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Experimental determination of creep properties of Beryllium irradiated to relevant fusion power reactor doses

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Abstract

A dead weight machine has been developed to measure creep in irradiated beryllium relevant to fusion power reactors. Due to the external compressive load, the material will creep and the specimen will shrink. However, the specimen also swells due to the combined effect of internal pressure in helium bubbles and creep. One of the major challenges is to unmask swelling and derive intrinsic creep properties. This has been achieved through appropriate pre-annealing experiments. Creep has been measured on irradiated and unirradiated specimens. The temperature and stress dependence is characterized and modeled using the product of an Arrhenius' law for the temperature dependence and a power law for the stress dependence. Irradiation increases the sensitivity to creep but the irradiation effects can be rationalized by taking into account the irradiation-induced porosity. Experimental evidence supports dislocation climb by vacancy absorption to be the most plausible intrinsic creep mechanism.

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1. Introduction

Beryllium has been selected for the helium cooled pebble bed blanket (HCPB) in the European fusion technology long term program. Beryllium acts as a neutron multiplier that will allow tritium production in the lithium ceramic breeder. Before using the HCPB concept in a power reactor (e.g. DEMO) it has to be fully qualified and tested in experimental programs and in a fusion reactor (e.g. ITER).

The thermomechanical behavior of the pebble bed is a key element for safe routine operation and in case of an accident. For example, tritium production and retention is a potential safety hazard and waste issue that is closely related to the local thermal and stress conditions. Appropriate heat removal will also depend on the mechanical contact between the pebbles. Another important issue is the structural integrity of the HCPB. Undesirable loads on structural components can result from differential thermal expansion, temperature gradients and swelling due to helium produced by irradiation and its migration into bubbles. Thermal creep can play an important role to reduce and redistribute stresses in the pebble bed. However, most of the creep data available in the literature were produced in the 1960s and are not relevant to the material condition at the end of life in a fusion power reactor. Therefore, an extensive

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experimental program has been executed. The results of this program are reported in the present paper and discussed in the light of data available in the literature.

2. Materials

Beryllium is used as a structural component, moderator and neutron multiplier in the water-cooled high flux BR2 material testing reactor of SCK·CEN. A large amount of highly irradiated beryllium is available, since the second beryllium matrix has been replaced in 1997 after 15 years of operation. The factors limiting the life time of the matrix are embrittlement, cracking and helium production resulting from neutron irradiation. The irradiation temperature is 50 °C, and the neutron fluence is 4.6×10^{22} n/cm^2 ($E > 1$ MeV) corresponding to a measured helium build-up due to the nuclear reactions of 22 500 appm.

The elements of the matrix were delivered by American Beryllium Company Inc. which received the raw product from Kawecki Berylco Industries. The beryllium is a structural grade S200-E (similar to S200-F) produced by the vacuum hot press (VHP) process [1].

The chemical composition is pure 98.4% beryllium with some limited impurities: 1.9% BeO, less than 1600 ppm for Fe and C, less than 500 ppm for Al, Si, Ni, Mn and less than 200 ppm for the remaining elements. The average grain size varies from 11 to 13 μm and is lower than in typical candidate pebble material to be used in a fusion reactor [2,3]. The yield strength, ultimate tensile strength, total elongation and fracture toughness are 310 MPa, 410 MPa, 3.8% and 10–12 MPa$\sqrt{m}$, respectively.

Specimens for creep and microstructure investigations were extracted from broken pieces of a hexagonal channel from the BR2 matrix [4]. Three specimens with dimension $4 \times 4 \times 20$ mm were cut using a Struers Accutom-50 and rectified by polishing. Challenges related to specimen preparation are toxicity, neutron activation, hardness and brittleness of the material. In addition, about 10 specimens of the same geometry were machined from unirradiated material representative of the second matrix.

To prevent swelling from occurring concurrently with creep, the three irradiated specimens were first annealed in vacuum at 750 °C for 200 h. Heating and cooling rates were limited to 20 °C/h to avoid any cracking and burst release of helium. It is shown in [5], that this treatment produces helium migration into bubbles and is adequate to reach a stabilized swelling.

Dimensional measurements prior and after annealing gave a volumetric swelling of 30.7%, 33.8% and 33.7% for specimen 1, 2 and 3, respectively.

Annealing produced an unexpected bending of specimen 1 only. The maximum deflection was 0.43 mm. Specimen 1 was re-machined in two sub-specimens, 1A and 1B, with nominal dimensions $2 \times 2 \times 9$ mm.

3. Experimental setup

The creep experiment is performed in an alpha tight box on a dedicated machine that has been designed and qualified [6,7]. It is a dead weight machine (see Fig. 1) allowing us to load a cross section of 16 mm^2 in compression from 2 MPa to 20 MPa. The specimens can be heated in a controlled manner up to 900 °C. To avoid possible oxidation, the creep tests are performed under argon atmosphere. Specimen shortening is measured using a linear variable differential transformer (LVDT) which is located in a cold area.

In addition to continuous measurement with the LVDT, total creep can be evaluated from dimensional measurement before and after the test and from the final LVDT value after the test when the system has stabilized back to room temperature.

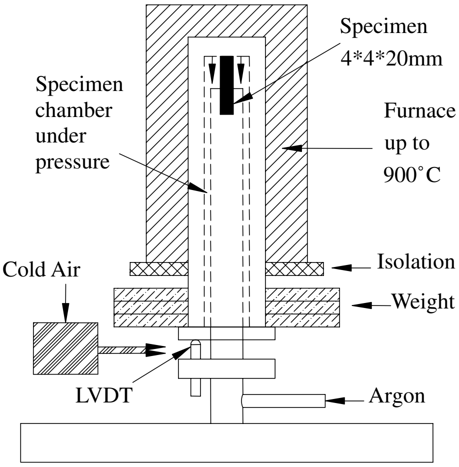

Fig. 1. Schematic of the creep dead weight system.

4. Results

Results from a typical creep measurement are given in Fig. 2. Between the initial and final transient, primary and secondary creep can easily be identified. Total and secondary creep measurements are summarized in Table 1. The primary creep duration is observed to be less than 35 h, therefore to measure the secondary creep the LVDT measurements were taken 35 h and 85 h after the temperature has reached the set point.

No particular problems were encountered during the test except that some specimens show some limited oxidation, probably due to residual adsorbed water vapor.

Specimen 11 exceeded the measuring range available. The combination of high temperature and stress induced a combination of creep and plastic deformation.

Fig. 3 compares the different techniques used to measure the creep. Total creep measurements using the LVDT lead to larger scatter and it is suspected to be a less accurate method. Total creep evaluated through dimensional measurement is reasonably well correlated to secondary creep. Three negative total creep values reported in Table 1 are probably

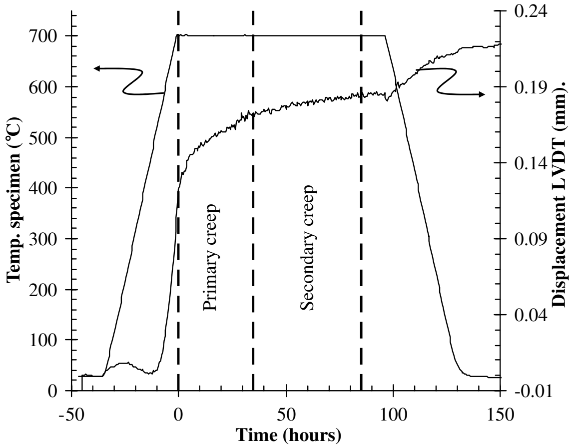

Fig. 2. Typical online measurement of creep. Unirradiated specimen 7 tested at 700 °C under a stress of 15 MPa.

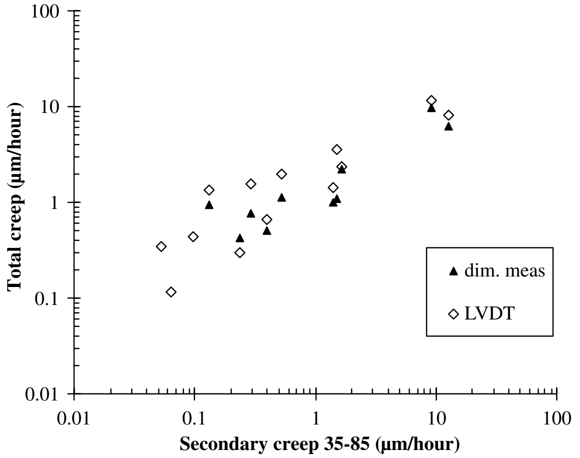

Fig. 3. Comparison of the different techniques to measure creep.

Table 1
Creep measurement

Specimen id.	Condition	Temperature (°C)	Stress (MPa)	Total creep		Secondary creep 35–85 (µm/h)
				Dimensional measurement (µm/h)	LVDT (µm/h)	
1a	i.a.[a]	650	16.4	−0.0491	0.115	0.0644
1b	i.a.	650	20.0	1.141	1.995	0.522
2	i.a.	650	9.0	−0.242	0.443	0.097
2[b]	i.a	750	9.0	0.50	0.67	0.40
3	i.a.	750	12.0	6.246	8.030	12.735
4	a.r.[a]	770	14.9	1.004	1.414	1.402
5	a.r.	700	12.0	2.243	2.376	1.666
6	a.r.	770	11.9	0.955	1.677	0.131
7	a.r.	700	15.0	0.775	1.538	0.293
8	a.r.	770	11.9	0.427	0.301	0.235
9	a.r.	700	12.0	−0.0169	0.348	0.0533
10	a.r.	800	12.0	1.102	3.513	1.504
11[c]	a.r.	840	18.0	>600[c]	>600	>600
12	a.r.	800	15.0	9.644	11.54	9.058

[a] i.a. ≡ irradiated and annealed, a.r. ≡ as received.

[b] Specimen 2 is reused to produce additional data.

[c] Measurement out of range more than 2 mm deformation.

due to measurement accuracy and small variation of specimen dimensions.

5. Discussion

To evaluate the consistency of the results, one needs to compare the different tests. Three parameters need to be evaluated: the effect of temperature, stress and irradiation. This is illustrated in Fig. 4 where a small increase in temperature or stress substantially increases the creep rate. The susceptibility to creep is clearly enhanced by irradiation. The method used to evaluate creep does not substantially change the observed trend.

To rationalize the data, an existing model [8] has been applied. Secondary creep is expressed as a simple formula that includes porosity, stress and temperature effects:

$$\varepsilon_c = A(1 - p^{2/3})^{-n} \exp(-Q/T)\sigma^n, \qquad (1)$$

where A, Q and n are material constants defined as the proportional factor, the creep activation energy and the stress exponent, respectively, p is the porosity volume fraction, T the temperature in Kelvin, σ the stress in MPa and ε_c the creep rate in s^{-1}.

The previous equation is the classical activation energy–stress exponent formulation applicable to a large range of metals [9], where the applied stress has been modified to account for the material porosity. The stress correction is a good engineering approximation but does not take into account local stress concentration in the vicinity of a void. The stress exponent, n, is constant and experimentally found to be about 5 for a large range of materials [9]. It should be noted that for many materials a one to one correlation exists between the creep activation energy and the lattice self diffusion activation energy [9].

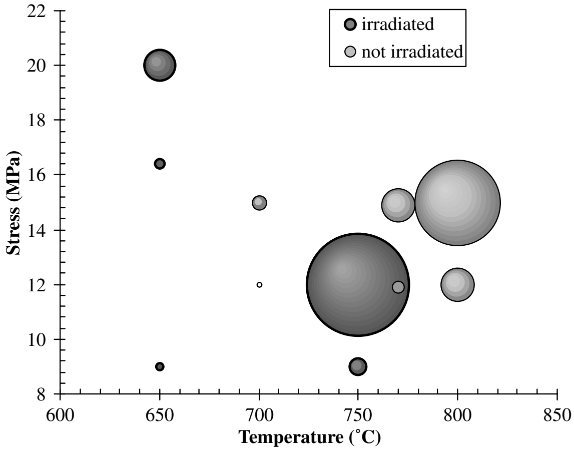

Fig. 4. Effect of temperature stress and irradiation on the secondary creep measured between time 35 and 85 h. The area of the symbol is proportional to the creep rate.

This means that, whatever the mechanism for creep, the criterion for it to be triggered is sufficiently high atomic and point defect mobility. Out of the possible mechanisms proposed in the literature, dislocation movement out of the glide plane through climb induced by the absorption of vacancies is the most widely accepted one. Two possible climb mechanisms (climb of edge dislocation and climb controlled glide) are proposed in [10]. The stress exponent is theoretically derived to be 3 and 4.5, respectively. In both models, the creep rate is independent of the material grain size and the creep activation energy is equal or greater than the self diffusion activation energy.

The experimental work presented here is too limited to determine the model parameters accurately. Therefore, additional data on beryllium creep found in [11–14] has been examined.

In the 1960s [11,12], 15 data points were obtained on several grades of hot pressed beryllium tested in the range 400–700 °C and 8–100 MPa.

In [14], creep experiments are done on pebble bed candidate materials representative of those for a fusion reactor. Since the actual creep actual was not evaluated, the packing density (~60%) was taken into account to compute the effective stress through an equivalent porosity factor, p, of 40%. This approach is rather brute force as it does not take into account high local contact pressure between the pebbles [15]. Preliminary investigations [15] show that the yield stress is even reached locally leading to local plasticity and enhanced creep. To allow us to rationalize those data, a porosity factor of 97.8% had to be used.

Data on the effect of irradiation is found in [13] where three grades of hot isostatically pressed (HIP) beryllium were tested in as received and irradiated condition up to a fluence of 2.4×10^{21} n/cm^2 ($E > 0.1$ MeV) at 750 °C. Creep on irradiated beryllium was higher than on unirradiated material. However, under those conditions the volumetric swelling is about 50% [13] which corresponds to a porosity factor of 33%. The relation between the volumetric swelling and porosity is given by $p = v/(1 + v)$, where p is the porosity volume fraction and v the volumetric swelling.

For the data on irradiated beryllium generated in this work, a porosity factor of 25% has been taken into account for the 33% volumetric swelling.

Taking the logarithm of the creep law, it is possible to perform a simple linear regression to identify the parameters of the creep law. The best linear fit gives $A = 7.21 \times 10^{-3}\ \mathrm{s}^{-1}\ \mathrm{MPa}^{-n}$, $Q = 19470$ K, $n = 2.43$ and a correlation coefficient of 0.858.

The results produced in this work are compared to data found in the literature in Figs. 5 and 6. Despite the scatter, the model describes the temperature and stress effect relatively well. The data produced in this work compare reasonably well with the other data. Having a closer look at Figs. 5 and 6, the irradiated data produced in this work and in [13] do not demonstrate a higher creep rate than the unirradiated data. This could seem paradoxical, but in both Figs. 5 and 6, the effective stress is used, allowing one to take into account the porosity. Therefore, the enhanced creep rate for the irradiated material is predominantly due to the porosity that increases the effective stress in the material.

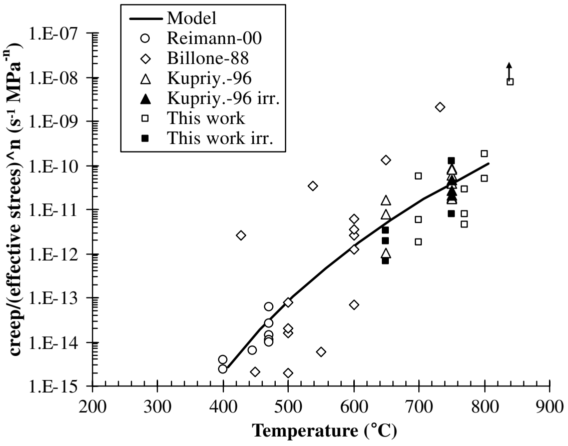

Fig. 5. Effect of temperature on the secondary creep. Creep is normalized by the effective stress to a power 2.43.

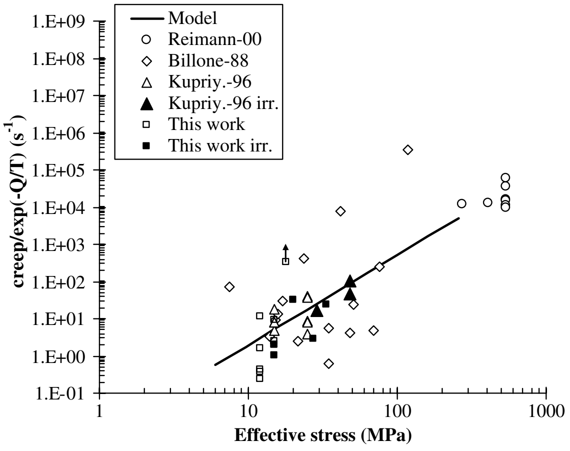

Fig. 6. Effect of stress on the secondary creep. Creep is normalized by the temperature effect with an activation energy of 19470 K.

Creep parameters A, Q and n were evaluated in [8] on a more limited database. They found $0.751\ \mathrm{s}^{-1}\ \mathrm{MPa}^{-n}$, 26000 K and 3.6, respectively. One needs to be aware that several sets of parameters can give equally good fits of the data. To evaluate the sensitivity, several sets of parameters were determined that lead to a correlation coefficient equal or better than 0.836 which corresponds to a 90% confidence level (errors are supposed to be normally distributed). The sets of parameters are bound by an ellipsoid [16]. Its projections on the n–Q and A–Q plane are given in Fig. 7. The size of the ellipse is relatively large, meaning that the accuracy of the parameters of the model is relatively weak. However, relations between parameters exist; n increases with increasing Q and for Q fixed, A decreases with increasing n.

The difficulties in obtaining accurate parameters for the creep model come from the following:

- A small error in applied temperature or stress greatly affects creep.
- Measurement of small specimen elongation under extreme temperature conditions is not straightforward. Attempting to make remote displacement measurements can result in substantial errors.

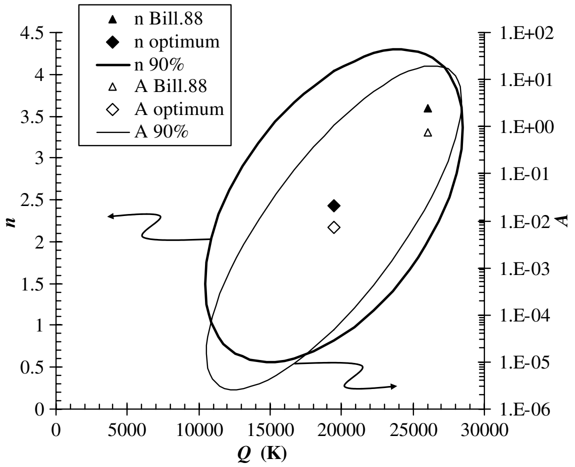

Fig. 7. Creep parameters are within the domain with a 90% confidence level.

- To derive the n and Q parameters accurately, measurements of creep over five orders of magnitude are needed, which is difficult to achieve with a single test set up and results in a wide range of measurement durations.

The self diffusion activation energy has been measured using a ^{7}Be tracer [17]. The value of the activation energy perpendicular and parallel to the basal plane was found to be 18 900 and 19 800 K, respectively. The ratio between the activation energy and the melting temperature is 12.4 and is lower than the ratio generally observed on hexagonal close packed (HCP) and face centered cubic (FCC) materials [17,18], which is between 17 and 20. It is interesting to note that notwithstanding experimental uncertainties, the creep activation energy found in this work (19 470 K) is in perfect agreement with the self diffusion activation energy. Therefore, the experimentally determined creep activation energy and stress exponent both support climb to be the main mechanism for beryllium creep.

6. Conclusions

Experimental determination of creep has been performed on irradiated and non-irradiated beryllium. The main findings of this work are summarized as follows:

- Creep rate increases substantially with stress and temperature.
- Irradiation increases the sensitivity to creep. However, the increase in creep rate is primarily due to irradiation-induced porosity that increases the effective stress.
- Data from the literature are in general agreement with our experimental results.
- Secondary creep can be modeled using the product of an Arrhenius' law for the temperature dependence and a power law for the stress dependence. The identified parameters are $A = 7.21 \times 10^{-3}\ \mathrm{s}^{-1}\ \mathrm{MPa}^{-n}$, $Q = 19470\ \mathrm{K}$ and $n = 2.43$. The uncertainties on those parameters are rather large and result from the experimental difficulties to measure creep accurately.
- The creep activation energy is equal to the self diffusion activation energy and the stress exponent is large. Therefore, the plausible intrinsic creep mechanism is dislocation climb by vacancy absorption. This mechanism is insensitive to grain size and irradiation.

Acknowledgments

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