⁸ y	4.5 x 10 ⁵ y
Absorption	2.75	687	585
Activation	2.8	107	52
Fission per fission	2.46	2.46	2.31

201.2 Fabrication
The range of fission products in natural uranium is 6.8 microns (2.7 x 10⁻⁴ in.)

Casting	
Vacuum melt and cast in Zirconia washed and outgassed graphite crucible. Metal temperature 1300 C.	
Rolling	
Generally alpha phase, room temperature to 600 C. Large reductions per pass possible.	
Extrusion	
Alpha or Gamma phase. Extrusion constant 150,000 to 55,000 psi as uranium temperature is raised from 200 to 600 C. Area of rod	
Forging	
Heat in atmosphere 500 to 650 C. Forge in air.	
Swaging	
Hot or cold, up to 85% R.A. possible.	
Machinability	
Carbide tip tool, similar to 304 stainless steel plus the weld in inert atmosphere or vacuum (electron beam).	
Joining	
Weld in inert atmosphere or vacuum (electron beam).	

201.1.3 Physical Properties
Density (1) 19.05 ± 0.02 g/cm³ (25 C) (High purity)
Wrought material 19.7 - 19.08 g/cm³ (25 C)
Irradiation at high temperature causes a density decrease known as swelling. The parameter "R" used to describe swelling data in the figure below does not take into account flux dependence and incubation periods, but it is a convenient parameter for comparing experimental data.

SWELLING DATA
R = 168 g
m = 8 ± 0.427 T - 300
+ if T < 300

Thermal Expansion
Beta Heat Treated Uranium

$$\alpha = \frac{dL}{L dT} = (13.4 + 0.0132 T) \times 10^{-6} / ^\circ C$$
$$\alpha = (7.33 + 0.00407 T) \times 10^{-6} / ^\circ F, T = ^\circ F$$
$$\alpha_{avg} = [13.4 + 0.0132 T_1 + 0.0066 (T_2 - T_1)] \times 10^{-6} / ^\circ C$$
$$\alpha_{avg} = [7.33 + 0.0407 T_1 + 0.00203 (T_2 - T_1)] \times 10^{-6} / ^\circ F$$

Heat Capacity (4)

$$At 25 C (77 F) - 6.612 \pm 0.007 \text{ cal/deg mole } (0.278 \pm 0.003 \text{ Btu/lb } ^\circ F)$$
$$\alpha, C_p = 3.15 + 8.44 \times 10^{-3} T + 0.80 \times 10^{-5} T^2$$
$$\beta, C_p = 10.38$$
$$\gamma, C_p = 8.10$$
$$T = ^\circ \text{ Kelvin}$$

201.1.5 Mechanical Properties
Tensile Properties (2)

The Effect of Exposure and Exposure Temperature on the Room Temperature Properties of Uranium			
As rolled	Gamma heat treated	Beta heat treated	As cast
85-160	65-80	65-105	59-75
UTS (psi x 10 ⁻³)			
29-90	15-29	29-31	28
YS (psi x 10 ⁻³)			
10-30	8-15	8-17	7-10
Elongation (%)			
10-30	8-15	8-17	7-10
RA (%)			
10-30	8-15	8-17	7-10

Plastic behavior of beta quenched uranium is approximated by the expression

$$\epsilon = \frac{3}{2} (6.40 \times 10^{-6}) [1 + A(T)] \left(\frac{1000}{T} \right)^4$$
$$\text{where } \log_{10} A(T) = -6.105 + 0.184 \left(\frac{100}{T} \right) + 0.0355 \left(\frac{100}{T} \right)^2$$
$$\epsilon = \% \text{ strain, } \sigma = \text{psi stress and } T = ^\circ C.$$

Primary Creep Behavior:

$$\epsilon = \exp. k + d \left(\frac{\sigma}{G} \right) \epsilon^{-m} \left(\frac{30}{T} \right)$$
$$\text{where } G = 70,000 \text{ psi/\%}$$
$$m = 8 \pm 0.427 T - 300$$
$$+ \text{ if } T < 300$$

Phase transformation points⁽¹⁾

Alpha-Beta	668 C
Beta-Gamma	774 C
Melting Point	1132 ± 1 C
Boiling Point	3818 C

Lattice constants⁽¹⁾

Alpha phase, Orthorhombic, Space group cmcm
4 atoms per unit cell

25 C

a_o	$= 2.8541 \pm 0.0003$
b_o	$= 5.8692 \pm 0.0015$
c_o	$= 4.9563 \pm 0.0004$

Beta phase, Tetragonal, Space group Pr/mnm
30 atoms per unit cell

720 C

a_o	$= 10.759 \pm 0.001$
c_o	$= 5.856 \pm 0.001$

Gamma phase, body centered cubic, 2 atoms per unit cell

805 C $a_o = 3.50 \pm 3$

Elastic Properties⁽²⁾

Modulus of elasticity (25.5 to 30) $\times 10^6$ psi
Shear modulus (10.6 to 12.1) $\times 10^6$ psi

Minimum Recrystallization Temperature

Cold worked alpha uranium. 400°C grain coarsening of beta quenched material occurs above 600 C

Predominant Deformation Mechanism⁽¹⁾

- Slip on the {010} plane in the [100] direction and, below 350 C, {130} twinning.

201.1.4 Thermal Properties

Thermal Conductivity = $15.6 + 7.15 \times 10^{-6} T^2$ Btu/hr ft $^{\circ}F$ $0 < T < 740 F$
= $16.8 + 5.13 \times 10^{-6} T^2$ Btu/hr ft $^{\circ}F$ $740 < 1620 F$

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$$k = 8.24 - \frac{8.45 \times 10^4}{T + 273} + \frac{1.19 \times 10^7}{(T + 273)^2}$$

Secondary creep data shows a wide scatter. Typical results are given in Reference 8.

20.1.1.6 Corrosion of Uranium

Reaction with Metals: Uranium is insoluble, and is therefore not corroded by pure liquid metals of the alkali or alkaline earth groups except beryllium.

In general there is little or no solid state reaction between uranium and other metals at temperatures below 0.25 to 0.30 times the absolute melting point of the other metal.

A summary of available data on the diffusion of uranium and other metals is contained in Reference 9.

Reactions of Uranium with Various Gases are summarized below:

Temperature (°C)	Penetration Rate (cm/h)			
	Air	Steam	CO ₂	Hydrogen
200	7×10^{-5}	5.5×10^{-3}	1.7×10^{-6}	2.7×10^{-1}
250	4.4×10^{-4}	3.1×10^{-2}	2.9×10^{-6}	3.0×10^{-1}
300	1.1×10^{-3}	2.8×10^{-2}	4.3×10^{-6}	1.9×10^{-1}
350	3.7×10^{-3}	3.5×10^{-2}	5.5×10^{-6}	--
400	1.1×10^{-2}	3.1×10^{-2}	5.6×10^{-6}	--
450	--	2.0×10^{-2}	1.0×10^{-5}	--
400	--	2.0×10^{-2}	5.7×10^{-5}	--
550	--	--	--	--
600	--	1.8×10^{-2}	--	--

* extrapolated data

The reaction of uranium with hydrogen to form UH₃ is most rapid ($\sim 7.5 \times 10^{-4}$ g atom hydrogen/cm²/min) at 225 C and above this temperature the rate decreases as the decomposition pressure of the hydride approaches the externally applied pressure.

Temperature (°C)	Dissociation Pressure of Uranium Hydride	
	(mm Hg)	(psia)
400	350	6.8
500	2580	49.5
600	11910	230.3

The diffusion coefficient for hydrogen in uranium is:

Temperature (°C)	Diffusion Coefficient (in ² /h)
566	0.0040
593	0.0064
640	0.0125

Uranium also reacts readily with nitrogen although in oxygen-nitrogen mixtures the oxide is formed preferentially.

Temperature (°C)	Increase in wt (g) of specimens 2 cm dia. x 2 cm long after 24 h in nitrogen
300	0.0006
400	0.0022
500	0.0074
600	0.0315

Reaction with pure nitrogen⁽¹⁰⁾

$$(\text{Nitrogen consumed})^2 = 202 \exp. \frac{(-25,500)}{RT} \frac{\text{Ml}}{\text{cm}^2 \text{ sec}}^2$$

Reaction with liquids:

Water: Rate of reaction with hydrogen saturated water is linear. The temperature dependence is represented by the relation⁽¹⁾

$$\text{Corrosion rate} = 1.3 \times 10^{10} \exp. \frac{(-16,600)}{RT} \text{ mg/cm}^2/\text{hr}$$

where T = °K

Organic Coolants: (Terphenyls, biphenyl and monoisopropylbiphenyl)
No reaction up to temperatures at which pyrolytic decomposition of the organic is excessive. Hydrogen generated by decomposition of organic causes corrosion to proceed as if hydrogen were present alone at a total pressure equal to its partial pressure in the organic.⁽¹¹⁾
Irradiation to 0.06 a/o burnup has little effect on uranium corrosion in organic coolants.⁽¹²⁾

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201 URANIUM - R. G. Wheeler

Unalloyed uranium is a reactor fuel that has many desirable properties. In the unalloyed condition the density, thermal conductivity and ductility of uranium are at a maximum. Complications do arise with the use of metallic uranium because of its highly anisotropic room temperature (alpha phase) structure.

Properties of uranium which can be stated briefly without qualification are condensed in the Table as a quick source of data. Other properties which require qualifying comments are discussed in the following pages.

201.1.1 Composition and Reactivity

- I. Unirradiated (see the Table)
- II. Irradiated

The composition of uranium under irradiation changes with time as a function of the rate of fission in a unit volume of the uranium and the diffusion of fission products. The composition of fission products during or after irradiation may be obtained from reference (14). The effect of fission product atoms on the properties of uranium is heterogeneous in massive fuel elements because self shielding effects result in a higher concentration of fission events near the surface than in the center. (15)

201.1.2 Fabrication

Cast uranium fuel elements are used extensively in Great Britain whereas in the United States natural uranium fuel elements are made almost exclusively of wrought material. Recent improvements in vacuum casting techniques and equipment may place casting in an economically competitive position for many types of fuel elements. Irradiation behavior of cast fuel elements in HAP0 reactors is currently under study. (16)

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Uranium can be converted to wrought shapes by most of the conventional forming methods⁽¹⁷⁾ such as extrusion, drawing, rolling, forging, pressing, and swaging. In the alpha phase, high forces (comparable to those used for steel) are necessary to form uranium. The beta phase is 2 to 3 times tougher than the high alpha phase and the temperature range of this phase is so narrow that it is generally avoided during forming. Die wear, lubrication and protection against oxidation are the major problems associated with uranium extrusion. Glass is used successfully as a lubricant for γ phase extrusion.⁽¹⁶⁾ Warping of the soft gamma extruded material is reduced by extruding vertically downward. A number of jacketing techniques are based on processes in which the jacket material is deformed with the uranium to produce a bond between the two metals.⁽¹⁸⁾

Heat treatment through the $\alpha - \beta$ phase transformation is used to control grain orientation and grain size. Certain conditions tend to produce the fine randomly oriented grain structure required for stable irradiation behavior at temperatures below 400 C. These conditions are: small initial grain size in the as wrought or cast material, rapid quenching through the phase transformation temperature, short hold time in the beta phase and uniform cooling. Obviously it becomes more difficult to meet these conditions as the uranium volume to surface ratio increases. The small initial grain size condition is difficult to obtain in cast and gamma extruded uranium.

201.1.3 Physical Properties

I. Unirradiated

a. Density and Thermal Expansion

The thermal expansion coefficients in the three crystallographic directions of alpha uranium crystals are different. They are negative along the b axis and positive along the a and c axes. These differences

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in thermal expansion supply the driving force for growth as a result of thermal cycling.⁽²¹⁾ This growth may be 500 to 600 microinches/inch/cycle for alpha-rolled rods but the effect becomes negligibly small for fine grained beta-heat-treated uranium subjected to the number of cycles encountered in most reactor applications.

The average value for the coefficient of thermal expansion of beta-heat-treated uranium from room temperature to 600 C is 17.5×10^{-6} . This value varies from 15 to $20 \times 10^{-6} / ^\circ \text{C}$ ^(19, 20) because of the difficulty in obtaining stress-free and randomly oriented polycrystalline beta-heat-treated uranium. Variations of uranium density cause thermal stresses and distortions in a fuel element, and cause forces between the jacket and the core. These limit the maximum size of a billet that can be heat treated without cracking. Density variations are directly related to thermal expansion but may also be caused by irradiation effects.

Figure 1 shows the per cent decrease in density of uranium with temperature. Discontinuities in density occur at the beta and gamma phase transformation temperatures.

b. Elastic Properties

The tensile stress-strain curve for uranium exhibits little or no proportional range, and marked hysteresis effects are observed on unloading and reloading. The modulus obtained from tensile data is subject to error and it varies with metal fabrication history, hence the large range in the values reported in the Table.

II. Irradiated

a. Swelling

The term swelling is used to describe density changes in a fuel material. Swelling is generally considered to be caused by formation of fission product gas bubbles in the uranium. A

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convenient unit for comparing swelling data from various sources is the "R value" defined as:

$$R = \frac{(100) \text{ change in volume/initial volume}}{\text{Per cent burnup}}$$

Swelling is reduced or its inception delayed by application of a constraining force on the volume of metal under consideration. Conditions under which there is little constraining force on the specimen are obtained in flat plates, cored cylinders, small specimens surrounded by liquid metal and small gas cooled specimens.

R values of 40 to 70 were observed by Minor⁽²³⁾ and Goffard⁽²⁴⁾ in insulated cylindrical elements cored so that the hottest uranium (that most likely to swell) was unconstrained. The central temperature in these irradiations were at least into the beta phase. A compilation of swelling data on essentially unconstrained unalloyed uranium is shown in the Table.⁽²⁵⁾ Volume increases due to swelling cause design limits on temperatures and exposures because the increased fuel volume can restrict the flow of coolant or cause cladding failures.

b. Growth

The term, growth, is used to describe changes in dimensions where the volume or density remains constant. The reader is referred to a review of the subject by Bush.⁽²⁶⁾ Irradiation of single crystals of uranium causes growth in the (010) direction, shrinkage in the (100) direction, and little change in the (001) direction. Fuel elements consisting of fine randomly oriented uranium grains do not grow when irradiated because the growth of individual crystals is effectively cancelled by the random orientation. Irradiation growth is small or absent below -100 C and above 400 C. Warping, bumping, or fuel element elongation are design limitations are encountered if irradiation growth is not eliminated.

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201.1.4 Thermal Properties

I. Effects of Irradiation

a. Thermal Conductivity

The limited data available (27, 28, 29) indicate a decrease in thermal conductivity of about 10-15 per cent as a result of irradiation. Since the predicted accuracy of the methods used to obtain these data are probably ± 20 per cent at best, the validity of the results in an absolute sense is dubious. Qualitatively, however, the results indicate that there is no gross change in thermal conductivity of uranium during or after irradiation.

b. Thermal Expansion

No reference to measurements of thermal expansion of uranium during or after irradiation were found in the literature, although experimentally, more difficult measurements are made, i. e., post-irradiation stress vs. strain measurements at high temperature. Experience with production reactors operating with long columns of fuel would indicate that there is at least no gross change in the thermal expansion of beta-heat-treated uranium with exposure.

201.1.5 Mechanical Properties

I. Unirradiated

a. Tensile Data

Uranium shows a range of tensile properties that depends on fabrication history and impurity content. A typical range of values for different forming processes is shown in the Table. Tensile stress-strain curves for beta-heat-treated uranium tested at various temperatures are represented by equations in the Table and shown graphically in Figure 2. These curves are in fair agreement with similar data obtained at BMI. (30)

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FIGURE 1
Per Cent Decrease in Uranium Density vs Temperature

FIGURE 2
Stress-Strain Curves of Beta Heat-Treated Uranium
at Various Temperatures(5)

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b. Creep

The effect of thermal cycling an anisotropic metal plus the experimental difficulties associated with creep testing such a reactive metal as uranium may explain the absence of agreement in creep data. The following table shows the significant effect of small thermal cycles on creep test results:⁽³⁸⁾

Effect of Thermal Cycling on Creep of Uranium

Material	Creep strain (%) after 400 hours on thermally cycling through range shown			Stress lb/sq in
	472-500 C	493-500 C	499 1/4-500 3/4 C	
Rolled at 600 C then alpha-annealed	5.1	2.2	0.55	1,500

Figure 3 shows that the creep resistance in a sagging bar and and strut test at 500 C is an order of magnitude less than in a tensile creep test.

The data sheet in reference (8) summarizes the creep behavior of uranium fabricated by a number of different methods. Large total strains before rupture are obtained (> 25% elongation) in short time stress rupture tests on beta-heat-treated uranium⁽³⁰⁾ but for long time tensile creep tests the total strain is about 0.1 per cent elongation.

c. Strain Cycling

A ΔT of 25 C from surface to center of a cylindrical uranium fuel produces plastic strains in the uranium. Cycling the power generation of a fuel element operating at greater than 25 C ΔT therefore strain cycles the uranium. Coffin⁽³¹⁾ has shown that for most metals the number of plastic strain cycles to failure can be predicted from the reduction in cross sectional area produced by fracturing a specimen in tension. For a given amount of plastic strain per cycle the number of cycles to failure is assumed to be proportional to $\ln \frac{A_0}{A}$, where A_0

is the initial area of the cross section and A is the area after fracture. The complete relationship is $N \Delta \epsilon_p^n = \ln \frac{A_0}{A}$ where N is the number of cycles to failure and $\Delta \epsilon_p$ is the total plastic strain per cycle. For most metals the exponent "n" is 2 (slope of $\log \Delta \epsilon_p$ vs. $\log N = \frac{1}{2}$). Figure 4 shows the strain fatigue behavior of beta-quenched uranium. The slope of $-\frac{1}{2}$ is a reasonable fit for the data (Figure 4) but does not extrapolate to the 1/4 cycle (a tensile) test. At present, strain cycling or thermal fatigue data is used as a criterion for mechanical failure in ductile materials subject to cyclic thermal stresses such as are encountered in fuel elements.

II. Irradiated

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a. Tensile

Heuschen, Kemper and Kelly have investigated the effects of burnup, irradiation temperature, U^{235} content, post-irradiation annealing, and testing temperature on the properties of irradiated natural uranium. ⁽⁵⁾ The ductility of irradiated uranium as indicated by fracture strain rapidly reaches a damage plateau at a burnup as low as 0.02 a/o. (See Figure 5).

Post-irradiation annealing increases fracture elongation providing the heating and cooling rates above 600 C are less than 10 C/hr. An increase in fracture elongation from less than one per cent in as-irradiated 0.075 a/o burnup specimen to 12.4 per cent, and to 4.6 per cent from 0.55 per cent in the case of 0.1 a/o burnup specimens was observed. ⁽⁵⁾ Lower temperature anneals also produced some recovery of ductility.

Elevated temperature tensile test data for specimens of natural uranium irradiated to 0.075 a/o burnup at 150 C is shown in Figure 6.

The effect of elevated temperature irradiation on tensile properties is shown in the Table. These data give an indication of properties to be expected of exposed uranium during a reactor startup but they may not represent the properties of uranium during irradiation.

A

B

FIGURE 3

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FIGURE 4

Strain Fatigue Behavior of Beta-Quenched Uranium

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FIGURE 5

Room Temperature Properties of Irradiated Uranium - Effect of Exposure

FIGURE 6

Elevated Temperature Stress-Strain Curves of Irradiated Uranium

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b. Creep

The effect of irradiation on the creep rate of uranium at very low stresses (200 to 2000 psi) at a 100 C test temperature and a thermal flux of 10^{12} nv was measured by Roberts and Cottrell⁽³⁰⁾ and was found to be in the order of three per cent per year at 1000 psi. No creep of uranium would be expected at this stress and temperature in the absence of irradiation and thermal cycling. Since Cottrell attributes this creep to indirect effects of irradiation growth, this irradiation effect should be absent above 400 C (the temperature above which little growth is observed).

Some upsetting of end loaded fuel elements and sagging of horizontal fuel elements has been observed in the British reactors.⁽⁸⁾ These data indicate that, with maximum fiber stresses under 2000 psi, up to 1.5 elastic deflections per MWD/T burnup may occur. Available data indicate that under irradiation uranium may not be relied on to supply structural support.

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c. Strain Cycling

Chapter 500 shows that strain cycling of uranium resulting from power cycling of the fuel is a logical explanation for many core failures. There is little or no information in the literature concerning direct measurements of the effect of exposure or thermal neutron flux on the strain cycling behavior of uranium. Indirect evidence of fuel element failure as a result of strain cycling is present in the correlation of core splitting with reactor operation.⁽³²⁾

d. Property Indicating Cracking Tendency

Post-irradiation evidence such as elongation at fracture of tensile specimens, maximum deflector of fuel elements fractured in bending and the observed presence of microcracks indicates that exposure makes uranium brittle. Bend tests on exposed HAPO fuel elements show that the bend strength is essentially that of the jacket and the uranium is

either fractured or has no bend strength. Post-irradiation optical and electron micrographs⁽²⁶⁾ show the presence of microcracks produced by a .03 per cent burnup at less than 300 C.

The work of Bierlein⁽²⁶⁾ shows that fracturing unirradiated uranium produces extensive plastic deformation of the fractured grains, unless the break is made by impact at -190 C. Impact fractures made at this low temperature are of a cleavage type and almost identical in appearance with the fracture surface of 0.1 a/o burnup uranium broken in tension at room temperature after annealing. This appearance of cleavage fracture in irradiated (0.1 a/o burnup) uranium under conditions which favor ductile fracture indicates little resistance to impact loads.

201.1.6 Chemical Reactions

The rate of reaction between uranium and water depends more on the pressure and temperature of the water than on the temperature of the uranium. Minor et al.⁽³⁹⁾ found that, when resistance heated uranium was suddenly exposed to 300 C 2000 psi water, the greatest amount of attack appeared at the cold ends of the uranium rod. Some of their corrosion results are tabulated below:

Corrosion Rates for Heated and Unheated Uranium Specimens

<u>Initial Specimen Temperature</u>	<u>Water Temp.</u>	<u>Time in Minutes</u>	<u>Penetration Inches/min x 1000</u>
Unheated	300 C	20	1.5
600 C	300	20	1.5
700	300	20	1.25
Unheated	200	200	0.11
700	200	200	0.095

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Uranium in massive form does not react violently with gas or liquid coolants even at gamma phase temperatures. Subdivided or molten uranium may burn or explode when it is suddenly flushed with reducible coolant.

201. 2 Design Limits

201. 2. 1 Fabrication

Experience has shown a direct correlation between irradiation behavior and uranium metal quality. Analytical, metallographic and non-destructive testing control over metal quality is essential to any fuel element specification. (33)

Uranium fuel elements that are to operate at a minimum temperature below 400 C should have randomly oriented grain structure to prevent growth and fine grains to prevent bumping. (34, 35)

The fabrication and heat treatment process should be symmetrical about the long axis of the fuel element to minimize warping.

201. 2. 2 Physical Properties

Operation of uranium so that two phases are present (core temperature above 668 C) is generally avoided in production reactors. Maximum thermal strains are in the order of 0.5 to 1 per cent, and transition of the thermal center of a fuel element from the alpha to beta phase produces a one per cent strain which is additive to the thermal effect. Cycling through a phase transformation also accelerates the release of fission product gases. (37) Limited experience with two phase operation indicates that rupture rates are not affected significantly if the rate and number of power and temperature cycles is controlled. (32) Beta phase operation is a potentially rewarding goal that may be achieved when the core fracture phenomenon is well understood.

Gamma phase (774 to 1130 C) operation and even melting of the core is not impossible providing internal space is available for the associated volume changes and account is taken of possible sublimation and redistribution of liquid metal.

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Swelling is a potential design limit when a significant portion of the uranium operates above 400 C. The sharp rise in the swelling data curve in the Table would be modified by external constraint on the uranium, but quantitative evaluation of the parameters affecting swelling of natural uranium is not advanced to the point where design for a swelling limit is possible.

Density changes under conditions not conducive to swelling (maximum temperature <400 C) are not a serious limit. The density decrease in unalloyed uranium is about one-half the atom per cent burnup. ⁽³⁵⁾

201. 2. 3 Mechanical Properties

On the basis of British in-reactor creep data little if any structural support can be expected from uranium under irradiation. Where column or beam loading of the fuel is encountered, conservative design would obtain the required strength from the jacket until irradiation experience shows that uranium contributes some strength to the particular fuel assembly.

Strain cycling could well be a design limit both for the fuel and for reactor operation; but, unfortunately the only data concerning strain cycling resistance of uranium under irradiation is inferred from fuel element rupture data.

201. 2. 4 Chemical Reactions

The rate of uranium corrosion in the event of cladding failure can impose a design limit on the ratio of coolant volume to fuel volume and the space distribution of these volumes. The risk of overheating because of corrosion product blockage of coolant flow is reduced by reducing the cross sectional area of individual fuel rods and by increasing the cross sectional area of the coolant. Other possible limits based on uranium corrosion are sticking of the fuel charge and contamination of the primary coolant system.

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The reaction of uranium with water and other coolants containing hydrogen atoms releases hydrogen. Excessive hydriding of zirconium process tubes is possible if the coolant flow is blocked so effectively that dry hydrogen is exposed to the zirconium. ⁽³⁶⁾

201.3 Needed Data

There is some question regarding the effect of impurities on the swelling of uranium. A comparison of the swelling of production and electrolytic uranium under controlled conditions should help clarify this problem. Other needed data include:

- experimental and theoretical data which will enable one to predict swelling as a function of irradiation temperature, exposure, flux, and constraint;
- a comparison of high quality cast fuel elements with standard wrought fuel elements in U. S. production reactors;
- the value of a minimum uranium surface temperature of irradiation above which it is unnecessary to use randomly oriented uranium fuel elements to prevent growth;
- in reactor creep, strain cycling behavior, and the crack propagation tendency;
- the rate of reaction of uranium with coolants under conditions simulating the uranium power generation and coolant temperature, pressure and flow rates of reactor operation.

201.4 General Comments

The thermal and mechanical properties are all temperature dependent therefore, uranium temperature is one of the important variables subject to control by the designer. Control of temperature and evaluation of the thermal strain cycling of the fuel requires knowledge of thermal conductivity as a function of exposure. Pre-and post-irradiation tensile and creep test

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data do not adequately represent the mechanical properties of uranium under irradiation. The filling of a hollow core in uranium by swelling involves enormous strains yet the post-irradiation tensile tests indicate such strains would completely fracture the metal. The number of strain cycles necessary to produce failure and the residual ductility after a certain number of strain cycles is the kind of data that most nearly represents service conditions and can therefore be interpreted in terms of fuel life and reactor operation.

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202 URANIUM ALLOYS - J. W. GoffardIntroduction

Uranium may be alloyed by the addition of other metals for improvement of its characteristics or properties. The principal characteristics or properties of uranium that may require improvement for nuclear fuel applications are those of corrosion resistance, irradiation behavior, mechanical strength, and formability or fabricability. The necessary material changes to be obtained by alloying will depend upon the application of the fuel elements.

Nuclear Considerations

Alloying uranium has the detrimental effect of diluting the concentration of the fissionable fuel (U^{235}) and the fertile absorber (U^{238}) with a parasitic neutron absorber. This dilution is significant in reactors utilizing neutrons in the thermal energy range where efficient conversion of fertile U^{238} to fissionable Pu^{239} is desired. Thus, the neutron reproduction factor, the ratio of fast neutrons produced to thermal neutrons captured, is an important nuclear characteristic of an uranium alloy fuel which specifies one of the nuclear effects of alloying the uranium. The neutron reproduction factor, η , is determined by the following equation:

$$\eta = \frac{\text{neutrons produced}}{\text{thermal neutrons captured in fuel material}}$$
$$= \frac{\sum \eta^i \sigma^i N^i}{\sum \sigma^i N^i}$$

η^i = neutron reproduction factor for nuclide "i"

σ^i = thermal neutron absorption cross section for nuclide "i"

N^i = atoms of nuclide "i" per cc fuel material

Tables of η values of fissionable nuclides and the σ values of alloying additions follow:

η Values of Fissionable Nuclides and Natural U

<u>Fissionable Nuclides</u>	<u>η, Neutron Reproduction Factor⁽¹⁾</u>
U ²³³	2.31 $\pm$ 0.03
U ²³⁵	2.08 $\pm$ 0.02
P ²³⁹	2.03 $\pm$ 0.03
U Natural	1.34 $\pm$ 0.02

σ Values of Alloying Additions

<u>Alloying Addition</u>	<u>Integrated Thermal Neutron Absorption Cross Sections, Barns⁽¹⁾</u> (1 Barn = 10 ⁻²⁴ cm ²)
Be	0.010 $\pm$.001
C	0.0032 $\pm$ 0.001
Mg	0.063 $\pm$ 0.004
Al	0.230 $\pm$ 0.005
Si	0.13 $\pm$ 0.03
Ti	5.6 $\pm$ 0.4
V	5.1 $\pm$ 0.2
Cr	2.9 $\pm$ 0.2
Fe	2.53 $\pm$ 0.06
Ni	4.6 $\pm$ 0.2
Cu	3.69 $\pm$ 0.12
Zr	0.180 $\pm$ 0.004
Nb	1.1 $\pm$ 0.1
Mo	2.5 $\pm$ 0.2
Ru	2.46 $\pm$ 0.12
Th	7.0 $\pm$ 0.4
U (natural)	7.68 $\pm$ 0.07
U ²³³	585 $\pm$ 10
U ²³⁵	687 $\pm$ 7
U ²³⁸	2.75 $\pm$ 0.04
Pu ²³⁹	1065 $\pm$ 15

The η value of a fuel alloy may be adjusted to some desired or required degree by the use of enrichment with some fissionable nuclide. This provides the fuel element designer and the reactor physicist with a degree of freedom in the selection of fuel alloys.

Organization

The uranium alloy fuel materials section has the following sub-divisions:

202.1 Low Alloying Additions

This section includes alloys of natural uranium with low concentrations of the alloying additions where low is based upon the relative values of η for the alloy system as compared to that of natural uranium.

202.2 Phase Stabilized Alloys

This section includes alloys with sufficient concentrations of those alloying additions which will stabilize a certain crystallographic structure of the uranium. η values of these alloys with natural U will be less than those of alloys of low additions.

202.3 Dispersion Systems

This section covers alloys in which the fissionable phase is dispersed as discrete particles in a continuous and non-fissionable matrix or structural phase. Relative η values of these alloys will be less than those of alloys of low additions, but may be comparable with phase stabilized alloys.

202.4 General Alloy Systems

This section will include fuel alloys which may not fit into any of the above categories.

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202.1.1 LOW ALLOYS OF URANIUM-SILICON

202.1.1.1 Remarks on Tables of Properties

I. Composition

The phase diagram of interest in this section covers the composition range to about 4 w/o or 25 a/o Si. An eutectic occurs at about 1 w/o or 8 a/o Si with a melting point of 985 C. Solubility of Si in uranium is low, especially in the alpha phase temperature range. Generally, alloys in this region of the diagram will consist of two phases, one phase being U with some dissolved Si and the other being a high uranium intermetallic compound. The distribution and size of the intermetallic compound in the uranium matrix can be controlled to some extent by fabrication and heat treatment procedures.

II. Fabrication

Alloy preparation on a semi-works scale has been accomplished by co-reduction using pure dehydrated silicic acid with a Si recovery of 60-65 per cent.⁽⁴⁾ Direct casting may be employed and would be most suitable for alloys of 0.5 w/o Si and greater. Centrifugal casting to a finished shape is possible.⁽⁶⁾ The Si alloys of less than 0.5 w/o may be fabricated as is uranium, however the procedures may require changes to satisfy the properties of the alloy being worked. For example, 900 C is required for primary breakdown of an as-cast 0.08 w/o Si alloy, rather than the 600 C conventionally used for uranium.⁽³⁾ Uranium - 0.5 w/o Si alloy has been extrusion clad with Zircaloy in the gamma phase temperature range without grain growth.

III. Physical Properties

U-Si alloy with 0.21 a/o Si have been observed to be only slightly finer grained than pure U. Alloys with 0.46 and 0.93 a/o Si exhibited a grain size of 0.065 mm. Increased alloy content to 1.62 a/o Si did not further reduce the grain size.⁽⁷⁾

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IV. Thermal Properties

The thermal properties of U-1.4 a/o Si alloy are very similar to those of pure uranium⁽⁹⁾ and for practical purposes the established values for uranium may be used.

V. Mechanical Properties

The tensile strength of U-1.4 a/o Si alloy is about 30 per cent greater than that of arc or induction melted U. The ductility is about 70 per cent of that of arc-melted U.

VI. Corrosion Characteristics

The aqueous corrosion resistance of unirradiated U-2 a/o Si alloy has been studied at BMI.⁽⁸⁾ The tabulated data below show the effect of the heat treatment, and further indicates that low additions of Si to U have a negligible effect on the corrosion rate.

<u>Sample</u>	<u>Wt Change, mg/cm² After 24 Hours</u>	<u>Remarks</u>
U-2 a/o Si	--	100 C distilled water, 24 hrs 0.27 w/o Si, 0.042 w/o C, 0.009 w/o W
A	-19.4	4 hrs at 600 C, furnace cooled
B	- 7.69	1 hr at 700 C, water quench
G	-30.3	1 hr at 815 C, water quench
GA	-10.3	G followed by A

Since improvement of corrosion resistance is a primary purpose of the alloy addition, significant improvement in this respect must be made before other properties and irradiation behaviors require evaluation.

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Burning curves⁽¹¹⁾ of U-Si alloys indicate that alloys up to about 1.5 a/o Si burn more readily in air than does pure uranium. The burning tendencies are: 0.45 a/o Si - most readily, 2.46 a/o Si - intermediate, 0.92 a/o Si - least readily.

202.1.1.2 Design Limits

1. As the Si content increases in these alloys the manner in which fuel shapes may be fabricated becomes limiting. With Si additions of less than 0.5 w/o, conventional uranium fabrication practices may be used, but if the Si content is increased, fuel shapes may have to be cast.
2. The lowering of the solidus line by Si addition represents an upper temperature limit since this temperature is about 150 C below the melting point of pure uranium.

202.1.1.3 Needed Data

1. More corrosion data on uranium alloys in the range of Si addition to 4 w/o is needed to see if significant reductions in corrosion rate can be obtained.
2. If enough improvement in corrosion resistance can be obtained, suitable fabrication techniques must be developed so that representative mechanical properties can be determined.

202.1.1.4 General Comment

At present, lack of significant data prevents a conclusive evaluation of this alloy system.

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203 CERAMIC FUELS - W. E. Roake

Ceramics are chemical compounds, or mixtures thereof, generally made into useful shapes by processes requiring unusually high temperatures. Ceramics have high compressive strength, low tensile strength, high melting points, poor thermal conductivity, low ductility, low atom density, and poor thermal shock resistance. These properties can be "improved" (made more like metals) by additions of metallic fibers or infiltrants. Such products are conveniently called cermets.

All of the commonly used fissionable elements can exist as refractory chemical compounds such as oxides, carbides, nitrides. These compounds can be fabricated by modifications of conventional ceramic forming techniques. They are also useful in granular form as dispersions in non-fissile matrices, or in powder containing elements such as those produced by rotary swaging or by isostatic pressing. Non-fissile ceramic cladding materials and diluents are also used, particularly in gas cooled and military reactor fuel elements.

More degrees of freedom are possible in designing ceramic fuel elements than in designing metallic fuel elements. Ceramic ware can be sintered to densities varying from 100 per cent to 70 per cent or less, with an accompanying wide variation of physical properties. The physical and mechanical properties of ceramics and the common occurrence of thermal fracture requires dependence on the cladding material for structural integrity.

Ceramic nuclear fuels should always be considered with respect to their own peculiar and desirable properties. They should in no way be considered as a substitute for a metallic fuel.

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203.1.1 TABLES OF PROPERTIES OF URANIUM OXIDES

203.1.1.1 Composition

Phase Diagram for Uranium Oxides
From UO_2 to U_5O_{13}

203.1.1.4 Thermal Properties

- Linear Thermal Expansion (80 to 2300 F)⁽⁴⁾
 $\alpha = 1.539 \times 10^{-2} + 4.637 \times 10^{-4} T + 6.630 \times 10^{-8} T^2$
 where T = degrees Fahrenheit
 α = per cent thermal expansion based on unit length
 at 80 F = $\left[\frac{\text{in.}}{\text{in.}} \right]_{\text{F}}$
- Specific Heat (0-200 C)⁽⁵⁾
 $0.056 \text{ Btu lb}^{-1} \text{ F}^{-1}$
- Thermal Conductivity (range 0 - 1800 F)⁽⁶⁾
 $K_{\text{UO}_2} = \frac{4530 \rho}{(T+600) \rho_0} \text{ Btu/hr} \cdot \text{ft} \cdot \text{F}$
 where ρ = density, gm cm^{-3}
 ρ_0 = crystallographic density, 10.97 gm cm^{-3}

203.1.1.5 Mechanical Properties

- Tensile Strength
5000 psi at 25 C
- Compressive Strength
200,000 psi
- Dynamic Elastic Properties⁽⁷⁾ (Room Temperature)
 (Strongly dependent on fabrication history)
 - Young's Modulus - 26×10^6 psi
 - Shear Modulus - 10^7 psi
 - Bulk Modulus - 21×10^6 psi
 - Poisson's Ratio - 0.291

203.1.1.2 Fabrication Methods⁽¹⁾

- Ceramic Forming Processes
 - Cold dry pressing
 - Isostatic pressing
 - Extrusion
 - Slip casting
 - Soft plastic molding
 - Hot dry pressing (pressure sintering)
 - Rolling
- Sintering Atmospheres
 - Hydrogen
 - Inert gas
 - Vacuum
 - Steam
- Compaction for Non-sintered Fuel Bodies
 - Tamping
 - Vibratory packing
 - Rotary swaging
 - Cold die pressing
 - Isostatic pressing

203.1.1.3 Physical Properties

- Crystal density: 10.97 gm cm^{-3}
- Lattice constants
 - Face centered cubic⁽²⁾
 - Lattice constant in $\text{\AA}$ ⁽²⁾
 $\text{\AA} = 5.69 - 0.111 r$
 where $r = \frac{\text{moles O}}{\text{moles U}}$
 valid in range $r = 1.9$ to 2.35
- Phase transformation points
 - M. P. $2750 \pm 10 \text{ C}$ ⁽³⁾
 - B. P. Estimated 3150 C

203.1.1.6 Chemical Reactions

- Solids
 - $\text{UO}_2 + \text{Al}$; reacts measurably above 450 C to produce intermetallic compounds.
 - $\text{UO}_2 + \text{Zr}$; reacts measurably above 600 C.
 - $\text{UO}_2 + \text{Stainless Steel}$; reacts measurably above 1200 C.
 - $\text{UO}_2 + \text{C}$; forms carbides and metal above about 1600 C.
- Liquids
 - $\text{UO}_2 + \text{H}_2\text{O}$; stable to at least 2750 C/
 - $\text{UO}_2 + \text{NaK}$; stable.
 - $\text{UO}_2 + \text{monoisopropyl biphenyl}$; stable at 400 C.
- Gases
 - $\text{UO}_2 + \text{H}_2$; stable.
 - $\text{UO}_2 + \text{steam}$; stable in equilibrium system.
 - $\text{UO}_2 + \text{Halogen}$; reacts.
 - $\text{UO}_2 + \text{Halogenated organics}$; reacts with many.
 - $\text{UO}_2 + \text{N}_2$; stable.
 - $\text{UO}_2 + \text{O}_2$; oxidizes to U_3O_8 , UO_3 , U_4O_7 , etc., depending on temperatures.

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203.1 URANIUM OXIDE - W. E. Roake

Among the ceramic fuel materials, the uranium oxides have received the most attention. The important advantages of uranium oxides are chemical stability toward high temperature coolants and ability to withstand irradiation exposures of at least 3×10^{21} nvt without lattice distortion.

203.1.1 Comments on the Table of Properties

203.1.1.1 Composition

Uranium commonly exists in compound form in tri-, tetra-, and hexavalent states. Oxides are commonly found in the latter two valence states and are reported in lower and intermediate states such as in the compounds U_4O_9 and UO . The high temperature stable form most useful as a nuclear fuel is UO_2 .

203.1.1.2 Fabrication

Conventional techniques which are generally applicable to uranium dioxide fabrication include cold die pressing, isostatic pressing, extrusion, and slip casting, each followed by sintering to achieve a chosen density and porosity. Pressure sintering also has been studied. Sintering is ordinarily accomplished at temperatures between 1200 and 1800 C in an inert or a reducing atmosphere. Relatively low sintering temperatures can be used in steam atmosphere, but the product contains excess oxygen which reportedly seriously affects fission gas retention, and chemical and mechanical stability.

Less conventional compaction techniques (such as rotary swaging, vibratory packing, tamping, and isostatic pressing of clad powders) show promise of fuel element manufacturing economies. Such elements must be engineered to accommodate extensive fission gas release and fuel relocation which may occur during irradiation if further sintering or melting takes place.

The success of each of these methods depends primarily on the properties of the raw materials. Methods are developed for producing, characterizing, and activating uranium oxides for each method. ⁽⁹⁾

203.1.1.3 Physical Properties

Density: UO_2 has a maximum density of 10.97 gm cm^{-3} . Thus, far fewer uranium atoms can be packed in a unit volume than in the case of U metal. A virtue is found here, however, in that the crystal lattice is sufficiently open that it will accommodate as much as 20 per cent excess oxygen without crystal lattice expansion. In fact, the lattice constant actually decreases slightly with the excess oxygen addition. This open lattice permits retention of relatively large quantities of rare gas fission products, which have atomic dimensions similar to those of oxygen. The crystal structure of UO_2 is face-centered cubic. Because the material is isotropic, excellent dimensional stability during irradiation is expected and observed. No dimensional changes have been observed in specimens irradiated to greater than 8.5×10^{21} nvt. No phase transitions have been observed in UO_2 below the melting point, 2750 C.

203.1.1.4 Thermal Conductivity

Two separate measurements have been made of the thermal conductivity of sintered UO_2 as a function of temperature. Kingery et al. ⁽⁶⁾ utilized a comparative linear flow apparatus on a one-inch cube of UO_2 prepared by slip casting and vacuum sintering to 73.3 per cent of theoretical, crystallographic density. Extrapolation to 100 per cent density was made according to a theoretical analysis by Loeb and demonstrated by Kingery and Franci. ⁽⁶⁾ Measurements were made to 1000 C. Other measurements by Hedge ⁽¹⁰⁾ are not considered acceptable. Hence, the Kingery data are accepted as more accurate material values. Quite recent (January, 1958) measurements by Kingery ⁽¹¹⁾ on high (90-95%) density UO_2 confirm his earlier

measurements. Thermal conductivities are being measured by a linear flow method at BMI on samples prepared at HAPO by extrusion, isostatic pressing, and hydrogen sintering to densities in the range 85 to 97 per cent of the theoretical, crystallographic density. Measurements will be made on similar samples irradiated at low temperatures (50-200 C) to 2.3×10^{21} nvt. Since maximum temperatures may be a limiting design parameter for some oxide fuel elements, thermal conductivity values useful in estimating temperatures in operating fuel elements which may have internal cracking are necessary design information.

203.1.1.6 Chemical Reactions

Among uranium bearing fuel materials UO_2 is noted for its chemical stability. It does not react with common coolants or any reducing gas. The chemical reactions are well covered in volume VIII-5 of the National Nuclear Energy Series, "The Chemistry of Uranium" by Katz and Rabinowitch. UO_2 is chemically stable in high temperature water and, for most practical purposes, is compatible with cladding materials such as stainless steel, zirconium and aluminum. It is reduced at high temperatures by the alkaline earth metals, but only by lithium among the alkalis. It reacts at high temperatures with aluminum, a potentially useful matrix material. It has possible advantages in chemical separations because of its oxidation state.

Air oxidation of UO_2 at elevated temperatures requires precautions in handling and treatment. It does not bond easily to metallic cladding materials. Low tensile strength permits thermal cracking.

203.1.2 Design Limits

1. UO_2 - cladding interface temperature

(a) UO_2 - Al; 450 C

(b) UO_2 - Zircaloy; 600 C

(c) UO_2 - Stainless Steel; 1200 C

2. Molten phase operation because of volume expansion on melting estimated 5%.

3. Comments

Design limits are imposed on the use of a material in two ways: first, by known mechanisms of failure under certain conditions, and second, by ignorance and fear of what may happen if current design conditions are exceeded. Examples of the first type, in the case of UO_2 , are core-clad interface temperatures at which measurable chemical reaction begins. An example of the second type is fear of rupture of the cladding due to excessive internal fission gas accumulation. A current summary of fission gas retention has been published by Eichenberg et al.⁽⁸⁾ In summary, they show that observed amounts of gas release arise from a bulk diffusion mechanism sensitive to operational and structural features of the particular UO_2 bodies being irradiated. It is not yet possible to accurately calculate and predict such release data. A currently useful generalization is that UO_2 sintered to at least 94 per cent density will retain 99 per cent or more of the generated fission gas atoms if no phase change occurs. Lower density, or powdered UO_2 will release a large fraction (i. e. 30 per cent).

There is considerable reluctance toward operating a UO_2 fuel element with a portion of the core molten. Primarily, this is because there is no knowledge of the expansion of UO_2 on melting. Currently this is estimated to be about 5 per cent or less. Complete fission gas release should be expected from the molten zone. The fuel element must, of course, be engineered to accommodate the internal pressure.

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203.1. 3 Needed Data

There are three main areas in which data are needed. These are:

1. Effect of irradiation on UO_2 thermal conductivity
2. Effect of non-stoichiometry on:
 - (a) thermal conductivity
 - (b) irradiation stability
 - (c) fission gas retention
3. Rates of fission gas release in non-sintered UO_2 fuel elements

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1. The first step in the process is to identify the problem or issue that needs to be addressed. This involves gathering information and understanding the context of the problem.

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204 PLUTONIUM AND PLUTONIUM ALLOYS - W. J. Bailey

Plutonium has a very prominent role to play in the field of nuclear power production and it is becoming increasingly evident that we must endeavor to enlarge our knowledge of the usage of plutonium and its alloys as reactor fuel materials.

Experimentation with plutonium and plutonium alloys as reactor fuel materials has been quite limited up to the present time. The plutonium-containing materials which have been employed were not in the proper composition range for direct application to fast reactor systems. Engineering heat transfer limitations in power reactors severely restrict the use of fuel elements with high concentrations of plutonium and focus attention on the dilute plutonium alloys as the prime fuel material source for reactor systems. An incentive for using plutonium as a fuel is created because of the possibility of breeding (breeding ratio of at least 1.5 for Pu^{239} fuel -- U^{238} blanket) in fast reactors. Also, since plutonium is separated from uranium by chemical means and can be used as a source of fuel enrichment, the use of plutonium fuels would relieve the need for diffusion plants to create enriched fuel materials.

This section reports some of the characteristics and properties of plutonium and plutonium alloys which are needed to design fuel elements to meet the requirements of both fast and thermal reactors.

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Abstract

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204.1.1 TABLE OF PROPERTIES OF PLUTONIUM (Pu)

204.1.1.1 Composition and Reactivity

Mass Number - most abundant isotopes 239, 240, 241
Source - produced mainly by neutron capture by ^{238}U in nuclear reactors

Nuclear Properties of the Isotopes of Plutonium⁽⁵⁾

Nuclide	Half-life	Decay and energy, Mev	Thermal Neutron Data			
			σ_a	σ_f	σ_c	σ_{tr}
Pu^{239}	24,360 y	α , 5.15, 5.14, 5.10 γ , .384, .124, .10, .039, .05	1,022	731	291	2.9
Pu^{240}	6,600 y	α , 5.16, 5.12, 5.01 γ , 0.05	350	4	346	3
Pu^{241}	13 y	β , (99%) 0.02	1,336	971	365	3
Pu^{242}	4×10^5 y	α , 4.90, 4.85	22.9	--	22.9	--

σ = No. of neutrons per fission

204.1.1.2 Methods of Fabrication

α - phase by hydraulic pressing
 δ - phase (ductile) by forging, pressing and extrusion
 β and γ - phases by warm working
Casting is often used for remote fabrication.

204.1.1.3 Physical Properties

Melting Point - $1159.6 \pm 12.6^\circ\text{F}$ ⁽³⁾
Boiling Point - 5846°F ⁽²⁴⁾
Heat of Vaporization - $595 \pm 2.5 \text{ Btu/lb}$ ⁽³⁾
Vapor Pressure - $\log_{10} P = \frac{-(17,587 \pm 73)}{T(^{\circ}\text{K})} + (7.895 \pm 0.047) \text{ mm Hg}$ ⁽³⁾

Phase-Transformation Data on Plutonium Metal^(3,4)

Transformation	Temperature, $^{\circ}\text{F}$	% Volume Change
$\alpha - \beta$	252	8.9
$\beta - \alpha$	397	2.4
$\gamma - \delta$	603	6.7
$\delta - \delta'$	847	-0.4
$\delta' - \epsilon$	891	-3.0
$\epsilon - \text{liq}$	1183.1 ± 3.6	

-- On heating only

Crystal Structure Data for the Plutonium Phases⁽⁴⁾

Phase	Crystal lattice	Unit cell dimensions, $\text{\AA}$	No. of atoms in unit cell	Calculated density (g/cm^3)
Alpha	Monoclinic	At 70 F $a = 6.1835 \pm 0.0005$ $b = 4.8244 \pm 0.0005$ $c = 10.673 \pm 0.001$ $\beta = 101.81^\circ \pm 0.02^\circ$	16	19.818 ± 0.006
Beta	Crystal structure unknown			
Gamma	Face-centered orthorhombic	At 455 F $a = 3.1587 \pm 0.0004$ $b = 5.7682 \pm 0.0004$ $c = 10.162 \pm 0.002$	8	17.14 ± 0.01
Delta	Face-centered cubic	At 608 F $a = 4.6371 \pm 0.0004$	4	15.92
Delta-Prime	Body-centered	At 859 F $a = 3.327 \pm 0.003$ $c = 4.482 \pm 0.007$	2	16.00
Epsilon	Body-centered cubic	At 914 F $a = 3.6361 \pm 0.0004$	2	16.51

204.1.1.4 Thermal Properties

Coefficients of Thermal Expansion of the Plutonium Phases⁽⁴⁾

Phase	Temperature range ($^{\circ}\text{F}$)	Coefficient of linear expansion ($\text{in/in}/^{\circ}\text{F}$)
Alpha	-303 to 212	$\bar{\alpha} = (26.2 \pm 0.03 + 0.056 t) \times 10^{-6}$
Beta	271 to 397	$\bar{\alpha} = (18.7 \pm 0.6) \times 10^{-6}$
Gamma	410 to 590	$\bar{\alpha} = (11.0 \pm 0.65) \times 10^{-6}$ $\bar{\alpha}_a = (23.0 \pm 0.35) \times 10^{-6}$ $\bar{\alpha}_b = (44.8 \pm 0.1) \times 10^{-6}$ $\bar{\alpha}_c = (19.3 \pm 0.4) \times 10^{-6}$
Delta	608 to 824	$\bar{\alpha} = (4.8 \pm 0.2) \times 10^{-6}$
Delta-Prime	869 to 905	$\bar{\alpha} = (170 \pm 20) \times 10^{-6}$ $\bar{\alpha}_a = (-366 \pm 40) \times 10^{-6}$ $\bar{\alpha}_b = (-9 \pm 15) \times 10^{-6}$
Epsilon	914 to 1022	$\bar{\alpha} = (20.3 \pm 0.4) \times 10^{-6}$
Liquid	at 1229	Vol. coefficient = $(28 \pm 14) \times 10^{-6}$

Specific Heat⁽⁴⁾

Alpha Pu (at 77 F)	Beta Pu (at 320 F)
$C_p = 0.033 \text{ Btu/lb-}^{\circ}\text{F}$	$C_p = 0.038 \text{ Btu/lb-}^{\circ}\text{F}$
$C_v = 0.030 \text{ Btu/lb-}^{\circ}\text{F}$	$C_v = 0.036 \text{ Btu/lb-}^{\circ}\text{F}$

Thermal Conductivity

Thermal Conductivity of High Purity Plutonium⁽²⁸⁾

Phase	Temperature ($^{\circ}\text{F}$)	k (Btu/hr-ft- $^{\circ}\text{F}$)
α	59 to 230	3.15 ± 0.053
β	270 - 282	8.71

204.1.1.5 Mechanical

Property	Value	Alpha Plutonium	
		Pressure coefficient	Temperature coefficient
		($\times 10^{-6}$ /psi)	($10^{-5}/^{\circ}\text{F}$)
Young's Modulus ⁽⁴⁾	14.4×10^6 psi	+1.24	-0.71
Shear Modulus ⁽⁴⁾	6.29×10^6 psi	+0.96	-0.81
Poisson's Ratio ⁽⁴⁾	0.15	+1.8	+0.77
Micro-hardness ⁽⁴⁾	233 to 250 (kg/mm 2)		
Ultimate Strength ⁽²²⁾	61,500 psi	--	--
Yield Strength ⁽²²⁾	38,200 psi	--	--
(0.02% offset)			

204.1.1.6 Chemical Reactions

Compatibility of Plutonium Metal with Gases

Gas	Product of Reaction	Rate of Reaction
H_2	$\text{PuH}_{2.00}$ to $\text{PuH}_{3.00}$ ⁽⁵⁾	Slow at 77-122 F, very rapid at 392 F.
O_2	PuO , Pu_2O_3 , PuO_2 ⁽⁵⁾	Slow at room temperature. Protective oxide film is formed. Reaction increases rapidly with temperature. Presence of moisture accelerates oxidation.
CO_2	PuO_2 (mainly) ⁽⁵⁾	Rapid at comparatively low temperatures.
N_2	PuN ⁽⁵⁾	Very slow at low temperature. Slow at temperatures in range 1472-1632 F. ⁽⁵⁾

F_2 , Cl_2 , Br_2 , I_2

Appropriate halides in which valency of Pu may vary between 3 and 6.

Reaction with all halogens.

Air

Oxide ^(5,23,24)

Oxidizes slowly in air up to 212 F. Oxide film forms immediately on exposure to air and even in vacuums of 10^{-6} mm Hg. Moisture accelerates reaction.

Inorganic Gases, Halogen Acid Gases

--

Vigorously attacked ⁽⁵⁾

Compatibility of Plutonium Metal with Liquids

Liquid	Rate of Reaction
NaK	Not dissolved. ⁽²⁵⁾ Rate $0.04 \text{ mg/cm}^2 \text{ hr}$ for Pu wires ⁽²⁶⁾ at 760 F.
Water, acids ⁽⁵⁾	Smoothly

Compatibility of Plutonium Metal with Metals or Solids

Metal	Rate of Reaction
Mo, Ti, Zr, V	No reactions in solid state, all reacted when Pu was molten. ⁽²⁷⁾
Hg, Al, Mg, Bi, Mo, Sn ⁽⁵⁾	Form compounds
Li ^(4,5)	Insoluble
C ^(4,5)	Forms PuC , Pu_2C_3 . Isostructural with uranium carbides.
Halogens, Sulfur ⁽⁵⁾ Compounds	Vigorously attacked

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204.1 PLUTONIUM

The metal plutonium is the product or a by-product of almost every nuclear reactor. The fertile isotope of uranium is transformed to plutonium by neutron absorption according to the reaction: ^(5,6)

A portion of the plutonium is fissioned, in situ, and acts as a stabilizer of reactivity by adding its energy to that of the U^{235} , however, a large quantity of the plutonium that is generated is ejected unburned in the used or spent fuel elements, even in the reactors of high efficiency. ⁽²⁾ The plutonium obtained from fuel elements which have been subjected to relatively short irradiation exposure is almost completely all Pu^{239} . By extending the irradiation period of the fuel element, the isotope Pu^{239} absorbs neutrons and is converted progressively to the higher plutonium isotopes (up to mass number 243) which are more or less stable. ⁽⁵⁾

A natural or slightly enriched uranium fuel element under irradiation becomes alloyed with increasing quantities of plutonium as the in-reactor exposure progresses. The plutonium may be extracted by chemical or pyrometallurgical processing techniques.

204.1.1 Comments on Properties of Plutonium204.1.1.1 Composition and ReactivityNeutron Reproduction Factor, η

Pure Pu^{239} has a high fission yield; however, the η value, number of neutrons produced/neutrons absorbed, is scarcely sufficient to insure breeding in a thermal reactor system. In a thermal reactor system when the Pu^{239} is contaminated with the non-fissile isotopes Pu^{240} , Pu^{242} and Pu^{243} , which occurs with Pu^{239} after prolonged irradiation, the η value falls below 2.0. Raising the moderator temperature, which increases the

average neutron velocity, causes the value of η to decrease still further. In a fast neutron system all plutonium isotopes are fissile and the value of α (ratio of capture to fission cross sections) is quite low; hence, the η values become attractive for breeding operations.

204.1.1.2 Fabrication

Non-Remote Fabrication⁽⁵⁾

The alpha phase of plutonium is brittle. It is not easily deformed except by hydraulic pressing. The delta phase of plutonium is quite ductile and may be formed by conventional methods such as forging, pressing, and extrusion. The beta and gamma phases will probably require warm working for forming since they are brittle in nature. Deforming or shaping of plutonium is most satisfactorily performed at elevated temperatures, however, one should strive to use the lowest practicable temperature in order to reduce the amount of oxidation and to minimize the compatability problems. Distortion, due to the significant volume changes encountered on cooling, may be a problem of considerable concern on plutonium specimens finished at elevated temperatures. Castings of plutonium may be produced with level or flat tops because the metal does not shrink when it solidifies. Plutonium metal may be subjected to cutting, grinding, or machining, however, the physical and metallurgical properties must be given careful consideration. Since plutonium, especially in finely divided form, is pyrophoric and extremely toxic, it must be emphasized that all forming operations performed on the material, particularly at high temperatures, involve serious hazards and must be conducted under adequately controlled conditions.

Actual manual handling times must be minimized in order to avoid excessive radiation exposure to the hands. When it is necessary to perform operations with long exposure plutonium from power reactors, the high activity levels necessitate heavy shielding and remote manipulators.

Remote Fabrication

A method has been described by Shuck⁽⁸⁾ for the fabrication by remote means of solid fuel elements of Pu and U²³³. It involves the injection of the molten fuel into precision ceramic molds. Dry runs have produced fuel cylinders two feet long and 0.140 inches in diameter with a diametral tolerance of 0.001 inches.

Plutonium handling facilities are in general located in hoods or sealed glove boxes which are maintained at a slightly negative pressure. The exhaust from plutonium-handling hoods is filtered and care is taken to see that the concentration of plutonium in the exhaust does not exceed 2×10^{-12} $\mu\text{c/ml}$.

Criticality Hazards

Plutonium has a critical mass of only a few pounds. If the mass is increased beyond the critical value, the associated radiation hazard from gamma rays and neutrons is tremendously increased. The more serious risk of an explosion may also be introduced. All proposed operations involving plutonium should be reviewed and cleared by an experienced nuclear physicist before work is initiated and from a criticality standpoint satisfactory means should be provided for metal storage, fabrication and transportation.

Health Hazard^(2, 5, 9)

The health hazard associated with plutonium handling is of prime importance since it is relatively independent of the amounts of plutonium involved. Plutonium is an alpha emitter and has a half-life of 24,300 years. The particles can inflict serious damage to the body tissue in the immediate vicinity surrounding the point of deposition of the plutonium. Some plutonium isotopes also emit high energy X and low energy gamma radiation. Should plutonium enter the body bloodstream, it is rapidly and primarily deposited in and on the bone surfaces. The

biological half-life of plutonium in the body is approximately 100 years. The major danger in this event is the interference with the production of white corpuscles. The most important health hazard is the inhalation and consequent absorption through the lungs of plutonium and its compounds. Plutonium has an ingestion tolerance of 1×10^{-7} of 2×10^{-6} Mgm/kg and its air concentration tolerance is only one billionth that of chlorine.

For plutonium fixed in bone matter the present maximum permissible body burden has been established at 0.04 microcuries or about 0.5 micrograms. At HAPO, an employee is restricted from work with plutonium or a biologically similar isotope if his body burden has been determined to be in excess of one-fifth of the maximum allowable limit.

The stringent tolerances require exceedingly fine care in the fabrication and operation of plutonium handling facilities. A major reason for such care is the extreme mobility of plutonium, believed to be due to the formation of microscopic oxide particles.

Pyrometallurgical Processing

Pyrometallurgical techniques have been investigated and recent developments have been made to the extent that plutonium metal, from which the more serious effects of irradiation-induced damage have been removed, may be returned or recycled directly for formation into fuel elements by remotized fabrication methods. ⁽⁵⁾

Essentially the pyrometallurgical process ^(5, 17) consists of melting the irradiated fuel material in a vacuum to extract the more volatile elements. The elimination of or reduction in the quantity of undesirable metals, in particular the rare earths, in the melt is accomplished by slagging.

As an alternative method, a plutonium-uranium alloy of higher concentration and with a rather low radioactivity level may be obtained by treating the decontaminated melt with a fused salt or a liquid metal such as silver. ⁽¹⁸⁾ The resultant alloy can be subjected to additional processing fairly inexpensively or may be applied immediately as an enrichment material. ⁽⁵⁾

204.1.1.3 Physical Properties

Phase Transformations

Slightly different values have been obtained by investigators for the phase transformation temperatures and volumetric changes associated with transformation^(12,13,14) even though the experimental techniques were quite similar. In general the Los Alamos data have been preferred because the Los Alamos metal was 99.8 w/o Pu while the metal available elsewhere was on the order of 99 w/o Pu. It should be noted that the phase transition temperatures shown on the data sheet were obtained on heating at a slow rate.

Density

The density values listed were calculated from X-ray data with the single exception of the beta phase. The dilatometric values are thought⁽⁵⁾ to be two-three per cent lower than the X-ray values partially because of micro-void formation associated with phase changes⁽⁴⁾ and partially due to retention of higher temperature phases. For this reason one may encounter appreciable volumetric contractions in alpha-phase plutonium which is allowed to stand at room temperature. Density changes encountered during the heating of plutonium are notable, especially from α to δ phase (20 per cent decrease) and from δ to ϵ phase (3 per cent increase).

204.1.1.4 Thermal Properties

Thermal Expansion

The values for the coefficient of thermal expansion, which were calculated from X-ray data, are not in complete agreement with the dilatometric values. The δ and δ' s phases of plutonium are unique in comparison to other isotropic metals in that they have a negative coefficient of thermal expansion. This fact has not yet been satisfactorily explained.⁽⁵⁾ It should be observed that plutonium undergoes practically no volume change on melting or solidification.

204.1.1.5 Mechanical Properties

On the declassified elastic moduli for plutonium, Bagley⁽⁵⁾ observes that:

- (1) The most consistent set of values for the elastic constants have been obtained by Lacquer.⁽⁴⁾
- (2) A. E. Kay and B. W. Lowthian⁽¹⁵⁾ conducted room temperature compression tests and determined a value of 14.4×10^6 psi for Young's Modulus and 0.4 for Poisson's ratio.
- (3) The compressibility data are in close agreement with values obtained by P. W. Bridgeman.⁽⁴⁾

Recent declassified data by Gardner⁽²²⁾ on the mechanical properties of pure plutonium are shown below:

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<u>Pu Tensile Specimen</u>		<u>Average Tensile Properties</u>							
Size Diameter (in)	Treatment	Ultimate Strength (psi)	Yield Strength (0.02% offset) (psi)	Modulus of Elasticity (10 ⁶ psi)	Modulus of Resilience (in-lb/cu in)	Hardness (10 kg load) (DPH)	Density (g/cc)	Temp. of Test (° F)	Total Impurity content (ppm)
0.250	As cast and cold treated.	62,800	39,300	13.6	23.5	262	19.51	87.8	975
0.250	As cast, delta annealed and cold treated	61,000	38,700	12.8	28.6	259	19.51	87.8	1091
0.500	As cast and cold treated	59,100	35,300	14.1	16.3	269	19.49	93.2	1514
0.500	As cast, delta annealed and cold treated	63,100	38,900	13.6	18.2	270	19.33	93.8	1254

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204.1.1.6 Chemical ReactionsCommentsUnirradiated

Bagley⁽⁵⁾ reports that plutonium is apparently more reactive chemically than uranium. Plutonium oxidizes slowly in dry air at 122 F forming PuO which may be protective.⁽¹⁵⁾ If moisture is introduced into the air the oxidation rate is accelerated and non-protective oxide layers (PuO₂) are formed. Bagley points out that under the same temperature environment the plutonium will oxidize more rapidly than uranium and that plutonium metal will ignite in air at about 572 F.

At 392 F, plutonium rapidly reacts with hydrogen to form PuH₃. The reaction occurs at an appreciable rate in the temperature range 75.2 - 122 F.⁽¹⁹⁾ The reaction is believed to proceed in two steps and the products are PuH₂ and PuH₃ which normally exist in solid solution. The solid solution has a reactivity very much like that of plutonium metal.⁽²⁰⁾

No data are available on the compatibility of plutonium with the alkali metals; however, lithium has been employed in the reduction of some compounds of plutonium.⁽⁵⁾ Formation of solutions or intermetallic compounds has not been noted. Bagley also observes that it is quite certain that in a circulating system plutonium will reduce any oxide of potassium, sodium, etc., present in the system. This could result in serious corrosion attack of the metal unless strict purity standards are imposed.

It is known that the low melting point metals, Al, Mg, Bi, Pb, Sn, and Hg, form plutonium compounds.⁽⁵⁾ When in contact with Hg or Al in air, the metallic Pu will grow fronds of PuO₂. No liquid metals, except for the alkali metals and alkaline earth metals, are likely to be compatible with plutonium.

The intermetallic compound formation by plutonium with high melting point metals is very analogous with that of uranium. Bagley observes that application of the Hume-Rothery 15 per cent rule indicates that size factor considerations favor extensive solid solubility between delta or epsilon plutonium and Zn, Hf, Ta, Nb, Ti, W and Mo.

Strictly speaking, these metals are incompatible with plutonium and it may be predicted that these metals will contain plutonium for reasonable long periods at temperatures approaching the melting point, if it is assumed the diffusion rates are slow.

Melting and casting of plutonium present perplexing problems because of the extreme reactivity of plutonium. As Bagley notes, graphite reacts with plutonium and most of the refractory oxides, even those which appear to be thermodynamically more stable than plutonium oxide, e. g. , BeO, are decomposed by plutonium with the formation of plutonium oxide and intermetallic compounds. Tantalum or tungsten may offer a short-term solution, they are expensive and difficult to fabricate.

204.1.2 Design Limits

Some limiting factors on plutonium fuel element design are:

- (a) low melting point (11696 F)
- (b) marked shape, volume and density changes associated with phase transformations, and
- (c) low thermal conductivity.

Plutonium metal appears to be more chemically reactive than uranium, will oxidize more rapidly than uranium at the same temperatures, and will ignite in air at about 572 F. ⁽⁵⁾ The metal oxidizes rapidly in air and the rate is accelerated by the presence of moisture. ⁽²⁾

204.1.3 Needed Data

- (1) Thermal conductivity values for plutonium phases other than alpha and beta need to be obtained.
- (2) More information is required on the irradiation behavior (diffusion, corrosion, dimensional stability, etc.) of pure plutonium (solid or molten) fuel materials with the associated jacketing or container materials.
- (3) Corrosion rates in water should be obtained.
- (4) Biological and criticality hazards for irradiated or recycled plutonium fuel elements should be established.

204.1.4 General Comments

The primary function of plutonium as a reactor constituent has been as the nuclear reactor product. The Los Alamos Fast Plutonium Reactor, Clementine, has been the single exception since it operated with plutonium as a fuel material.^(10, 21) In this particular case the fuel elements were pure plutonium rods encased in mild steel. The reactor operated from 1946 until 1952, at which time a fuel element ruptured, and was disassembled in 1953.⁽²⁾

When one considers the highly toxic nature of plutonium, the criticality hazards, and the unique nuclear, chemical, and metallurgical properties, it appears that the use of pure plutonium as a fuel rod will be very limited.^(5, 22) With such a high cross section to thermal neutrons, pure plutonium in the form of a fuel element with other than a geometrically thin cross section would absorb all the neutrons in the outer layer and the interior would be shielded and essentially not used. For this reason, pure plutonium fuel elements of any size must of necessity be restricted to fast reactor systems, but here the significant problems of unusual metallurgical properties and rather low melting point

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definitely limit the application of pure plutonium fuel elements to reactors like the Zephyr where the amount of heat developed is negligible. (22)

As Wyatt⁽²²⁾ and Bagley⁽⁵⁾ point out, if the experience gained from uranium experiments is any criterion, attempts to obtain heat from irradiation of pure plutonium will yield extraordinary perimetric and volumetric changes, due partially to the accumulation of fission products, to structure alterations due to irradiation, and to effects produced by thermal cycling. Unless provisions are made to maintain a low fuel element surface temperature, the rather low thermal conductivity for pure plutonium may lead to melting at the specimen center. There appears to be little doubt that plutonium-containing fuel elements with quite reasonable irradiation behavior could be produced by combining or diluting the plutonium with uranium, thorium, or non-fissile metals of low neutron absorption cross section like aluminum, zirconium, iron, or nickel with which plutonium is known to form stable alloys.

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301.1.1 TABLE OF PROPERTIES OF 2S ALUMINUM

(Aluminum Assn. No. 1100, Commercial 2S, ASTM designation 990 A.)

301.1.1.1 Composition⁽¹⁾

Element	Al	Cu	Fe + Si	Mn	Zn	Others
w/o	99 min. 0.20 max.		1.00 max.	0.05 max.	0.10 max.	0.15 max.

Nuclear Constants⁽²⁾

- a) Microscopic thermal neutron cross section (σ) 0.23 $\frac{\text{barns}}{\text{at}}$
 b) Macroscopic thermal neutron cross section (Σ) 0.012 cm^{-1}

301.1.1.2 Fabrication⁽³⁾

The alloy aluminum 1100 is noted for its versatility and ease of fabrication. However, it should be noted that 1100 has poor casting qualities and is therefore classed as a wrought alloy.

301.1.1.3 Physical Properties

Density 0.098 [lb/in^3] (68 F)
 Melting Point Range 1180 - 1215 F

301.1.1.4 Thermal Properties

Thermal Conductivity (78 F) 2.09 [$\text{Btu/hr} \cdot \text{F} \cdot \text{ft}$]
 Avg. Coefficient of Linear Thermal Expansion $\times [10^{-6} \frac{\text{in/in}}{\text{F}}] = \alpha$
 Temp. Range 68 - 212 F 68 - 392 F 68 - 572 F
 α 13.1 13.7 14.2
 Specific Heat (212 F) 0.414 Btu/lb

301.1.1.5 Mechanical Properties⁽¹⁾ 1100-0 (annealed condition)

a) Tensile Strength	d) Compressive Yield Strength	g) Modulus of Elasticity (tension)
13,000 psi	~5,000 psi	75 F 10 ⁴ psi
b) Yield Strength (tension)	e) Torsional Yield Strength	212 F 9.7 x 10 ⁶
5,000 psi	~2,750 psi	300 F 9.5 x 10 ⁶
c) Elongation in 2 inches	f) Torsional Ultimate Strength	400 F 9.0 x 10 ⁶
35% (1/16 inch thick specimen)	~3,250 psi	500 F 8.5 x 10 ⁶
		600 F 6.5 x 10 ⁶
		700 F 4.0 x 10 ⁶
h) Modulus of Elasticity (compression)		
2% higher than in tension		
i) Modulus of Rigidity (shear modulus)		
3.9 x 10 ⁶ psi		
j) Poisson's Ratio		
0.33		

Effect of Cold Work (R. T.)⁽³⁾

Temper (a)	U. T. S. psi	Y. S. psi	Elongation (%) (b) (c)	Shearing Strength %	Brinell Hardness (d)	Endurance Limit (e) psi
1100-0	13,000	5,000	35 45	9,000	23	5,000
1100-H12	15,000	15,000	12 25	10,000	28	6,000
1100-H14	17,000	17,000	9 20	11,000	32	7,000
1100-H16	20,000	20,000	6 17	12,000	38	9,000
1100-H18	24,000	22,000	5 15	13,000	44	9,000

- a) -0 represents the fully annealed state; -H18 fully cold worked (hard); -H12, -H14, -H16 are intermediate degrees of cold working between -0 and -H18.

b) Sheet specimens 1/16 inch thick.

c) round specimens, 1/2 inch in diameter.

d) 500 kg load, 10 mm ball.

e) based on 500,000,000 cycles using R. R. Moore type rotating beam machine.

Cyclic Strain Behavior⁽⁶⁾

Annealed 2S Aluminum

$$N^{\frac{1}{2}} \Delta \epsilon_p = 0.80 \quad .1 < N < 10,000 \quad (\text{range of actual data})$$

20% Prestrain

$$N^{\frac{1}{2}} \Delta \epsilon_p = 0.88 \quad .1 < N < 400$$

40% Prestrain

$$N^{\frac{1}{2}} \Delta \epsilon_p = 0.65 \quad .1 < N < 400$$

Where N = number of cycles to failure, $\Delta \epsilon_p$ = plastic strain range.

301.1.1.6 Uniform Aqueous Corrosion Behavior in Reactor⁽¹⁴⁾

$$\log C. R. = 2.680 - \frac{2602}{T} + 0.036 P + 0.515 \text{ pH}$$

C. R. = corrosion rate

P = K.W. per 8 inch slug

T = temperature, ° K.

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301.1 2S ALUMINUM (No. 1100) - D. L. Gray

The use of aluminum alloy 1100 as a cladding material is desirable because of its low macroscopic thermal neutron cross section, its good heat conducting properties, ductility, availability and ease of fabrication. It is used in water cooled thermal reactors and low temperature gas-cooled reactors, and is a good candidate for organic reactors. The major limiting factors for its use are corrosion resistance and mechanical properties at operating temperatures.

301.1.1 Discussion of Table of Properties30.1.1.5 Mechanical Properties

a.) Tensile Properties: Since aluminum 1100 is a non-heat-treatable alloy, it is usually cold-worked to obtain a desired combination of properties. The range of properties so obtained with various alloy tempers is in the data sheet. These properties recover in the temperature range near 350 F to 400 F (see Figure 1). If strength is required at higher temperatures, 1100 aluminum is not a suitable choice as a cladding material. Under certain circumstances irradiation will cause an increase in tensile strength and yield strength together with a decrease in ductility (see the following table).

The effects are usually more marked for annealed material than for cold worked material. Although ductility is decreased, the decrease is not likely to limit the use of aluminum 1100 as a cladding material.

This irradiation data applies only below 150 F. No data is available concerning irradiation at higher temperatures.

EFFECT OF NEUTRON FLUX ON TENSILE PROPERTIES

<u>Temper</u>	<u>Irradiation Condition</u>	<u>Tensile Strength 1000 psi</u>	<u>0.2% Yield Strength 1000 psi</u>	<u>Per Cent Elongation</u>
1100-0 ⁽⁴⁾	1. 26×10^{21} nvt 150 F			
	Before	13.6	6.8	38
	After	26.0	17.1	21
	% Change	91	150	45
1100-H14 ⁽⁴⁾	1. 26×10^{21} nvt 150 F			
	Before	17.3	16.6	6
	After	26.0	24.0	5.5
	% Change	50	60	8
1100-H16 ⁽⁵⁾	7. 6×10^{20} nvt			
	Before	22.4	20.1	6
	After	28.3	24.4	6
	% Change	26	21	0
	Total 4×10^{20} nvt fast (68 to 140 F)			

b.) Hardness (also see Table III): Hot hardness tests on 1100 aluminum show that it has better resistance to indentation than pure magnesium, but is not as good as APMP 257 and APMP 457.⁽⁷⁾

Irradiation at 68 to 140 F tends to increase hardness. The hardness of aluminum 1100-H16 irradiated to a total integrated flux of 7.6×10^{20} nvt increased 15.5 per cent.⁽⁵⁾

c.) Creep: Although the creep of aluminum has been investigated in some detail, little information has been obtained at the low temperatures at which 2S usually finds application as a cladding material in water cooled reactors.

The temperature range of use however may be extended to temperatures as high as 400 C for organic coolants and even higher for gas atmospheres. Figures 2 to 5 present design curves for 1100-0 aluminum sheet at 400 C and 500 C. (8, 9)

Experiments on the in-reactor creep properties of aluminum cladding indicate that irradiation has little or no effect on creep. (10, 11, 12)

d.) Impact: Aluminum 1100 has an impact energy of 19 ft-lbs at 75 F. (1) It retains its notch toughness even at low temperatures. (At -292 F the impact energy is in fact increased to 27 ft-lbs.)

301.1.1.6 Corrosion and Chemical

a.) Aqueous Corrosion: Aqueous corrosion of aluminum below 200 C generally proceeds uniformly. Above this temperature for 1100 aluminum and practically all commercial aluminum alloys, a non-uniform corrosion process generally called "intergranular", becomes important.

Below 200 C the corrosion rate is affected by water composition and pH, water flow rate, temperature, irradiation, and other conditions. At a constant temperature, rates are rapid at first and then decrease to a low constant value nearly linear with time. This latter portion of the rate curve is used to predict corrosion behavior.

Rates observed for heat producing aluminum clad fuel elements are higher than those for isothermal loops at the same temperature. Corrosion in process water is considerably more rapid than in deionized water at the same temperature. Statistical treatment of a large body of in-reactor corrosion data⁽⁴⁾ produced the following corrosion expression (also in the data sheet),

$$\text{Log C. R.} = 2.680 - \frac{2602}{T^{\circ} \text{ K}} + 0.036P + 0.515 \text{ pH}$$

CR = corrosion rate

T = ° K

P = KW/"C" slug.

which is valid for uniform corrosion. No mention has been made yet of the effect of minor alloying constituents. For uniform corrosion, composition is a negligible factor⁽¹⁵⁾ in determining rates. However, above 200 C, where the so-called intergranular process is important, corrosion is quite sensitive to alloy composition. This process is highly dependent on temperature, and will cause failure in 2S aluminum in 350 C water in approximately 3 hours.⁽¹⁶⁾ Application of aluminum in high temperature water service is restricted to those alloys which are resistant to intergranular corrosion.

b.) Organics: No reaction has been noted between aluminum and any of the organic compounds considered for reactor coolant use up to the breakdown temperature of the organics (~450 C). The hydrogen formed by thermal or radiolytic breakdown of the organic has no detrimental effect on aluminum.

c.) Gases: In dry gas atmospheres at elevated temperatures, aluminum is highly resistant to corrosion. This includes air, oxygen, nitrogen, hydrogen, and carbon dioxide. Possibly the only exceptions would be the halogens and their compounds.

To temperatures as high as 350 C there is little increase in corrosion due to the presence of water vapor.⁽¹⁷⁾

d.) Liquid Metals: The Liquid Metals Handbook⁽¹⁸⁾ gives a summary of the resistance of aluminum alloys to attack by liquid metals. At 300 C only the resistance in liquid Bi-Pb, Bi, Ph, and Tl was listed as "good". Apparently aluminum tends to alloy with most liquid metals, hence its application in a liquid-metal system would definitely be limited.

Information concerning aluminum in NaK indicates that below 210 C long time use may be considered. (19)

301.1.2 Design Limits

a.) Mechanical Properties: Above 300 F the tensile properties produced by cold work begin to recover rapidly (Figure 1). If strength or creep resistance is required for applications of 1100 as cladding, temperature will be a limiting factor.

b.) Effect of Coolant on Cladding Corrosion: The aqueous corrosion properties of 1100 are fairly good up to 150 C. In air and gases (except the halogens) the corrosion resistance is good at elevated temperatures. In this case, however, excess water content of the gas may be limiting.

301.1.4 General Characteristics

Aluminum 1100 alloy is classed as a wrought, non-heat-treatable alloy. Changes in properties are obtained by strain hardening.

This alloy has good ductility and is used where high strength is not required.

It has good heat conductivity, a low cross section for thermal neutrons and good aqueous corrosion resistance at low temperatures.

The alloy shows promise for use in organics since the hydrogen pickup causes no deleterious reaction.

Aluminum 1100 also is easy to fabricate and has good welding and joining characteristics.

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FIGURE 1

Effect of Temperature on Tensile Properties

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FIGURE 2

Design Curves for 1100-0 Aluminum Sheet at 400 C

FIGURE 3

Design Curves for 1100-0 Aluminum Sheet at 450 C

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FIGURE 4

Design Curves for 1100-0 Aluminum Sheet at 500 C

FIGURE 5

Design Curves for 1100-0 Aluminum Sheet at 550 C

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302.1 TABLES OF PROPERTIES OF ZIRCALOY-2

302.1.1.1 Composition and Reactivity (Weight %)

Sn	Fe	Cr	Ni	N	O	Zr
1.2-1.7	0.07-0.20	0.05-0.15	0.03-0.08	0.006 max.	0.17 avg.	Remainder

Thermal Neutron Absorption Cross Section (Macroscopic) - 0.0208 in⁻¹

302.1.1.2 Methods of Fabrication

Zircaloy-2 can be fabricated by most standard methods, but because of limiting ductility, considerable development work must be done before drawn shapes can be produced by quantity production methods.

Forms Available and Experienced Manufacturers⁽¹⁾

1. Standard sizes of rounds, bars and billets, wire, strip, sheet, and plate	Allegheny-Ludlum Co.
Firth Sterling, Inc.
2. Structural Shapes	Babcock and Wilcox Co.
3. Drawn shells and tubing	Bridgeport Brass Co.
Superior Tube Co.
Nuclear Metals, Inc.
Chase Brass Co.
4. Coextruded shapes	Nuclear Metals Co.

Processing Conditions

Fabricating

Forging temperature: 1800 F (980 C)
Rolling temperature: 1550 F (845 C)

Annealing

Hot-rolled material: 1550 F (845 C), 1 hour in vacuum, furnace cool.
Solidification Shrinkage: 1.5 per cent

Welding

With adequate precautions, satisfactory ductile welds can be obtained with Zircaloy-2 by conventional methods.⁽¹⁾ A necessary precaution is the shielding of the material with argon, helium, or vacuum during heating to prevent oxygen and nitrogen pickup. Otherwise, the corrosion resistance and ductility will be greatly impaired.

302.1.1.3 Physical Properties

Density - 6.55 g/cc cm, 0.237 lb/cu in. (room temperature)
Modulus of Elasticity - 14.0 x 10⁶ psi (dE/dT = 5500 psi per °F)
Poisson's Ratio - 0.31
Crystal Lattice - Hexagonal close packed below 1650 F (900 C), Body Centered Cubic above 1630 F (1000 C)
Melting Temperature - 3310 F (-1820 C)

302.1.1.4 Thermal Properties

Thermal Coefficient of Linear Expansion - 3.6 x 10⁻⁶ per °F (Room Temperature to 1290 F)

Thermal Conductivity, (Btu per hr-ft-°F)

77 F (25 C) - 8.41 (extrapolated)
212 F (100 C) - 8.12
752 F (400 C) - 8.19

302.1.1.5 Mechanical (Reactor Grade)

	I. Unirradiated (Room Temperature)		II. Irradiated (100-140 F)
	Annealed	Cold Worked, hard	1.41x10 ²¹ nvt thermal
1. Tensile Strength, psi	70,000-80,000	100,000-120,000	78,000-85,000
2. Yield Strength, psi	45,000-55,000	90,000-110,000	66,000-70,000
3. Elongation (% in 2 in.)	20-25	6	14-18
4. Rockwell Hardness	79-95 B	25-29 C	--
5. Bend Test	120° (dia 2t)	--	--
6. Unirradiated Creep Rate = $\dot{\epsilon}$ [in/in/hr] (650 F)			
	$\log_{10} \dot{\epsilon} = 9.75 + 0.215 \times \frac{\sigma}{1,000}$ for $14 < \frac{\sigma}{1,000} < 27$		
	$\log_{10} \dot{\epsilon} = -246.95 + 9.0 \times \frac{\sigma}{1,000}$ for $27 < \frac{\sigma}{1,000} < \text{tensile strength where}$		
	σ [psi] = tensile stress.		

302.1.1.6 Corrosion and Chemical

TABLE I

Empirical Equations for the Corrosion Behavior of Zircaloy-2 in Water and Steam⁽⁸⁾

°F	°C	psi	Pressure Equation		Time (days)
			m in Mg/cm ²	T in days	
550	288	750	0.01 log m	= 0.005 + 0.003 log T	
600	316	1553	0.01 log m	= 0.0074 + 0.0026 log T	Up to 1150 days
			0.01 (m-34)	= 0.00065 (T-1150)	After 1150 days
680	360	2705	0.01 log m	= 0.0076 + 0.0038 log T	Up to 112 days
			0.01 (m-34)	= 0.0037 (T-112)	After 112 days
750	400	1500	0.01 log m	= 0.011 + 0.0032 log T	Up to 41 days
			0.01 (m-41)	= 0.0127 (T-41)	After 41 days

TABLE II

Values of A and Q in the Arrhenius Equation for Zircaloy-2 Reaction in Dry Air⁽⁹⁾

$$\text{Rate Constant } k = A \exp \frac{-Q}{RT}$$

Initial Reaction		After Transition	
A, (mg/cm ²)	Q, cal/mole	A, (mg/cm ²)/hr	Q, cal/mole
1.1 x 10 ³	3.94 x 10 ⁴	8.5 x 10 ⁶	3.10 x 10 ⁴

Extrapolated corrosion data are given in the following figure.

Extrapolated Curves for Weight Gain and Penetration⁽¹⁹⁾ Versus Time at Various Temperatures for Zircaloy-2

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302.1 ZIRCALOY-2 - A. L. Bement and D. W. Shannon

One of the strong candidates for a fuel element cladding material for high-temperature and/or high-pressure water, steam, and gas-cooled reactors is Zircaloy-2, because of its excellent aqueous corrosion resistance and mechanical strength below 750 F, low thermal-neutron cross section, and excellent weldability. Among Zircaloy-2's disadvantages are high process costs, hydrogen pickup and embrittlement, and only fair fabricability. This section contains data which are useful for determining the design limitations of Zircaloy-2 for fuel element cladding.

302.1.1 Comments on Table of Properties

302.1.1.5 Mechanical Properties

Strength

For most reactor fuel element applications, the ultimate and yield strengths of Zircaloy-2 in tension are adequate. However, for the design analysis of unbonded claddings or supporting devices the strength of the cladding may be required. Therefore, the following review of the factors which influence the strength of Zircaloy-2 is presented:

The yield strength of base-annealed Zircaloy-2 ranges from 45,000 to 55,000 psi, and the tensile strength ranges from 70,000 to 80,000 psi. For estimating purposes, the average yield and tensile strength for sponge-base Zircaloy-2 may be taken as 44,000 and 69,000 psi, respectively.

Among the many variables which influence the strength of Zircaloy-2, the more important are temperature, cold work, hot work, oxygen content, anisotropy, heat treatment, and neutron damage. The degree to which these variables affect the strength of Zircaloy-2 is illustrated in Figure 1.^(5,6,7) This plot compares the relative magnitude of these variables within the limits expressed, and is useful only for broad comparisons among different conditions.

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Contrary to the normal behavior of metals, Zircaloy-2 has a profound memory for hot work.⁽⁷⁾ This can be seen by the increase in strength above the base-annealed condition caused by hot rolling. For this reason and because oxygen content affects strength, the melting and fabrication processes should be taken into account when designing for strength.

Finally, it is important to note that the effect of irradiation on the strength of Zircaloy-2 at relatively high exposures does not greatly exceed other effects plotted in Figure 1. About 90 per cent recovery of original strength has been reported for Zircaloy-2 irradiated in the annealed state when given a post irradiation anneal at temperatures comparable to those used for cold-work recovery.

Ductility

The power law, $\sigma = k\delta^n$, serves as a good approximation for the plastic behavior of Zircaloy-2.⁽⁸⁾ Referring to Figure 2,⁽⁹⁾ an increase in temperature or cold work decreases the uniform elongation as well as the work hardening exponent "n". The low values of per cent elongation in hot-rolled Zircaloy-2 at elevated temperature, particularly in the transverse direction, has been attributed to strain aging.⁽⁹⁾ This type of aging, causing embrittlement, is commonly related to gaseous constituents present in metals.

If strain aging is prevalent in Zircaloy-2, some design allowance must be made for a loss in ductility during reactor service. Factors which contribute to strain aging are (1) impurities that form interstitial solid solutions, (2) residual cold work, (3) neutron damage, and (4) elevated temperatures below the recovery range. Hydrogen in solution may be particularly detrimental in this respect.

* σ = true stress; δ = natural plastic strain; k and n are constants.

Beta-quenching* Zircaloy-2 reduces the uniform elongation,⁽⁸⁾ and during deformation this material tends to separate along prior beta grain boundaries. Similar separation caused by shrinkage stresses has been encountered upon welding. Mechanically, this material behaves as if it had a grain size larger than the apparent alpha grain size.

The effects of neutron radiation on the ductility of Zircaloy-2 and zirconium are similar and are illustrated in Figures 3⁽⁴⁾ and 4.⁽¹⁰⁾ In general, the uniform elongation for both materials is reduced with increased exposure; whereas, the necking strain remains about the same. At high exposures, the material not only begins to "neck down" immediately upon yielding, but the strain hardening exponent is also reduced resulting in a critical state of impending fracture. As shown in Figure 3, the total elongation of annealed Zircaloy-2 is reduced by 35 per cent upon irradiation (1.4×10^{21} nvt thermal). This compares to 50 per cent prior to irradiation. Some recovery of uniform elongation occurs at annealing temperatures in the range used for cold work recovery⁽⁴⁾ (575-750 F). This behavior would affect designs which magnify strain; e. g., fuel elements with bonded cladding but with unbonded end caps.

Impact Strength

The impact strength is a useful property for evaluating the causes of embrittlement, and the change in impact strength illustrates hydrogen embrittlement in Zircaloy-2.

Depending upon the state of cold work, the ductile to brittle transition temperature for Zircaloy-2 containing about 50 ppm hydrogen occurs at about 400 F (205 C).⁽⁶⁾ ** Increasing the hydrogen content to about 400 ppm shifts the transition temperature to nearly 650 F (345 C). This loss of ductility is attributed to the precipitation

* Heat treating in the beta phase followed by water quenching.

** Based upon a notch-bar (Charpy), impact strength of 30 ft-lbs.

of zirconium hydride which at the usual concentration of 40 to 50 ppm is taken into solution at about 750 F (400 C).⁽⁸⁾

In the beta-quenched condition the transition temperature (50 ppm H_2) is low; however, upon aging (750 F for 3 hours and furnace cooled) the transition temperature increases to between 300 and 400 F.⁽⁵⁾ Again, this embrittlement is accompanied with hydride formation as observed metallographically, and is independent of (a) tin content, (b) type of base zirconium used for the alloy, and (c) melting procedures.⁽¹⁵⁾

For normal reactor applications below 400 F, 50 ppm hydrogen concentration should not be exceeded during service, since hydride formation during slow cooling accompanying shutdown may be dangerous at higher concentrations.⁽¹⁾ It is reported that Zircaloy-2 is extremely sensitive to embrittlement with over 10 ppm hydrogen⁽¹⁶⁾ (which is an average equilibrium concentration value obtainable by vacuum annealing). For this reason service conditions leading to hydrogen pickup in Zircaloy-2 must be considered in any design where strain is likely to occur.

The effect of oxygen on the embrittlement of Zircaloy-2 seems to be in doubt. One report indicates that 1.0 atomic per cent oxygen will reduce the ductility of zirconium from thirty to less than ten per cent elongation.⁽¹⁾ However, it is also reported that the difference in oxygen content between crystal-bar and sponge-base Zircaloy-2 has no significant effect on the percent reduction in area or the transition temperature.⁽⁷⁾

Notch Sensitivity

Notch tensile tests reveal that the ductility of vacuum-annealed Zircaloy-2 is sufficiently high to provide for notch strengthening at sub-zero temperatures.⁽⁶⁾ On the other hand, Zircaloy-2 is particularly sensitive to notches in the rolling plane.⁽¹⁾ For this reason, surface defects on tubing or cans will markedly reduce rupture resistance to impact loading.

Creep

Extensive testing programs are presently being conducted to measure creep in Zircaloy-2. Creep failures are sensitive to many variables, not all of which have been evaluated. For this reason extreme case must be used in extrapolating the relationships listed in Table 30.1.1.12. It is recommended, when designing for creep, that current information be reviewed to find a similar application. A few references to reports of work in progress are listed in the references. (11, 12, 13, 14)

302.1.1.6 Corrosion and ChemicalGeneral

Chemically, Zircaloy-2 is a very reactive metal. Normally, its chemical reactions are severely inhibited by the inert zirconium oxide (ZrO_2) film which forms instantly on metal exposed to air, water or steam. The rate of any chemical reaction with Zircaloy-2 depends on the diffusion rates through the protective oxide film. Anything which is done to the metal to alter this film, such as machining, welding abrading, etching, heating in air or water, etc., will alter the corrosion performance of the Zircaloy-2 surface. The final surface treatment which results in the lowest corrosion rate in 680 F (360 C) water is the preparation of a smooth, clean surface by machining, abrading, or vapor blasting. This is followed by the removal of about 2 mils of metal in an etch bath at 120-130 F (50-55 C) made up by volume; 45% concentrated (70%) nitric acid, 5% concentrated (48%) hydrofluoric acid and 50% water.

Corrosion in Water and Steam

As Zircaloy-2 corrodes, the ZrO_2 film grows in thickness. On corrosion resistant material this film is blue-black in color. On material with poor corrosion resistance a white oxide forms which

is non-adherent and spalls off. The appearance of this white film signifies the end of useful life for the material. During the first 112 days in 680 F water Zircaloy-2 corrodes at a very low rate of about $0.01 \text{ mg/cm}^2/\text{day}$. After this initial period a slightly higher corrosion rate begins which is linear with time. Empirical corrosion equations for Zircaloy-2 are given in the data sheet. ⁽⁸⁾

The following factors seem to have little effect on corrosion performance of Zircaloy-2 in 680 F water: ⁽¹⁷⁾

1. Cold or hot working of the metal (provided final etched surface is prepared).
2. Most dissolved gases including oxygen and hydrogen but excluding nitrogen.
3. Dissolved impurities in the water in small concentrations including most ions except fluoride ion, F^- .
4. High water velocity up to 30 ft/sec.
5. Galvanic coupling with other metals.
6. Extensive thermal cycling.
7. Zircaloy-2 does not seem to be affected by stress corrosion.

The following factors seem to have an adverse effect on the corrosion performance of Zircaloy-2 in 680 F water: ⁽¹⁷⁾

1. Final preparation by machining, abrading or otherwise mechanically distorting the surface without a final etch treatment.
2. Traces of fluoride ion in the water as low as 200 ppm. Crevice corrosion becomes severe in the presence of fluoride ion.
3. Failure to completely rinse the surface of Zircaloy-2 in hot water after etching in HNO_3 , HF , H_2O bath. This will result in erratic corrosion performance.
4. Exposure of Zircaloy-2 in high concentrations of dissolved nitrogen may impair corrosion resistance.

5. Nitrogen pickup in the metal during welding or other high temperature exposure can destroy corrosion resistance.
6. Surface contamination such as fingerprints may result in local attack.
7. Surfaces impregnated with hydrogen show poor corrosion performance and hydrogen contents above 2000 ppm can impair corrosion resistance.
8. Corrosion rates at temperatures above 750 F become high.

Accelerated corrosion of a Zircaloy-2 fuel element jacket will result in a buildup of the ZrO_2 film with a subsequent loss of heat transfer properties of the jacket. This will result in higher jacket temperature in an operating fuel element which will in turn cause higher corrosion rates. The danger of jacket penetration by corrosion is not nearly as important a reason for requiring low corrosion rates as is this loss of heat transfer properties. ⁽⁸⁾

Gas Reactions

While most of the studies of gas reactions have been on zirconium, the reactions of Zircaloy-2 are similar.

At elevated temperatures zirconium reacts with most gases except the noble gases. With the exception of hydrogen, no appreciable attack occurs below 750 F within a period of a few hundred hours (on material with a normal oxide film). Above 750 F zirconium reacts with oxygen, nitrogen, air, carbon dioxide, carbon monoxide, steam, propane, ammonia and sulfur dioxide. In all cases the reaction rates increase rapidly with temperature. The reaction products are generally soluble in the metal and influence its physical properties. The use of zirconium or Zircaloy-2 is not recommended above 750 F when exposed in non-inert atmospheres for prolonged periods. ⁽¹⁸⁾

The reaction of Zircaloy-2 in dry air proceeds in two stages: initially the rate decreases with exposure time approximating a cubic relationship. After sufficient exposure the rate becomes a linear function of time. The rate constants have been calculated from experimental data and can be expressed by the Arrhenius equation $k = A \exp (-Q/RT)$ where:

A is a constant

Q is the activation energy

R is the gas constant

T is the absolute temperatures

Values for A and Q are given in Table II. (19)

The water corrosion performance of Zircaloy-2 is influenced by very small additions of nitrogen. Heat treatments above 750 F or welding will result in nitrogen pickup unless performed in a vacuum or inert atmosphere. If only surfaces are contaminated, this corrosion-prone material can be removed by etching.

Zircaloy-2 depends on its oxide film for protection against hydrogen. Without the protective film Zircaloy-2 reacts instantly with hydrogen from room temperature to at least 930 F. Zircaloy-2 is normally covered with an oxide film so the reaction is governed by the diffusion rate of hydrogen to the metal surface. Conditions where hydriding can occur are:

- (1) When the protective oxide film dissolves into the metal at 930 F or above and the surroundings are oxygen deficient, so the film is not reformed (i. e. insufficient air, oxygen, water, or steam present).
- (2) In completely dry hydrogen atmosphere where molecular hydrogen gas can diffuse through the ZrO_2 film by way of defects and cracks in the film. Any water or steam present seems to "heal" these defects and to prevent hydrogen pickup.

- (3) During corrosion where about 10-20 per cent of the atomic hydrogen produced by the corrosion reaction,

$$\text{Zr} + \text{H}_2\text{O} \longrightarrow \text{ZrO}_2 + 4\text{H}$$
 is picked up by the metal. Addition of hydrogen to the corroding water seems to accelerate hydrogen pickup somewhat.
- (4) In the presence of nickel where there is superficial evidence that catalytic hydrogen pickup can result. Nickel should be used with caution until this catalytic pickup is investigated.
- (5) In the presence of uranium in a Zircaloy-2 water system where the uranium (a) can provide hydrogen from a water-uranium reaction, (b) dehydrate this hydrogen, thus removing water that would inhibit the hydriding reaction, and (c) furnish heat to speed up the reactions. The literature of the hydriding of zirconium and Zircaloy-2 is discussed in detail in HW-55460. (20)

Corrosion of Zircaloy-2 by Sodium and NaK (Sodium-potassium)

Zirconium and Zircaloy-2 appear to be resistant to attack by oxygen free sodium or NaK below 930 F under static and dynamic conditions. (21) The corrosion resistance of zirconium and Zircaloy-2 is very sensitive to the oxide content of the sodium or NaK (see Table III). Other contaminants in the sodium or NaK may have an adverse effect.

TABLE III

Corrosion Rates of Zirconium in Liquid Sodium (21)

Static Tests
1 mo. at 500 C

Nominal Oxide
Conc. as Na_2O

Corrosion Rate
 $\text{mg/cm}^2/\text{mo.}$

0.01
0.05
0.10

+ 0.12
+ 0.47
+ 0.74

Rotating Specimen
(2" dia. 1725 RPM 14 i. p. s.)
1 mo. at 450 C

0.0
0.01
0.50

+ 0.10
+ 0.23
+ 0.94

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Corrosion of Zircaloy-2 in Organic Heat Exchange Media

Zircaloy-2 is essentially inert to biphenyl and isopropyl biphenyl under 800 F when the water content is less than 200 ppm. Two types of attack are found when Zircaloy-2 is exposed in organics: (1) oxidation by water contamination in organic fluids, and (2) hydriding from hydrogen added or generated by decomposition of the organic material. Zircaloy-2 is severely corroded at 800 F by biphenyl containing 400 ppm water. ⁽²²⁾ Hydrogen pickup occurs at low hydrogen partial pressures. However, by venting the system to maintain a very low hydrogen partial pressure (less than 1 psi at 575 F) hydriding can be retarded. Work is now in progress on the effects of organics on Zircaloy-2 and quantitative data are not yet available.

302.1.2 Design Limits

- 1) Application of Zircaloy-2 should be restricted to temperatures below 750 F because it reacts with most gases and the reaction products influence physical properties.
- 2) The hydrogen concentration should not exceed 50 ppm because of the marked reduction in ductility caused by hydrides.
- 3) In operation above 650 F, improvement of yield strength due to cold work may be lost because of recovery.

301.1.3 Needed Data

Among the important areas for which data are seriously needed for Zircaloy-2 are (1) hydride formation, (2) effects of neutron radiation, and (3) creep. Some specific programs within these areas of study are outlined below:

1. Hydride formation

- a. The rate of hydride pickup during aqueous corrosion.
- b. Reaction with dissolved hydrogen in water.
- c. Inhibition of hydrogen pickup by moist and wet environments and the rate of hydrogen pickup as a function of the dewpoint of air.

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- d. The degree and rate of hydride redistribution from uranium to zirconium in coextruded sections.
- 2. Effects of neutron irradiation
 - a. The instantaneous and cumulative effects of neutron radiation on the creep rates, thermal conductivity, and thermal expansion. (To be measured in-reactor by instrumented capsules.)
 - b. Recovery of cold work at water temperatures above 200 C and neutron fluxes above 1×10^{14} nv.
- 3. Creep
 - a. The effects of heat treatments and cold work on reducing the amount of first-stage creep.
 - b. The influence of fabrication and reactor atmospheres on notch sensitivity and rupture-life expectancy.
 - c. The nature and cause of second-stage failures.

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FIGURE 1

Effect of Production and Environment Variables on the Strength of Zircaloy-2

FIGURE 2

Change in Uniform Elongation with Temperature of Zircaloy-2

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FIGURE 3

Radiation Effect on Elongation of Zircaloy-2⁽⁴⁾

FIGURE 4

Radiation Effect on Elongation of Zirconium⁽¹⁰⁾

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500 MECHANICAL DESIGN - K. R. Merckx

Since a fuel element must exist in a reactor environment, the interactions of this environment with the fuel element must be considered. In the previous chapters, design limits were based on the state of the component in question, although the direct interaction of coolant with the cladding or fuel material was considered for corrosion limits. In this chapter, the mechanical effects of temperature variations, gravity forces, and coolant flow induced loadings are considered and appropriate mechanical theories are used to determine geometrical or operational limits. The limits of these theoretical methods of analysis and appropriate mechanical testing methods are also included.

501 Thermally Induced Rupture of the Fuel Material

The thermal energy created by the fission process within reactor fuel elements must be transferred to the reactor coolant. To transmit this heat to the coolant, temperature gradients must exist within the fuel material. A fuel material with temperature gradients undergoes thermal expansions which must be partially suppressed if the material is to remain solid or free of voids.

Discussed in this section are the mechanical failure problems associated with the incompatible thermal expansions caused by the operation of the fuel material in a neutron flux with spatial and time variations of temperature.

501.1 Thermal Stress Calculations

Since the strain incompatibilities caused by the variation of the temperature are limited to the maximum thermal expansion caused by the temperature gradients, the strains which can induce stresses are limited. Hence, stresses which result from the incompatible strains depend upon stress-strain relationships of the fuel materials. In the discussion which follows, the thermal

stresses caused by the thermal expansions in reactor fuel elements are determined with various material models. The accuracy of the material model in predicting the stress dependence on strains is increased in elastic, plastic, and time dependent models, and the effects of the different material models on calculated values of the thermal stresses and strains is emphasized.

501.1.1 Elastic Solutions

Assuming that the thermal expansions are large enough to cause mechanical failure or splitting of a metallic fuel element, realistic values of thermal stresses and strains cannot be calculated with elastic types of solutions where the stress is assumed to be proportional to the strain. Elastic calculations can be used to initiate investigations of mechanical behavior of fuel materials and the simplicity of such solutions⁽¹⁾ is very appealing. For example, the axial stress, σ_ϕ , at the surface of a hollow cylinder, assuming radial symmetry and plane strain conditions, is given by the equation:

$$\sigma_\phi = \sigma_z = \frac{\alpha E}{1 - \nu} (T_{\text{avg}} - T_{\text{surface}})$$

where

$$T_{\text{avg}} = \frac{2}{1 - u_i^2} \int_{u_i}^1 T(u) u du$$

α = linear coefficient of thermal expansion

E = elastic modulus

u_i = ratio of inner to outer radius

ν = Poisson's Ratio.

Hence, the solution for the stresses under transient thermal conditions or for various methods of cooling are reduced to the solution of the heat transfer problem. The more involved solutions

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which account for non-symmetric conditions⁽²⁾ or axial variations^(3,4) can be used to guide the intuition on the problems where more realistic solutions⁽⁵⁾ require prohibitively lengthy calculations.

For brittle fuel materials such as uranium oxide, elastic solutions predict failure of the oxides under reactor operating conditions and such failures do occur; but reactor designers rely upon the ductile metallic cladding material to contain the oxide fragments. Hence, the design of oxide fuel elements is not limited by failure of the fuel material. The mechanical properties of irradiated metallic fuel materials do indicate that the strain to fracture is decreased with increasing irradiation, but some ability to plastically deform or creep still exists during exposure to radiation, especially at high temperatures.

501.1.2 Plastic Solutions

Since elastic calculations for the thermal stresses in metallic reactor fuel elements predict stresses which could not realistically accompany the calculated values of the strains, some attempt to incorporate actual material behavior into thermal stress calculations must be made. The theory of plastic deformations⁽⁶⁾ allows for non-linear relations between the stresses and strains, but does not allow for time dependent effects of plastic deformations such as creep or stress relaxation.

Calculations⁽⁷⁾ based on a material law which approximated actual tensile test curves of unirradiated uranium have been made. These calculations showed that the thermal stress values predicted by elastic theory for uranium fuel elements operating with large temperature gradients were about three times larger than those predicted with the plastic material model. Comparative analyses⁽⁸⁾ of solid, externally-cooled hollow cylinders (cored) and internally-externally cooled hollow cylinders (I and E) producing equal power

predicted small percentage reductions in stresses for the cored and a slightly larger reduction for the I and E fuel elements while a large reduction in the amount of plastic straining occurred.

In another method of solution,⁽⁹⁾ it was assumed that the material was a perfect plastic-elastic material with the yield strength varying as a function of the radius. This method was applied to cored fuel elements to approximate conditions after periods of constant temperature operation. From calculations made with the theory, it was predicted that very large central strains would develop if the holes in cored fuel elements were not large enough.

Because plastic solutions are non-linear and extremely temperature dependent, simple proportionality relations between stresses and strains in fuel elements under various operating conditions are non-existent. For any fuel element operating at high temperatures (<450 C for uranium) the relaxation or creep effects cannot be neglected. Because the stress levels within metallic reactor fuel elements (especially massive elements with temperature drops of 200 C or more) are well into the plastic range, creep effects will cause reductions of 10 to 40 per cent of the stress values calculated by models evaluated with simple tensile tests. Since the dependence is not included in the above plastic deformation model, estimates of the effects of rapid startups or reactor scrams cannot be predicted.

501.1.3 Time Dependent Solutions

If thermal stress calculations are to be used to determine limitations of reactor fuel element service conditions, the stress calculations for high temperature fuel elements have to include the time dependent effects of plastic deformation. An extensive⁽¹⁰⁾ set of thermal stress calculations have been made for cylindrical shapes using various temperature dependent visco-elastic models. (Visco-elastic models assume linear relations between stresses, stress rates,

strains and strain rates.) These solutions are not directly applicable for actual fuel element problems because metallic fuel materials have creep rates which are exponentially dependent upon stress. (11)

A general model of material behavior can be used with the equations of plastic flow. (12) These equations relate the strain rates to the stress rates, (the elastic portion) plus the product of the strain hardening coefficient (function of stress, S , strain, E , and temperature, T) and the stress (the plastic portion of the strain). This material law has the form:

$$\dot{e}_i - \dot{e}_T = \dot{s}_i + f(S, E, T)s_i,$$

where

$$\dot{e}_i - \dot{e}_T = \text{strain deviation rate}$$

$$\dot{s}_i = \text{stress deviation rate}/2G,$$

$$f = \text{strain hardening coefficient.}$$

The above law of material behavior is dependent on how the loads and temperatures are applied; hence, any solution of an actual problem is incremental. The above form of a strain hardening coefficient has been evaluated (13) with creep data from unirradiated uranium. (14) Calculations (15) using this strain-hardening coefficient were made to compare the effects of high and low temperature operation upon thermal stress values. The thermal stresses calculated by elastic, plastic deformation, (7) and plastic flow theory (15) for solid fuel elements which reached their maximum temperature in twenty-five minutes are given in the following table:

THE DEPENDENCE OF THE EQUIVALENT TENSILE STRESS UPON
ASSUMED MATERIAL MODEL AND TEMPERATURE OF OPERATION

	<u>Case I</u>	<u>Case II</u>
Surface Temperature (C)	100	350
Central Temperature (t = 25 min)	350	600
Elastic Solution		
Surface Stress (psi)	90,000	30,200
Central Stress (psi)	45,100	15,100
Plastic Déformation		
Surface Stress (psi)	39,400	22,600
Central Stress (psi)	19,700	11,300
Plastic Flow Theory		
Surface Stress (t = 25 min)	36,400	15,600
Central Stress (t = 25 min)	18,400	920
Surface Stress (t = 722 min)	31,200	11,700
Central Stress (t = 722 min)	14,300	0

The plastic strain per cycle has been suggested⁽¹⁶⁾ as a measure of the relative likelihood of failure under cycling conditions. Maximum calculated values of the plastic strain for the above cases at time t = 722 min are a surface plastic strain of 0.48 per cent for the 100-350 C case and a central-plastic strain of 0.34 per cent for the 350-600 C case. The position of maximum plastic strain changed from the surface to center of the fuel material as the temperature increased.

The amount of plastic strain occurring in a fuel element is dependent upon the geometry and the method of cooling (the shape of the temperature distribution curve) as well as the mean temperature and total temperature drop. In order to show this dependence on the method of cooling, the thermal stresses and strains for solid, cored, I & E, and internally cooled fuel elements were calculated⁽³⁹⁾

using the material model⁽¹³⁾ described above. In all the cases considered in this set of calculations the surface near the coolant remained at 200 C and the maximum temperature increased in one-half hour to 600 C and then remained constant. Figures 1 and 2 show how the equivalent plastic strain increases at the cooled surfaces and at the hottest portion of the fuel elements. For these examples, only the cored case had its maximum plastic strain at the hottest surface. Calculations were also made for the various methods of cooling cylindrical fuel elements with equal power generation. The results of these calculations are tabulated below:

THERMAL STRESS AND STRAIN VALUES CALCULATED
FOR CYLINDRICAL FUEL ELEMENTS
 (Fuel Surface Near Coolant at 200 C)

Fuel Element Type	Max. Temp. (C)	Max. Heat Generation (kw/ft)	Max. Equivalent Stress (psi)	Max. Equivalent Plastic Strain (%)
Solid Element	600	50	34,500	0.563 (u = 1.0) *
Cored Element (u _i = 0.4) **	600	76.8	33,800	1.35 (u = 0.4)
	460	50	31,500	0.450 (u = 0.4)
Internally-Externally Cooled	600	228	36,800	0.734 (u = 0.4)
Element (u _i = 0.4)	290	50	22,300	0.138 (u = 0.4)
Internally Cooled Element (u _i = 0.4)	600	42.8	36,700	0.785 (u = 0.4)

* u = ratio radial distance to outer radius

** u_i = Ratio core radius to outer radius.

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FIGURE 1

Plastic Strain at the Interior (600 C Max.) of Fuel Elements Cooled by Various Methods

(Initial and Cooling surface temperature = 200 C)

(Maximum temperature = 600 C obtained at $t = 0.5$ hr)

FIGURE 2

Plastic Strain at the Cooled Surface (200 C) of Fuel Elements Cooled by Various Methods

(Maximum Temperature = 600 C obtained at $t = 0.5$ hr)