AAEC/E133

AAEC/E 133

AUSTRALIAN ATOMIC ENERGY COMMISSION
RESEARCH ESTABLISHMENT
LUCAS HEIGHTS

EFFECT OF IRRADIATION ON THE MECHANICAL PROPERTIES OF BERYLLIUM METAL

by

G. T. STEVENS

B. S. HICKMAN

Issued Sydney, January 1965

AUSTRALIAN ATOMIC ENERGY COMMISSION

RESEARCH ESTABLISHMENT

LUCAS HEIGHTS

EFFECT OF IRRADIATION ON THE MECHANICAL PROPERTIES OF BERYLLIUM METAL

by

G. T. STEVENS

B. S. HICKMAN

ABSTRACT

Results are presented of mechanical property measurements on beryllium metal irradiated up to 6 x 10^{20} nvt > 1 MeV at 75 – 100 °C and 3.2 x 10^{20} nvt at 550 °C. The property changes are interpreted in terms of helium distribution in the material.

CONTENTS

1. INTRODUCTION

Beryllium metal has potential applications as moderator, canning material, and fuel matrix in nuclear reactors. At Lucas Heights, a programme on the effects of neutron irradiation on the properties of the material has been carried out. This work was done on material fabricated at Lucas Heights and the results have been summarised by Hickman and Stevens (1963). In addition work aimed at assessing the suitability of beryllium as a canning material for the Advanced Gas Cooled Reactor was carried out under contract to the U.K.A.E.A. (Hickman et al. 1962; Hickman and Bannister 1963). With the loss of interest in beryllium metal of both the A.A.E.C. and the U.K.A.E.A. these programmes were terminated. However some work was continued, extending the previous measurements to higher doses and different materials. The work reported here is concerned primarily with mechanical property studies.

2. EXPERIMENTAL METHODS

2.1 Materials

Beryllium fabricated by various routes was used as follows:

(i) Material A

Pechiney powder was vacuum hot pressed at 1050 – 1100 °C at 1500 p.s.i. and then extruded at 1050 °C with a 16:1 reduction ratio.

(ii) Material B

Pechiney powder was pre-oxidized by heating in an oxygen atmosphere in a fluidized bed at 775 °C to give an oxygen content of 1.5 – 2.0 per cent. The oxidized powder was then fabricated in a similar fashion to material A.

(iii) Material C

Pechiney powder containing an alloying addition of 0.4 per cent. calcium was vacuum hot pressed at 1030 °C, hot extruded at 1050 °C, and then warm extruded at 750 °C.

(iv) Material D

Pechiney powder was hot pressed at 1050 °C, hot extruded at 1050 °C, and warm extruded at 800 °C.

Materials A and B were fabricated at Lucas Heights, and materials C and D were fabricated by the U.K.A.E.A. Springfields Laboratories. Typical analyses are given in Table 1.

Materials B and C were included in the programme because other work had shown that these materials exhibited superior corrosion resistance to that of standard materials in moist CO_2 at high temperatures.

The materials were all irradiated as tensile specimens, materials A and B with a 0.2-inch gauge diameter and materials C and D with an 0.126-inch gauge diameter. Materials A and B were etched in a chromic-sulphuric-phosphoric acid solution before irradiation.

2.2 Irradiation Techniques

Materials A and B were irradiated to doses in the range 3 to 6 x 10^{20} nvt > 1 MeV at pile temperatures (75 – 100 °C) in the manner described by Hickman and Stevens (1963).

Materials C and D were irradiated to doses of 2.2 to 3.2 x 10^{20} nvt > 1 MeV at elevated temperatures (550 °C, 600 °C, and 700 °C) in a rig (X-81) almost identical with that described by Hickman et al. (1962).

Flux monitoring techniques were as described by Hickman and Stevens (1962). The doses quoted throughout this report are integrated doses of neutrons with energies greater than 1 MeV.

2.3 Testing Procedure

The specimens were all tested in air on a 10,000 lb Instrom machine using a cross head speed of 0.002in./min. The extension was measured from the reassembled specimen after fracture. Three groups of materials C and D were tested, namely as received, as irradiated, and heat treated specimens. The last group had been subjected to the same thermal history as was received by the irradiated specimens.

Electron microscope examinations of fracture surfaces were made using replica techniques.

3. RESULTS

3.1 Pile Temperature Irradiation

Material A was tested in the temperature range 25 – 700 °C after irradiation to 3.4×10^{20} and 6.0×10^{20} and the results are shown in Table 2. Changes observed were in general similar to but more marked than those reported by Hickman and Stevens (1963) for lower doses; at low testing temperatures there was an increase in yield strength and a decrease in ductility, with the changes becoming less marked as the testing temperature was raised. At both doses there was no measurable ductility at room temperature. After 3.4×10^{20} some ductility was observable at 200 °C but in the high dose material, ductility was only significant at temperatures of 500 °C and above. The increases in yield strength at room temperature, together with data reported earlier, are shown in Figure 1. A steady increase in yield strength is observed up to the highest dose measured.

The fracture characteristics showed normal behaviour. In the unirradiated material the fracture was predominantly cleavage at room temperature with increasing amounts of ductile shear failure as the temperature was raised. The change–over to intergranular failure occurred at 500 °C. In the material irradiated to 3.1×10^{20} nvt there was a smaller proportion of shear failure and at the higher dose (6.0×10^{20} nvt) failure below 500 °C was completely by cleavage. The change–over to intergranular failure occurred at the same temperature in all cases.

Material B was tested in the range 25 – 700 °C after irradiation to 3.1×10^{20} nvt. Although this material exhibited a higher strength than material in the as–received condition the changes due to irradiation were similar in both cases.

The recovery of irradiation damage in material A was investigated by annealing samples irradiated to 3.4×10^{20} and 6.0×10^{20} for one hour at successively higher temperatures and then testing at room temperature.

After 3.4×10^{20} nvt significant recovery of the yield strength and ductility was not observed until annealing temperatures of 700 °C, when a sharp change occurred. Recovery was not complete at 1000 °C. After 6.0×10^{20} nvt an annealing temperature of 800 – 900 °C was required for any observable recovery of the properties. The recovery is illustrated in Figure 2. Fracture in all cases was 100 per cent. cleavage. Sharp yield point effects were observed in most of the irradiated samples.

3.2 Elevated Temperature Irradiation

Considerable oxidation had occurred in specimens of both materials C and D at 600 °C and 700 °C and meaningful results could not be obtained from these specimens. The 0.4 per cent. calcium addition did not appear to have given any marked improvement in oxidation resistance under the particular conditions encountered in this test. The specimens irradiated at 550 °C were tested over the temperature range 25 – 700 °C. The results from samples of materials C and D irradiated at 550 °C, and the as–received and heat treated control samples, are given in Table 3. Comparison of the results from heat treated and as–irradiated specimens shows that in material C irradiation caused very little change in properties apart from some loss of ductility and increase in yield strength at testing temperatures of 200 ° C and below. Changes

at testing temperatures of 500°C and above were negligible. Material D on the other hand showed some loss of ductility at most testing temperatures, although considerable ductility still remained. The variation in ductility with testing temperature is illustrated in Figures 3 and 4.

Replica electron microscope examinations were carried out on samples of material C and D irradiated at 550°C and 600°C. After irradiation at 550°C, material C showed very fine helium bubbles 100 – 150 Å in diameter on some grain surfaces. After irradiation at 600°C material C again showed a very fine uniform helium bubble distribution at grain surfaces with most bubbles 100 – 200 Å in diameter. In material D the helium bubbles were somewhat coarser than in material C. After irradiation at 550°C they ranged from 100 – 250 Å and at 600°C from 200 – 750 Å. Typical electron micrographs are shown in Figure 5.

4. DISCUSSION

The results on material irradiated at pile temperatures are largely as expected from earlier work at lower doses and from hardness measurements (Hickman and Stevens 1963). The increased yield strength and loss of ductility is almost certainly due to solid solution hardening by helium. As the testing temperature is raised the effect is reduced by agglomeration of the helium into bubbles, and also by thermal activation, which reduces the effectiveness of the helium as a barrier to dislocation movement. The recovery of the room temperature properties with annealing is a consequence of helium bubble formation. The observation of sharp yield phenomena, also seen previously, is presumably a consequence of nucleation of helium bubbles at dislocations, which causes locking.

The behaviour of the materials fabricated in the U.K. is of particular interest when compared with the behaviour of nominally similar materials irradiated previously to similar doses (Hickman et al. 1963; Hickman and Bannister 1963). In the earlier experiments considerable loss of high temperature ductility occurred which has been attributed (Hickman et al. 1963; Hyam and Sumner 1962) to helium bubble formation at grain boundaries, which promotes grain boundary cavitation during deformation. The present material C showed no significant loss of high temperature ductility and the loss of ductility in material D was much less than that observed previously. The reason for this behaviour becomes obvious when the observed helium bubble sizes in the two experiments are compared. In the present experiments after irradiation at 550°C the bubbles in material C were very small (100 – 150 Å), but at the same temperature in the previous experiments the bubbles were generally 300 – 1000 Å in diameter and ranged up to 10,000 Å in some cases. In material C therefore the bubbles do not appear to have reached the necessary critical size to assist grain boundary cavitation. The bubbles in material D were somewhat coarser than in material C and resulted in some loss of ductility.

It has previously been suggested that the scale of bubble nucleation in beryllium metal is controlled by impurity precipitates (Hickman and Chute 1963). More recently Barnes and Mazey (1963) have demonstrated that helium bubbles in copper can migrate as bubbles and coalesce. It appears now that the role of the precipitate may not necessarily be to control the nucleation stage but rather to capture and trap moving bubbles and prevent coalescence and hence growth. Whichever mechanism is operative the actual number, and hence size, of the helium bubbles will depend on the scale of the impurity precipitation. Although material D in the present investigation was nominally identical with the previous material, small differences in heat treatment history are known to produce different impurity precipitate distributions, particularly of iron, and this has evidently occurred in this case. The superior behaviour of the 0.4 per cent. calcium alloy may be due to the calcium distribution controlling the bubble distribution or again to different iron distributions. Insufficient is known about the distribution of the calcium to determine this point.

5. ACKNOWLEDGMENTS

The irradiation of the U.K.A.E.A. specimens was planned and initiated by Mr. J.C. Bell of the U.K.A.E.A. Reactor Group whilst attached to Lucas Heights. Acknowledgment is due to the Fuel Element Development Section for the provision of specimens of materials A and B, to the Rig Group and Hot Cells for provision and handling of the irradiation rigs, and to Mr. R. Blake for the electron microscopy.

6. REFERENCES

Barnes, R.S., and Mazey, D.J. (1963). – A.E.R.E. R-4223.

Hickman, B.S., and Bannister, G. (1963). – AAEC/E115.

Hickman, B.S., and Chute, J. (1963). – J. Aust. Inst. Metals 8:2.

Hickman, B.S., and Stevens, G. (1963). – AAEC/E109.

Hickman, B.S., Bannister, G., Chute, J.H., McCracken, J.G., Smith, R., and Bell, J.C. (1963). – TRG Report 540 (S/X).

Hyam, E.D., and Sumner, G. (1962). – Radiation Damage in Solids, p.323. I.A.E.A., Vienna.

TABLE 1

DETAILS OF MATERIALS USED IN THE INVESTIGATION

Material	Fabrication Method	Density (g/cm^3)	Analysis								
			Fe (p.p.m.)	Al (p.p.m.)	Si (p.p.m.)	Ni (p.p.m.)	N (p.p.m.)	Cl (p.p.m.)	C (p.p.m.)	Ca (%)	O (%)
A.A.E.C. Material A	Vacuum hot pressed at 1050–1100 °C at 1500 p.s.i. Hot extruded at 1050 °C at 16:1 or 14:1 reduction ratio	1.846 to 1.854	220 to 700	450 to 600	100 to 200	ND *	33 to 160	ND	180 to 360	ND	0.7 to 0.8
A.A.E.C. Material B	Powder oxidized in fluidized bed for 1 hour at 775 °C Vacuum hot pressed at 1080 °C and 800 p.s.i. Hot extruded at 1050 °C at 30:1 reduction ratio	1.855 to 1.867	180	570	190	ND	33	ND	ND	ND	1.82
U.K.A.E.A. Material C	Vacuum hot pressed at 1030 °C Hot extruded at 1050 °C at 13:1 reduction ratio Warm extruded at 750 °C at 9.8:1 reduction ratio	1.85 to 1.86	765	295	160	175	ND	> 20	675	0.36	0.32
U.K.A.E.A. Material D	Vacuum hot pressed at 1050 °C Hot extruded at 1050 °C at 13:1 reduction ratio Warm extruded at 800 °C at 9.8:1 reduction ratio Stress relieved at 750 °C	1.85 to 1.86	480	380	135	155	ND	ND	415	ND	0.50

* ND – Not Determined

TABLE 2

RESULTS OF MECHANICAL PROPERTY MEASUREMENTS ON MATERIALS A AND B IRRADIATED AT 75 – 100°C AND TESTED AT 20–700°C

Material	Test Temp. (°C)	U.T.S. (p.s.i.)			0.1% Proof Stress (p.s.i.)			Elongation (%)			Reduction in Area (%)			Fracture *		
		Unirrad.	3.1×10^{20}	6×10^{20}	Unirrad.	3.1×10^{20}	6×10^{20}	Unirrad.	3.1×10^{20}	6×10^{20}	Unirrad.	3.1×10^{20}	6×10^{20}	Unirrad.	3.1×10^{20}	6×10^{20}
A	Room	91,000	68,500	79,600	43,000	61,200	78,700	14.3	0.7	0.2	13.0	0.0	0.0	C	C	C
	100	77,600	80,100	71,300	42,300	55,500	70,700	29.7	1.8	0.2	26.4	2.0	0.3	C–95% S–5%	C	C
	200	60,100	65,700	70,400	35,700	45,100	62,100	30.4	10.9	0.9	37.8	26.0	0.9	C–95% S–10%	C	C
	300	48,500	61,100	63,200	30,100	45,200	56,800	25.1	15.6	0.7	39.9	24.7	0.6	C–60% S–40%	C–90% S–10%	C
	400	37,700	48,200	53,900	27,000	42,900	52,800	14.7	9.4	0.7	33.7	1.5	0.4	C–5% S–95%	C	C
	500	20,600	28,700	31,200	18,900	27,200	27,900	9.8	5.2	2.8	8.0	2.8	2.0	I	I	C +I
	600	7,650	20,200	10,900	7,270	19,400	10,400	13.7	4.7	6.1	8.3	4.8	3.7	I	I	I
	700	3,790	–	4,720	3,660	–	4,560	16.9	7.3	6.1	10.9	–	2.9	I	I	I
B	Room	59,200	86,100	–	54,700	74,600	–	0.2	0.8	–	0.0	1.6	–	C	C	–
	100	–	86,900	–	–	70,800	–	–	2.2	–	–	1.5	–	–	C	–
	200	73,100	87,400	–	47,800	64,200	–	22.0	6.0	–	27.7	6.3	–	C–90% S–10	C–95% S–5%	–
	300	55,900	74,000	–	43,000	56,500	–	29.6	7.9	–	33.9	11.8	–	C–50% S–50%	C–90% S–10%	–
	400	43,200	57,500	–	35,900	50,800	–	13.0	7.4	–	22.6	14.5	–	C–5% S–90%	C–50% S–50%	–
	500	27,900	34,700	–	26,000	33,600	–	9.0	1.9	–	5.6	1.5	–	I	C–50% I–50%	–
	600	14,600	20,700	–	13,400	20,300	–	4.4	2.7	–	1.1	1.7	–	I	I	–

* C = cleavage
S = shear
I = intergranular

TABLE 3

RESULTS OF MECHANICAL PROPERTY MEASUREMENTS IN THE RANGE 20 – 700°C ON MATERIALS C AND D IN THE FOLLOWING CONDITIONS (i) AS RECEIVED (ii) HEAT TREATED (iii) IRRADIATED TO 3.1–3.2 x 10^{20} nvt > 1 MeV AT 550°C

Material	Test Temp. (°C)	U.T.S. (p.s.i.)			0.1% Proof Stress (p.s.i.)			Elongation (%)			Reduction of Area (%)			Fracture *		
		As Received	Heat Treated	Irradiated	As Received	Heat Treated	Irradiated	As Received	Heat Treated	Irradiated	As Received	Heat Treated	Irradiated	As Received	Heat Treated	Irradiated
C	25	125,400	79,200	99,500	84,500	47,700	97,100	8.9	6.9	1.4	7.5	5	1	C	C	C
	200	58,500	60,100	86,600	39,500	36,600	74,600	45	51	25	57	58	37	C	C	C
	300	60,100	71,700	46,300	48,500	58,300	36,000	44	42	40	59	52	42	C–95% S–5%	C–30% S–70%	C–85% S–15%
	400	43,900	56,000	28,200	37,500	52,200	26,000	30	28	25	38	41	28	S	S	C–55% S–45%
	500	28,600	29,200	34,600	26,000	26,200	32,500	30.5	30	17	36	25	16	S	I	I
	600	10,950	14,600	12,400	8,500	12,900	10,600	25	19	16	18	12	12	I	I	I
	700	4,100	5,700	6,150	3,400	4,700	5,280	21	13	7	15	6	13	I	I	I
D	25	89,100	78,800	58,800	42,400	38,200	41,700	9	8	7	8.5	8	12	C	C	C
	200	60,300	67,700	57,600	34,600	51,000	38,800	38	40	20	51	59	19	C–95% S–5%	C	C
	300	38,400	42,900	42,800	24,400	29,000	32,600	49	65	39	60	76	48	C–70% S–30%	C–75% S–25%	C–50% S–50%
	400	37,600	27,200	29,700	28,000	27,700	26,400	39	82	40	64	79	46	S	S	S
	500	20,000	16,400	20,700	17,400	15,300	20,000	37	65	37	37	38	43	S/I	S	S
	600	9,600	9,620	12,600	8,300	8,500	11,100	28	38	10	18	29	9	I	I	I
	700	3,750	4,510	3,900	2,950	3,700	3,560	26	14	12	18	10	6	I	I	I

* C = cleavage
S = shear
I = intergranular

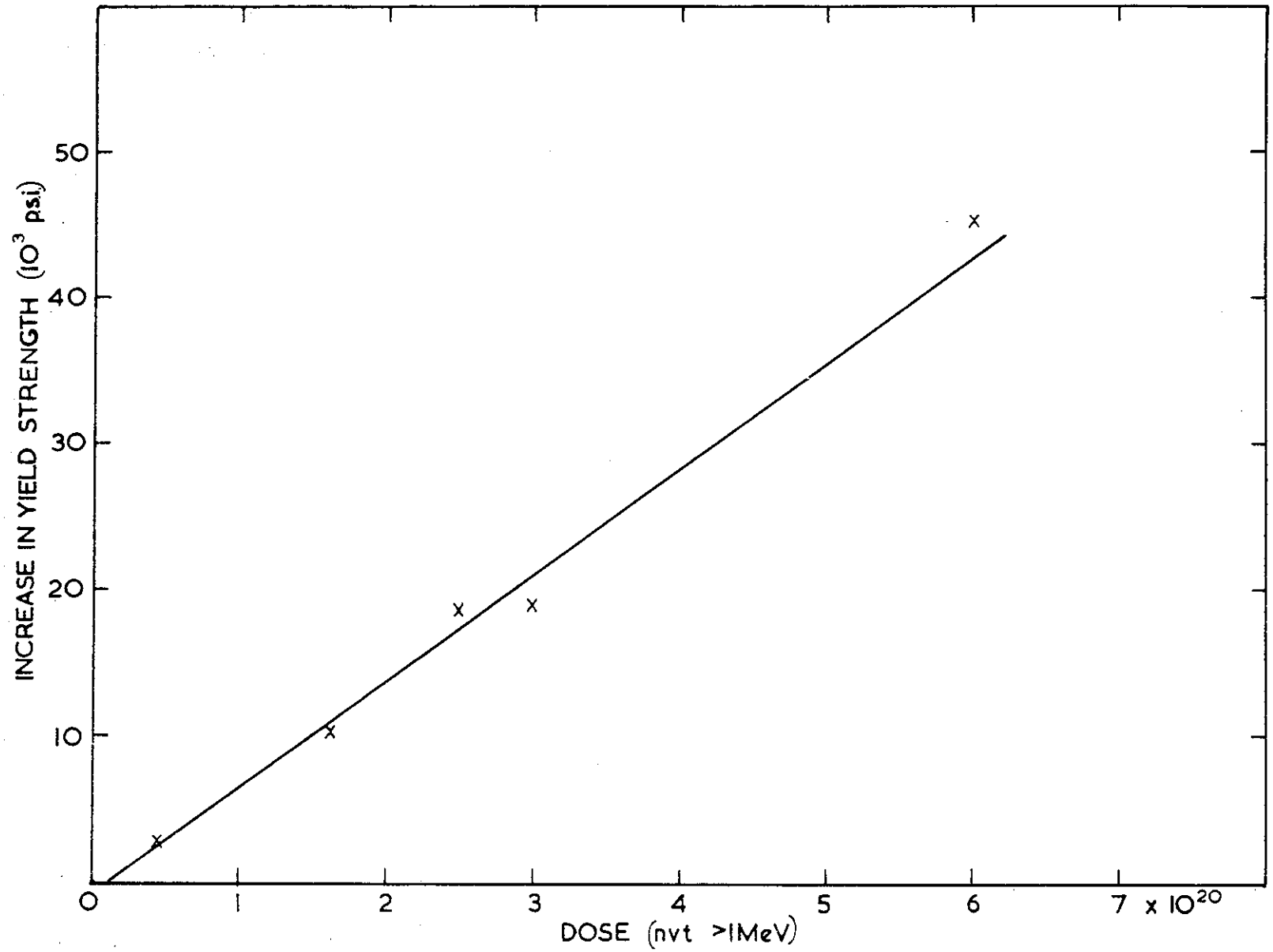


FIGURE 1 VARIATION OF YIELD STRENGTH WITH DOSE AFTER IRRADIATION AT 75-100 °C

P692

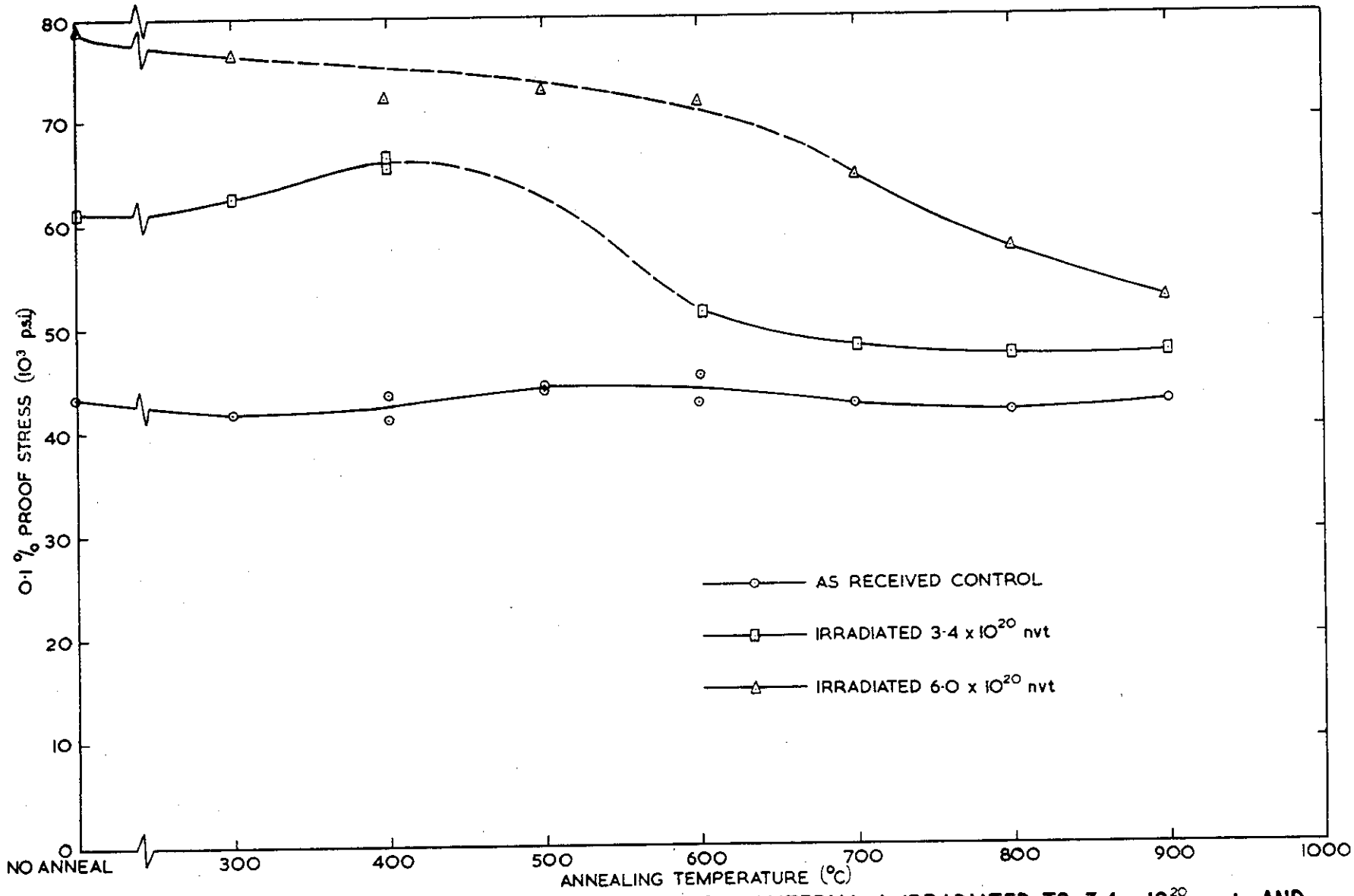


FIGURE 2a RECOVERY OF YIELD STRENGTH WITH ANNEALING IN MATERIAL A IRRADIATED TO $3{\cdot}4 \times 10^{20}$ nvt AND $6{\cdot}0 \times 10^{20}$ nvt >1MeV

P692

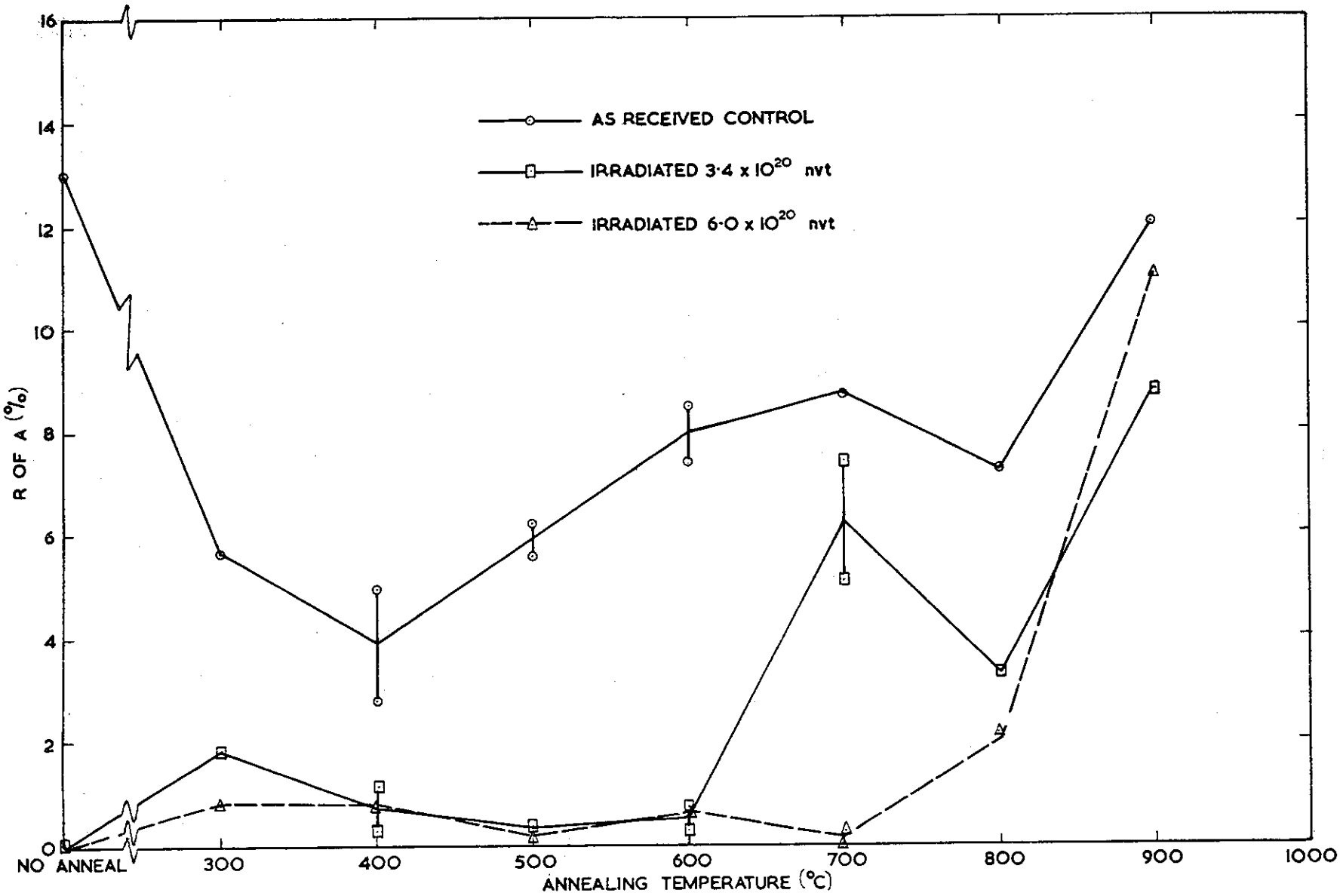


FIGURE 2b RECOVERY OF DUCTILITY WITH ANNEALING IN MATERIAL A IRRADIATED TO 3·4 x 10^{20} nvt AND 6·0 x 10^{20} nvt >1MeV

P692

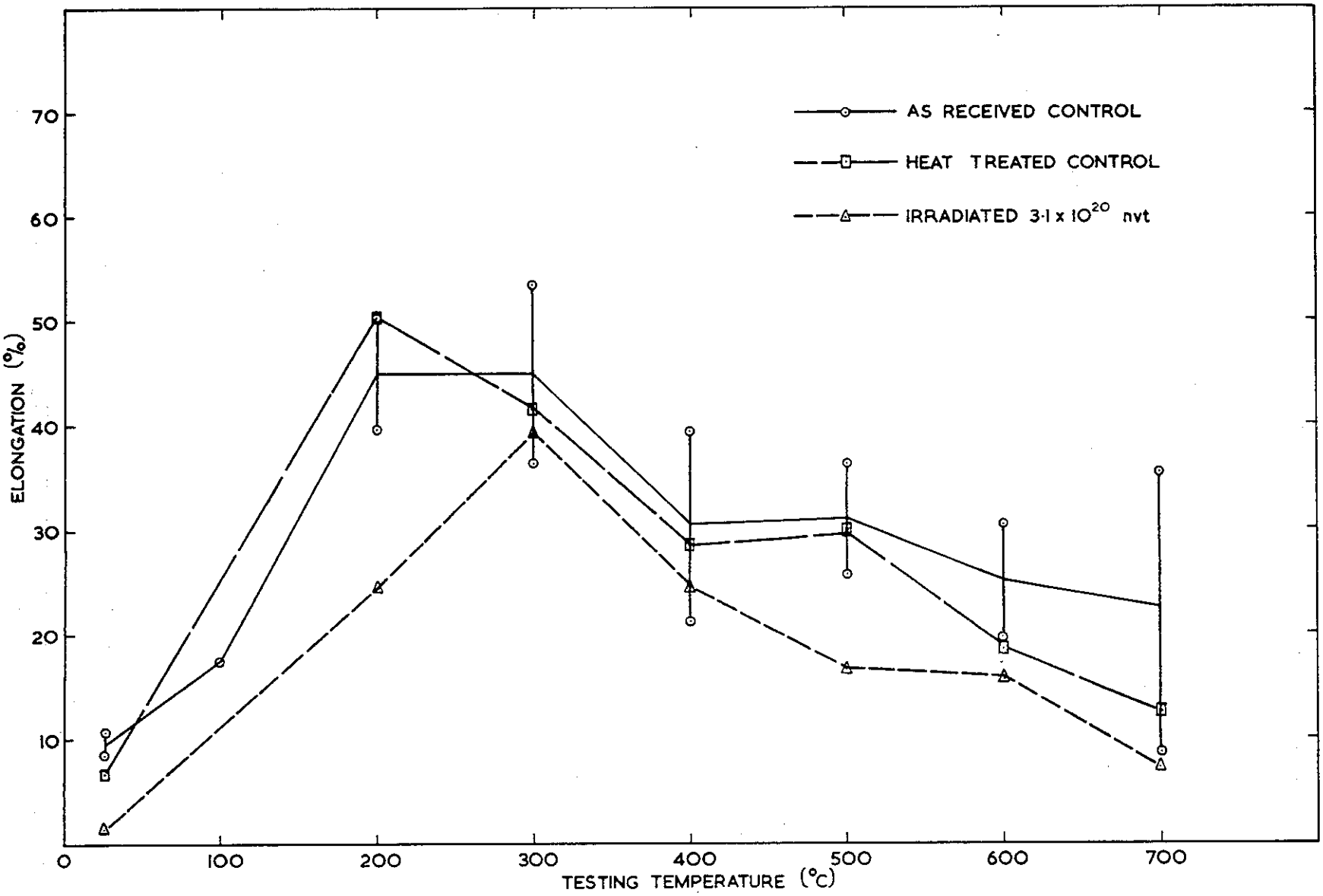


FIGURE 3 DUCTILITY OF MATERIAL C AS A FUNCTION OF TESTING TEMPERATURE

P692

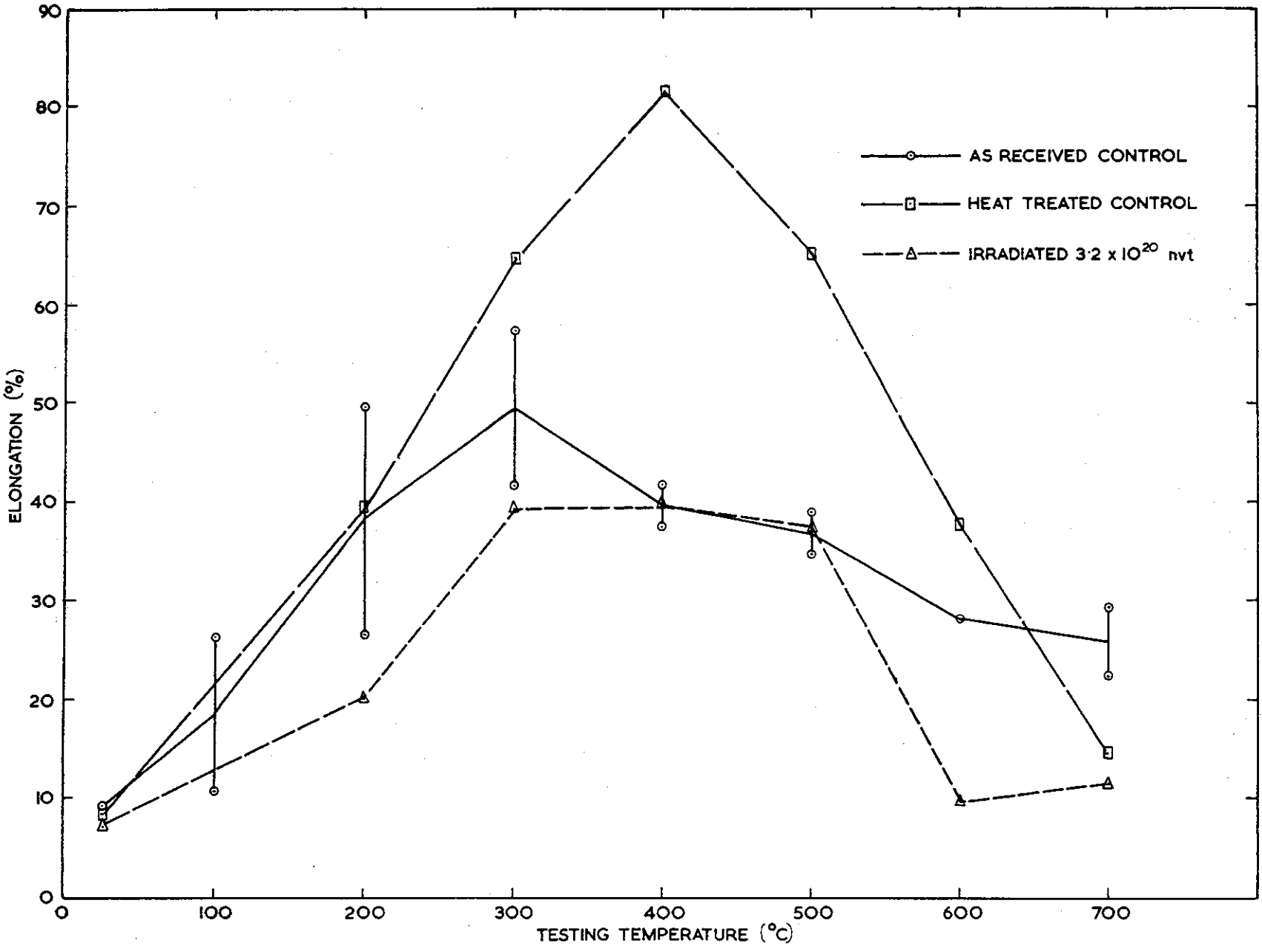
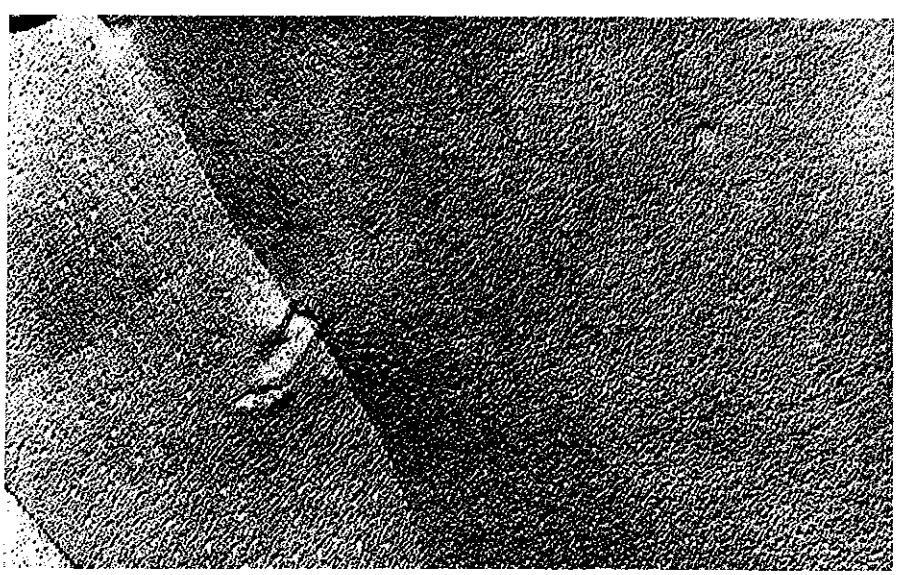


FIGURE 4 DUCTILITY OF MATERIAL D AS A FUNCTION OF TESTING TEMPERATURE

P692

X 30,000

(a) Material C



X 30,000

(b) Material D

FIGURE 5. ELECTRON MICROGRAPHS OF REPLICAS OF MATERIALS C AND D AFTER IRRADIATION AT 550°C