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COMPARISON OF COMPRESSION PROPERTIES AND SWELLING OF BERYLLIUM IRRADIATED AT VARIOUS TEMPERATURES

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J. M. BEESTON and L. G. MILLER

EG&G Idaho, Inc., Idaho Falls, Idaho 83415

E. L. WOOD, Jr.

Argonne National Laboratories, West, Idaho Falls, Idaho 83401

R. W. MOIR

Lawrence Livermore National Laboratory, P.O. Box 5511 L-644 Livermore, CA 94550

MASTER

A beryllium cylinder irradiated in Experimental Breeder Reactor (EBR-II) for four years at 700 to 760 K to a neutron fluence of 8.13×10^{22} n/cm^2 (total) or 1×10^{22} n/cm^2 ($E > 1$ MeV) was cut into samples and tested. Yield strength and plastic strain was determined in compression tests at 300, 723, 823 K and after annealing at 1173 K for one hour. The immersion density and helium content were measured on samples. An equation for swelling was derived from the data by regression analysis. The microstructure showed agglomeration of helium in voids or bubbles at the grain boundaries.

1. INTRODUCTION

A beryllium cylinder irradiated in EBR-II (position 8E2) from 9-10-72 to 7-27-76 at 700 to 760 K to a neutron fluence of 8.13×10^{22} n/cm^2 (total) or 1×10^{22} n/cm^2 ($E > 1$ MeV) was cut into samples and tested. The stainless steel encapsulated beryllium was subjected to a flux of 1.1×10^{15} n/cm^{2}s with a ±25 percent gradient from side to side of the cylinder. The cylinder had an outside diameter of 5.08 cm and an inside diameter of 3.2 cm. The cylinder was cut into sections with an abrasive saw using coolant. The properties determined were compression yield and failure (cracking) strength, ductility, immersion density, microhardness, helium content, and microstructure. Comparison of the EBR-II beryllium compression properties and swelling is made.[1,2,3,4,5,6,7] The elevated temperature properties of beryllium are important to the lifetime analysis of beryllium as a neutron multiplier in conventional and hybrid fusion blankets.[8,9] The cylinder was hot pressed beryllium (type HP-20) with 2% BeO and a density of 1.85 g/cm^2. The cylinder was irradiated in EBR-II with the bottom of the cylinder at about 700 K and the top at 760 K. The fluences for all samples were estimated by considering the flux gradients within the reactor at position 8E2 for the cylinder.

2. COMPRESSION STRENGTH

The compression yield and failure (cracking or barreling) strength was measured at 300, 723, and 823 K temperatures on 2.8 cm long prism sectors, Figure 1, cut from the cylinder. The tests were conducted on an Instron at a cross head speed of 0.05 in./min. The data are presented in Table 1. In the ambient temperature tests the irradiated beryllium failed with considerable release of energy and produced fine shards and particulate as shown in Figure 1. In the 723 and 823 K tests the irradiated beryllium underwent much more plastic deformation than in the ambient tests. After the 1173 K anneal for 1 hour, the specimens tested at 723 K and 823 K experienced such extensive plastic deformation in the compression tests that barreling occurred before failure as shown in Figure 1. The data is presented in Table 2.

3. DUCTILITY

The irradiated beryllium ductility is seen to be affected by the temperature at which it is loaded, by the fluence or helium content, and by the high temperature annealing, Table 1 and Table 2. The higher ductility at higher loading temperature (723 and 823 K) is clearly seen; since the plastic strain at ambient temperature is about 3 percent for the 5 tests with fluence above 1 x 10^{22} n/cm^2, Table 1 or Figure 1, while the plastic strain at 723 or 823 K is about 14 percent for 7 tests above 1 x 10^{22} n/cm^2. The effect of fluence or helium content is also discernable with lower plastic strain at higher fluence in the ambient, 723, and 823 K tests, Table 1. The effect of the high temperature annealing was to reduce the strength and increase the ductility (Figure 1, Table 1 and Table 2).

4. IMMERSION DENSITY

The specimens as in Figure 1 were cut from the cylinder and ultrasonically cleaned and the immersion density measured per ASTM B 311. The preirradiated density was taken as 1.85 g/cm^3. The immersion density was measured using a wetting agent. The density of the irradiated specimens is given in Table 3, and the calculated swelling is taken as the negative density change. The equation which best fit the negative density change (swelling) by regression analysis with a correlation coefficient, r = 0.89 and multiple correlation coefficient $r^2 = 0.79$, was $\Delta V/V = -0.002 + 1.83 \times 10^{-58} \phi^2 T^4$, where ϕ is the fluence in n/cm^2 (E > 1 MeV). The high regression coefficient indicates a very good fit of the data, although the range of temperature and fluence is small. The swelling, thus, increases as a function of fluence and irradiation temperature. The apparent fit of the data is indicated in Table 3 by the comparison of the calculated swelling with the measured swelling, and is indicated in Figure 2 at a constant fluence of 1 x 10^{22} n/cm^2 and in Figure 3 at a constant temperature of 750 K. The data points in Figure 2 or 3 are from Table 3 at the nearest constant fixed value of fluence or temperature, respectively.

Because of the physical constraint that no swelling occurs at zero values of the parameters, the equation recommended for calculation of swelling in irradiated beryllium is $\frac{\Delta V}{V} = 1.83 \times 10^{-58} \phi^2 T^4$.

5. MICROHARDNESS

The microhardness was measured in samples at the top, center, and bottom of the cylinder. The diamond pyramid hardness measured on two samples in the center which had received a fluence of 1.2 x 10^{22} n/cm^2 (E > 1 MeV) was 221 ± 5 DPH (14 measurements). The microhardness in the bottom sample which had received a fluence of 0.3 x 10^{22} n/cm^2, (E > 1 MeV) was 175 ± 5 DPH and in the top sample with a fluence of 0.8 x 10^{22} n/cm^2 was 195 ± 8 DPH. Intermediate samples had intermediate hardness. The highest microhardness occurs in the highest fluence center samples with the highest helium content. The microhardness at these temperatures can be represented by the equation, $DPH = 135 + 2.98 \times 10^{-6} \phi^{1/3}$, where ϕ is the fluence in n/cm^2 (E > 1 MeV).

Microhardness on post-irradiated annealed material (773 K) was given by Hickman[10] as 192 DPH.

6. HELIUM CONTENT

Two beryllium fragments from the compression tests were analyzed[11] for helium content. The fluence for these samples 6-3 and 9-3 was estimated as 1.2 x 10^{22} n/cm^2 (E > 1 MeV). The fragments weighed 25.5 and 26.0 mg, respectively. The fragments were ultrasonically cleaned in acetone, dried under a heat lamp, and examined under a low-power optical microscope to check for adhesions or surface irregularities. None were observed. Smaller individual samples, labeled A and B, were cut from each fragment for duplicate analysis. The helium content of each sample was determined by mass spectrometry by vaporizing each sample in a resistance-heated graphite crucible in a high-temperature furnace. The absolute amount of ^{4}He released was measured relative to a known quantity of added ^{3}He "spike". Reproducibility in the duplicate analyses was excellent, averaging 0.7%. Absolute uncertainty in the results is estimated to be <1.0% of the concentration values. Conversion from total helium to helium concentration was based on the calculated number of beryllium atoms per gram of sample. The value used was 6.6821 x 10^{22} atoms per gram. The results of the helium measurements are given in Table 4, and are listed as total atoms of helium released, and helium concentration in atomic parts per million (10^{-6} atom fraction).

It is of interest to extrapolate the effect of EBR-II irradiation of beryllium to other reactors. The assumption is made that the helium content is the controlling damage mechanism. From the helium content results, Table 4 the larger helium content will be taken to correspond to the fluence of 1.2 x 10^{22} n/cm^2 (E > 1 MeV). The fluence for the other sample would be 0.96 x 10^{22} n/cm^2 (E > 1 MeV), which is a reasonable result for the other fragment considering the size of the compression specimen, position in the cylinder, and flux gradient in the reactor. Thus, for the center flux spectrum in position 8E2 in EBR-II, the helium production is about 1849/1.2 x 1^{22} n/cm^2 = 1540 a ppm helium per 10^{22} n/cm^2 (E > 1 MeV). In the Advanced Test Reactor (ATR) this fluence of 1.2 x 10^{22} n/cm^2 (E > 1 MeV) would produce a helium content of about 4440 appm helium content and in the Tandem Mirror Hybrid Reactor (TMHR) concept near the first blanket zone wall this fluence of 1.2 x 10^{22} n/cm^2, (E > 1 MeV) would produce 8,400 appm helium content in beryllium. Thus, the equivalent damage ratio for helium production for TMHR/ATR/EBR-II in beryllium is 8400/4440/1849 or 4.5/2.4/1. The corresponding atomic displacement (dpa) ratios for E > 1 MeV would be 0.25/0.22/1. In terms of wall loading a TMHR fluence of 1.2 x 10^{22} n/cm^2 E > MeV corresponds to a first wall loading of 3.4 MW·y/m^2, and t e EBR-II fluence of 1.2 x 10^{22} n/cm^2 E > MeV corresponds to a TMHR first wall loading of 3.4 ÷ 4.5 or 0.76 MW·y/m^2.

7. MICROSTRUCTURE

The microstructure of the irradiated beryllium was examined by optical microscopy, scanning electron microscopy, and by transmission electron microscopy.

The optical microscopy revealed a fine-grained structure of about ASTM Grain Size 6 (diameter of 51 microns). Transverse and longitudinal sections showed the structure was equiaxed representative of hot-pressed powder beryllium. Some porosity is present in the optical photograph apparently due to the BeO pullout during polishing, the helium swelling, and to porosity (Figure 4a) which is enhanced in the scanning electron micrograph, Figure 4b. No effect of the canning in stainless steel was observed.

Transmission electron microscopy on a thin section irradiated at about 710 K to $\sim 1 \times 10^{22}$ n/cm^2 (E > 1 MeV) done on a JEOL 200 B microscope showed most of the beryllium matrix not to contain voids or bubbles. There were voids on the grain boundaries. Figure 4c shows 0.03 μm sized voids distributed uniformly on a grain boundary with a density of 6×10^{15}/m^2. Note the anisotropic shape which depends on the angle of observance. The voids were distributed very inhomogeneously and generally accompanied by a precipitate. The hexagonal crystallographic shape indicated the voids may not be filled with helium.

8. COMPARISON OF EBR-II IRRADIATED BERYLLIUM WITH OTHER DATA

Beryllium irradiated and tested at temperatures above 623 K generally exhibits a small increase in yield strength and small decrease in ductility.[1,2,4,5] As the fluence (helium content) increases the temperature to produce complete annealing appears to increase[4] and the temperature to produce 1% swelling appears to decrease.[6] Beryllium irradiated and tested at temperatures < 373 K exhibits nil ductility with an increase in yield strength.[3,7]

The EBR-II beryllium irradiated and tested at temperatures of about 740 K containing 1850 appm helium can undergo appreciable compressive plastic strain with an increase in yield strength, Figures 5 and 6. The data from Table 1 for cylinder 6 (irradiated at 740 K, Table 3) and for cylinder 9 (irradiated at 755 K Table 3) have been averaged and plotted in Figures 5 and 6. Although the effect of irradiation temperature on swelling was sufficient to affect the correlation coefficient, the temperature difference is too small to see an effect on plastic strain or yield strength, Figures 5 and 6. Annealing of the irradiation produced defects and some agglomeration of helium together with additional available slip systems[12] at temperatures of 500-750 K are responsible for the increased plastic strain.

A comparison of the swelling with the post-irradiation swelling[8] (Table 5) indicates that at intermediate temperatures (673 to 873 K) and intermediate fluences (based on equivalent helium content) the calculated swelling by the equation $\frac{\Delta V}{V} = 1.83 \times 10^{-58} \phi^2 T^4$ is equivalent to the estimate[8] of $\frac{\Delta V}{V} = 0.00549 \, \phi^{1.035} + 7.8 \times 10^{-4} (2.5)^{\chi}$.

9. CONCLUSIONS

Beryllium irradiated in EBR II and tested at temperatures of 700 to 823 K to fluences of 1.2×10^{22} n/cm^2 (E > MeV) (helium contents of 1850 appm) can undergo compressive plastic strains greater than 10%. Irradiation increases the compressive yield strength at these temperatures (700 to 823 K) to 280 MPa at 723 K. The plastic strain at 723 K was 13%. Annealing at 1173 K increased the ductility such that at 723 K the plastic strain increased to 30%.

The swelling of irradiated beryllium at these temperatures can be represented by the equation $\frac{\Delta V}{V} = 1.83 \times 10^{-58} \phi^2 T^4$, where ϕ is the fluence, n/cm^2 (E > 1 MeV), EBR-II--8E2 spectrum, and T is the temperature, K. At these irradiation temperatures (700-760 K) the swelling is less than 0.9%, and agglomerated helium is mostly at the grain boundaries. Electron transmission microscopy did not reveal extensive voids or bubbles in the beryllium grain matrix. The equivalent damage ratio for helium production for TMHR blanket/ATR/EBR-II--8E2 was 4.5/2.4/1.

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TABLE 1. COMPRESSION YIELD AND FAILURE STRENGTH, AND PLASTIC STRAIN OF EBR-II BERYLLIUM SAMPLES

Spec. No.	Fluence n/cm^2 E > 1 Mev ($\times 10^{22}$)	Average Cross Section (m^2) ($\times 10^{-4}$)	Test Temp. °K	Yield Load N ($\times 10^3$)	Max. Load N ($\times 10^3$)	Plastic[a] Strain in 1.2 in. Yield to Max. Load (in./in.)	Compressive[b] Yield Strength MPa	Failure[c] Strength MPa
6-1	0.76	1.458	300	58.3	77.4	0.067	400	531
6-2	0.93	1.877	300	65.8	82.3	0.046	351	439
9-1	0.93	1.310	300	53.4	64.9	0.053	408	495
6-3	1.20	1.238	300	56.5	64.9	0.030	456	524
9-2	1.10	1.239	300	47.6	53.6	0.037	384	433
6-4	1.27	1.297	300	52.0	57.2	0.028	401	441
9-3	1.27	1.529	300	48.5	55.2	0.027	317	361
9-4	1.27	1.781	300	56.5	73.6	0.036	317	413
6-8	0.76	1.394	723	42.3	56.0	0.305	303	402
6-7	0.93	1.310	723	42.7	50.7	0.160	326	387
9-5	1.10	1.503	723	44.5	48.0	0.094	296	319
9-10	1.10	1.342	723	35.1	40.0	0.165	262	298
6-11	1.27	1.426	723	40.0	41.8	0.162	281	293
9-9	0.93	1.484	823	35.6	45.8	0.164	240	309
6-6	1.10	1.219	823	34.7	35.1	0.115	285	288
6-14	1.10	1.245	823	32.9	35.8	0.189	264	288
6-5	1.27	1.335	823	34.7	35.6	0.097	260	267
6-12	1.27	1.181	823	35.1	35.8	0.123	297	303

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a. Plastic strain is the strain to maximum load from elastic-extension-line.

b. Yield strength is derived from 0.2% offset.

c. Failure strength is derived from the maximum load at which cracking or barreling occurred.

TABLE 2. COMPRESSION YIELD AND FAILURE STRENGTH, AND PLASTIC STRAIN OF EBR-II BERYLLIUM SAMPLES ANNEALED AT 1173 K FOR ONE HOUR

Spec. No.	Fluence n/cm^2 E > 1 Mev $(\times 10^{22})$	Average Cross Section (m^2) $(\times 10^{-4})$	Test Temp. °K	Yield Load N $(\times 10^3)$	Max.[a] Load N $(\times 10^3)$	Plastic[b] Strain at Max. Load	Yield[c] Strength MPa	Failure[a] Strength MPa
6-10	0.93	1.600	723	27.1	44.5	0.204	169.4	278.1
9-14	0.93	1.477	723	16.5	44.5	0.302	103.1	278.1
9-11	1.27	1.613	723	10.2	44.5	0.379	63.8	278.1
9-15	0.76	1.465	823	19.6	44.5	0.252	122.5	278.1
9-6	0.93	1.406	823	12.5	44.5	0.291	78.1	278.1
9-13	1.10	1.581	823	12.5	44.5	0.416	78.1	278.1

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a. Due to extensive plastic deformation an arbitrary load of 10,000 lb at which excessive barreling occurred was used to define the maximum load and failure strength in compression.

b. Plastic strain is the strain to maximum load from elastic-extension-line.

c. Yield strength is derived from 0.2% offset.

TABLE 3. DENSITY AND SWELLING OF EBR-II IRRADIATED BERYLLIUM

Specimen No.	Fluence (n/cm^2 E > 1 MeV) ($\times 10^{22}$)	Density (g/cm^3)	Irradiation Temperature (K)	Measured Swelling $\Delta V/V$	Calculated Swelling by Equation[a]
1-A	0.7	1.8490	700	0.0005	0.0002
-B	0.9	1.8474	700	0.0014	0.0016
-C	1.1	1.8446	700	0.0029	0.0033
4-B	1.2	1.8445	730	0.0030	0.0055
-C	0.8	1.8478	730	0.0012	0.0013
-D	1.0	1.8461	730	0.0021	0.0032
7-A	1.0	1.8412	750	0.0048	0.0038
-B	0.8	1.8471	750	0.0016	0.0017
-C	1.0	1.8403	750	0.0052	0.0038
-D	1.3	1.8340	750	0.0086	0.0078
10-A	0.8	1.8478	760	0.0012	0.0019
-B	1.0	1.8422	760	0.0042	0.0041
-C	1.2	1.8353	760	0.0079	0.0068
-D	1.0	1.8434	760	0.0036	0.0041
13-A	1.1	1.8434	760	0.0036	0.0054
-B	0.8	1.8483	760	0.0009	0.0019
-C	0.7	1.8491	760	0.0005	0.0010
-D	0.8	1.8476	760	0.0013	0.0019
6-5	1.27	1.8386	740	0.0062	0.0069
-6	1.10	1.8375	740	0.0068	0.0046
-7	0.93	1.8420	740	0.0043	0.0027
-8	0.76	1.8455	740	0.0024	0.0012
9-13	1.10	1.8408	755	0.0050	0.0050
-14	0.93	1.8454	755	0.0025	0.0030
-15	0.76	1.8473	755	0.0015	0.0013
-16	0.76	1.8470	755	0.0016	0.0013

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[a] Equation for calculating swelling, $\Delta V/V = -0.002 + 1.83 \times 10^{-58} \phi^2 T^4$, where ϕ = fluence, n/cm^2 E > 1 MeV, and T = temperature, K. The regression coefficients were $r = 0.89$ and $r^2 = 0.79$.

TABLE 4. HELIUM PRODUCTION IN BERYLLIUM SAMPLES

			Helium Atoms Measured (10^{15} atoms)			Helium Concentration (appm)[b]			
Fragment	Sample Name	Sample Mass[a] (mg)	^{3}He	^{4}He	^{4}He/^{3}He Ratio	^{3}He		^{4}He	
6-3	A	0.3103	0.162	38.21	236	7.81	7.87	1843	1849[c]
	B	0.5066	0.268	62.78	234	7.92	±0.08	1855	±8
9-3	A	0.5001	0.225	49.87	222	6.73	6.67	1492	1483[c]
	B	0.5568	0.246	54.85	223	6.61	±0.08	1474	±13

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a. Mass uncertainty is ±0.3 μg.

b. Measured helium concentration in atomic parts per million (10^{-6} atom fraction with respect to beryllium) and mean values and 1σ standard deviations.

c. Mean of duplicates A and B.

TABLE 5. COMPARISON OF CALCULATED SWELLING FROM RECOMMENDED EQUATION WITH ESTIMATED POST-IRRADIATION EQUATION

Fluence ϕ n/cm^2 E > 1 MeV ($\times 10^{22}$)	Temperature T (K)	Swelling $(\Delta V/V)^a$	Equivalent Fluence for Helium Production ϕ	Temperature Parameter X $(\frac{T°C}{100})$	Swelling $(\Delta V/V)^b$	Ratio $\frac{(\Delta V/V)^b}{(\Delta V/V)^a}$
1	673	0.0037	2.4	4.0	0.044	12.0
	773	0.0065		5.0	0.090	14.0
	873	0.0071		6.0	0.204	29.0
2	673	0.015	4.8	4.0	0.058	3.9
	773	0.026		5.0	0.104	4.0
	873	0.043		6.0	0.218	5.1
3	673	0.034	7.2	4.0	0.073	2.1
	773	0.059		5.0	0.119	2.0
	873	0.096		6.0	0.233	2.5
4	673	0.060	9.6	4.0	0.088	1.5
	773	0.105		5.0	0.133	1.3
	873	0.170		6.0	0.247	1.5
5	673	0.094	12.0	4.0	0.102	1.1
	773	0.163		5.0	0.148	0.9
	873	0.266		6.0	0.262	1.0
6	673	0.135	14.4	4.0	0.116	0.9
	773	0.235		5.0	0.162	0.7
	873	0.383		6.0	0.276	0.7
8	673	0.24	19.2	4.0	0.147	0.6
	773	0.42		5.0	0.193	0.5
	873	0.68		6.0	0.307	0.5

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a. Equation is $\Delta V/V = 1.83 \times 10^{-58} \phi^2 T^4$.

b. Equation is $\Delta V/V = 0.00549\, \phi^{1.035} + 7.8 \times 10^{-4} (2.5)^X$, where $X = \frac{T°C}{100}$, and ϕ is equivalent fluence in units of 10^{22} n/cm^2, E > 1 MeV.

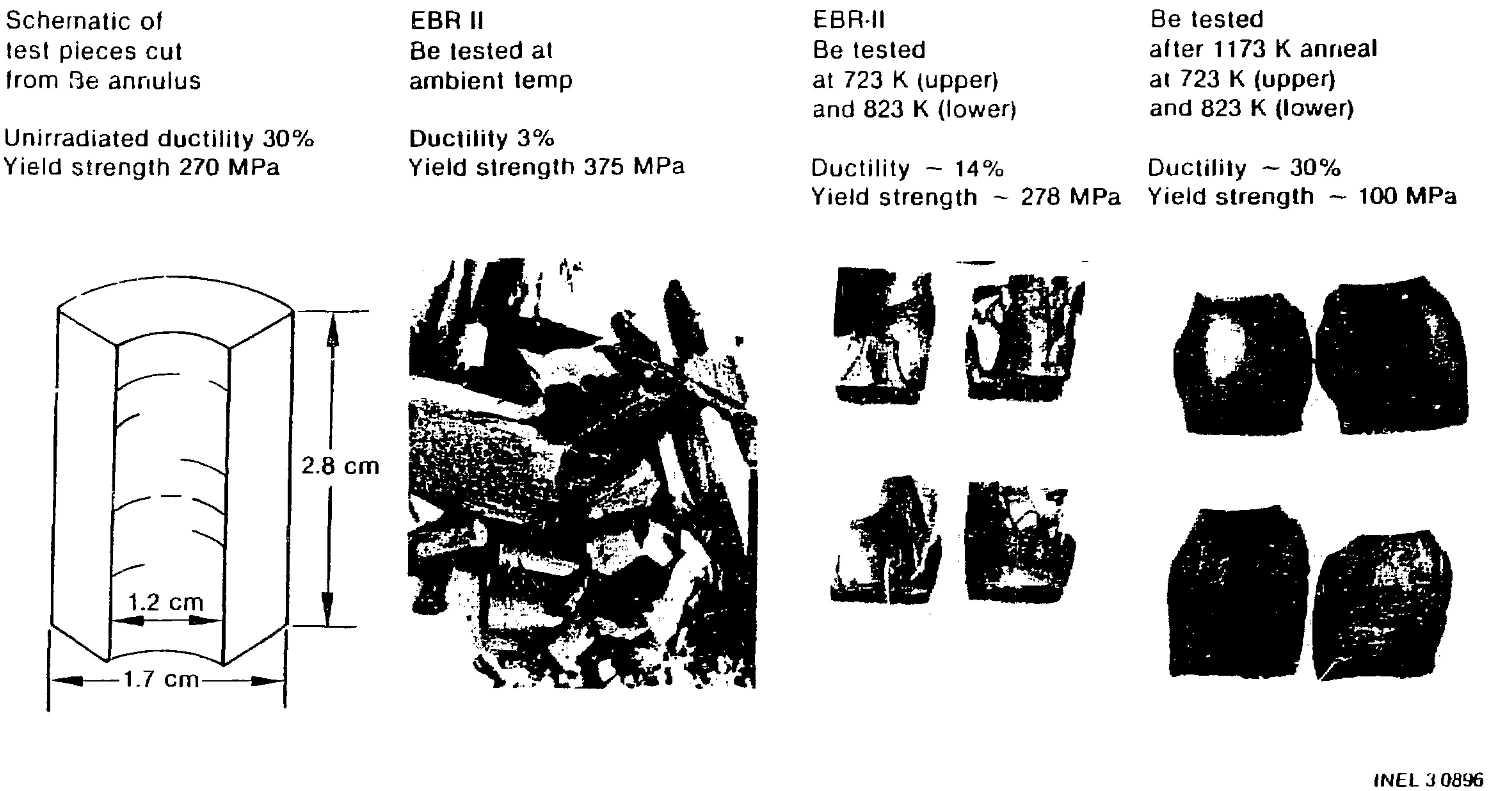

Figure 1. Beryllium compression tests after 10^{22} n/cm^2(E>1MeV) fluence at 700 to 760 K irradiation temperature.

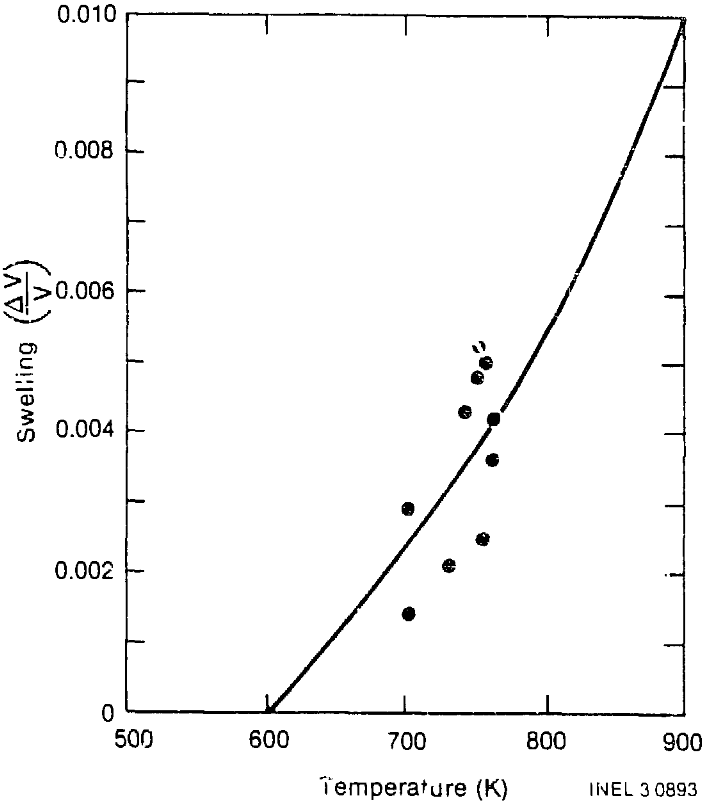

Figure 2. Swelling versus temperature (line represents swelling equation while points are at a constant fluence of $1x10^{22}$ n/cm2),

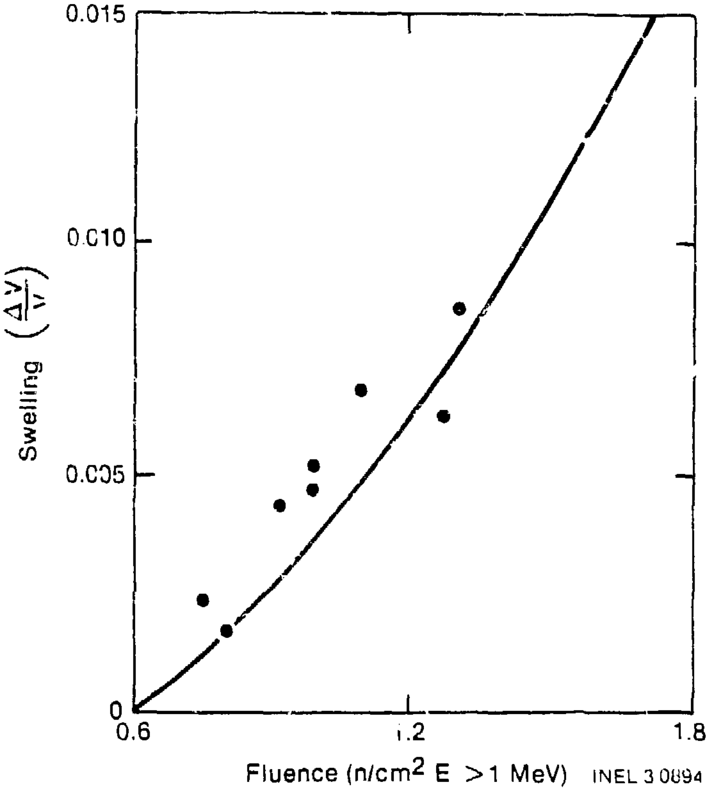

Figure 3. Swelling versus fluence (line represents swelling equation while points are at a constant temperature of 750 K).

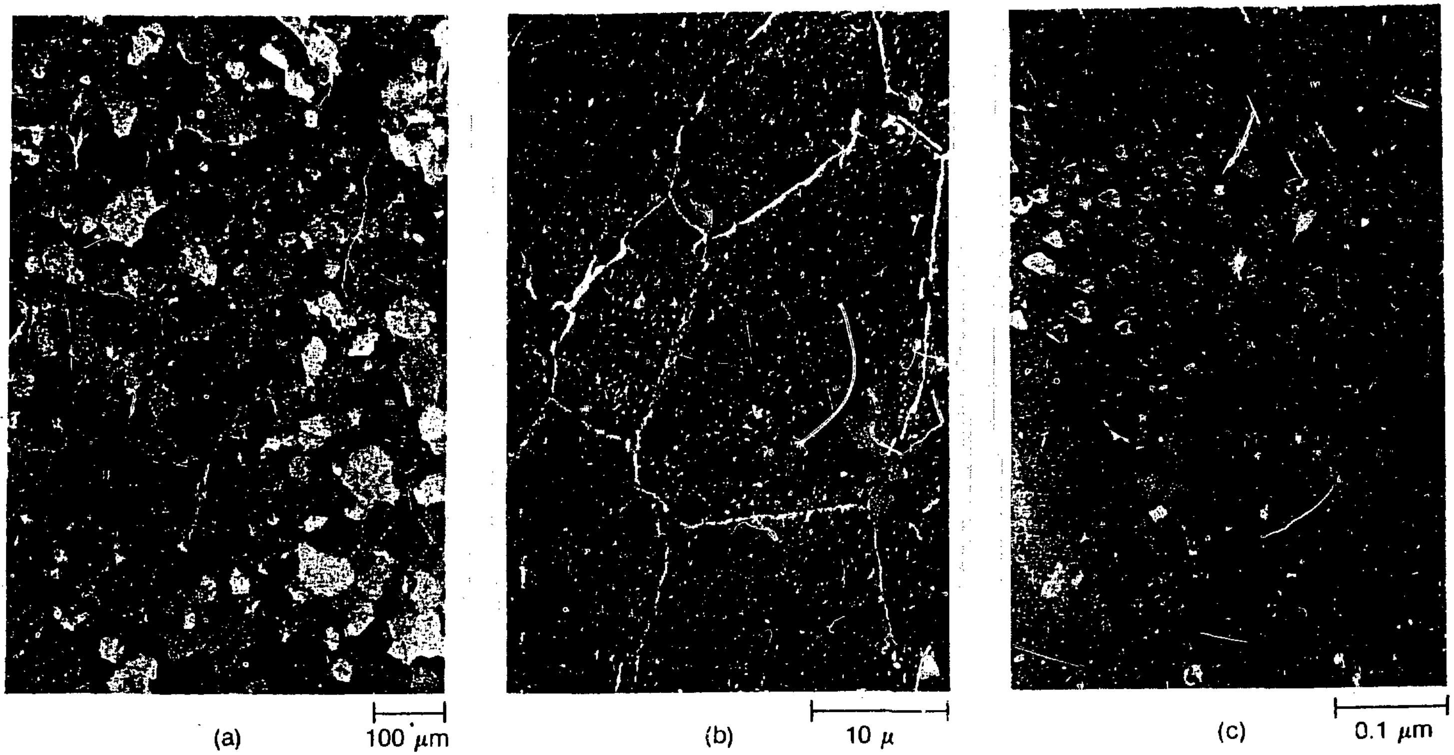

Figure 4. Microstructure shows equiaxed grains with some porosity and voids: (a) optical, Pd light, (b) SEM, (c) TEM with voids on grain boundaries. Note anisotropic shapes in matrix of grains.

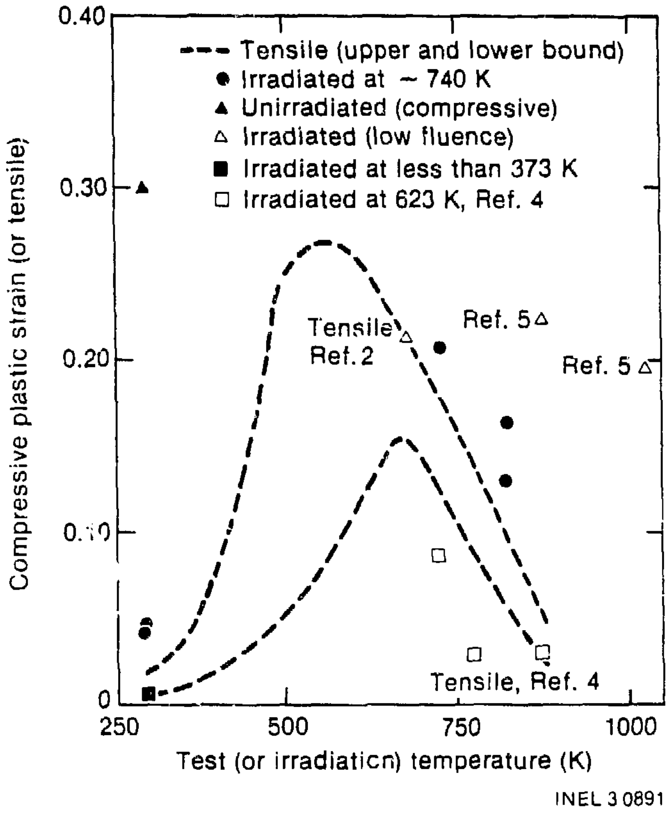

Figure 5. Plastic strain of beryllium as function of test or irradiation temperature.

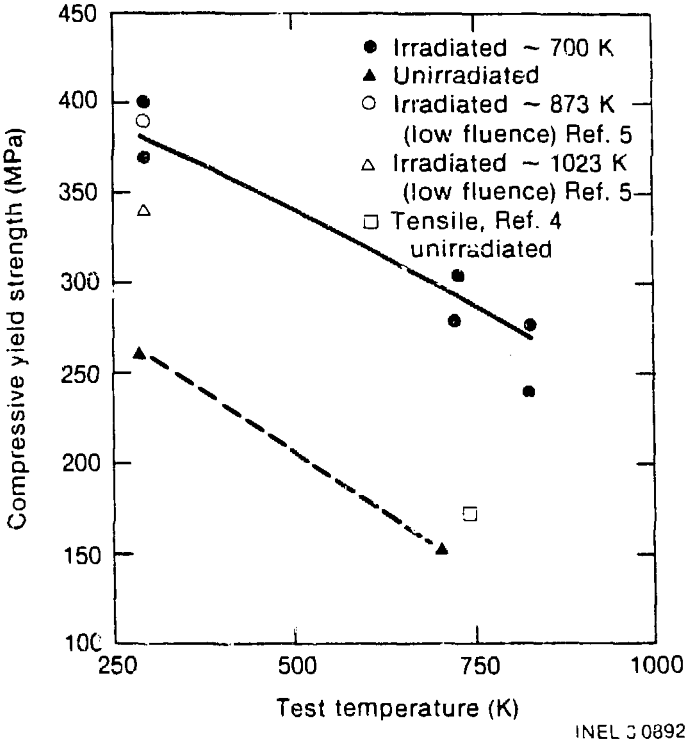

Figure 6. Yield strength of beryllium as function of test temperature.