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LUCAS HEIGHTS

THE RELATIONSHIP BETWEEN MICROCRACKING AND MECHANICAL PROPERTIES IN NEUTRON-IRRADIATED BERYLLIUM OXIDE

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ABSTRACT

Beryllium oxide bend test specimens of two grain sizes, 1 – 2 microns and 8 – 35 microns, were irradiated to fast neutron doses of up to 5.6 x 10^{20} nvt (above 1 MeV) at 75 – 100 °C. Specimens were examined by X-ray line breadth (30.0 reflection), modulus of rupture, elastic modulus, open porosity, and lattice parameter measurements.

The results show that there is no significant change in mechanical properties up to the dose at which microcracking is first observed i.e. 3.5 x 10^{20} nvt in the case of the fine grain size material and 1.2 x 10^{20} nvt in the case of the coarse grain size material. Above these doses the modulus of rupture and the apparent elastic constants fall rapidly. Microcracking occurred at an earlier dose in both materials than would have been expected from earlier work.

CONTENTS

1. INTRODUCTION

The most serious effect of fast neutron irradiation on polycrystalline beryllium oxide is the microcracking which results from anisotropic expansion of the crystal lattice. It would be expected that a deterioration in mechanical strength would accompany microcracking but early results on hot pressed material (Hickman 1962) suggested that there would be little change in mechanical properties up to the dose at which microcracking is first observed. Recent work by the General Electric Company (1964) has confirmed that there is little significant change in properties up to the dose at which microcracking was thought to occur but that drastic loss of strength occurs above the dose. Clarke (1963) investigated the problem theoretically, and from strain energy considerations deduced a relation between mechanical strength and anisotropic growth which suggested that the anisotropic growth strains would add to the externally imposed strains and cause some loss of strength before microcracking actually occurred.

Previous work at this laboratory has shown relief of the broadening of the (h, k, o) reflections to be a very sensitive method for detecting the onset of microcracking (Hickman, Sabine, and Coyle 1962; Walker, Mayer, and Hickman 1963). The purpose of the work described in this report was to relate the changes in mechanical properties to the internal strain and onset of microcracking as measured by the X-ray technique as well as by volume and porosity changes and microstructural changes.

2. EXPERIMENTAL

2.1 Materials

Specimens of two grain sizes were prepared from Brush UOX beryllium oxide powder. Material with a mean grain size of 1 to 2 microns was prepared by isostatic pressing at 20 t.s.i. and sintering for one hour at 1525°C in nitrogen. Material with a larger grain size was prepared by die pressing with a P.V.A. binder and sintering for four hours at 1500°C in nitrogen followed by a second sintering under the same conditions. The grain size of this material was not uniform; there were regions having a mean grain size of about 8 microns and regions having a mean grain size of about 35 microns. The volumetric ratio of these regions varied in the specimens which were ultimately cut from the as-fabricated compacts.

Specimens in the form of bend test specimens measuring 1 inch by 0.24 inch by 0.16 inch were cut from the larger as-fabricated blocks. They were finished by surface grinding after which they were heated to 800°C for 4 hours to remove the cement used to hold them during grinding.

2.2 Techniques

Irradiation was done at 75 – 100°C in a hollow fuel element in HIFAR giving integrated fast neutron doses (above 1 MeV) of 1.2×10^{20}, 2.2×10^{20}, 3.5×10^{20} and 5.6×10^{20} nvt.

Bulk densities were calculated from specimen weights and dimensions (obtained with a vernier micrometer) and impregnated densities by hydrostatic weighings in n-octanol. From these measurements open porosities and their changes on irradiation were calculated.

Grain sizes were measured by the linear intercept method on polished and etched sections taken from broken control specimens. Optical microscopy was used for the coarse-grained material and replica electron microscopy for the fine-grained material.

Integral breadths of the (30.0) X-ray reflections were measured from recorded graphs obtained with a Philips diffractometer. As a further check on the behaviour of the material, and to enable comparison with previous work, lattice parameters were measured on crushed samples using powder photographs and a least squares refinement programme for the IBM 1620 computer (Walker 1963).

Moduli of rupture were measured in a four point bend test with an Instron Tensile Testing machine using a cross head speed of 0.05 inch per minute (strain rate of 6.7 per cent. per minute) and a 1000 pound load cell.

Strain measurements for the determination of the elastic modulus were obtained with strain gauges glued to the under-side of bend test specimens with an epoxy cement. Measurements were made on four specimens of each grain size in the unirradiated condition and two of each type at each irradiation level. Where possible the specimens were not broken in the test and stress - strain curves were plotted for both the loading and unloading cycles.

Metallographic evidence of microcracking was sought by examining selected specimens in the polished but unetched condition. Coarse-grained material was examined optically and the fine-grained material by the replication technique with an electron microscope.

3. RESULTS

Results are tabulated in Table 1 and presented graphically in Figures 1 to 5. For convenience, they are described separately for the fine- and coarse-grained materials.

3.1 Fine-Grained Material

The average modulus of rupture (see Figure 1) for unirradiated specimens was 35,000 p.s.i. Average values after neutron doses of 1.2×10^{20} nvt and 2.2×10^{20} nvt were 32,000 p.s.i. and 34,300 p.s.i. respectively with standard deviations of about 4,000 p.s.i. After a dose of 3.5×10^{20}nvt the modulus dropped to 18,000 p.s.i. and at 5.6×10^{20} nvt to 4,300 p.s.i.

The elastic modulus (Figure 2) showed no significant change from the unirradiated value of 52×10^6 p.s.i. (range, 48 to 54×10^6 p.s.i.) at doses up to 2.2×10^{20} nvt. At 3.5×10^{20} nvt the value dropped to 26×10^6 p.s.i. and at 5.6×10^{20} nvt to 8.6×10^6 p.s.i.

The breadth of the (30.0) reflection (Figure 3) increased with dose to 0.56 degree θ at 3.5×10^{20} nvt but at 5.6×10^{20} nvt was only 0.44 degree θ. However, the broadening at 3.5×10^{20} was some 18 per cent. lower than would be expected from extrapolation from the lower doses and, when the broadening is plotted against $\underset{\sim}{c}$ parameter change to remove the uncertainties associated with dose measurement, the broadening at 3.5×10^{20} is 25 per cent. lower than would be expected. This is a clear indication that some relief of strain by microcracking had occurred at this dose.

Open porosity did not change significantly at doses up to 2.2×10^{20} nvt but increased rapidly above this dose (Figure 5), again indicating that microcracking commenced at 3.5×10^{20}nvt. At 5.6×10^{20} nvt, the open porosity was 5 per cent. compared to the value of 0.5 per cent. in unirradiated material.

Metallographic examination failed to show microcracking in all specimens. Grain pull-out was evident at all doses and at 5.6×10^{20} nvt it was severe enough to prevent a useful examination.

3.2 Coarse-Grained Material

The average modulus of rupture in the unirradiated condition was 23,000 p.s.i. (with a standard deviation of 2,400 p.s.i.). The strength decreased with irradiation (Figure 1) to 17,700 p.s.i. (± 4,200 p.s.i.) at 1.2×10^{20} nvt, 10,400 p.s.i. (± 2,400 p.s.i.) at 2.2×10^{20} nvt, and 2,000 p.s.i. (± 700 p.s.i.) at 3.5×10^{20} nvt. Specimens irradiated to 5.6×10^{20} nvt crumbled on handling and could not be tested.

The elastic modulus fell sharply on irradiation (Figure 2), even at 1.2×10^{20} nvt. The unirradiated value was 51×10^6 p.s.i. and values of 27×10^6 p.s.i. at 1.2×10^{20}, 30×10^6 at 2.2×10^{20}, and 6.7×10^6 at 3.5×10^{20} nvt were measured after irradiation. Unfortunately, owing to the fragility of these specimens, measurements were obtained on one specimen only at doses of 1.2×10^{20} and 3.5×10^{20} nvt.

The breadth of the (30.0) reflection increased with neutron dose (Figure 3) to 0.22 degree θ at 2.2×10^{20} nvt but at 3.5×10^{20} nvt no broadening at all was evident. As is apparent from Figure 3 the rate of broadening was less than with the fine-grained material and even at 1.2×10^{20}nvt was considerably less than for the fine grain size material, indicating that microcracking had commenced at this dose.

Open porosity (Figure 5) increased by an amount of little or no significance at 1.2 x 10^{20} and 3.3 per cent. at 3.5 x 10^{20} nvt. Evidently insufficient microcracking had occurred at 1.2 x 10^{10} to affect the open porosity. Examination of the microstructure at 1.2 x 10^{20} nvt revealed slight microcracking in the regions of 35 micron grains but none in the 8 micron grain regions. Microcracking was noticeably greater at 2.2 x 10^{20} nvt and was very pronounced at 3.5 x 10^{20} nvt. The microcracking when present was predominantly intergranular with little or no transgranular cracking.

3.3 Lattice Parameter Measurements

The percentage changes in lattice parameters as functions of dose are plotted in Figure 4. The change in the $\underset{\sim}{a}$ parameter increased linearly with dose to 0.16 per cent. at 5.6 x 20^{20} nvt. The $\underset{\sim}{c}$ parameter showed the typical two-stage increase (gradual increase up to about 2 x 10^{20} nvt followed by a higher rate of increase at higher doses) and had increased by 2 per cent. at 5.6 x 10^{20} nvt.

4. DISCUSSION

The results confirm previous observations that microcracking causes severe loss of strength as measured by modulus of rupture in irradiated beryllium oxide (Hickman 1962; General Electric Co. 1964). In the coarse-grained material the modulus of rupture was reduced by 24 per cent. at the lowest dose at which microcracking was observed and in the fine-grained material the corresponding decrease was 48 per cent. In the fine-grained material there was no significant change in strength up to and including 2.2 x 10^{20} nvt, at which dose no microcracking could be observed by other techniques. No measurements were made on coarse-grained material in which no microcracking had occurred. It can be concluded that provided microcracking cannot be detected by X-ray or other techniques then the beryllium oxide will retain its unirradiated strength. It does not appear that the high degree of internal strain which exists in the material before microcracking causes any significant loss of strength. A small effect, however, could exist and be obscured by the scatter in results.

The apparent large decreases in elastic moduli in microcracked samples are not considered to be due to a real change in the elastic constants. The value of the elastic modulus as measured in the bend test is probably quite meaningless once the material has microcracked as the specimen, rather than experiencing a true elastic strain, probably separates at the cracks. This is supported by the fact that the strain in microcracked specimens did not fully recover on unloading; in both materials the permanent set (measured as the residual strain per pound of applied load) increased with increasing neutron dose, that is, with increasing degree of microcracking. However, it can be concluded from the results on fine-grained material that there is no change at all in the elastic modulus if no microcracking has occurred. This is supported by measurements of elastic moduli made by the General Electric Co. (1964) using a resonant frequency technique. They observed no change up to the doses at which microcracking occurred. Above this dose they could not obtain reproducible results.

A disturbing feature of the results is the low dose at which microcracking occurred in both materials when compared with previous results on cold pressed and sintered material (Hickman and Pryor 1963). On the basis of previous work and assuming an inverse square root relation between grain size and the dose for the onset of microcracking (Clarke 1963), microcracking would not have been expected in the coarse-grained material until ≈ 2 x 10^{20} nvt and in the fine-grained material until ≈ 10^{21} nvt whereas it was actually observed at one half to one third of these doses. The present results show microcracking at doses very similar to that which would be predicted from the previously observed behaviour of hot pressed material (Hickman and Pryor 1963). The only known difference between the present material and the C.P. and S. material investigated previously is that the powder used for the production of the latter material was subjected to an homogenizing treatment before pressing which involved grinding with alumina balls in a porcelain mill. Microcracking observed in the earlier C.P. and S. material was predominantly transgranular whereas the present material and the hot pressed material investigated previously cracked almost entirely intergranularly. Evidently the grain boundaries were stronger in the early C.P. and S. material and this may be associated with impurity pick-up during the homogenizing operation. However any mechanism by which this could occur is not at all clear and the problem is being investigated further. Fine grain size material still performed better than coarse-grained material but it appears that although fine grain size is a necessary condition for good irradiation stability other factors are also important.

One further feature of the present results is that the c parameter change at a given dose is less than the value quoted by Walker et al. (1963), as can be seen from Figure 4. The only explanation which can be offered for this is that the measured doses were incorrect and higher than the actual doses. The same monitoring techniques were used in both series of experiments and it is difficult to see how the discrepancy has arisen.

5. CONCLUSIONS

1. The modulus of rupture and elastic modulus of C.P. and S. beryllium oxide are substantially unaffected by neutron irradiation in the absence of microcracking.

2. When microcracking is first detected by X-ray techniques severe decreases in modulus of rupture occur. An apparent decrease in elastic modulus also occurs at this stage but this is not thought to be a real effect.

3. The material used in this investigation microcracked at a lower dose than would have been expected from previous data. This was associated with a change from predominantly transgranular cracking in the previous material to intergranular cracking in the present material and hence is associated with weaker grain boundaries.

6. ACKNOWLEDGMENTS

The assistance of R. J. Hemphill (density and dimension measurements), K. G. Watson (lattice parameter measurements), R. G. Blake (electron microscopy), and J. G. Flanigan (metallographic preparation and grain size measurements) is gratefully acknowledged.

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TABLE 1

MECHANICAL PROPERTIES OF NEUTRON-IRRADIATED BERYLLIUM OXIDE

Mean Grain Size (microns)	Neutron Dose (nvt > 1MeV)	Broadening of 30.0 Reflection (°θ)	Lattice * Expansion		Modulus of Rupture		Youngs Modulus x 10^6 p.s.i.	Open Porosity %		Metallographic Observations
			$\Delta a/a$ %	$\Delta c/c$ %	No. of Specimens	Modulus of Rupture (p.s.i.) and r.m.s. Deviation		Before	After	
1 – 2	Nil	–			7	35,000 ± 3,600	51.9$^{\neq}$			
	1.2	0.16			7	32,500 ± 4,600	48.1	0.3	0.04	Grain pull–out.
		0.16								
	2.2	0.39			7	34,300 ± 4,200	53.7		0.01	Grain pull–out.
		0.39								
	3.5	0.53			5	18,000 ± 2,500	25.9	0.4	1.5	Severe grain pull–out.
		0.59								
	5.6	0.48			7	4,280 ± 450	8.6	0.4	5.0	Pronounced grain pull–out.
		0.40								
8 – 35	Nil	–			16	23,000 ± 2,400	50.7$^{\neq}$			
	1.2	0.13	0.041	0.235	4	17,700 ± 4,200	27.3‡	0.6	0.5	Grain pull–out. Some micro-cracking in regions of 30μ grain size.
		0.11								
	2.2	0.21	0.063	0.482	6	10,400 ± 2,400	30.0		2.3	Microcracking and some grain pull–out.
		0.23								
	3.5	Nil	0.100	0.991	6	2,000 ± 700	6.7‡	0.5	3.3	Severe microcracking.
	5.6	–	0.163	1.97	Specimens powdered during irradiation			Not measured		

* Specimens Crushed.

$\neq$ Mean of 4 specimens)
) Other results are mean of two.
$\ddagger$ One specimen only)

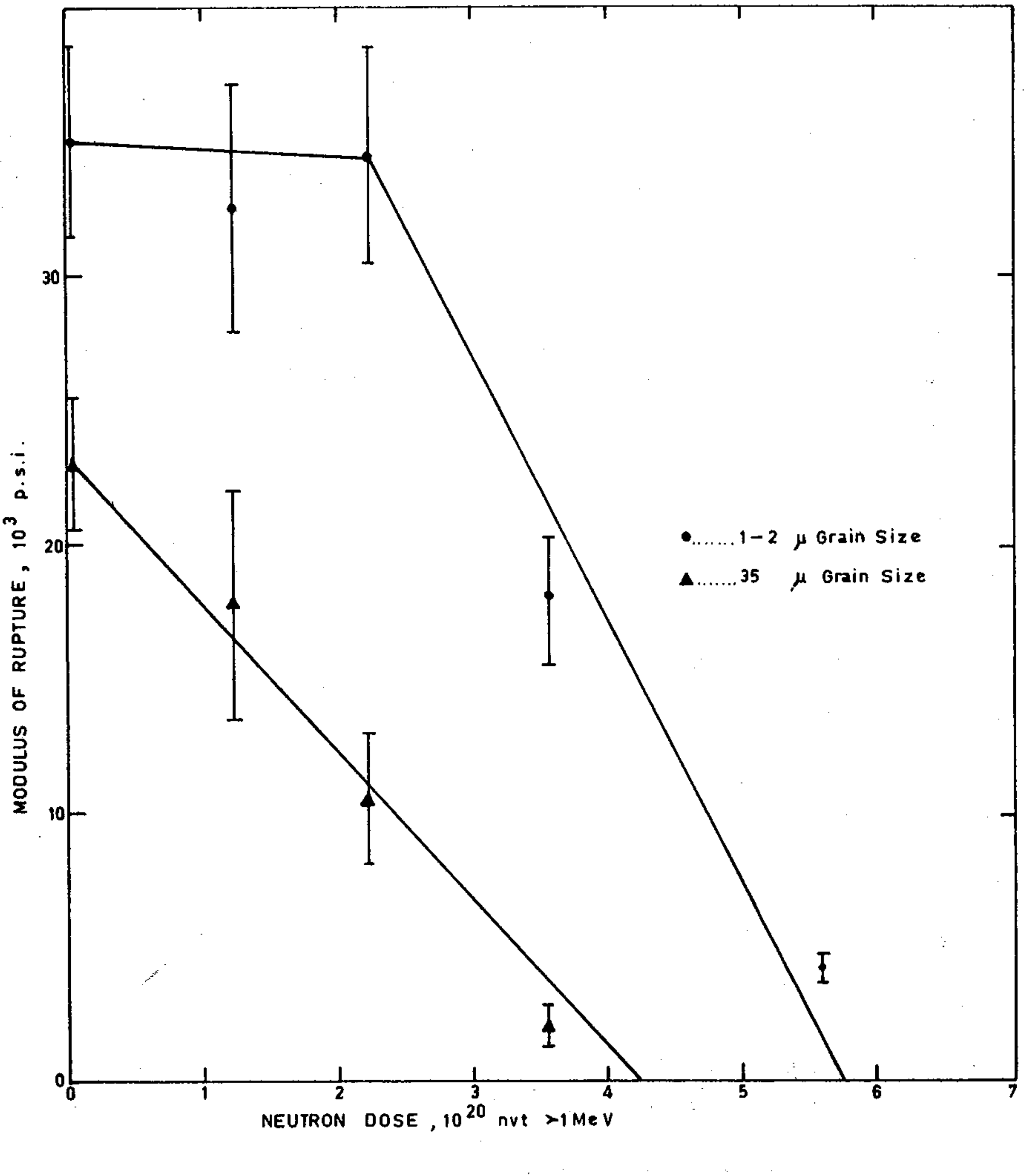

FIGURE 1. MODULUS OF RUPTURE

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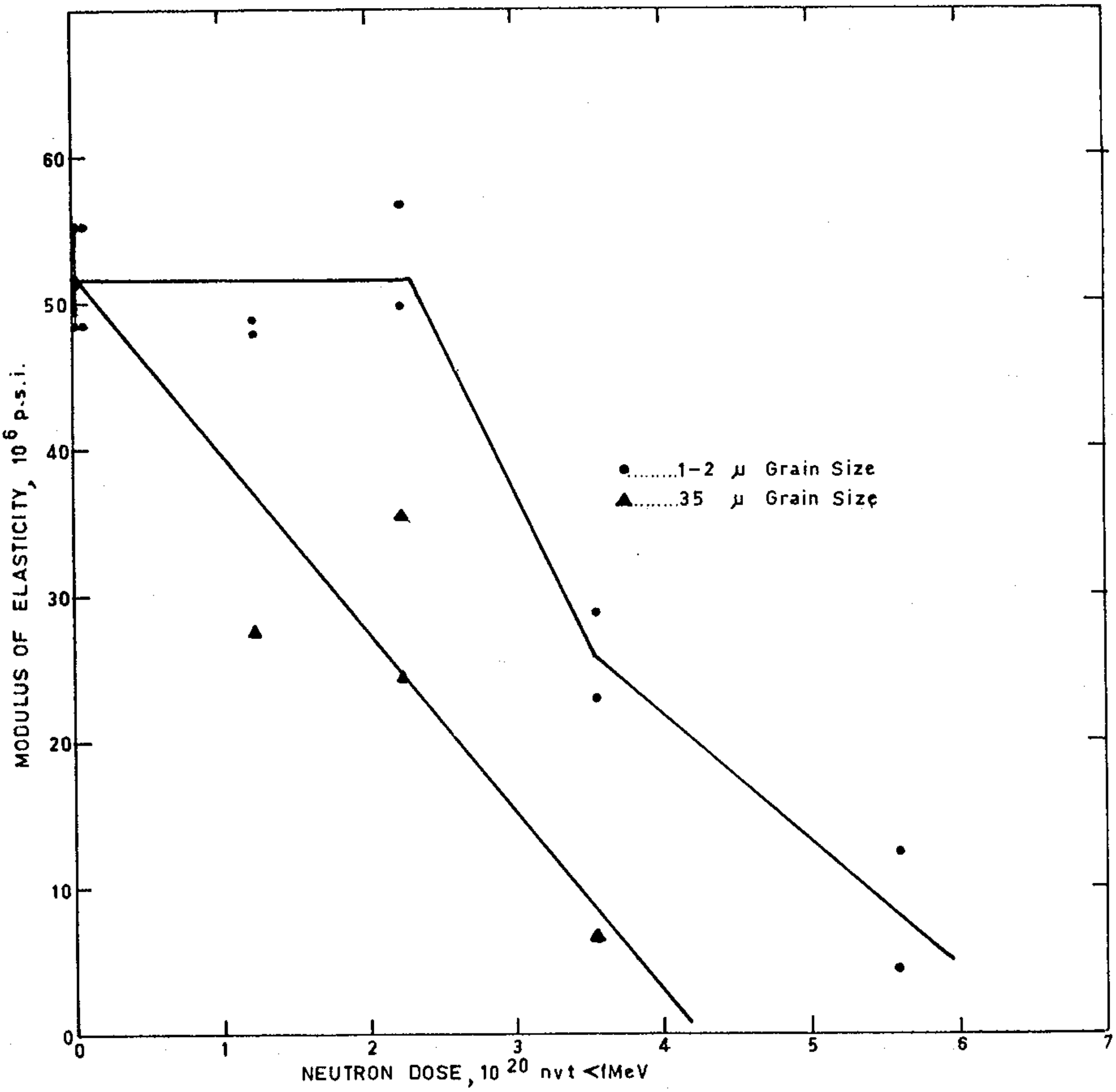

FIGURE 2. MODULUS OF ELASTICITY

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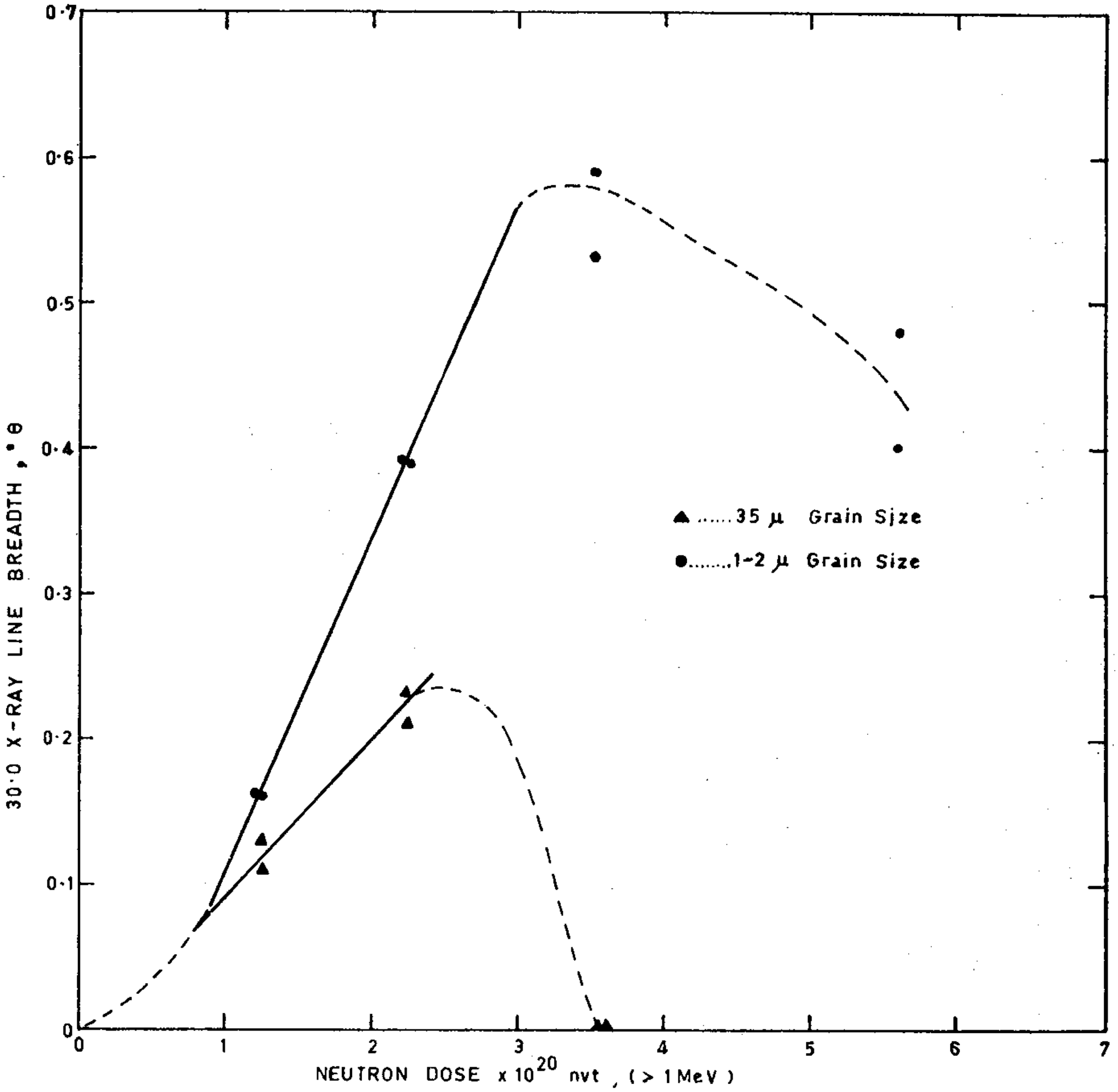

FIGURE 3 30·0 X – RAY LINE BREADTH

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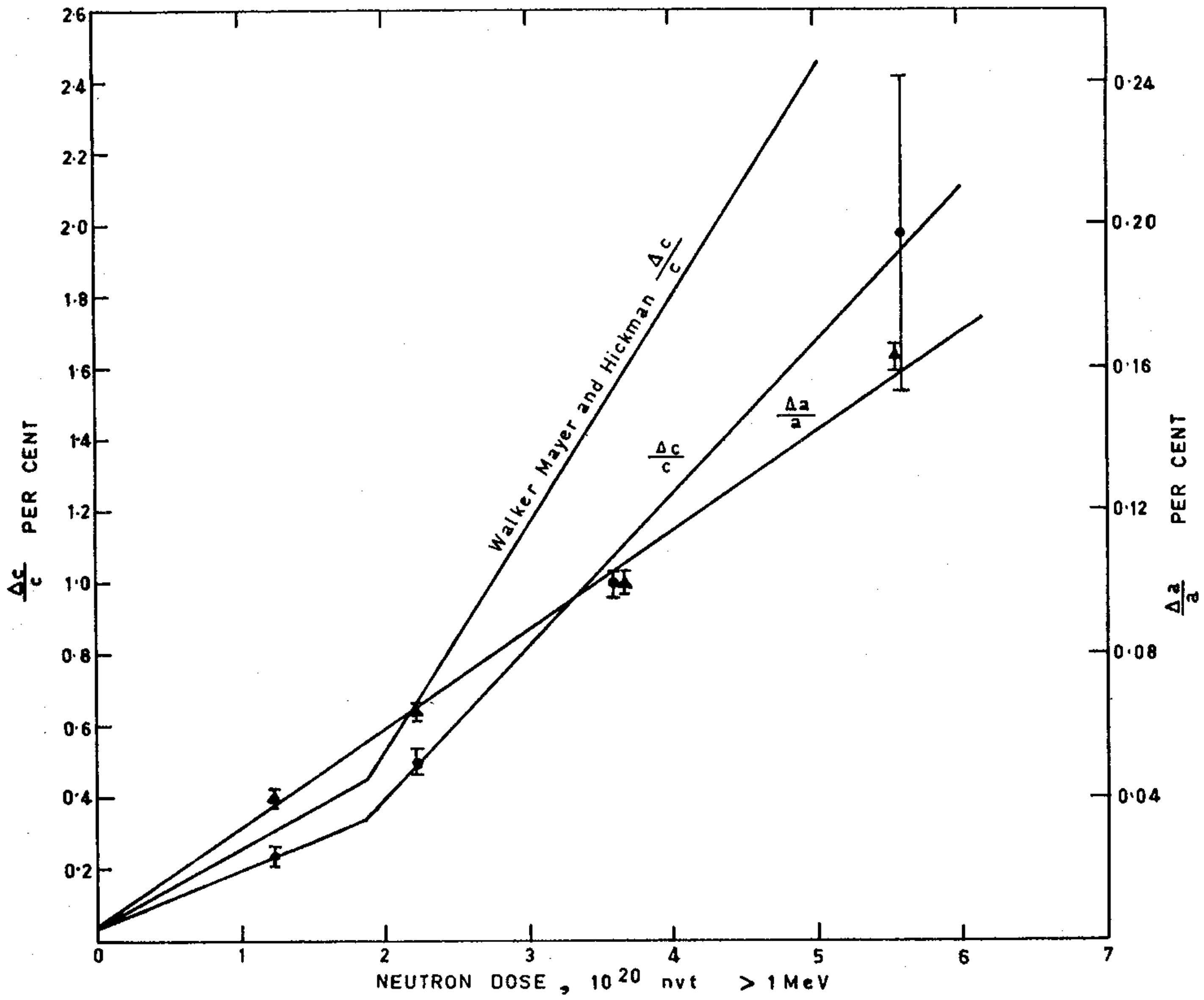

FIGURE 4. LATTICE PARAMETER

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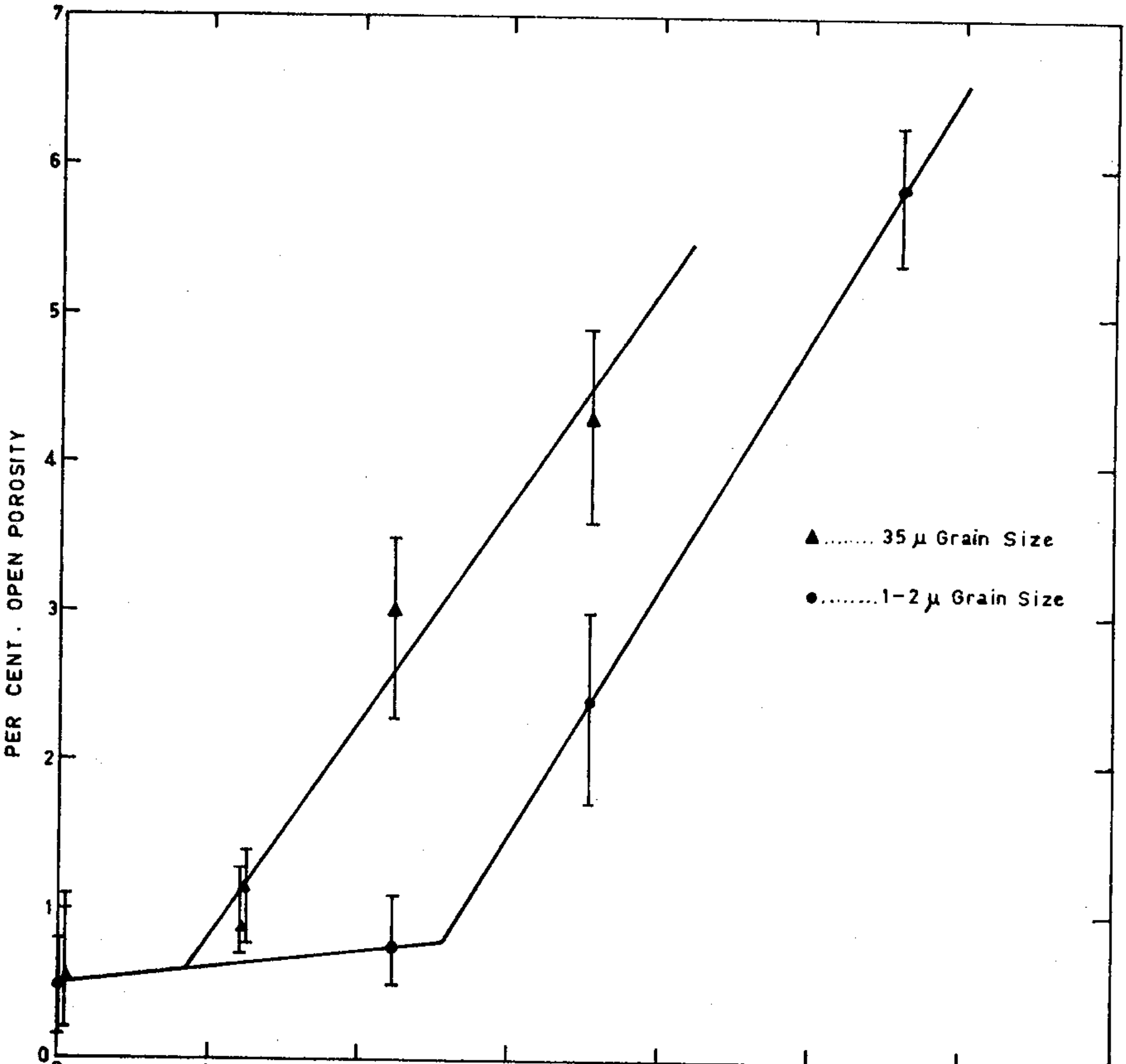

FIGURE 5. OPEN POROSITY

P641