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LUCAS HEIGHTS

THE STRENGTH OF IRRADIATED BERYLLIUM OXIDE FUELLED WITH UO_2-ThO_2

by

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Introduction and Table 1, for "Rig X–11" read "Rig X–12"

Page 3, para. 3, for "ThO_2, 9.3×10^{-6}" read "ThO_2, 9.75×10^{-6}".

This new value for the thermal expansion coefficient of ThO_2, which results from more recent (unpublished) work by W. B. Rotsey and D. N. Turner, does not affect the argument or the discussion.

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ABSTRACT

The mechanical strength of hot-pressed and cold-pressed and sintered dispersions of $(U,Th)O_2$ in BeO was measured before and after irradiation using the diametral compression test.

The addition of more than 5 volume per cent. $(U,Th)O_2$ lowered the unirradiated strength to about 60 per cent. of the strength of BeO. Irradiation resulted in a marked drop in strength of all coarse dispersions. Hot-pressed fine dispersions behaved similarly to coarse dispersions of the same composition but the cold-pressed fine dispersions exhibited an apparent increase of strength on irradiation.

High temperatures favoured the retention of strength during irradiation.

CONTENTS

1. INTRODUCTION

Irradiation experiments with dispersion fuels of beryllium oxide fuelled with solid solutions of urania and thoria have been described by Hanna, Hickman, and Hilditch (1963), Hanna and Hilditch (1964), and Hanna (1964). These reports were devoted to metrological, microstructural, and X-ray diffraction examinations and to measurements of total fission gas release. Recently, compression testing equipment was installed in the hot cell and specimens were tested for changes in mechanical strength using the diametral compression test. This test was chosen because :

(i) It is a simple test to perform remotely and failure occurs under a tensile stress;

(ii) the specimen shape is simple and specimens could be readily prepared in the hot cell from the cylindrical irradiation specimens; and

(iii) more specimens were made available than if the axial compression test were used.

The irradiation experiments, known as the X-11 and X-71 experiments, were done in a region outside the reactor core so that radiation damage was produced only by fission within the specimens themselves and not by exposure to the reactor fast flux.

Specimens used in the X-11 experiment were prepared by hot pressing (H.P.) and contained irregular-shaped fuel particles. The X-71 specimens were cold-pressed and sintered (C.P.S.) and contained spherical fuel particles in the coarse dispersions. Fuel particles in all coarse dispersions were in the size range 100 to 200 microns. Two specimens in which the fuel particles were mostly less than 10 microns and irregular in shape were included in each experiment. Details of specimens are given in Table 1.

It must be emphasised at the outset that the results of the experiments probably have no quantitative significance and must be regarded only as an indication of how the tensile strength of dispersion fuels is affected by irradiation. This is because only small numbers of specimens were available for testing (normally large numbers must be used with ceramic materials to allow adequate statistical analysis when wide scatters are encountered) and because the diametral compression test is less thoroughly understood and less frequently used than bend tests.

2. EXPERIMENTAL

Details of the irradiation techniques were reported by Hanna, Hickman, and Hilditch (1963) and Hanna and Hilditch (1964). Burnups and fission densities are given in Table 1.

Specimens were cylindrical in their original form and were slit into discs about 6 mm thick using 0.02 in thick silicon carbide wheels. Control specimens were prepared outside the cell by slitting unirradiated specimens with a diamond-impregnated wheel.

The faces of some discs cut from irradiated specimens were badly scored and were ground on a metallographic grinding machine until the marks were removed.

Compression testing was done on a 20-ton capacity, electrically-driven hydraulic press, using a two-ton load cell which had been calibrated outside the hot cell against an Instron Tensile Testing Machine. Fresh cardboard pads about 0.01 in thick (after Rudnick et al. 1963) were placed at the specimen's loading points for each test.

3. RESULTS

3.1 Coarse Dispersions

Tensile strengths measured in the diametral compression tests are given in Table 1 and in Figures 1 to 4 where they are expressed as residual strength, that is percentages of the unirradiated values, and plotted against various irradiation parameters.

Figure 1 is a plot of strength versus fuel content of unirradiated material. Owing to the small number of specimens available the as-fabricated and heat-treated control specimens were grouped together to obtain average strengths of unirradiated materials.

Both hot-pressed and cold-pressed and sintered materials showed a marked drop in strength in the range 0 to 5 volume per cent. fuel material. Above this, however, the strength became constant at a value of about 60 per cent. of the strength of unfuelled BeO.

The strength of H.P. material containing 4.4 volume per cent. fuel was independent of the composition of the fuel phase; values of 18,550, 19,140, and 19,350 p.s.i. were obtained for UO_2: ThO_2 ratios of 1:3, 1:1, and 3:1 respectively. In contrast, the strength of C.P.S. material containing 15 volume per cent. fuel decreased from 16,200 p.s.i. at U:Th = 1:3 to 14,820 p.s.i. at U:Th = 1:1 and 10,670 p.s.i. at U:Th = 3:1. There is reason to believe that the effect on the C.P.S. specimens is real (see Section 4) and so the residual strengths of irradiated C.P.S. specimens were normalised to the mean value for the unirradiated specimens of the same composition. Hot-pressed specimens, on the other hand, were normalised to the mean value for all unirradiated specimens having the appropriate fuel content (4.4 volum per cent).

No unirradiated specimens were available in the H.P., 25.8 volume per cent. fuel material and an "unirradiated" strength of 16,000 p.s.i. was obtained by extrapolation of the curve in Figure 1.

Irradiation reduced the strength of all coarse dispersion specimens. The strength of H.P., 3 volume per cent. material fell by 87 per cent. and that of 4.4 to 25.8 volume per cent material by 39 to 68 per cent. The strength of C.P.S. 5 volume per cent. material was 30 per cent. lower after irradiation and those of the 15 and 30 volume per cent. materials 52 to 65 per cent. lower.

The residual strength of H.P. specimens showed no correlation with fission density in either the fuel or whole specimens but the residual strength of the 4.4 volume per cent. specimens increased as the irradiation temperature increased (see Figure 2). The strength of C.P.S. specimens decreased as the fission density in the whole specimens increased (Figure 3); as the fuel content increased (Figure 4) there was no correlation with fission density in the fuel material. There were insufficient specimens of a given composition to obtain a strength versus temperature plot as obtained with the H.P. specimens.

3.2 Fine Dispersions

The H.P. material containing fine fuel particles (4.4 volume per cent.) had an unirradiated strength of 28,840 p.s.i. This fell by 54 per cent. on irradiation.

The fine C.P.S. dispersions (15 volume per cent). had a mean strength of 13,280 p.s.i. before irradiation and 20,700 p.s.i. afterwards. It should be noted here that only two unirradiated and three irradiated specimens were tested.

4. DISCUSSION

4.1 Unirradiated Material

Rotsey and Veevers (1964) have discussed the effect on the strength of BeO of introducing spherical fuel particles of $UO_2 - ThO_2$ solid solutions. When the fuel is not bonded to the matrix the particles will behave as holes. Under an applied stress the holes will cause local stress concentrations in the adjacent matrix and will therefore reduce the applied stress needed to cause failure. If, on the other hand, the particles are bonded to the matrix, fracture will initiate in the fuel material because it is weaker; Rotsey and Veevers deduced that typical BeO fuelled with 200 micron spheres of $(U,Th)O_2$ (strength 14,3000 p.s.i.) will fail under a tensile strength of 26,800 p.s.i. They measured the strength of dispersions containing 1.7 volume per cent. $(U,Th)O_2$ and found it to be 23,000 p.s.i. in material which fractured through, rather than around, the fuel particles. The difference between predicted and observed values was considered to be due to :

(a) stresses arising from differential contraction during cooling,

(b) slightly irregular particle shapes, and

(c) irregular packing of particles and the overlap of some stress fields adjacent to particles during testing.

If fuel particles are bonded to the matrix the strength of the material when subjected to a uniform tensile stress will not vary with fuel content because failure can occur in any fuel particle. However, if the testing technique yields a non–uniform stress, as in the bend and diametral compression tests, the strength should decrease with increasing fuel content owing to the increasing chance of a flaw lying in the region of maximum stress. This is essentially the behaviour observed in the work reported here (see Figure 1), when the strength fell rapidly in the range 0 to 5 volume per cent. fuel. The "saturation" strength (about 60 per cent. of that of unfuelled BeO) reached at higher fuel contents is lower than expected from Rotsey and Veevers' work but the difference can perhaps be accounted for by reasons similar to those suggested by Rotsey and Veevers (see above) to account for the difference between observed and predicted strengths.

The dependence of the strength of C.P.S. 15 volume per cent. dispersions on the composition of the fuel phase can be explained by differences in thermal expansion between the fuel and matrix phases if the fuel particles are bonded to the matrix. Microscopic examination of fracture faces revealed that fracture occurred predominantly through the particles thus indicating that most particles were in fact bonded. Values of the thermal expansion coefficients (from 20–1000 °C) of the three compounds concerned are : BeO, 9.3×10^{-6} ; ThO_2, 9.3×10^{-6} ; and UO_2, 10.1×10^{-6} (W.B. Rotsey, private communication). No data exist for solid solutions of UO_2 and ThO_2, but if one assumes a linear dependence on molar composition the tensile stresses which arise within the particles from differential contraction on cooling from the fabrication temperature will be greater in dispersions of UO_2 and UO_2–rich solid solutions. Hence, strength should decrease as the UO_2 content of the fuel phase increases, and this was observed in the C.P.S. specimens 158, 264, and 272.

Unfortunately the H.P. specimens do not support this picture since the strength of the 4.4 volume per cent. specimens did not change significantly with $UO_2 : ThO_2$ ratio. This suggests that in these specimens the particles were unbonded or cracked. However, the strength of H.P. material containing 15.7 volume per cent. of $(U,Th)O_2$ (1U : 3 Th) was the same as that of C.P.S. material of the same composition, indicating that the particles were bonded.

Rotsey and Veevers recommend that the fuel particle size be kept below four microns if the effects of stress concentration on crack propagation are to be minimised. Although this condition was not fully met in either of the fine dispersion materials used in this work (both contained some particles up to 30 microns in diameter), the strength of the fine H.P. material (Specimens 149, 150) was 28,850 p.s.i., which was comparable to that of unfuelled BeO. The strength of the C.P.S. material (13,300 p.s.i., specimen 283) was higher than that of coarse dispersions of the same composition (specimen 272), but it was weaker than pure BeO (18,900 p.s.i.). The value of 13,300 p.s.i. must be considered doubtful, however, as the irradiated specimens had a mean strength of 20,700 p.s.i. which compares favourably with that of the BeO.

4.2 Irradiated Material

There are three striking features of the results obtained with irradiated specimens:

(i) the marked fall in strength of all coarse dispersions on irradiation,

(ii) the temperature dependence of the residual strength of the coarse 4.4 volume per cent. H.P. material, and

(iii) the high strength of the C.P.S. fine dispersion specimens.

There can be little doubt that the loss of strength of coarse C.P.S. dispersions is associated with the cracking of fuel particles (see Figure 5, also Hanna and Hilditch 1964), but the reason for this cracking is not known. Cracking of fuel particles does not appear to be the whole explanation for loss of strength, however, as cracks were not observed in the fuel particles of irradiated H.P. material.

In the absence of cracked fuel particles there are three mechanisms which could conceivably lead to the observed deterioration of mechanical properties:

(i) Fast neutron damage to the BeO matrix arising from fission neutrons born within the specimens.

(ii) Swelling of fuel particles and the imposition of stresses on the matrix.

(iii) Damage to the fuel particles and the immediately adjacent matrix by fission fragment recoil.

The results do not indicate which of these effects are important.

Ideally, fission fragment recoil damage and fuel particle swelling should be reflected by a dependence of residual strength on fission density in the fuel material whereas the effects of fast neutron damage should vary with the fission density in the specimen as a whole.

There was no correlation of the strength of coarse H.P. material with any parameter other than temperature. The strength of coarse C.P.S. material, however, correlated reasonably well with fission density in the whole specimen but the scatter was large when strength was plotted against fission density in the fuel material. (Based on Figure 2 the scatter would be even larger if points could be normalised to the one temperature).

This, then, suggests that fast neutron damage might be important but the work of Lawrence (1963) has shown that the effective integrated fluxes received by the specimens was probably only 3×10^{19} to 2×10^{20} nvt. In the absence of other effects (for example fuel particle swelling) these doses at temperatures of 600 to 750 °C should not lead to microcracking or loss of strength in the BeO matrix. Furthermore, there was no metallographic evidence of microcracking.

Hanna (1964) suggested that strain detected by X-ray diffraction in coarse H.P. specimens was probably due to swelling of the fuel particles. Daniel and co-workers (1962) found that UO_2 irradiated with centre temperatures in the range 800 to 1200 °C swelled at the rate of 0.7% $\Delta v/v$ per 10^{20} fissions per cm^3 but that the apparent swelling rate up to 2×10^{21} fissions per cm^3 was only 0.16% $\Delta v/v$ per 10^{20} fissions per cm^3 owing to the elimination of porosity and voids within the sheath. Assuming that $UO_2 - ThO_2$ solutions swell at a similar rate, one can use the expression derived by Selsing (1961) to evaluate the stress generated in the matrix. The hydrostatic pressure P is given by:

$$P = \frac{\Delta r}{r} \, \frac{1}{\left(\dfrac{1 + \nu_m}{2 E_m}\right) + \left(\dfrac{1 - 2 \nu_t}{E_f}\right)} ,$$

where $\Delta r/r$ is the fractional radial misfit, ν is Poissons ratio, E is Youngs Modulus, and the subscripts m and f denote matrix and fuel respectively.

Taking as an example the C.P.S. specimens 158-160, where the burnup was 6×10^{20} fissions per cm^3 of fuel, the value of P is found to be 11,090 p.s.i. for a swelling rate of 0.16 per cent. The important component of the stress is the tensile component in the matrix, P_t, which acts tangentially at the particle boundary, and which will be additive to an externally applied stress, resulting in an apparent reduction in tensile strength. This is equal to P/2 and therefore has a value of 5500 p.s.i.

The highest burnup reached (specimen 272) was 18 x 10^{20} fissions per cm^3 which yields a value of 7900 p.s.i. for P_t .

The choice of 0.16 per cent. as the swelling rate may be conservative as there were no voids at the particle-matrix boundaries and the only free volume to be absorbed by the swelling particles was porosity within the particles. The imposed stresses may therefore have been higher than those estimated above. (If 0.7 per cent. per 10^{20} is chosen as the swelling rate the values of P_t are 240,000 and 346,000 p.s.i.). It is likely,then, that fuel particle swelling did contribute to the loss of strenth and may well be a very important factor at high burnups.

The importance of fission fragment recoil damage in BeO dispersions is not known at present. The fact that cracks appeared in the fuel particles of C.P.S. specimens indicated that drastic effects were produced in the particles themselves, but there was no evidence that the surrounding shell of BeO was severely damaged in any way. A count made of the fracture faces of specimen 272 showed that the fracture propagated across, rather than around, a significantly larger proportion of fuel particles after irradiation.

The effect of fuel particle size on fission fragment damage to the matrix is not straightforward, as the severity of the damage in the damaged regions decreases with decreasing particle size (White, Beard, and Willis 1957) whilst the volume fraction of matrix damaged increases. However, if we compare C.P.S. specimens 154 (coarse fuel) and 283 (fine fuel), we find that the fission fragment density in the damaged BeO is about twice as high in the fine dispersions as in the coarse and yet the strength of the fine was much greater.

Although this is a comparison of very few results it demonstrates that fission fragment damage to the matrix was not a dominant factor in causing the loss of strength on irradiation. The result may also highlight the importance of fuel particle swelling for a greater fraction of fission fragments escapes from fine fuel particles (95 per cent. from 10 μ particles) than from coarse (10 per cent. from 150 μ particles) and fine particles are therefore likely to swell less rapidly. A reasonably correct impression of fission fragment density in fuel particles should be given by comparing the fission density in the fuel of coarse dispersions (or 90 per cent. of it) to the average fission density for the whole specimen of the fine dispersion. On this basis, it will be seen that, in the C.P.S. specimens, the fission fragment density in the fuel of fine dispersions is lower than that in the coarse dispersions by a factor of from three to six.

This discussion leads to the inevitable conclusion that the fission density within the fuel particle should be the dominant parameter relating to the fall of strength, and the lack of a correlation in the results is disappointing.

It must be emphasised again that these experiments only indicate trends and should not be used for making quantitative deductions. The number of tests was very limited, and the materials were of various types, none of which was comparable to currently available fuel materials. An extensive series of investigations on the mechanical properties of irradiated fuels is at present in progress and should give quantitative data on the behaviour.

5. CONCLUSIONS

1. The strength of dispersions of $(U, Th)O_2$ particles in BeO is seriously reduced by irradiation in a thermal neutron flux which does not damage the BeO matrix.

2. Swelling of fuel particles can lead to severe stressing of the matrix and may be a major reason for loss of strength.

3. Damage of the matrix by fission fragment recoil does not appear to be a dominant factor in the loss of strength.

4. Dispersions containing fine fuel particles retain their strength as well as and possibly better than coarse dispersions of similar composition and burnup. This suggests that fission fragment damage to the matrix is not a serious problem.

5. More comprehensive experiments using modulus of rupture measurements are required before firm conclusions can be drawn on the mechanical properties of irradiated fuels.

6. ACKNOWLEDGEMENTS

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TABLE 1

SPECIMEN COMPOSITIONS, BURNUPS, AND STRENGTHS

Rig Number	Specimen Number	v/o $(U, Th)O_2$	U: Th Ratio	Fission Density		Irradiation Temperature (°C)	Pre-irradiation Strength		Post-irradiation Strength		Residual Strength (%)
				Fuel	Whole		Range (p.s.i.)	Mean (p.s.i.)	Range (p.s.i.)	Mean (p.s.i.)	
				x 10^{19}	x 10^{19}						
X-11	–	Pure BeO	–	–	–	–		28700 (5)			
	100	3	1:3	75	2	655	22000–22200	22100 (2)	2570–3470	2870 (4)	13
	101					655					
	107	4.4	1:3	80	3.5	675	16370–20880	19050	5680–7710	6690 (2)	35
	108					610			4190–6680	5430 (2)	28
	130	4.4	1:1	140	6	765	"	"	9920–14000	11700 (4)	61
	131					765					
	139	4.4	3.1	180	8	735	"	"	7875–10220	9270 (4)	49
	140					735					
	112	8,4	1:3	75	6.5	745	15900–18280	17080 (3)	4890–8420	6300 (4)	37
	113					745					
	118	15.7	1:3	65	10	695	12700–20250	16250 (4)	5620–8090	7090 (4)	44
	119					695					
	124	25.8	1:3	60	15	820	–	16000 *	880–2400	6720 (4)	42
	125					820					
	149 (fine $(U, Th)O_2$)	4.4	1:3	70	3	630	23000–36000	28850 (4)	12600–14050	13300 (4)	46
	150 (fine $(U, Th)O_2$)					600					
X71	–	Pure BeO	–	–	–	–	17550–20450	18900 (8)			
	154	5	1:3	65	3.3	610	12750–17400	15760 (4)	10300–11550	11000 (4)	70
	158	15	1:3	54	8.1	670	14730–20760	16200 (6)	6880–8860	7870 (2)	49
	264	15	1:1	94	14.0	720	13400–15320	14800 (6)	–	5200 (1)	35
	272	15	3:1	118	18	725	9280–12320	10700 (4)	4130–4780	4450 (2)	42
	279	30	1:3	46	14	730	10280–15100	12400 (3)	4840–7110	5970 (2)	48
	283 (fine $U, Th)O_2$)	15	3:1	110	17	720	12300–14250	13280 (2)	15900–23280	20700 (3)	(156)

* Value of 16,000 p.s.i. obtained by extrapolation.

Numbers in parenthesis are numbers of specimens tested.

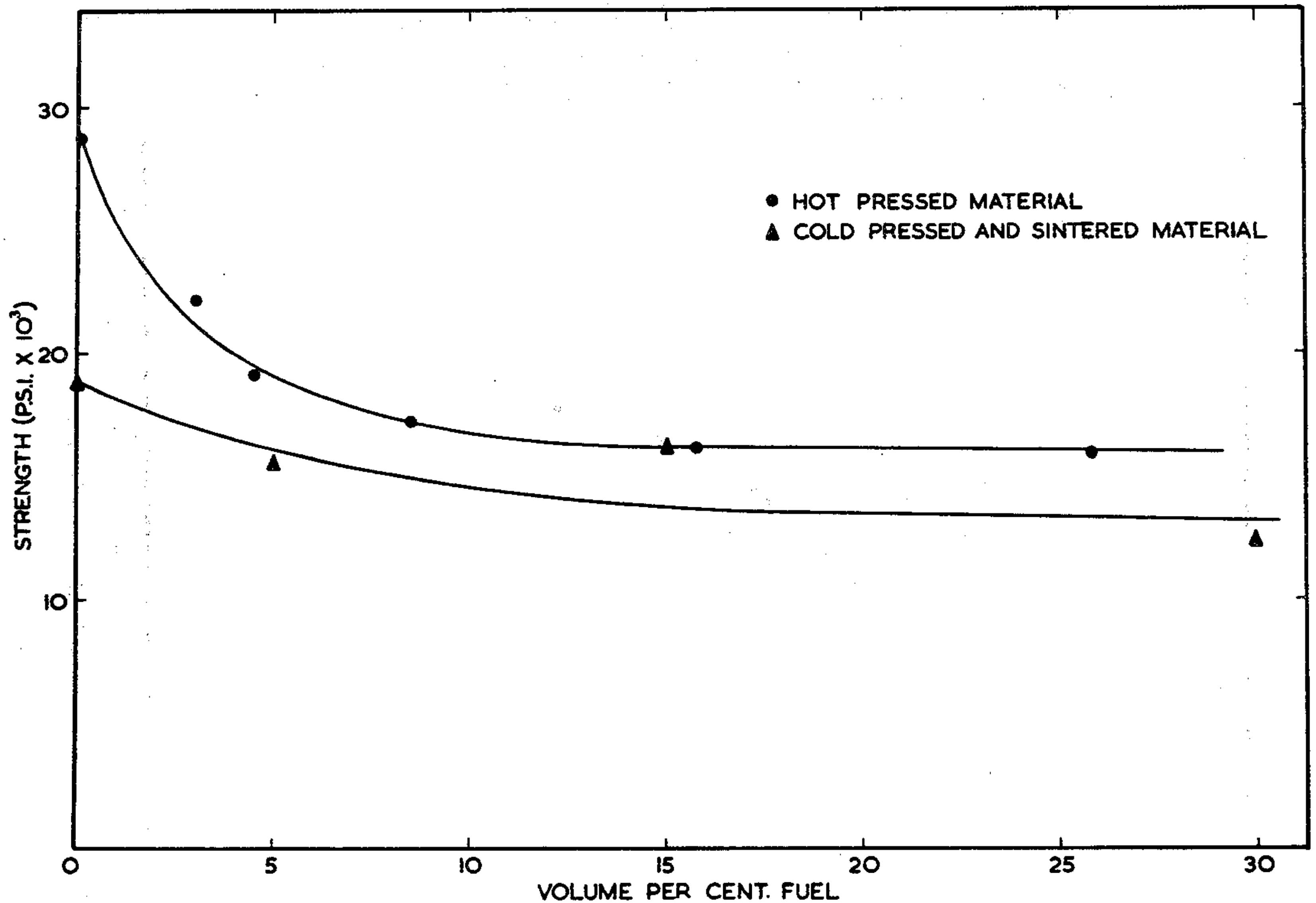

FIGURE I STRENGTH OF UNIRRADIATED FUEL DISPERSIONS

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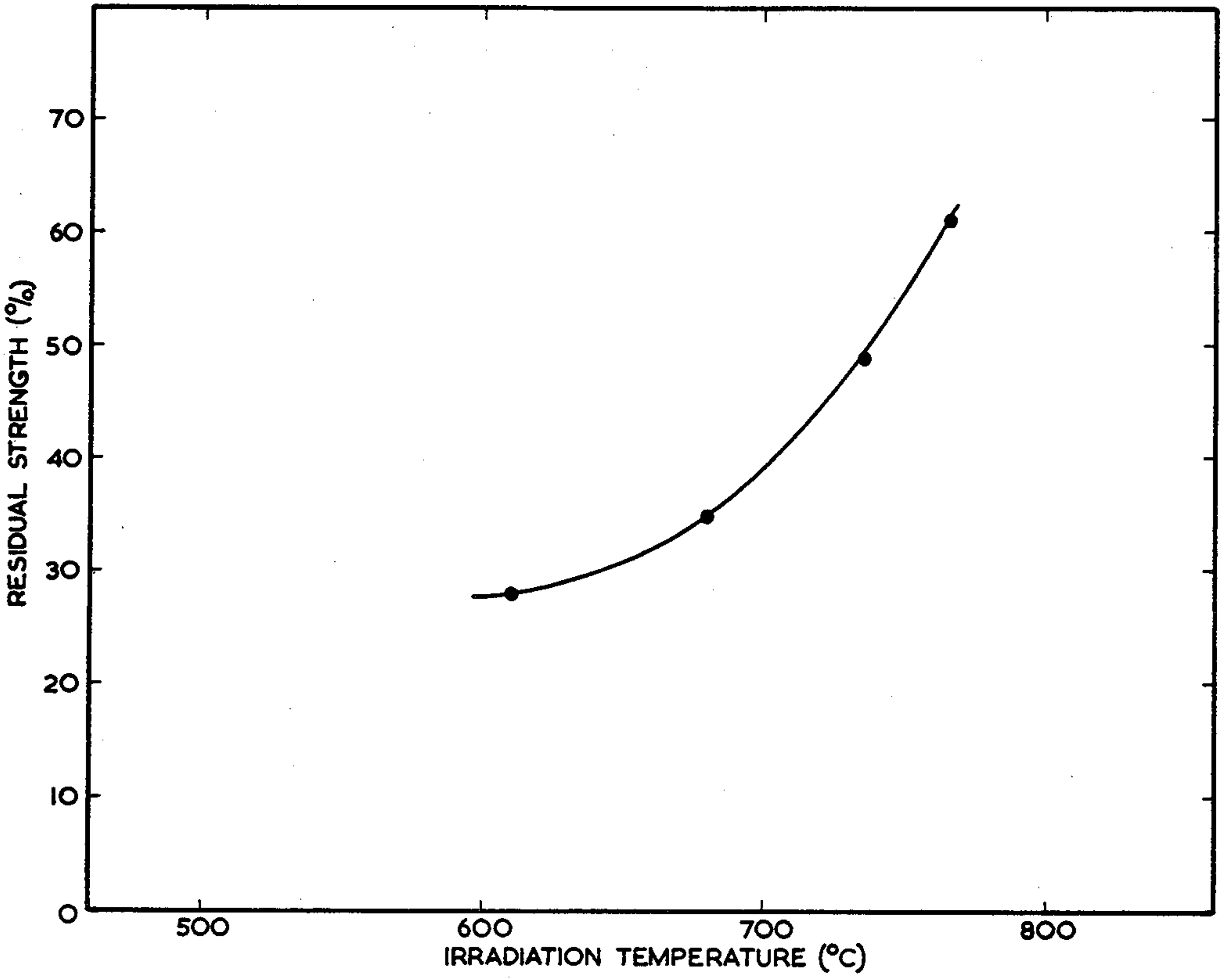

FIGURE 2 TEMPERATURE DEPENDENCE OF RESIDUAL STRENGTH AFTER IRRADIATION OF 4·4 VOLUME PER CENT. H.P. MATERIAL

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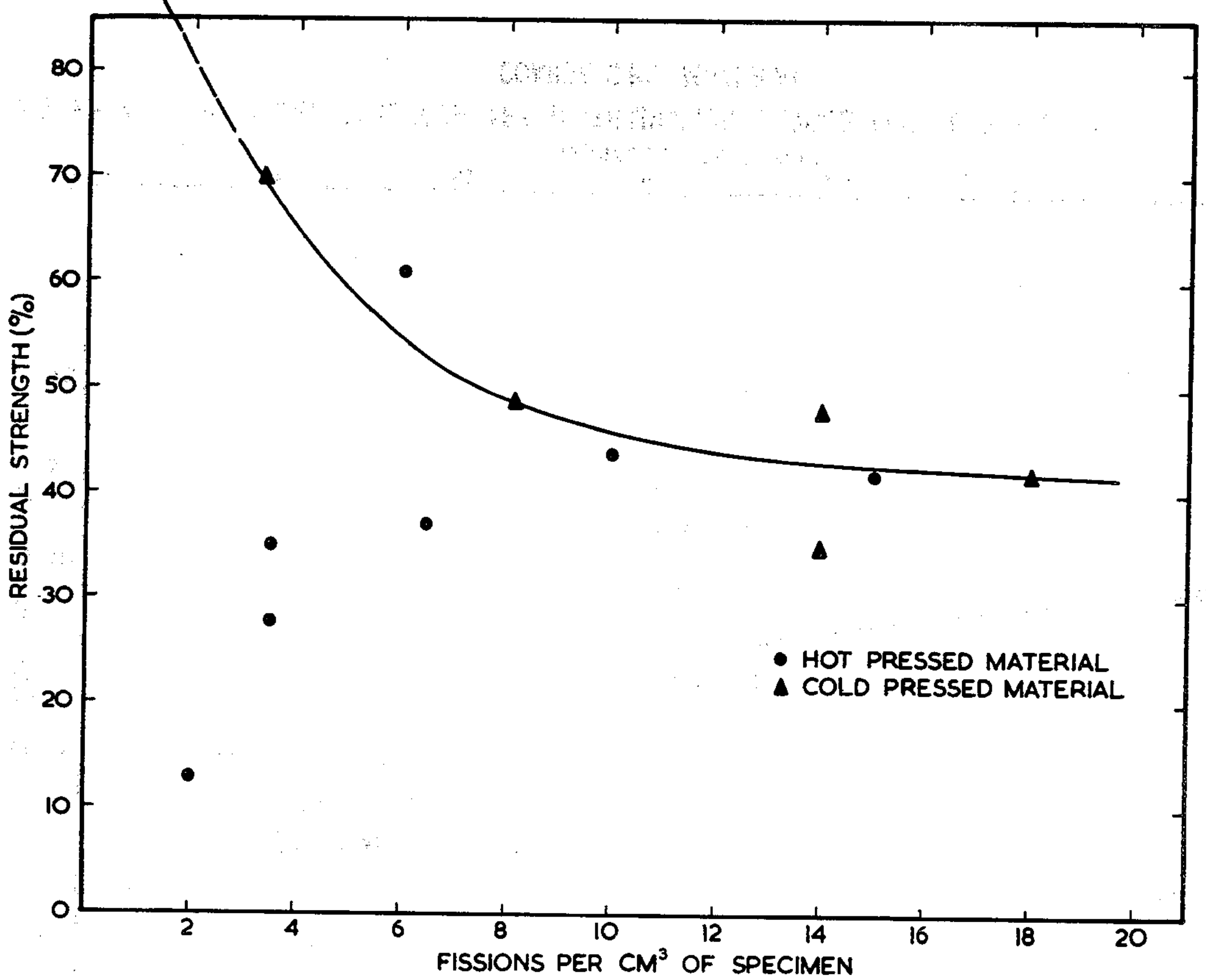

FIGURE 3 RESIDUAL STRENGTH AFTER IRRADIATION VERSUS FISSION DENSITY IN THE WHOLE SPECIMEN. COARSE DISPERSIONS ONLY

P814

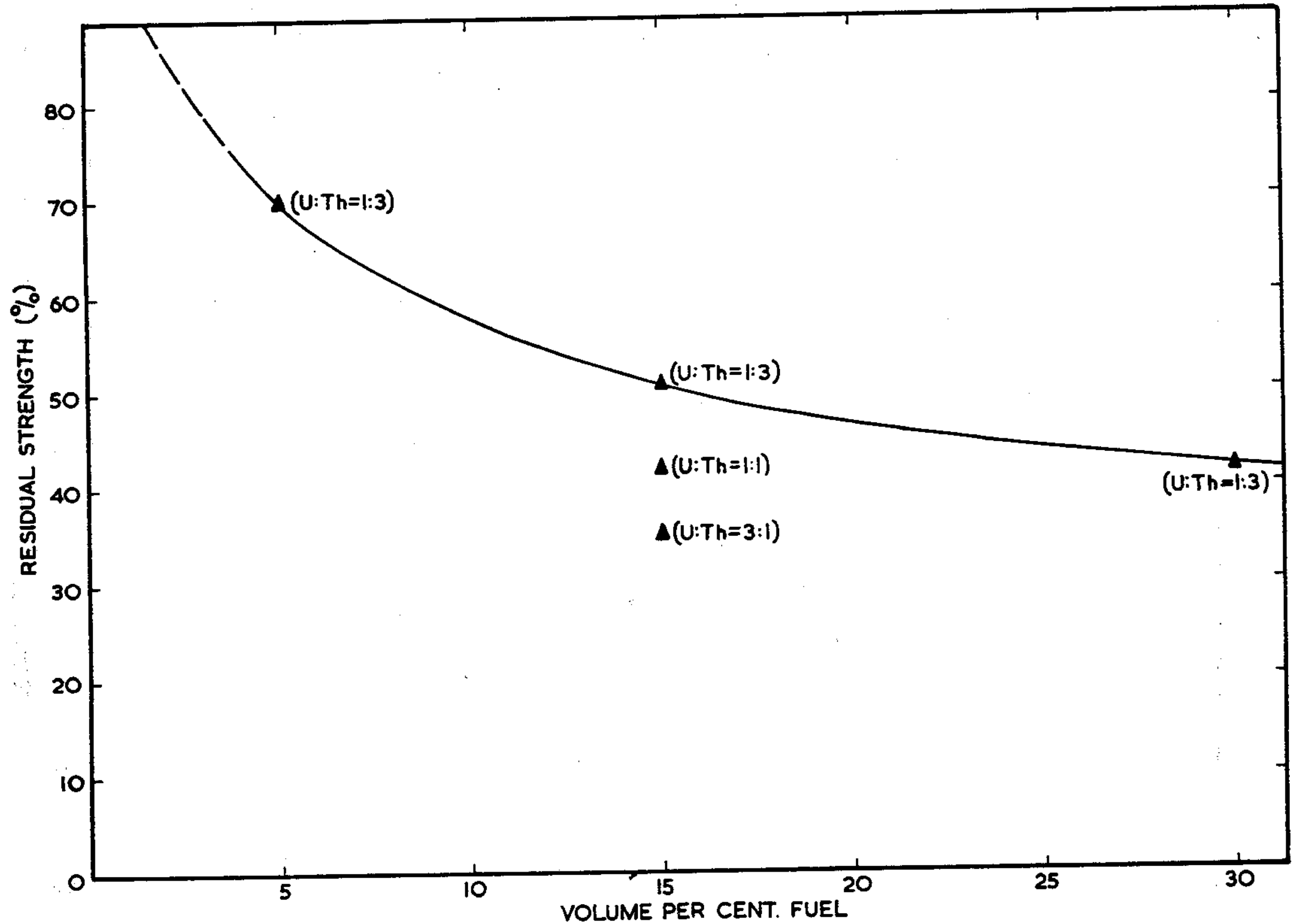

FIGURE 4 RESIDUAL STRENGTH AFTER IRRADIATION VERSUS FUEL CONTENT OF COARSE C.P.S. MATERIAL

PB14

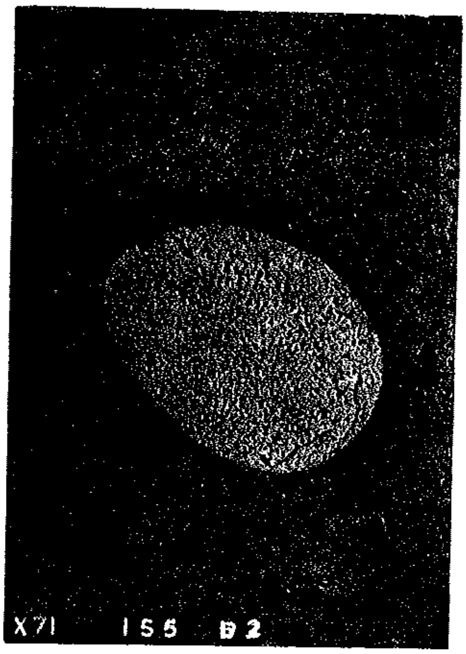

FIGURE 5 CRACKED FUEL PARTICLE TYPICAL
OF IRRADIATED C.P.S. MATERIAL