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IRRADIATION BEHAVIOUR OF SOLID AND HOLLOW U_3Si FUEL ELEMENTS: RESULTS TO 15,000 MWd/tonne U

Exp-NRX-52233

by

M.A. FERADAY, G.H. CHALDER and K.D. COTNAM

Chalk River, Ontario

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ABSTRACT

U_3Si fuel elements clad in zirconium alloy sheaths have been irradiated to burnups close to 15,000 MWd/tonne U in pressurized water at 220°C, 98 bars. The results show that the external swelling can be controlled by incorporating free volume in the element. The dimensional stability of such elements is adequate to permit their use in power reactor fuel bundles.

A diameter increase of 1.2% had occurred in an element initially containing 12.8% total free volume, after a burnup of 14,700 MWd/tonne U. There was no change in diameter between burnups of 5200 and 14,700 MWd/tonne U. Elements containing 3% total free volume had increased in diameter about 2.5% at 2000 MWd/tonne U compared to 0.2% at 9500 MWd/tonne U for elements containing 22% total free volume.

The observed swelling in the U_3Si is discussed in terms of possible mechanisms.

Chalk River Nuclear Laboratories
Fuel Materials Branch
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Comportement sous irradiation d'éléments combustibles en U_3Si, massifs et creux: résultats jusqu'à 15 000 MWd/tonne U

par M.A. Feraday, G.H. Chalder et K.D. Cotnam

Résumé

Des éléments combustibles en U_3Si enfermés dans des gaines en alliage de zirconium ont été irradiés jusqu'à des taux de combustion d'environ 15 000 MWd/tonne U dans de l'eau pressurisée à 220°C, 98 bars. Les résultats montrent que le gonflement extérieur peut être contrôlé en incorporant un volume libre dans l'élément. La stabilité dimensionnelle de ces éléments permet leur utilisation dans les faisceaux de combustible destinés aux réacteurs de centrale.

Une augmentation de diamètre de 1.2% s'est produite dans un élément contenant initialement un total de 12.8% de volume libre, après un taux de combustion de 14 700 MWd/tonne U. Le diamètre n'a pas changé entre les taux de combustion de 5 200 et 14 700 MWd/tonne U. Des éléments contenant un total de 3% de volume libre ont eu des augmentations de diamètre d'environ 2.5% à 2 000 MWd/tonne U comparé à 0.2% à 9 500 MWd/tonne U pour des éléments contenant un total de 22% de volume libre.

Le gonflement observé dans l'U_3Si est étudié en vue de déterminer les mécanismes qui le provoquent.

L'Energie Atomique du Canada, Limitée

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IRRADIATION BEHAVIOUR OF SOLID AND HOLLOW U_3Si FUEL ELEMENTS: RESULTS TO 15,000 MWd/tonne U

1. INTRODUCTION

In 1965, a series of irradiation tests was started to determine if the swelling previously observed[1,2,3] at low burnup in U_3Si could be controlled by suitable fuel element design. The objective was to provide economic burnup (~10,000 MWd/tonne U) with acceptable external dimensional changes (<1% on diameter and length) for use in CANDU-type power reactors. Experimental fuel elements, consisting of cylindrical rods of U_3Si clad in Zircaloy-2, were irradiated in a pressurized water loop in the NRX reactor.

Results already reported[4] from this series of tests showed that:

1) provision of an internal void along the axis of the U_3Si was effective in limiting increases in the external diameter of the fuel elements compared to earlier tests[1] in which the fuel had no void (Figure 1),

2) the rate of swelling of the U_3Si itself appeared to decrease with increasing burnup (Figure 2); extrapolation of the curve for 15 mm diameter elements clad in 0.7 mm thick Zircaloy sheaths indicated a fuel volume increase of 13% after a burnup of 10,000 MWd/tonne U,

3) the rate of swelling observed in U_3Si at low burnup was not consistent with the growth of fission gas bubbles alone; structural disordering, cavitation and the accumulation of solid fission products were suggested as other possible mechanisms,

4) fuel power output did not appear to have an effect on the swelling of U_3Si, at least in the range studied, and

5) a definite effect of sheath thickness on swelling was not established because the elements clad in thinner (0.45 mm) Zircaloy sheaths had not attained sufficient burnup for an accurate comparison to be made with those clad in 0.7 mm sheaths.

When these same results were published[5], the values of power output and burnup, previously reported from loop calorimetry, were revised downwards to correspond to more accurate measurements from chemical analysis, which had become available (see Section 5.1 for details).

Results are now available for those original elements after higher burnup, and for additional elements which have been included in the experiment more recently. Major variables in all elements are listed in Table 1, and results reported here include:

i) The effects of higher burnup on the original 15 mm diameter elements containing 9.6 to 12.8% total free volume and clad in 0.45 mm and 0.7 mm thick Zircaloy sheaths. (Types 1 and 2 in Table 1)

ii) The effect of different levels of free volume, approximately 3, 11 and 22%, on the swelling of 15 mm diameter elements clad in 0.7 mm thick zirconium-2.5 wt% niobium. (Types 3, 4 and 5, Table 1)

iii) The effect of sheath strength on swelling in elements containing approximately 15% free volume. Elements 15 mm in diameter were clad in 0.46 and 0.66 mm thick Zircaloy and duplicate elements of each type had an induction annealed zone in the cold worked sheath. (Types 7 and 8, Table 1)

iv) The effect on swelling of minor alloying additions to U_3Si. Alloys were irradiated containing 4 wt% silicon, with and without the addition of $\sim$350 ppm iron and 1500 ppm aluminum. (Types 2 and 9, Table 1)

2. FABRICATION OF FUEL ELEMENTS

Fuel rods were prepared by vacuum induction melting and casting into cored molds[6]. The cast rods were decored, then heat treated for 72 hours at 800°C in vacuum to transform the as-cast structure of uranium and U_3Si_2 to one of U_3Si containing some residual U_3Si_2. The heat-treated rods were centreless ground to the required diameter and machined to length. Fuel elements were assembled by slip fitting U_3Si rods into Zircaloy or Zr-2.5 wt% Nb sheaths which were then sealed by resistance welding end caps in place.

In elements AWY, AWZ, AYN, and AYP, a section of the Zircaloy sheath was beta-annealed by induction heating in vacuum prior to element assembly. The fuel element data are summarized in Table 2 and detailed chemical analyses of the fuel in Table 3.

3. IRRADIATION OF ELEMENTS

The fuel was irradiated in the X-5 pressurized water loop of the NRX reactor. The loop conditions were approximately as follows:

loop pressure - 98 bars

inlet temperature - 220°C

coolant flow (H_2O)- 0.5 ℓ/sec.

Table 4 summarizes the irradiation history of each of the elements during the period September 1965 to July 1968.

4. POST-IRRADIATION EXAMINATION

The techniques used in underwater and in-cell examinations have already been described[4]. Underwater examination in the NRX reactor bay consisted of a visual inspection followed by measurement of fuel element volume by water displacement. In-cell examination included more detailed visual examination of elements followed by length, diameter and bow measurements. Elements MJC, MJL and AWE were sectioned for metallography and for density measurements on the U_3Si fuel. Beta/gamma autoradiographs were also taken on the cut sections.

5. RESULTS

5.1 Power Outputs

The power outputs and burnups shown in Table 4 are lower than those reported previously(4) for these elements, which had been derived from loop calorimetry. The revised values are based on more recent chemical burnup analyses of two specimens irradiated to ~6000 MWd/tonne U and have been proportioned according to the known flux distribution in the loop.

The power outputs and burnups are both now quoted in terms of 185 MeV/fission, the heat released by the fuel to the coolant, whereas previously(4,5) they were quoted in terms of 199 MeV/fission - the total heat released by the fuel. The 185 MeV/fission values more closely represent the usable power outputs of fuel elements in a power reactor. The chemical burnup measurements were taken on samples from the mid-plane of the elements.

In general, the specimens experienced maximum power outputs early in their irradiation. Table 4 lists the range of power outputs and the time average power output of each element.

5.2 Temperature Distribution in the Fuel

In calculating temperature distribution in the fuel (Table 4) we used:

i) a constant fuel/sheath heat transfer coefficient of 5 W/cm^2°C which is considerably higher than the 1.2 W/cm^2°C used previously(4,5). Post-irradiation metallography has indicated that the U_3Si swells into intimate contact with the Zircaloy cladding fairly early in an irradiation (<1000 MWd/tonne U) and even before the central void is closed up. Based on this information and on other out-reactor work(7), we have estimated that the interface coefficient for U_3Si/Zircaloy would range from 0.8 W/cm^2°C for new fuel to 6 W/cm^2°C for highly irradiated fuel,

ii) an average thermal conductivity of 0.2 W/cm°C, based on a value of 0.15 W/cm°C measured on unirradiated U_3Si at 30°C(8) and assuming that an increase in conductivity with temperature occurs in U_3Si similar to that reported for uranium(9),

iii) the maximum power output of the fuel (Table 4) adjusted to 182 MeV/fission, the heat generated <u>in</u> the fuel.

The maximum fuel temperature determined in this way was 520°C.

5.3 Dimensional Changes

Diameter profiles of each element were measured in-cell at various stages during the irradiation. Average diameter increases were also calculated from more frequent underwater volume measurements by assuming that the volume change represented a uniform diameter increase of the element and that no length change had occurred. Measured length changes were small in all elements after irradiation (Table 4). The average diameter increase has been plotted against burnup for all elements in Figure 3, but for clarity, curves have been drawn only for elements of Types 1 and 2 (Table 1). In Figure 4, these two curves have been reproduced (without data points) along with the curves for each of the other element types.

5.4 Examination of the Fuel

Measurements of void filling, by destructive examination of low burnup elements, were confined to three elements of type 1[4] and showed that a 7 vol% void was entirely filled at a burnup of about 1500 MWd/tonne U (corrected value), i.e. at about the stage at which significant diameter increases began to occur in that type of element. The values of U_3Si volume increase in Figures 5 and 6 were plotted on the assumption that the total free volume in each type of element had been taken up by the time external diameter increases were measured. This method of calculating the volume change in U_3Si (ΔV_1) is explained in more detail in Appendix A.

The volume change in the U_3Si was also determined from density measurements on samples of the irradiated fuel (ΔV_2). This method also is detailed in Appendix A, and the results are given in Table 5 and Figure 6 (data points). As in an earlier test[10], the values obtained were in general higher than those obtained by dimensional measurements on complete elements (ΔV_1).

The values of U_3Si volume increase versus burnup plotted in Figures 5 and 6 bear the same relationship to one another as do those for diameter increase versus burnup shown in Figures 3 and 4, viz:

- Figure 5 shows all points but curves for elements of types 1 and 2 only,

- Figure 6 shows curves for all element types and the data points obtained from direct density measurements (see above).

Elements MJL, MJC and AWE were destructively examined after higher burnup (5490, 8435 and 9455 MWd/tonne U respectively) and examined as-sectioned, and after polishing and etching. Photographs of representative sections from each element and from unirradiated controls are shown in Figures 7-18.

The original axial void was found to be filled in all three elements (Figures 7-11). Micro-examination of the fuel in general confirmed the results obtained earlier on similar specimens irradiated to lower burnup[4]. Although the distribution of the U_3Si and U_3Si_2 phases appeared to be unchanged by irradiation (Figures 14 and 15), it was not possible to delineate grain boundaries in irradiated U_3Si, using the etchant which is effective on unirradiated material*. Fine pores or inclusions in the irradiated U_3Si phase appeared more numerous than before irradiation. The fuel and sheath were in intimate contact in all three irradiated elements but there was no evidence of metallurgical reaction (Figure 18).

Macro-examination of etched cross-sections (Figures 9-11) revealed dark bands at the periphery and central regions of the fuel which were not visible on polished sections. Beta/gamma autoradiographs of the same cross-sections showed a uniform darkening over the whole of the fuel cross-section, indicating that the dark bands are a structural effect rather than a concentration of fission products. No consistent difference in microstructure could be found to correspond to these dark bands although similar areas have been observed on both fractured and etched sections of fuel in another test[10].

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* 3.4 grams Citric Acid, 72 cm^3 Nitric Acid, 170 cm^3 Water and 1 cm^3 48% Hydrofluoric Acid.

6. DISCUSSION

The principal objective of the experiment was to determine whether external swelling of U_3Si fuel elements could be controlled adequately at terminal burnup to permit application of this material in power reactor fuel bundles. The limits tentatively set for this application were a diameter increase of less than 1% at a burnup of 10,000 MWd/tonne U.

A secondary objective was to determine the mechanism of the irradiation-induced swelling observed in U_3Si; methods might then be developed for reducing this swelling and thus the amount of free volume required to accommodate it, with consequent increase in effective uranium density in the fuel element. The results of the experiment are discussed below with respect to these objectives.

6.1 Dimensional Stability

We have considered the observed swelling behaviour of the different fuel element types tested with respect to the effects of:

- free volume in the element
- sheath restraint
- power output
- fuel composition.

Because of uncontrolled and coincident variations in these parameters in the specimens used in this test, it is not possible to establish with certainty the effects of each alone.

6.1.1 Free volume

In previous work[4] we found that total free volume i.e. including porosity in the fuel and assembly clearances, rather than just the axial void, appeared to be the important parameter in defining external swelling of the elements. The total free volume in each type of element was generally 3-5% greater than the nominal axial void volume (see Table 1).

One element of the original design used in this experiment (MJD, of type 2 and containing 12.8% total free volume) attained an exposure of 14,700 MWd/tonne U with a diameter increase of 1.2%. No net increase in diameter occurred since this element was reported on earlier[5] at an exposure of 5200 MWd/tonne U. Five other elements of this type attained exposures ranging from 5500 to 8500 MWd/tonne U and, after correction for small differences in initial free volume, their behaviour confirms that of MJD. Since it is known that the axial void in these elements is filled at an exposure below 5500 MWd/tonne U (Figures 7 and 9) it appears that there is no significant volume change in U_3Si, irradiated under these conditions, between burnups of 5200 and 14,700 MWd/tonne U.

Results in Figure 4 show that for elements of other types, having widely different amounts of free volume, the increase in diameter varies at a given burnup. For elements of type 7 containing a total free volume of 14-15%, a diameter increase of 0.7% was observed after a burnup of 6700 MWd/tonne, while for elements of type 5, containing 22% free volume, the diameter increase was 0.2% after 9400 MWd/tonne U. Based on the behaviour of element MJD we do not expect further significant diameter increases in either of these element types to a burnup of at least 14,700 MWd/tonne U. Thus, although the diameter increase can be reduced further by using greater free volume, it appears that a total of 15 vol% will be sufficient to maintain diameter increases below 1% in a burnup of at least 10,000 MWd/tonne U, under the conditions of this test.

The derived curves of U_3Si volume increase versus burnup (Figures 5 and 6) in general provide better agreement, between individual elements of each type, than that obtained with direct diameter measurements. This confirms earlier observations[4] and is attributed to small differences in porosity in the fuel which are allowed for in deriving fuel volume changes.

Figure 6 suggests there is a greater volume increase in the U_3Si at a given burnup when the initial free volume in the element is greater. One element of type 5 (AWE) was sectioned after a burnup of 9400 MWd/tonne U and the axial void found to be filled, confirming the assumptions made in deriving the fuel volume change.

6.1.2 Sheath restraint

The restraint in our specimens is the sum of

- self-restraint in the fuel,
- sheath restraint, and
- coolant pressure.

To a first approximation, fuel self-restraint and coolant pressure would be the same for all specimens in the present test; we have observed differences in external swelling of the specimens which are attributed to differences in sheath restraint. The sheathing variables studied were:

- sheath material; Zircaloy vs Zr-2.5 wt% Nb
- sheath thickness; in the range 0.45-0.7 mm
- sheath condition; cold worked vs β-annealed.

Comparison of the effect of sheath material, under otherwise similar conditions is possible between elements of types 3 and 6 (containing 3.0-3.8% free volume) and types 2 and 4 (containing 10.0-12.8% free volume). Elements clad in Zr-2.5 wt% Nb (type 3) swelled less than those clad in Zircaloy (type 6), both on diameter (Figure 4) and in fuel volume (Figure 6), when the available free volume in the element was low. However, because of the large strain developed in the sheaths of these elements, their irradiation was terminated at low burnup and the comparison is based on only two measurements. At higher levels of free volume, the diameter increase (Figure 4) in zirconium-niobium clad elements (type 4) was greater than those clad in Zircaloy (type 2) but after correction for initial free volume, fuel swelling was not significantly different (Figure 6) at corresponding burnup.

The effect of different thicknesses of Zircaloy sheathing is compared between elements of types 1 and 2 (9.6-12.8% free volume) and types 7 and 8 (14.1-17.7% free volume). There is about 2% greater diameter increase at corresponding burnup in type 1 elements compared to type 2 which persists, after correction for differences in initial free volume, in a 2% greater increase in fuel volume (Figure 6). For higher values of initial free volume (types 7 and 8) the effect of the sheath thickness is less; although the diameter increase is slightly greater for the thinner sheath (Figure 4) the apparent fuel volume increase is lower (Figure 6). There was an inadvertent difference in silicon concentration between fuel in type 7 (4.0 wt% Si) and type 8 (4.3 wt% Si) elements. In another test[10] fuel containing 4.3 wt% silicon was found to have increased in diameter before void filling was complete. Similar behaviour in fuel of type 8 elements of the present test could account for the apparently higher calculated fuel volume increases, compared with type 7 material.

The effect of sheath condition was examined within individual specimens by induction heating a 4 cm long section of the cold-worked Zircaloy sheath into the β range prior to element assembly. Elements having this feature were included in types 7 and 8 together with other elements having entirely cold-worked sheaths. As shown in Figure 19A, for elements of type 8 after a burnup of 7965 MWd/tonne U, no difference in diameter was detectable between sections of 0.7 mm thick sheath in the two conditions, and the diameter increase was comparable with that in otherwise similar elements having a completely cold-worked sheath. At a lower (0.46 mm) sheath thickness however, elements of type 7 exhibited enhanced swelling in the β annealed zones after a burnup of 6740 MWd/tonne U (Figure 19B) compared to otherwise similar elements having a completely cold-worked sheath (Figure 19C).

6.1.3 Power output

When individual elements underwent a step change in power output a reversible change was noted in diameter. This effect is most clearly illustrated by elements of types 1 and 2 (Figure 3) between burnups of 3000 and 7000 MWd/tonne U. A decrease in linear power output of element MJA from 500 W/cm to 350 W/cm near 4000 MWd/tonne U resulted in a diameter decrease of about 0.2%; similar effects can be noted for elements MJH, MJP and MJS.

6.1.4 Fuel composition

Fuel containing 350 ppm iron and 1500 ppm aluminum (type 9) exhibited similar diameter changes (Figure 4) to material of low iron and aluminum concentration (type 2) under otherwise similar conditions. After correction for differences in initial free volume however, the volume increase (Figure 6) in the material containing higher iron and aluminum concentrations appears to be significantly less; about 2 vol% at burnups between 4000 and 10,000 MWd/tonne U.

The higher calculated volume increase in fuel containing 4.3 wt% Si (see section 6.1.2), compared with material containing 4.0 wt% Si, has already been attributed[10] to a higher flow stress in the high silicon material. Attention should be given in future tests to silicon concentration of the fuel, since 4.0 wt% Si may not be the optimum.

6.2 Swelling Mechanism

From an analysis of earlier results from this experiment we concluded[4,5] that the growth of fission gas bubbles alone could not account for the rapid swelling observed in U_3Si at low burnup. X-ray diffraction studies by MacEwan and Bethune[11] have shown that U_3Si, irradiated to low exposures at a temperature of 60°C, transformed from a body centred tetragonal (b.c.t.) to a disordered face centred cubic structure (f.c.c.). Displacement density measurements revealed an accompanying volume increase of about 3%. Complete recovery of the b.c.t. structure was obtained by annealing after irradiation, at 250-500°C. Samples of irradiated fuel from elements MJB, MJE and MJL of the present test were examined by X-ray diffraction and no change from the pre-irradiated (b.c.t.) structure was detected[12]. Thus we conclude that any irradiation-induced structural changes of the type observed by MacEwan and Bethune were simultaneously recovered at the higher irradiation temperature (300-520°C) in our work.

The b.c.t. to f.c.c. transformation, and the accompanying 3% volume increase cannot directly explain the 13-22 vol% swelling observed in our specimens. MacEwan and Bethune[11] have however suggested a mechanism whereby large volume increases might be caused by the structural transformation b.c.t. → f.c.c. → b.c.t. Within a fission spike, material would be transformed to the f.c.c. structure with a volume increase of about 3% which could be accommodated locally by plastic deformation of the surrounding U_3Si. When the material reverted to the b.c.t. structure, a void might be formed as a consequence of the associated volume decrease. Voids formed in this way could give rise to bulk volume changes in the fuel several times that associated with a single transformation.

In addition to the structural changes found by MacEwan and Bethune, there are at least three other possible mechanisms by which voids could be formed in U_3Si. The peritectoid reaction, by which U_3Si transforms to uranium and U_3Si_2 above 930°C, involves a volume increase of ~4%. If, within the area affected by a fission spike, this reaction went in the forward direction, and if the reverse reaction to reform U_3Si followed, then similar voids to those proposed by MacEwan and Bethune could be produced. Alternatively, the slight anisotropy of the tetragonal lattice of U_3Si might give rise to cavitation tears of the type observed[13] in uranium metal irradiated at 400-500°C. As a further alternative, microtears might be developed in U_3Si of the type observed in uranium metal irradiated between 500 and 600°C, which have been attributed[14] to agglomeration of lattice defects formed by fission events.

Observations of the U_3Si used in this experiment are qualitatively in agreement with a swelling mechanism which involves the development of micro-voids in the fuel material. Optical micrographs (for example, Figure 17) of irradiated U_3Si exhibit small dark spots, near the limit of resolution, which might be pores of diameter <1 μm. More detailed optical[15] and electron microscopy[16] of samples of fuel from this test, are in progress and will be reported at a later date.

We can develop a qualitative hypothesis of the swelling behaviour of fuel elements containing U_3Si based on the observations from this test and assuming that the principal swelling mechanism is one involving void formation. During the early part of the irradiation, swelling of the fuel can be accommodated by flow of material into any voids present in the element under the influence of a combination of fuel self-restraint and external restraint. Provided these restraints are sufficiently high to overcome the flow stress of the fuel material, external dimensional changes to the element will be limited to those produced by creep under the net stress developed in the sheath. Although new voids are continually being produced as a consequence of fresh fission events, sintering forces will be acting to eliminate those already present, and will be augmented by the external restraint of the system, particularly after large voids initially present in the fuel are filled. The total volume increase in the fuel will represent an equilibrium between the instantaneous void volume produced in the fuel by the fission process, and the rate at which voids, formed by previous events, are eliminated.

The implications of such a mechanism, which are in general confirmed by our observations, are

1) the volume increase of the U_3Si at a given burnup is not a fixed quantity but depends on fission rate, restraint, etc.,

2) the volume increase of the U_3Si at a given burnup will be greater at higher fission rates under otherwise similar conditions,

3) the volume increase of the U_3Si will be greater at a given burnup in elements which initially contained greater free volume,

4) the initial free volume necessary to maintain dimensional stability of the element to high burnup is less, under otherwise similar conditions, when the external restraint is greater,

5) conversely, the restraint required to maintain dimensional stability of the element to high burnup is less, under otherwise similar conditions, when the initial free volume in the element is greater, and

6) when an equilibrium has been established between void formation and elimination, no further volume increase in the U_3Si with burnup will occur, except as a consequence of other mechanisms such as the buildup of solid fission products.

At the fission rate and coolant conditions of the present test, dimensional stability (i.e. 1% increase in diameter after 10,000 MWd/tonne U) is obtained for a fuel element clad in 0.7 mm thick Zircaloy by the use of about 13% initial free volume. For a fuel element clad in 0.45 mm thick Zircaloy, about 15% initial free volume appears necessary.

Other limiting combinations of sheath restraint and free volume obviously exist. Insufficient information is yet available to develop a quantitative relationship between them or to derive corresponding relationships for other fission rates and coolant conditions.

7. SUMMARY AND CONCLUSIONS

1. External swelling of fuel elements containing U_3Si can be controlled by the provision of free volume within the element. Under the conditions of this test, an element containing 12.8% initial free volume (type 2) increased 1.2% in diameter after a burnup of 14,700 MWd/tonne U. The diameter increase at a given burnup can be reduced by provision of greater initial free volume.

2. The volume increase in U_3Si under irradiation is greater, under otherwise similar conditions, when free volume within the fuel element is greater. After a burnup of 10,000 MWd/tonne U in this test, the U_3Si in an element containing 12.8% initial free volume had swelled 16.2%, whereas U_3Si in an element containing 22% initial free volume, had swelled 22.6%.

3. The rate of volume increase of U_3Si on irradiation decreases at higher burnup. No change in fuel volume was observed in an element initially containing 12.8% free volume between burnups of 5200 and 14,700 MWd/tonne U.

4. External diameter increases in U_3Si fuel elements are greater, under otherwise similar conditions, when the sheath restraint is lower. This effect is more apparent for elements containing less initial free volume.

5. External diameter increases are greater, for otherwise similar elements, operated at higher power.

6. Small additions of iron and aluminum appear to reduce the swelling of U_3Si.

7. The observed behaviour of U_3Si in this test is consistent with a swelling mechanism involving the formation of micro-voids resulting from fission events which eventually reach an equilibrium concentration depending on fission rate and external restraint.

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APPENDIX A

A COMPARISON OF TWO METHODS FOR CALCULATING THE VOLUME CHANGE IN U_3Si DURING IRRADIATION

A) Method I

General

If we assume that the theoretical density (ρ_{TD}) of U_3Si is a function of silicon concentration*, the irradiation induced volume change in U_3Si can be calculated from the following measurable parameters. For this method and for method II (below), the volume change is expressed as a percentage of the volume of theoretically dense U_3Si rather than as a percentage of the volume of porous fuel in the rod as was done previously[4,5].

Parameters

V_1 = vol. of theoretically dense (T.D.) U_3Si in a fuel slug = $\frac{\text{weight}}{\text{theoretical density}}$ (cm^3)

V_2 = external slug volume calculated from the outside diameter and length of a U_3Si slug before irradiation (cm^3).

V_3 = apparent volume of U_3Si slug by immersion in a suitable liquid (cm^3).

V_4 = axial void volume in the new fuel = $V_2 - V_3$ (cm^3).

V_5 = volume of porosity and microcracks in the new fuel = $V_3 - V_1$ (cm^3).

V_6 = cold clearances in a new element (cm^3).

V_7 = hot clearances in a new element (cm^3).

---

* Decreasing linearly from 15.51 g/cm^3 for material containing 3.8 wt% Si to 15.11 g/cm^3 for material of 4.2 wt% Si.

V_8 = the external volume change which occurs in an element during irradiation (cm^3).

V_9 = volume of cracks in the U_3Si fuel element after irradiation (cm^3) estimated for 3 dimensions from the fuel cross-sections.

<u>Calculation of Fuel Volume Changes</u>

1) Metallographic examination of fuel sections after irradiation shows that the fuel and sheath are bonded, the fuel is cracked and none of the original diametral clearances are visible. In calculating the fuel volume change, it is possible to make either of two assumptions:

   a) assume that the fuel/sheath bond causes the sheath to collapse as the fuel cools after irradiation. In this case it is necessary to use V_6 and V_9 in the calculation or

   b) assume that all the clearance taken up by differential thermal expansion of fuel and sheath reappears as cracks in the fuel after irradiation. In this case, V_7 is used instead of V_6 and V_9.

2) Assuming that 1(a) above is correct, then:

   a) the total volume inside a fuel can before irradiation is given by $= V_1 + (V_4 + V_5 + V_6)$

   b) the total swelling which occurs in V_1 (cm^3) of U_3Si fuel is

$$\Delta V_a = V_1 + xV_4 + xV_5 + xV_6 + V_8 - V_1 - V_9$$

$$\Delta V_a = xV_4 + xV_5 + xV_6 + V_8 - V_9$$

   where x = fraction of central void closed up, determined by metallography after irradiation in several cases[4] and/or by assuming that no significant diameter increases occurred until after the central void had filled in. It was also assumed that the porosity and clearance filled in the same fraction as the central void.

c) in elements MJC and MJL where the voids were closed up, $x = 1$ and from measurements:

	MJC	MJL
V_4 (central void)	1.44	1.44 (cm^3)
V_5 (porosity etc.)	0.19	0.30 (cm^3)
V_6 (cold clearance)	0.34	0.33 (cm^3)
V_8 (external change)	1.23	0.63 (cm^3)
V_9 (estimate of cracks after irradiation)	0.18	0.18 (cm^3)
ΔV_a	3.02	2.52 (cm^3)
V_1	17.54	17.54 (cm^3)
$\frac{\Delta V_a}{V_1}$ x 100(%)	17.2	14.4 (%)

3) Assuming that 1(b) is correct then

a) the total volume in the element at the start of irradiation

$$\Delta V_b = V_1 + V_4 + V_5 + V_7$$

b) total swelling in V_1 (cm^3) of U_3Si fuel is

$$\Delta V_b = V_1 + xV_4 + xV_5 + xV_7 + V_8 - V_1$$

$$\Delta V_b = xV_4 + xV_5 + xV_7 + V_8$$

c) For elements MJC and MJL with x = 1

	MJC	MJL	
V_7 (hot clearance)	0.13	0.13	(cm^3)
ΔV_b	2.99	2.50	
$\Delta V_1 = (\Delta V_b / V_1)\ 100$	17.0	14.2	(%)

4) Since both assumptions give essentially the same results, and since it is difficult to accurately measure the volume of cracks in the U_3Si, it was decided to use the method employing the hot clearances.

B) Method II

Volume Change from Density Measurements

Subsequent to irradiation, density determinations are done on sections of the U_3Si fuel, and the volume change which occurs in V_1 (cm^3) of fuel is determined as follows using MJC and MJL as examples:

	MJC	MJL	
$\Delta V_2 = \left(\frac{\rho_{TD}}{\rho_i} - 1 \right) \times 100 =$	18.7	16.7	(%)

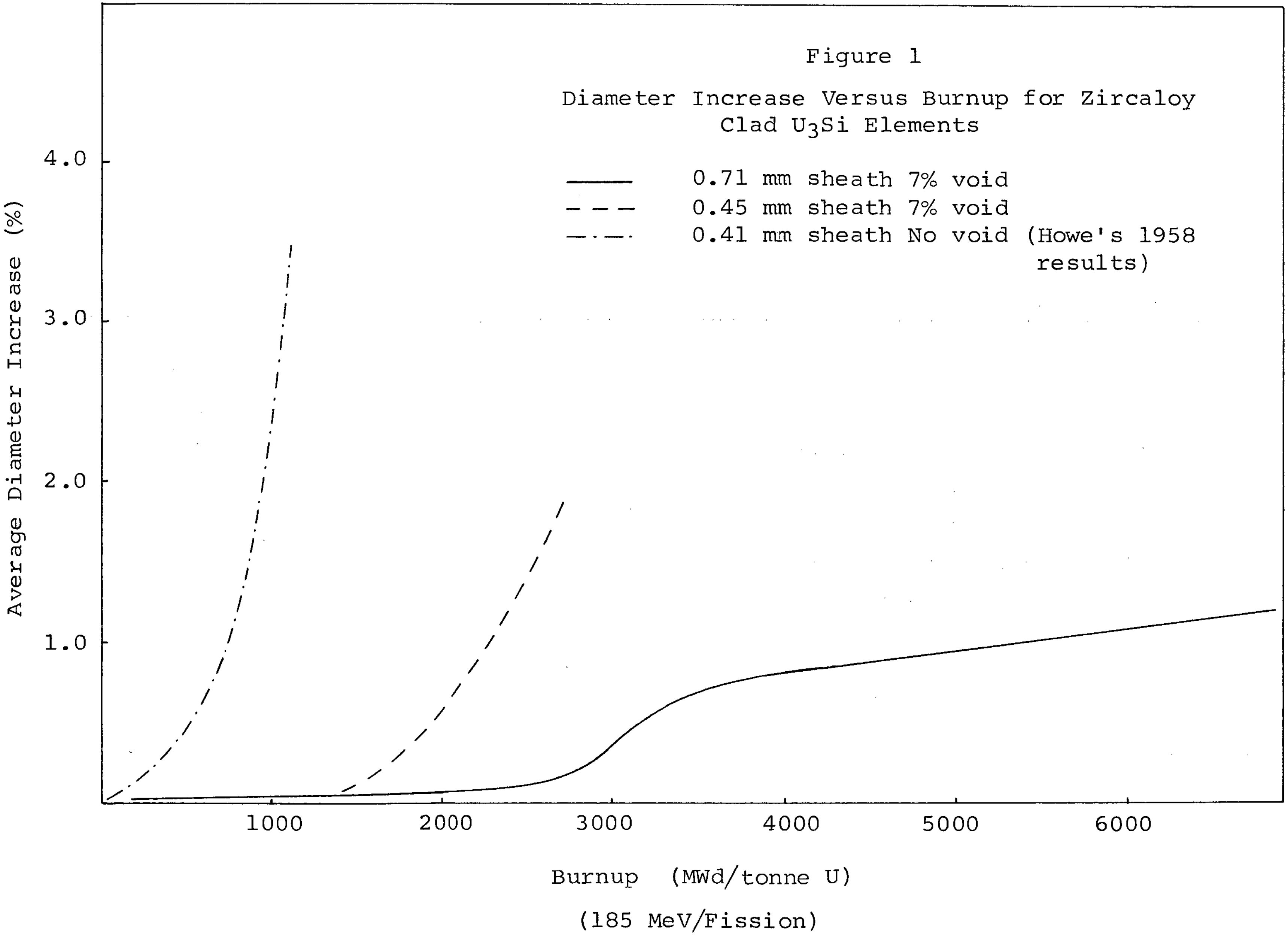


Figure 1

Diameter Increase Versus Burnup for Zircaloy Clad U_3Si Elements

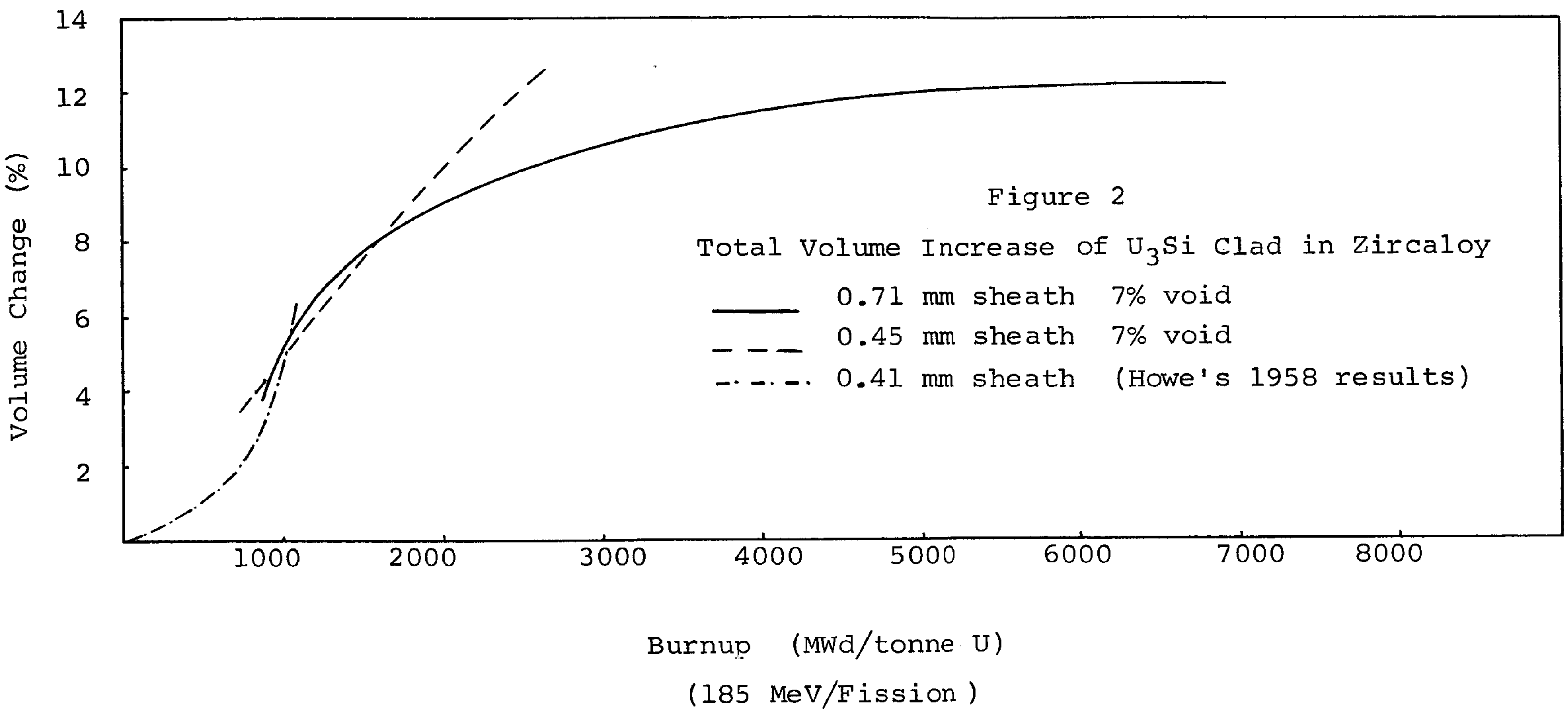


Figure 2

Total Volume Increase of U_3Si Clad in Zircaloy

FIGURE 3

VARIATION OF DIAMETER WITH BURNUP FOR EACH ELEMENT TYPE (SEE TABLE I) - DATA POINTS

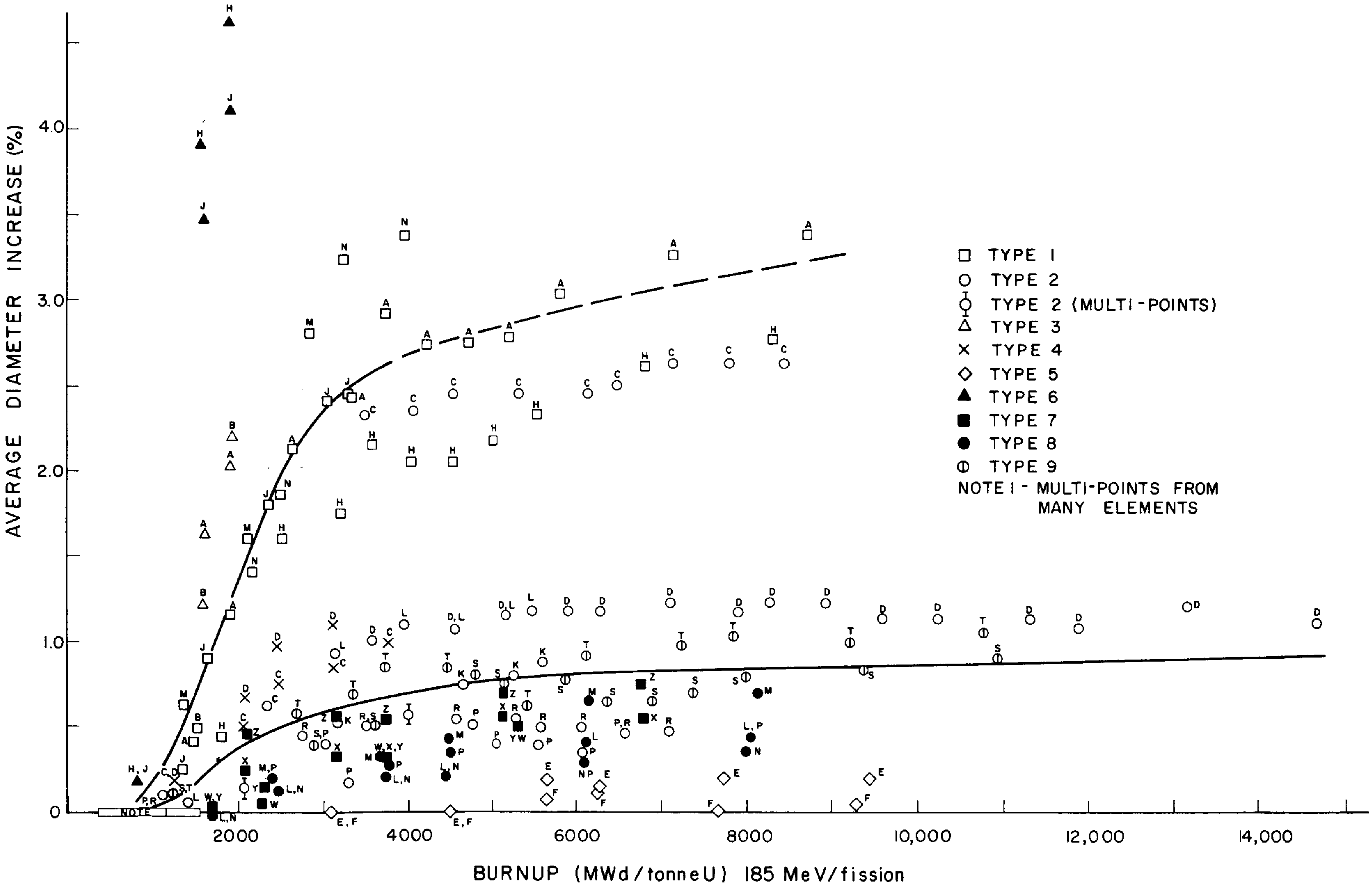
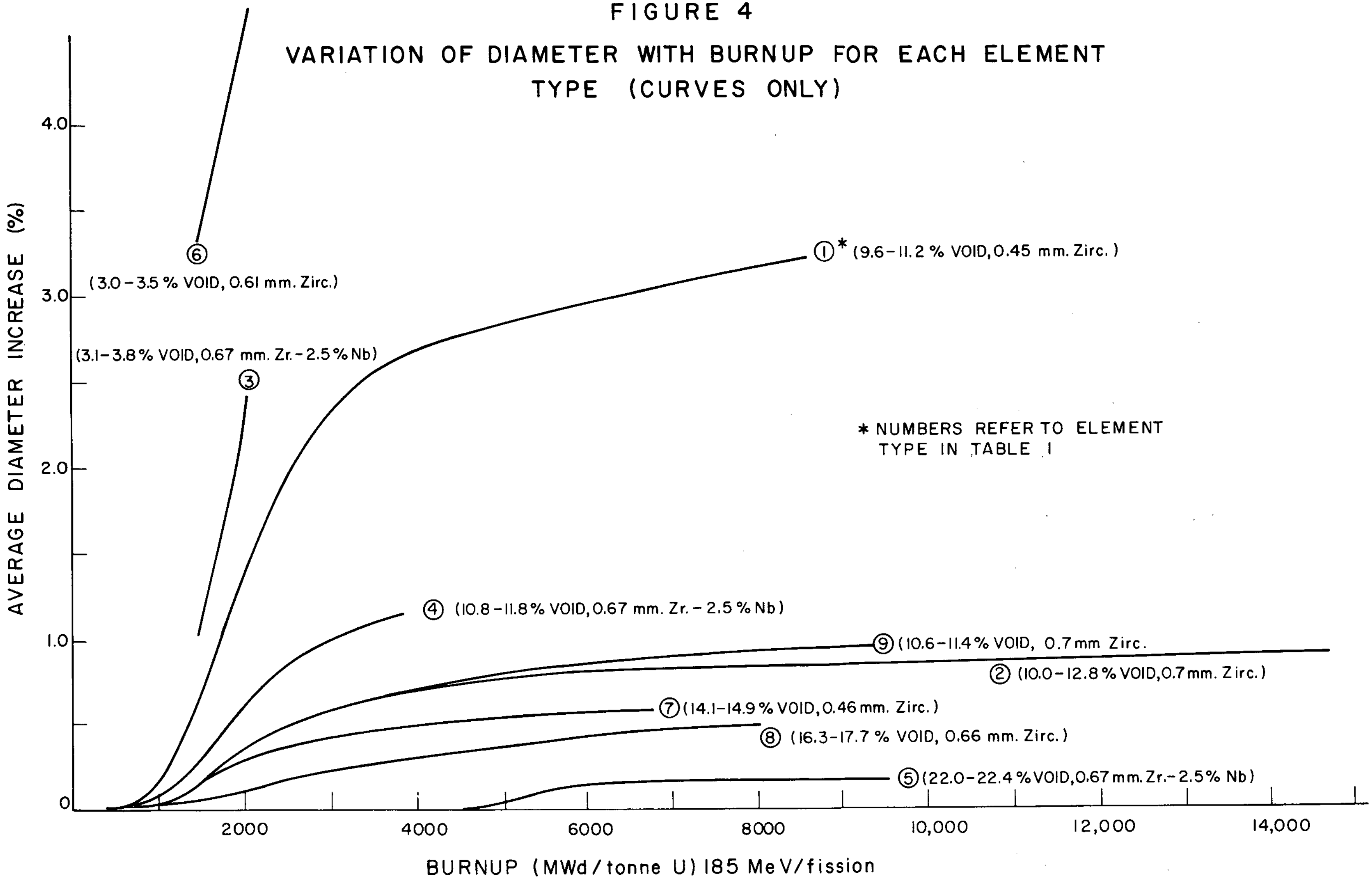


FIGURE 4

VARIATION OF DIAMETER WITH BURNUP FOR EACH ELEMENT TYPE (CURVES ONLY)



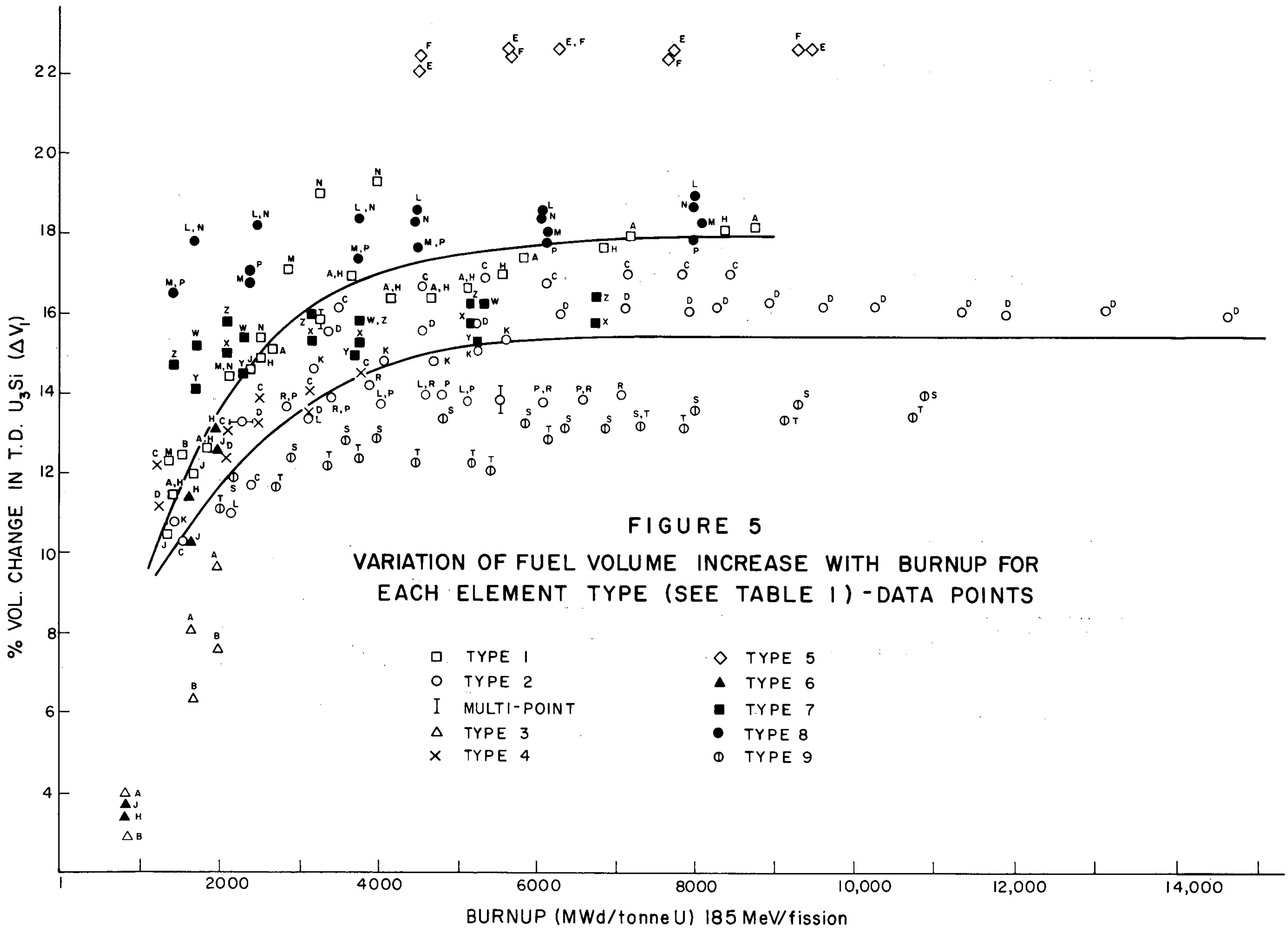


FIGURE 5

VARIATION OF FUEL VOLUME INCREASE WITH BURNUP FOR EACH ELEMENT TYPE (SEE TABLE 1) - DATA POINTS

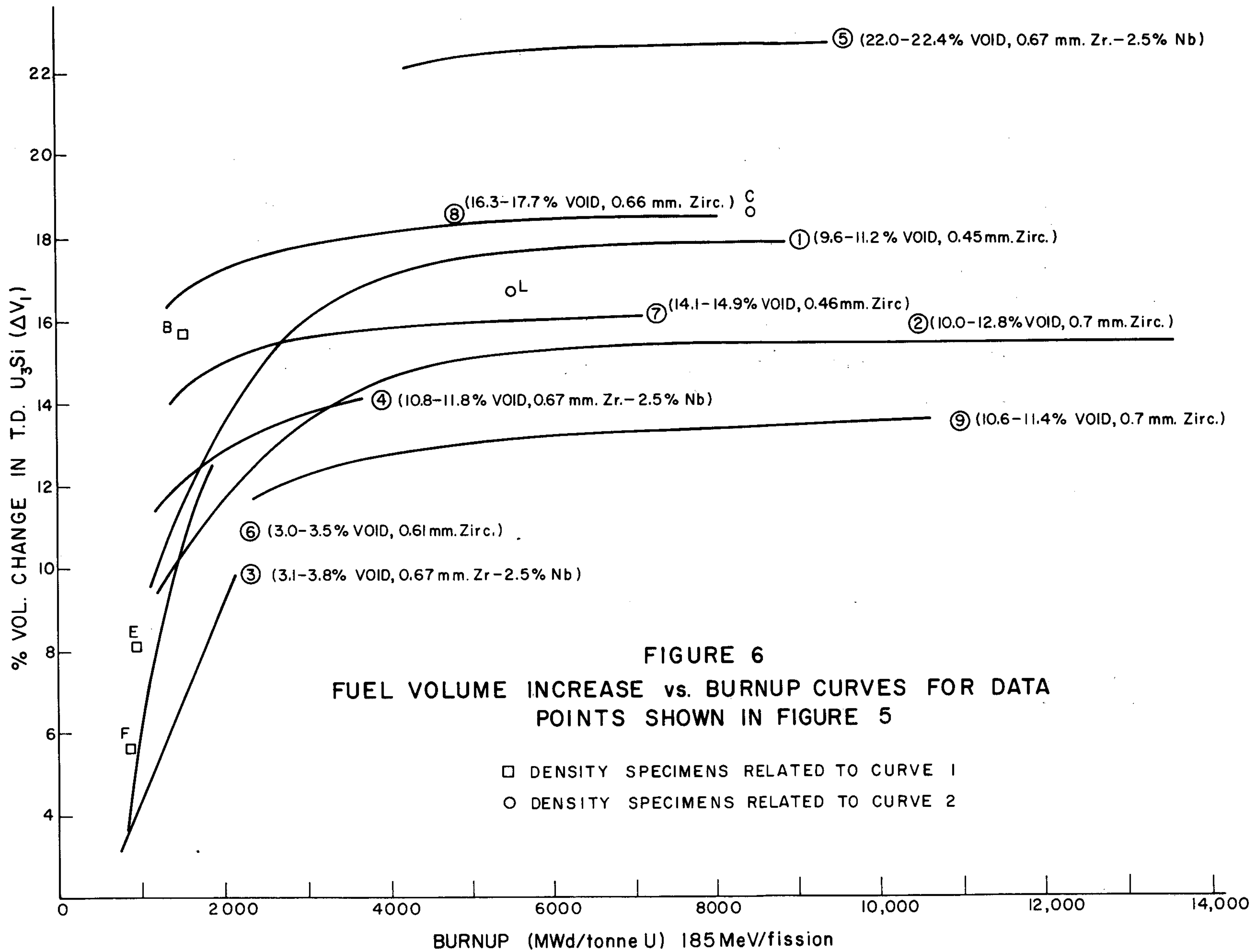
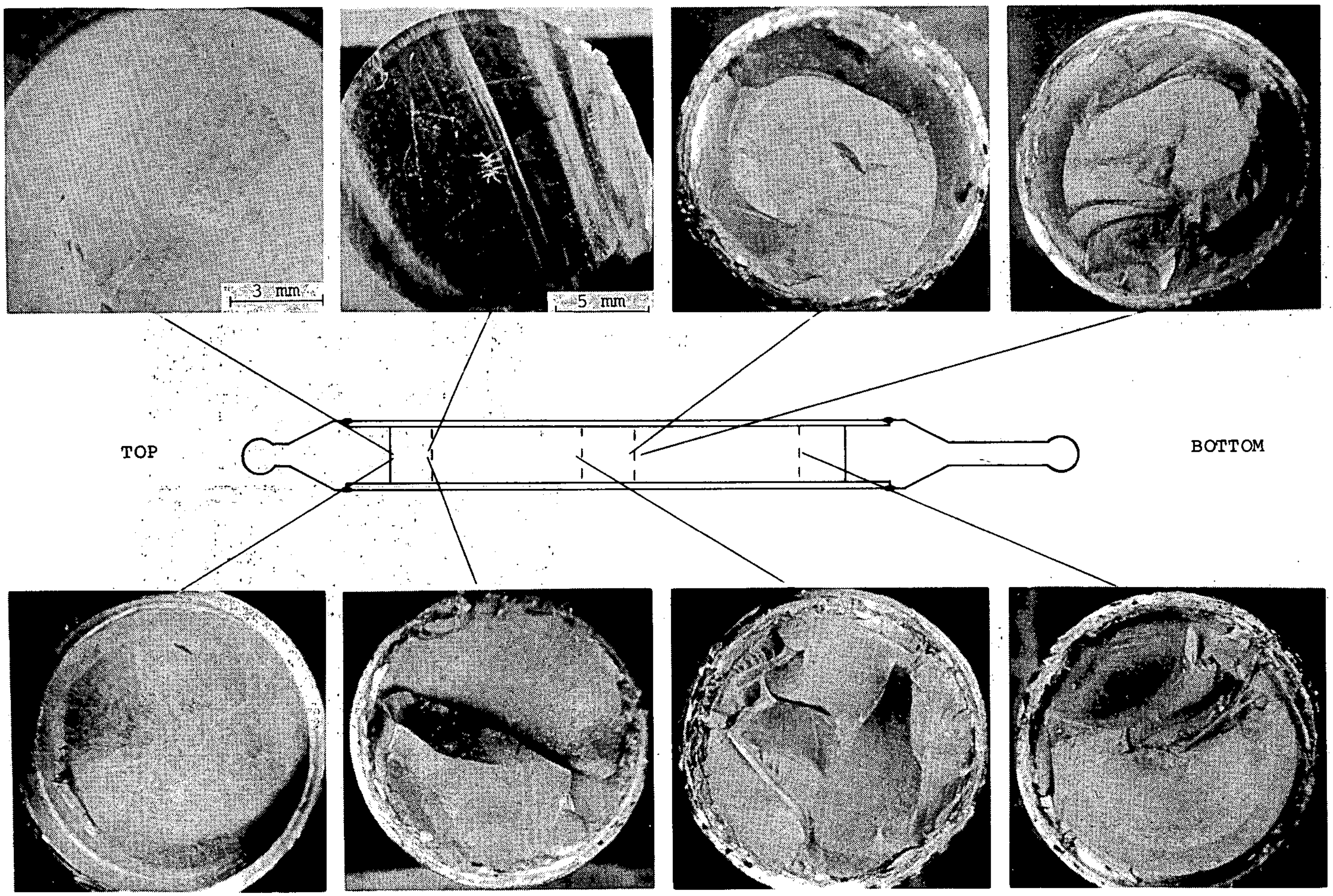


FIGURE 6

FUEL VOLUME INCREASE vs. BURNUP CURVES FOR DATA POINTS SHOWN IN FIGURE 5

End Face of Rod - A   Cut Section - B   Fractured Section - C   Fractured Section - D



End Face of Rod - A   Fractured Section - B   Fractured Section - C   Fractured Section - D

Figure 7

Element MJL after 5490 MWd/tonne U burnup.

Figure 8

Element MJC after 8435 MWd/tonne U burnup.

TYPICAL PHOTOGRAPHS SHOWING DARK ETCHING AREAS IN SECTIONS FROM ELEMENTS MJC AND MJL

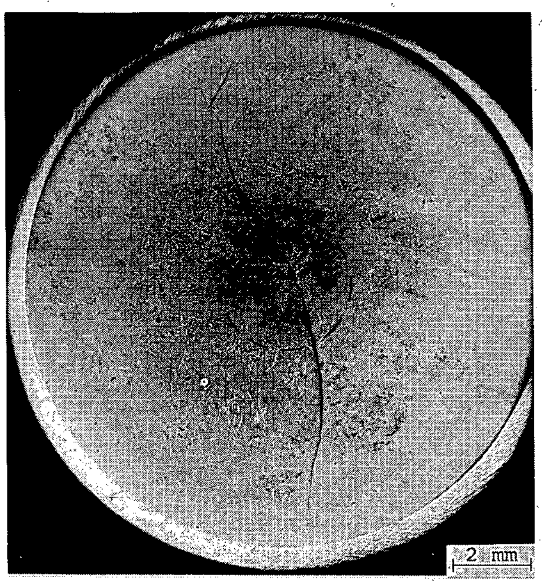


Figure 9 Element MJL
(R-29-A1)

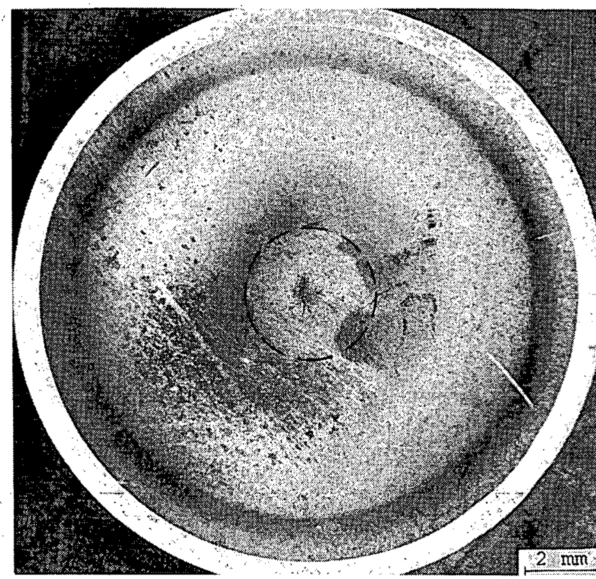


Figure 10 Element MJC - Mid-plane
(R-62-B9)

Figure 11 Element MJC - Top
(R-62-C8)

Dashed circles on cross sections show position and size of original central void

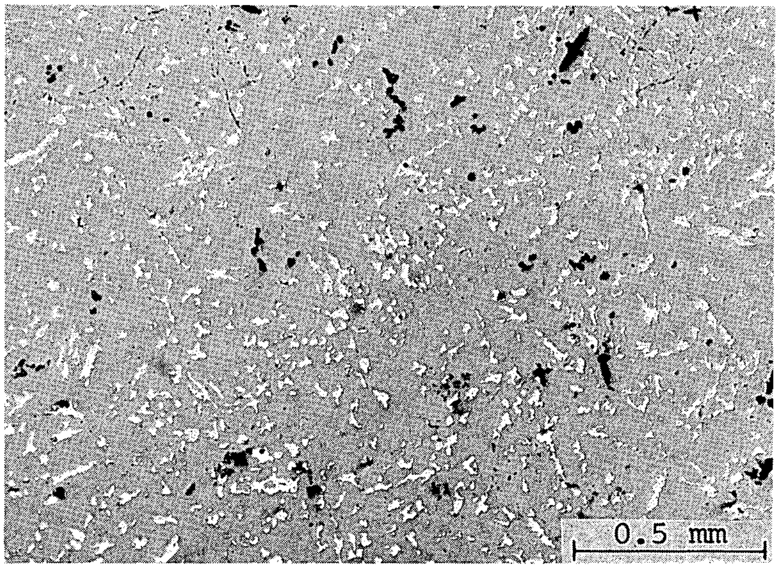


Figure 12 R-29-B4
(Etched)

Typical microstructure of the irradiated fuel showing U_3Si_2 (white) in the U_3Si matrix (grey) with some UO_2 (black). Periphery of element MJL.

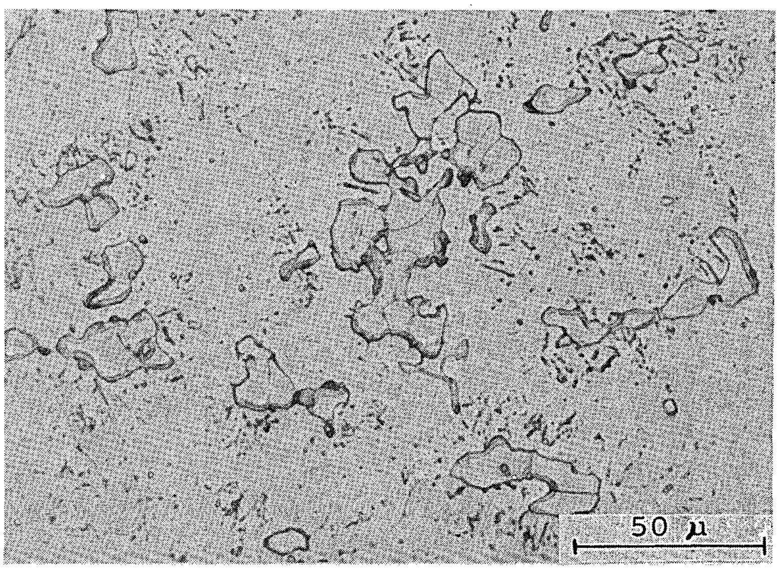


Figure 13 (Etched)

Typical grain size of the unirradiated U_3Si. Two-tone particles are U_3Si_2.

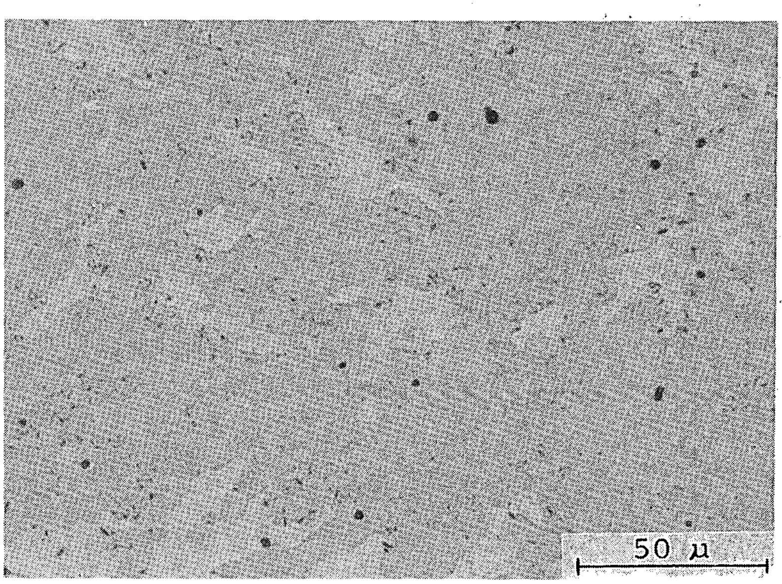


Figure 14 (Etched)

Typical microstructure of unirradiated fuel showing U_3Si (grey), U_3Si_2 (white), UO_2 (black) and unidentified spots.

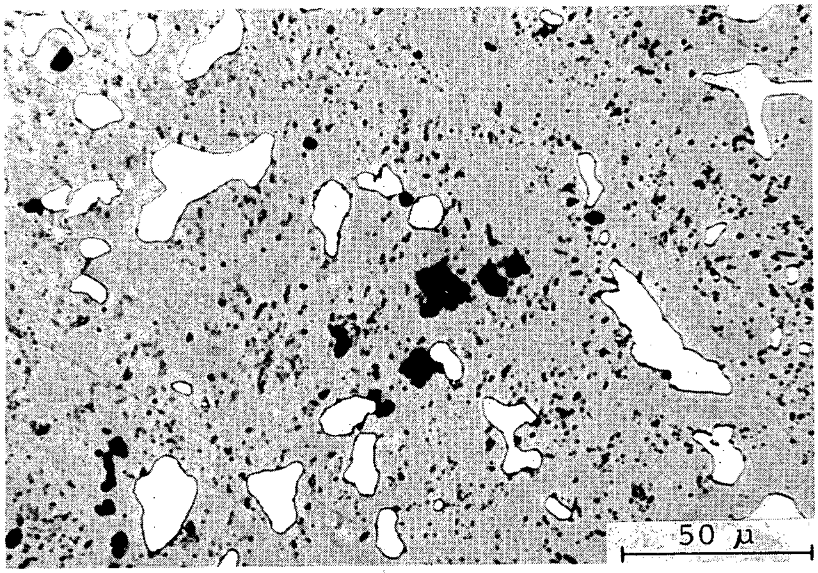


Figure 15 R-62-A7 (Etched)

Typical microstructure at the periphery of the irradiated fuel showing unidentified small black spots and some UO_2 (large black spots)
Element MJC.

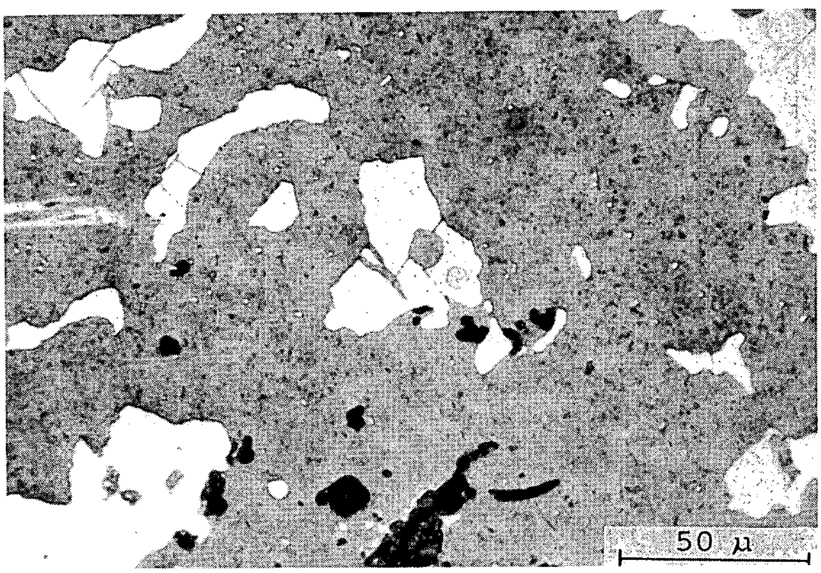


Figure 16 R-62-C11 (Etched)

Microstructure at the mid-radius of element MJC after irradiation showing U_3Si_2 (white) and UO_2 (black) in a dark grey U_3Si matrix.

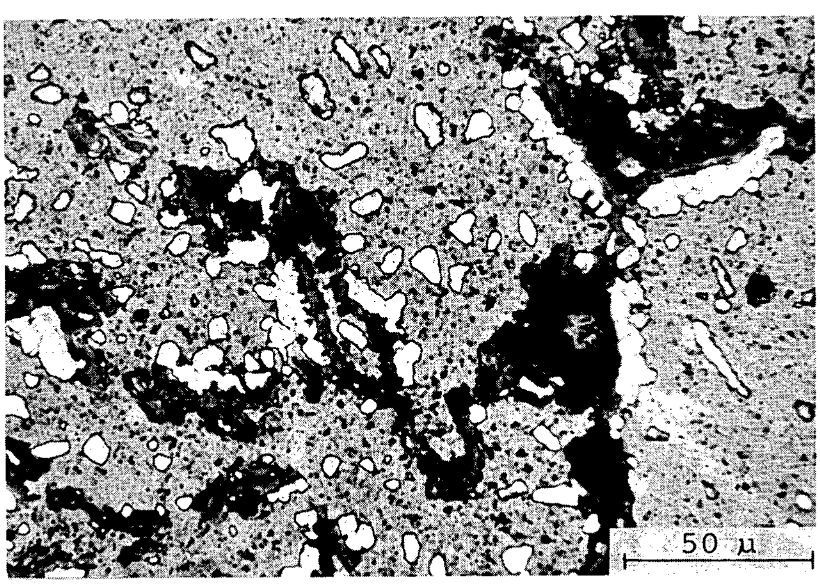


Figure 17 R-62-B2 (Etched)

Microstructure at the centre of element MJC showing remains of central void (black), U_3Si_2 (white), U_3Si matrix (grey).

Zircaloy sheath

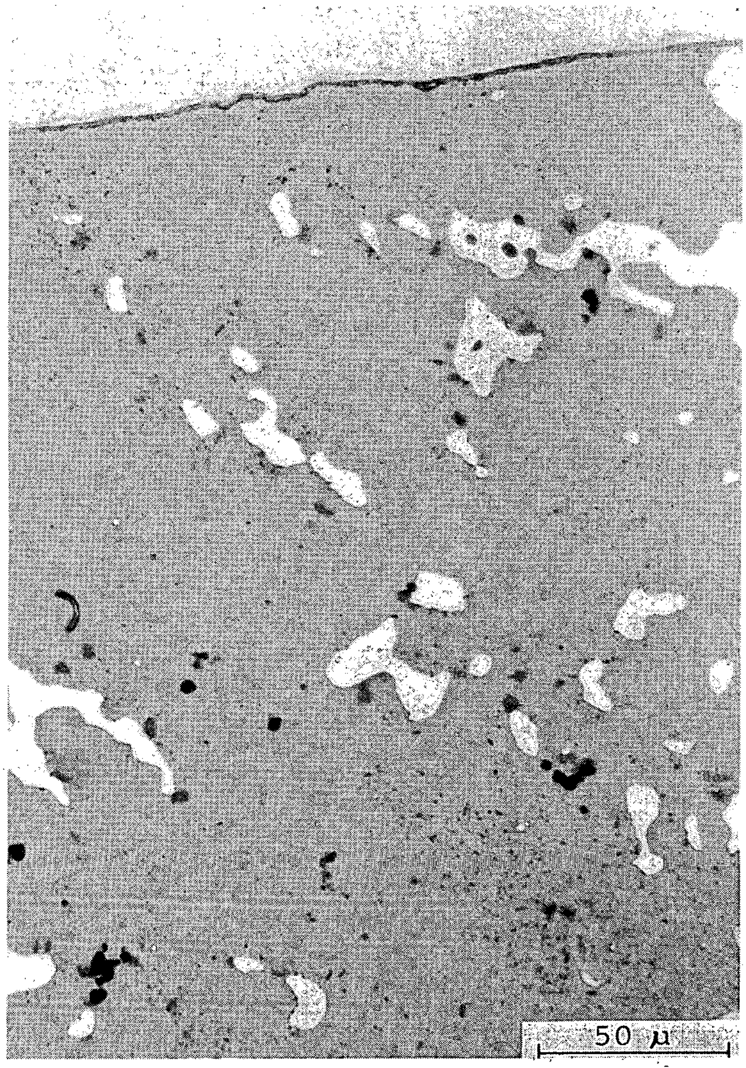


Figure 18 R-29-A4
(Etched)

Microstructure at the periphery of irradiated fuel in element MJL showing U_3Si_2 (white) and UO_2 (black) in U_3Si matrix (grey).

Figure 19

Diameter Profiles of Elements AWZ, AWX and AYN

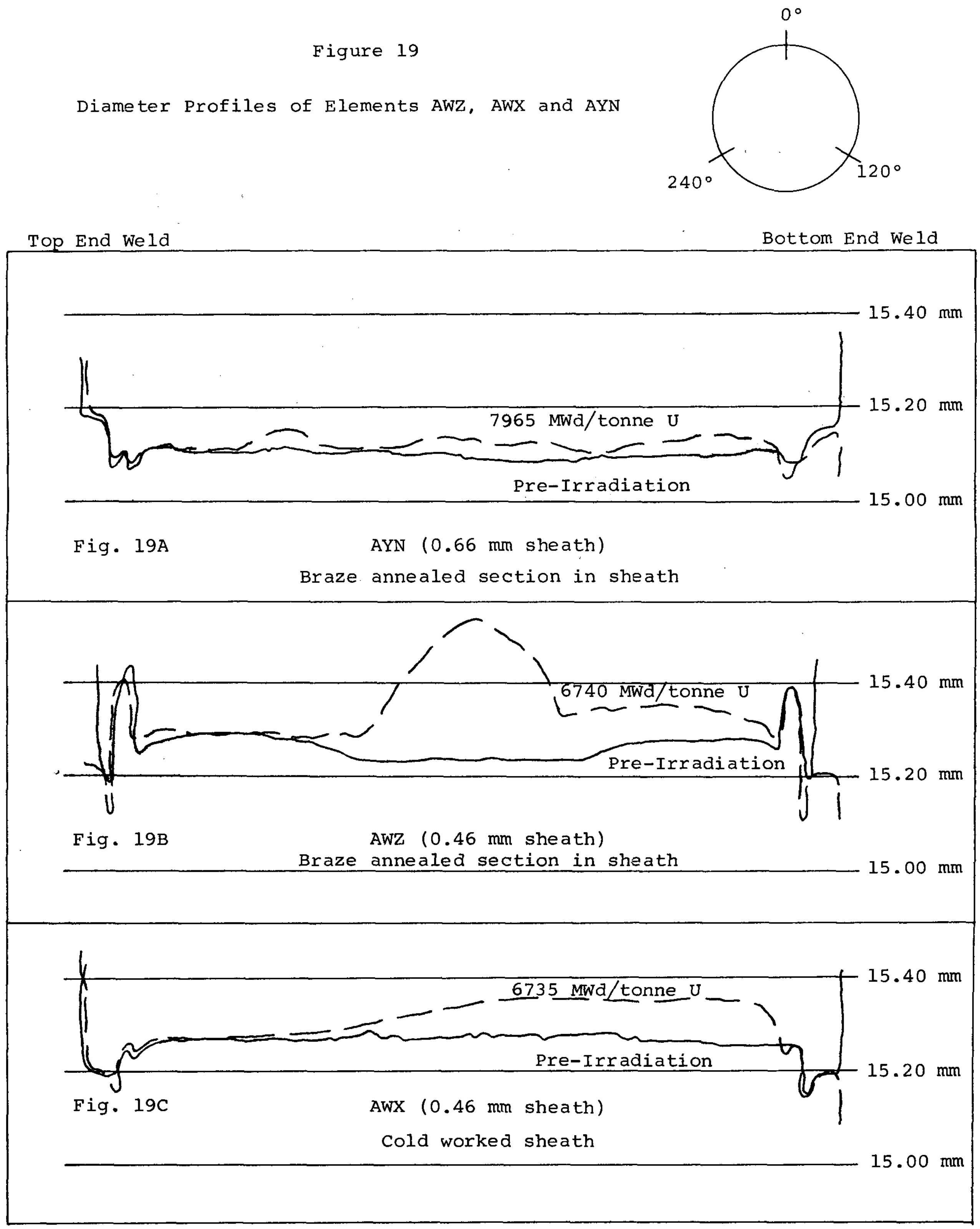


TABLE 1

SUMMARY OF FUEL** ELEMENT TYPES

Type	Sheath		Void (vol%)	
	Material	Thickness (mm)	Nominal Axial	Total*
1	Zirc-2	0.45	7	9.6-11.2
2	Zirc-2	0.71	7	10.0-12.8
3	Zr-2.5% Nb	0.67	0	3.1-3.8
4	Zr-2.5% Nb	0.67	7	10.8-11.8
5	Zr-2.5% Nb	0.67	16	22.0-22.4
6	Zirc-4	0.61	0	3.0-3.5
7	Zirc-2	0.46	10	14.1-14.9
8	Zirc-4	0.66	10	16.3-17.7
9†	Zirc-2	0.72	7	10.6-11.4

* Includes casting porosity and hot (calculated) clearances in fuel element and is expressed as a percentage of theoretically dense U_3Si present (see Appendix A).

** Nominal 4 wt% Si (see Table 3).

† Contained minor alloying additions (see Table 3).

TABLE 2

PRE-IRRADIATION DATA ON FUEL & ELEMENTS

Element		Fuel Rods (see Table 3 for analysis)					Sheath			
Type (Table 1)	No	Porosity V5* (%)	Central Void V4* (%)	O.D. (mm)	Length (mm)	Weight of U_3Si (g)	Material	O.D. (mm)	t (mm)	Total Free Vol/Element (V4 + V5 + V7)* (%)
1	MJA	1.9	7.4	14.28	129.5	290.63	Zirc-2	15.2	0.44	9.9
1	MJB	3.2	7.7	14.28	130.1	287.47	Zirc-2	15.2	0.45	11.0
2	MJC	1.1	8.2	13.71	129.8	268.30	Zirc-2	15.2	0.70	10.0
2	MJD	3.7	8.3	13.71	130.1	262.54	Zirc-2	15.2	0.72	12.8
1	MJH	3.5	7.0	14.25	131.8	291.15	Zirc-2	15.2	0.46	11.2
1	MJJ	1.7	7.2	14.25	131.2	294.22	Zirc-2	15.2	0.42	9.6
2	MJK	3.9	8.7	13.72	130.4	262.09	Zirc-2	15.2	0.72	13.0
2	MJL	1.7	8.2	13.71	130.6	268.45	Zirc-2	15.2	0.71	10.6
1	MJM	2.4	8.1	14.31	131.3	292.34	Zirc-2	15.2	0.45	10.9
1	MJN	2.7	7.7	14.31	130.7	291.22	Zirc-2	15.2	0.44	11.1
2	MJP	3.6	8.6	13.69	131.4	263.78	Zirc-2	15.2	0.72	12.7
2	MJR	3.9	8.2	13.69	130.8	262.72	Zirc-2	15.2	0.71	12.7
9	MJS	2.9	7.8	13.69	131.2	267.16	Zirc-2	15.2	0.72	11.4
9	MJT	2.4	7.4	13.69	129.7	266.63	Zirc-2	15.2	0.72	10.6
3	AWA	2.7	0	13.77	122.4	273.42	Zr-2.5% Nb	15.1	0.67	3.4
3	AWB	2.5	0	13.78	120.8	270.71	Zr-2.5% Nb	15.1	0.67	2.5
4	AWC	2.6	8.2	13.78	123.4	255.77	Zr-2.5% Nb	15.2	0.68	11.3
4	AWD	2.7	7.5	13.78	123.1	256.41	Zr-2.5% Nb	15.1	0.67	10.8
5	AWE	2.4	19.2	13.78	124.2	234.61	Zr-2.5% Nb	15.1	0.67	21.6
5	AWF	2.3	19.6	13.78	123.3	232.20	Zr-2.5% Nb	15.2	0.68	22.0
6	AWH	2.4	0	13.83	130.9	295.69	Zirc-4	15.1	0.61	2.9
6	AWJ	2.7	0	13.83	129.9	292.52	Zirc-4	15.1	0.61	3.2
7	AWW	2.5	11.2	14.30	126.7	273.90	Zirc-2	15.3	0.46	14.9
7	AWX	2.0	11.1	14.30	127.3	276.41	Zirc-2	15.3	0.46	14.4
7	AWY†	1.7	10.8	14.29	127.6	278.29	Zirc-2	15.3	0.46	13.9
7	AWZ†	2.2	11.1	14.31	126.5	274.63	Zirc-2	15.3	0.46	14.5
8	AYL	2.6	13.0	13.69	127.4	244.72	Zirc-4	15.1	0.66	17.9
8	AYM	2.0	12.5	13.69	127.2	245.26	Zirc-4	15.1	0.66	16.6
8	AYN†	2.6	12.5	13.69	127.8	246.65	Zirc-4	15.1	0.66	17.7
8	AYP†	1.8	12.5	13.69	127.0	245.35	Zirc-4	15.1	0.65	16.8

†These elements have an annealed zone in the cold worked sheath (see section 2)

*As a % of V_1, the volume of theoretically dense U_3Si - See appendix A.

TABLE 3

SUMMARY OF CHEMICAL & MASS ANALYSES DATA ON THE U_3Si

Element Type (see Table 1)	Si* wt% (±0.1)	Impurities (ppm) Fe**	Al**	Ni**	Zr**	C*	Enrichment wt% U^{235} in U
1,2	4.01	80	155	40	1	400	1.81
9	4.01	350	1500	100	160	375	1.81
3,4,5,6	3.92	330	420	80	500*	250	2.16
7	4.02	115	30	60	230	85	1.81
8	4.30	135	70	100	1	290	2.14

* Chemical Analysis

** Spectographic Analysis

TABLE 4

POST-IRRADIATION DATA ON THE U_3Si FUEL ELEMENTS

Element		Equivalent Full Power Days Irradiation as of July 1968	Power Outputs (W/cm) (185 MeV/fission)		Calculated Temperature* °C		Length Change (%)	Bowing (mm x 10^{-2})	Burnup as of 15 July 1968 (MWd/Te U)**	
Type	No		Range	Time Avg.	T_s	T_c			From Calorimetry	From Chemical Analysis
1	MJA	417	349-510	450	305	470	-0.39	41	8740	
1	MJB	63	500-510	505	305	470	NM	13	1505	1820
2	MJC	366	412-485	455	320	475	+0.05	32	8435	8980
2	MJD	639	407-485	445	320	475	-0.04	16	14,700	
1	MJH	417	332-503	425	305	465	-0.33	89	8350	
1	MJJ	162	334-468	440	300	450	-0.32	52	3305	
2	MJK	259	402-446	420	315	455	+0.01	2	5920	5990
2	MJL	259	402-446	420	315	455	-0.05	17	5490	5480
1	MJM	127	459-500	475	305	465	-0.36	170	2840	
1	MJN	191	424-479	445	300	455	-0.30	124	3960	
2	MJP	307	321-485	410	320	475	-0.05	11	6570	
2	MJR	336	318-497	405	325	480	-0.06	5	7085	
9	MJS	495	322-491	430	320	480	-0.02	20	10,950	
9	MJT	480	318-497	440	325	480	-0.02	11	10,780	
3	AWA	84	450-512	500	320	520	-0.20	34	1950	
3	AWB	84	450-512	498	320	520	-0.20	35	1950	
4	AWC	160	422-504	470	320	480	-0.02	13	3775	
4	AWD	160	422-504	470	320	480	-0.09	10	3760	
5	AWE	395	393-474	435	315	440	-0.12	8	9455	
5	AWF	395	393-474	425	315	440	-0.07	10	9275	
6	AWH	84	450-512	500	315	515	-0.16	85	1930	
6	AWJ	84	450-512	500	315	515	-0.16	85	1940	
7	AWW	235	446-479	470	300	445	-0.16	9	5320	
7	AWX	296	463-490	475	305	460	-0.24	15	6735	
7	AWY†	236	446-479	470	300	445	-0.18	8	5270	
7	AWZ†	296	463-490	475	305	460	-0.13	20	6740	
8	AYL	296	486-515	495	310	450	-0.11	3	8000	
8	AYM	296	447-548	505	320	450	-0.12	10	8085	
8	AYN†	296	486-515	495	310	450	-0.08	7	7965	
8	AYP†	296	447-548	505	320	450	-0.09	8	8075	

* See section 5.2

** Based on 185 MeV/fission heat generation by the fuel.

† These elements have an annealed zone in the cold worked sheath (see Section 2).

T_s and T_c are the calculated temperatures at the fuel surface and centre respectively.

NM - Not measured

TABLE 5

FUEL VOLUME CHANGES CALCULATED BY DIFFERENT METHODS

Element	Burnup (MWd/tonne U)	% Volume Change (see App. A)			
		ΔV_1*	$\Delta V2$**		
			Position of Sample†	Values	Average
MJF	840	4.5	Top Bottom Top	6.1 5.9 5.2	5.7
MJE	920	6.5	Bottom Top Top	11.7 7.0 5.6	8.1
MJB	1505	12.4	Top Top Bottom	18.6 16.7 11.7	15.7
MJL	5490	14.2	Top	16.7 16.7	16.7
MJC	8435	17.0	Bottom	18.7 18.7	18.7

* The data points on Fig. 5 are calculated using Method 1 (App. A)

** The volume changes calculated using Method 2 (App. A), i.e. the density samples, are plotted on Fig. 6.

† Top or bottom indicates the centre of the top or bottom fuel slug respectively.

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