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A revised description of graphite irradiation induced creep

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ABSTRACT

The UK fleet of advanced gas-cooled reactors (AGR) have been operating for a substantial period of time and rely on data obtained from material test reactor (MTR) programs, dating back to the 1960s and through to the end of the 1980s, to support current and future operation. Historically an empirical approach to the irradiation behaviour of graphite has been used and, due to the nature of the available MTR data and the lack of a consistent set of data containing all the relevant measurements, this is still largely the case at present. Differences in interpretation of the available MTR data can have a significant impact on the assessed integrity of core components. Consequently, a thorough review of the basis of the current models is being carried out, and new models are being developed as necessary. This paper presents some new interpretations of the available low fluence MTR data for irradiation creep and application of a revised model to some high fluence MTR data.

1. Introduction

Modern structural analysis codes, used to investigate the performance of irradiated graphite components, incorporate a constitutive material model that is complex, including solution dependent feedback mechanisms and significant numbers of input parameters. Ideally, the derivation would be based on a substantial amount of experimental data but, in reality, data are sparse and limited in extent. Consequently, material modelling becomes almost a black art rather than a science, often relying on empirical relationships and accepted wisdom rather than fundamental understanding accompanied by objective validation.

The precise interpretation of the experimental datasets has a bearing on the predictions of future core component integrity. As a result, a thorough re-examination of the sparse and disparate material test reactor (MTR) information is being undertaken in an attempt to develop improved models for irradiated graphite material properties. A review of the effect of structure on unstressed graphite material properties has been presented [1]. This paper reviews the existing UK graphite irradiation induced creep data and methodology and then presents some new interpretations of the available low fluence MTR data for irradiation creep. The new model is then applied to some high fluence MTR data.

2. Present UK graphite creep methodology

Irradiation induced creep is defined as the difference in length between an unstressed and a stressed specimen of the same (unirradiated) material properties subjected to the same irradiation environment. The currently employed formulation is based on Brocklehurst and Kelly [2], who proposed for constant stress at low fluence:

$$\varepsilon_c = \frac{\sigma}{E_0}\left(\alpha\left(1 - \exp^{-4\gamma}\right) + \beta\gamma\right). \tag{1}$$

The term outside the brackets is the initial elastic stain unit (esu), σ is the externally applied stress, E_0 is the static Young's modulus appropriate to the applied stress level. The first term inside the brackets in Eq. (1) is primary creep and is currently considered to saturate at 1 initial elastic strain unit for temperature ranges applicable to AGR, i.e. $\alpha = 1.0$. It is also fully recoverable with continued irradiation following load release [3]. The second component inside the bracket is secondary creep which is proportional to fluence and a coefficient, β, which is considered to be independent of irradiation temperature in the AGR temperature range. Secondary creep is considered to be irrecoverable with continued irradiation following load release.

Eq. (1) can be extended to higher irradiation fluence with varying stress [2]:

$$\varepsilon_c = \frac{4}{E_0}\exp^{-4\gamma^1}\int_0^{\gamma^1}\frac{\sigma}{SW}\exp^{4\gamma}\mathrm{d}\gamma + \frac{\beta}{E_0}\int_0^{\gamma^1}\frac{\sigma}{SW}\mathrm{d}\gamma. \tag{2}$$

The initial creep modulus is modified due to irradiation as a result of both structural (S) and radiolytic oxidation (W) changes to the Young's modulus.

Young's modulus itself has been shown to be weakly dependent on creep strain, however this is not taken into account in the present UK methodology. Both elastic and creep strain components are known to modify the coefficient of thermal expansion (CTE) [2,4],

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and the currently endorsed UK methodology incorporates this effect as a dependence on the sum of the elastic and creep strain components.

Based on an earlier theory [5], it was proposed that this strain induced change in CTE would, in turn, modify the underlying dimensional change rate of a stressed specimen [6]. Thus, 'true' creep strain was defined as the difference in length between an unstressed specimen and a stressed specimen with the same irradiated properties as the unstressed specimen, and so the observed 'apparent' creep strain is the sum of the 'true' creep strain and the 'interaction' strain:

$$\varepsilon_c^i = \int_0^\gamma \left(\frac{\alpha_x^1 - \alpha_x}{\alpha_c - \alpha_a}\right)\left(\frac{dX_T}{d\gamma}\right)d\gamma, \qquad (3)$$

where $\alpha_x^1 - \alpha_x$ is the difference between the CTE of the stressed and the unstressed specimens, $\alpha_c - \alpha_a$ is the crystal thermal expansion coefficients parallel and perpendicular to the basal planes and $\left(\frac{dX_T}{d\gamma}\right)$ is the crystal shape change parameter.

Finally, in the endorsed UK methodology the creep lateral strain ratio is assumed to be equivalent to Poisson's ratio for both primary and secondary creep and is independent of the accumulated creep strain.

2.1. Review of existing low fluence data

2.1.1. The dependence of the change in CTE on creep strain

Fig. 1 shows some of the UK BR-2 data for creep strain as a function of fluence [7]. Fig. 2 shows the CTE of the same, isotropic graphite, specimens. It can be seen that, whilst the 'apparent' creep strain continues to increase with fluence, the change in CTE appears to be saturating at a fluence of approximately 20–30 × 10^{20} n/cm^2 EDN indicating a lack of correlation between external strain induced change to CTE and 'apparent' creep strain.

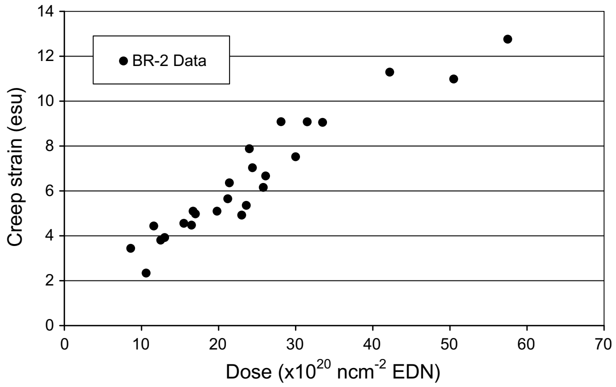

Fig. 1. BR-2 creep data.

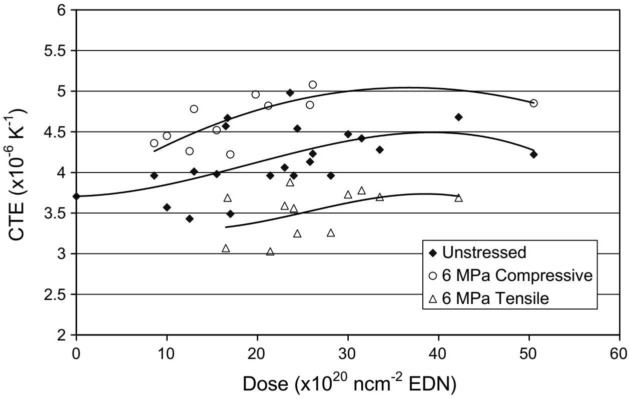

Fig. 2. BR-2 CTE (20–120) data.

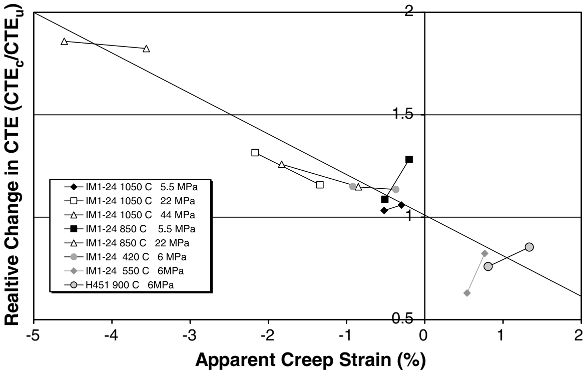

Fig. 3. CTE repeat measurements on IM1-24 graphite at 850/1050 °C.

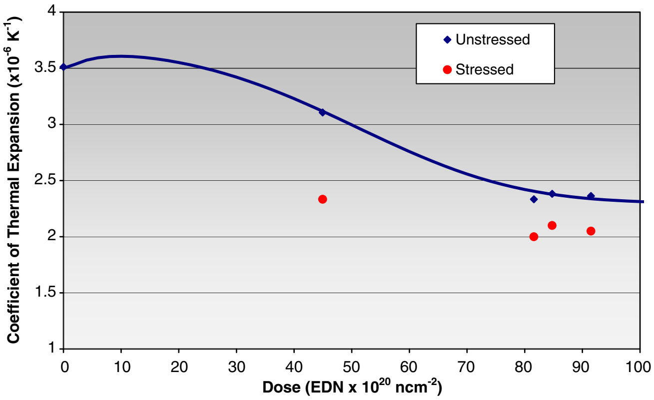

Fig. 4. CTE high fluence measurements on H451 irradiated at 900 °C [2].

Fig. 3 illustrates the results from experiments in which samples were subjected to more than one period of irradiation. These provide repeat measurements on CTE with increasing creep strain [8]. It can be seen that both tensile specimens indicate increasing CTE with increasing 'apparent' creep strain, and three of six compressive specimens indicate a reduction in CTE with increasing 'apparent' creep strain. Thus five out of eight measurements indicate a lack of correlation with 'apparent' creep strain.

Fig. 4 [2], which shows the effect of a tensile load on H451 graphite at 900 °C, reinforces this point. The effect of the externally applied tensile load (a reduction in CTE) decreases with increasing fluence, however during this increment in fluence the tensile creep strain would have increased significantly. Fig. 4 also suggests that the effect of an externally applied strain on CTE should be factorial and not additive.

In a separate part of the BR-2 experiments, tensile and compressive specimens along with their unstressed 'controls' were irradiated and subsequently thermally annealed [9]. Both the dimensional change and the CTE of the samples were measured during the thermal anneal and it was observed (Fig. 5) that the CTE in both the compressive and tensile samples recovered to the unstressed control specimen CTE at roughly the same annealing temperature[1] as that at which the dimensional change

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[1] It should be noted that the recovery of CTE during the thermal anneal occurs at different temperatures for the compressive and tensile samples.

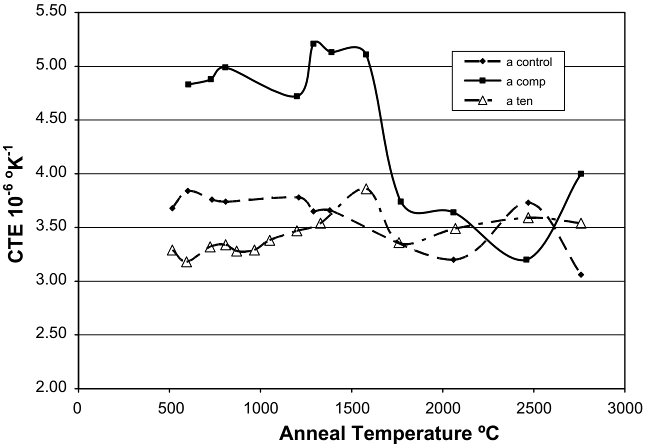

Fig. 5. Thermal anneal of tensile and compressive samples irradiated in BR-2.

(partially) recovered. This full recovery of CTE, albeit based on one or two specimens, suggests that the external strain induced change in CTE is dependent upon a strain component that is recoverable – i.e. it is a pseudo elastic phenomena and not dependent upon the irrecoverable secondary creep.

2.1.2. The level of recoverable creep strain

In the temperature ranges of interest to AGRs current creep models assume that only the primary creep component is recoverable and secondary creep is assumed to be irrecoverable. As noted above, there is evidence from the thermal annealing experiments that the strain recovery was in excess of the 1 esu that is associated with primary creep.

There is also evidence to suggest that when crept samples are subjected to an *irradiation* anneal, i.e. further irradiation with the external load removed, strain recovery significantly in excess of 1 esu is observed. Fig. 6 illustrates this and shows strain recovery in excess of 3 esu from a BR-2 sample initially irradiated under a 6.2 MPa compressive stress, from which the load was removed at $\sim 50 \times 10^{20}$ n/cm^2 EDN [9]. Unfortunately, the associated CTE data is not available on these samples and so it is not possible to confirm the CTE recovery that might be expected based on the results of the thermal annealing experiments. Inspection of Fig. 6 suggests that the fluence required for the full irradiation anneal strain recovery is somewhere in the region of $30–40 \times 10^{20}$ n/cm^2 EDN which is well in excess of the fluence required for primary creep alone to recover.

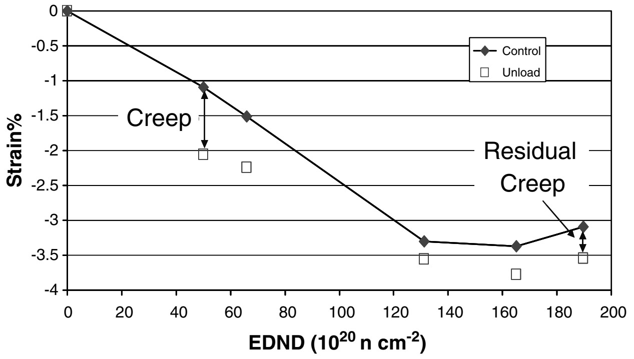

Fig. 6. Irradiation anneal strain recovery of a compressive sample irradiated in BR-2.

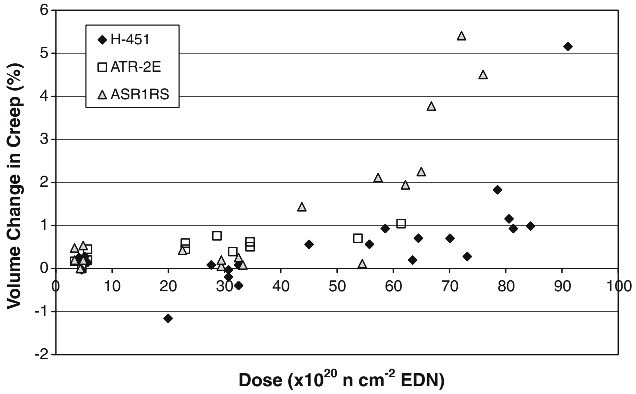

Fig. 7. Volume change of H451 graphite subject to tensile creep at 900 °C [2].

2.1.3. Lateral strain and creep-induced volume change

Fig. 7 shows US data for the volume change of graphite subjected to tensile creep, irradiated in the HFR at Petten to high fluence at 900 °C [2]. Inspection of Fig. 7 suggests that a small volume change occurs at low fluence followed by a period of creep at virtually constant or slightly diminishing volume followed by significant volume change at high fluence.

2.1.4. Summary of findings

Following the review of the available low fluence experimental data, any revised graphite irradiation induced creep model must be consistent with the following key behaviours:

1. Significant irradiation induced creep recovery well in excess of 1 esu.
2. Associated recovery of irradiation induced creep on CTE.
3. Extended fluence required to effect full strain and CTE recovery.
4. Diminishing effect of irradiation induced creep on CTE at higher fluences.
5. Creep at virtually constant volume during the secondary phase.

Additionally, temperature dependence of the creep coefficients is also required especially for operating temperatures higher than 600 °C.

3. Development of new model

The remainder of this paper presents an alternative interpretation of the available low fluence data which attempts to address the above key behaviours and is then applied to high fluence data.

3.1. Thermal anneal

As discussed earlier, tensile and compressive specimens along with their unstressed 'controls' were irradiated in BR-2 and subsequently thermally annealed [9]. Fig. 8 shows the residual strains following each stage of the thermal anneal[2]. These thermal annealing experiments suggest that a proportion of what has traditionally been assigned to secondary creep is, in fact, recoverable. This has significant implications for the fluence dependence of irradiation creep, since the irrecoverable strain, defined as the residual strain post thermal anneal, appears to exhibit a linear response with fluence,

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[2] Unfortunately, the control sample for the compressive specimen was not thermally annealed and an estimate of the residual strain has been obtained by scaling from the control dimensional change at 35×10^{20} n/cm^2 EDN.

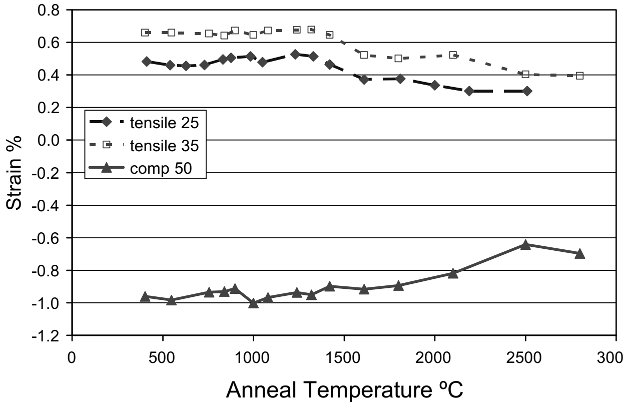

Fig. 8. Residual strain of two tensile specimens and one compressive specimen following thermal anneal.

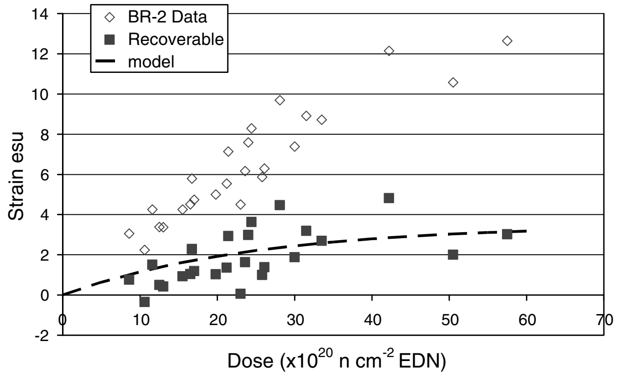

Fig. 9. Recoverable strain derived from BR-2 data.

equivalent to a reduced secondary creep coefficient of ~0.15 (c.f. $\beta = 0.23$).

The implied associated recoverable strain has been derived from the BR-2 data (Fig. 1) and is shown in Fig. 9. The recoverable strain appears to be saturating at approximately 3 initial esu beyond approximately 30x10^{20} n/cm^2 EDN. The trend shows a remarkable similarity to that for CTE (Fig. 2) and suggests that the mechanism inducing the change in CTE is linked to the component of strain that can be recovered by thermal annealing.

3.2. Alternative creep model

Historically, Irradiation induced creep data has been collected for a number of graphites for a range of fluence and irradiation temperature conditions. The data provide reasonable confirmation that, not only can irradiation induced creep be represented by a viscoelastic model, but also that the underlying creep phenomena is independent of graphite type [3]. By drawing together some of this information from UK and US creep experiments in this review, strong evidence has been found to suggest that:

- The strain induced change in CTE is not a function of secondary creep strain, but saturates after a dose of $\sim 30 \times 10^{20}$ n/cm^2 EDN.
- There is evidence, from both thermal and irradiation annealing, for a recoverable strain several times that of primary creep, and a lower associated secondary creep coefficient than has been hitherto assumed.
- The dose at which the recoverable strain saturates bears a striking similarity to that for the saturation of the CTE change.

An alternative creep model is therefore proposed without the need for interaction strain and containing one additional term, recoverable creep:

$$\varepsilon_c = \frac{\alpha k_1}{E_0}\exp^{-k_1\gamma 1}\int_0^{\gamma^1}\frac{\sigma}{SW}\exp^{k_1\gamma}d\gamma + \frac{\omega k_2}{E_0}\exp^{-k_2\gamma 1} \times \int_0^{\gamma^1}\frac{\sigma}{SW}\exp^{k_2\gamma}d\gamma + \frac{\beta}{E_0}\int_0^{\gamma^1}\frac{\sigma}{SW}d\gamma. \qquad (4)$$

All components are proportional to esu and the effects of structural changes and radiolytic oxidation are also included. The rates of saturation of the primary and recoverable creep components are controlled by the dose constants k_1 and k_2. The first and last terms in Eq. (4) are primary and secondary creep as before with the middle term being recoverable creep.

Primary creep is still fast acting but (in the AGR temperature range) appears to act on a longer fluence scale equivalent to that associated with the Young's Modulus pinning [1], $k_1 = 0.1$ and saturates at 1 esu ($\alpha = 1$). The irrecoverable creep is synonymous with secondary creep, but with a coefficient, β, derived from the irrecoverable strain post thermal anneal, as 0.15 per 10^{20} n/cm^2 EDN in the AGR temperature range.

As a first step, after inspection of Fig. 9 a similar mathematical form to primary creep has been chosen for the recoverable creep strain component. The recoverable strain as derived from BR-2 data, builds up (and recovers) over a much longer fluence scale, $k_2 = 0.06$ and saturates at 3 esu ($\omega = 1$).

The lateral creep strain ratios for primary and recoverable creep are assumed to be the Poisson's ratio and, in accordance with the suggestion of Fig. 7, secondary creep is assumed to occur at constant volume.

3.3. Alternative creep strain model predictions

Fig. 10 demonstrates, unsurprisingly, that the new creep model represents the original BR-2 data very well. A better test of the new model is Fig. 11, which presents the prediction of the behaviour of the BR-2 irradiation annealing experiment shown in Fig. 6. The prediction is obtained by adding the unstressed dimensional change data post load removal to the strain recovery of the loaded specimen. The prediction is a reasonable representation of the data but slightly underpredicts the recovery. The underprediction may be a result of the dimensional change data 'curve slipping' or an indication that the recoverable strain has been underestimated for this particular sample.

Fig. 12 shows the new model applied to some high dose ATR-2E tensile creep data irradiated at 500 °C in HFR, Petten [10]. The prediction matches the observation remarkably well up to significant

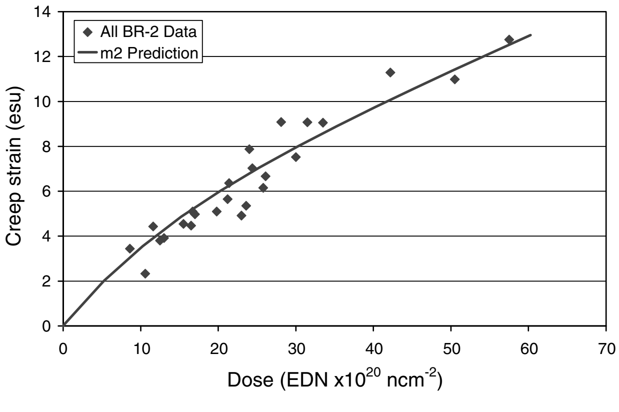

Fig. 10. Alternative creep model prediction with BR-2 data.

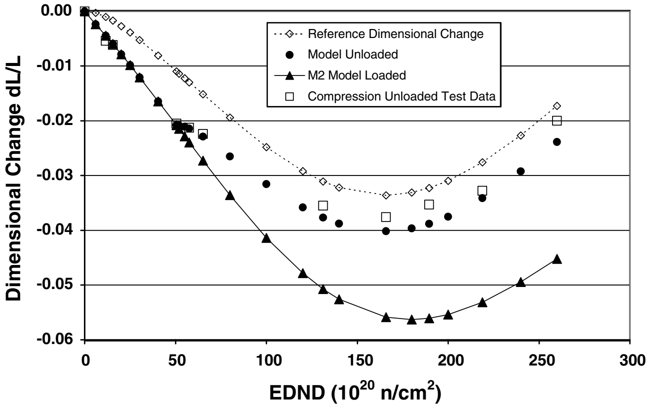

Fig. 11. Irradiation anneal strain recovery of a 6.2 MPa compressive BR-2 specimen.

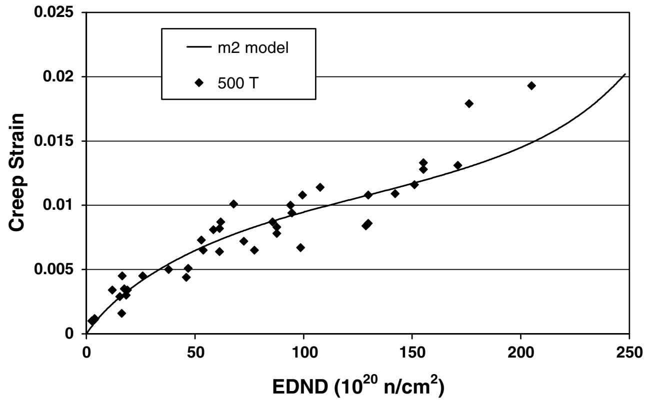

Fig. 12. ATR-2E tensile creep data irradiated at 500 °C in Petten.

fluence of approximately 160×10^{20} n/cm^2 EDN. Only beyond this fluence does the new model prediction deviate from the data (two points) with a delay in the increase in creep strain at high doses that is often referred to as the 'tertiary' creep phase.

Fig. 13 shows the corresponding compressive creep data, irradiated at 550 °C. The prediction overpredicts the data slightly but follows the trend remarkably well up to a significant fluence of approximately 160×10^{20} n/cm^2 EDN. Beyond this fluence the compressive prediction also indicates a 'tertiary' creep, but the data do not extend into this region. The data indicate a possible difference between tensile and compressive creep which requires further investigation, but may simply indicate that the effect of applied load on the static modulus has been overestimated for the compressive creep experiment.

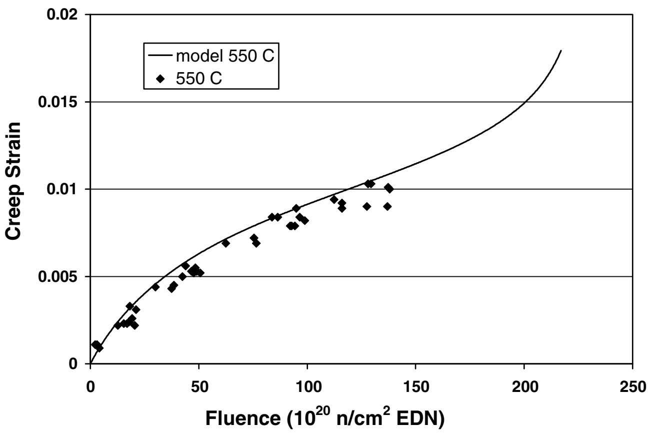

Fig. 13. ATR-2E compressive creep data irradiated at 550 °C in Petten.

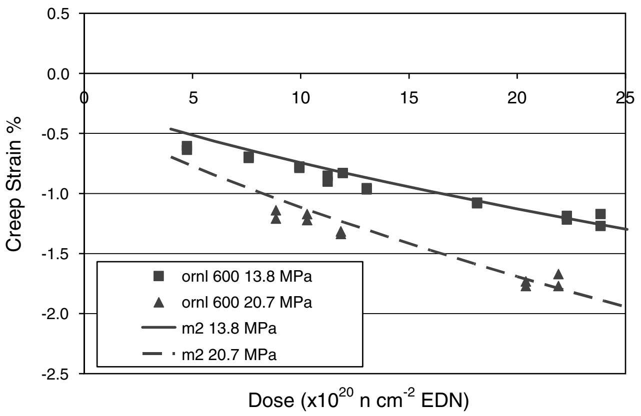

Fig. 14. H451 Graphite creep strain at 13.8 and 20.7 MPa irradiated at ORNL at 600 °C.

The new creep model has also been applied to data obtained on H451 graphite irradiated at ORNL [11]. Fig. 14 shows the creep strain prediction assuming the same constants as derived for the BR-2 and Petten data. Good agreement is obtained, albeit at low fluence.

3.4. Lateral creep strain ratios and volume change

Fig. 15 shows the lateral creep strain ratios obtained from IM1-24 samples irradiated at 1050 °C[3] and the prediction, which is asymptoting to 0.5 as the secondary creep component dominates. The prediction shows reasonable behaviour at low total creep strain but appears to overestimate the lateral strain ratio at higher creep strains (presuming the effect to be genuine, and not merely the result of measurement uncertainties).

Fig. 16 shows the H451 density change (or volume change assuming no mass loss during the irradiation) along with predictions from the new model using the parameters derived from the BR-2 experiments. The model underpredicts the volume change, although the form of the measured volume change data is well represented. Significant improvement in the volume change predictions can be obtained by adjustment of the model parameters (increasing recoverable strain) without compromising the fit to creep strain.

Although the agreement between the revised model and experimental data is encouraging, it is clear that this aspect of the creep behaviour requires some further investigation.

3.5. Effect of externally applied strain on CTE using new model

Fig. 2 suggests that the change in CTE due to an external applied strain does not monotonically increase with increasing 'apparent' creep strain and also suggests that the change in CTE due to an external applied strain occurs on a similar fluence timescale to the build up of recoverable strain. Fig. 4 suggests that the effect of an applied external strain on CTE should be factorial and not additive. UK and US CTE data [7,8,11] have therefore been re-examined using factorial functions. Fig. 17 shows the measured factorial change in CTE as a function of primary and recoverable strain (assumed to be saturated at 3 esu). There is good consistency

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[3] For this comparison, the constants used in the new model have had to be modified due to the high irradiation temperature.

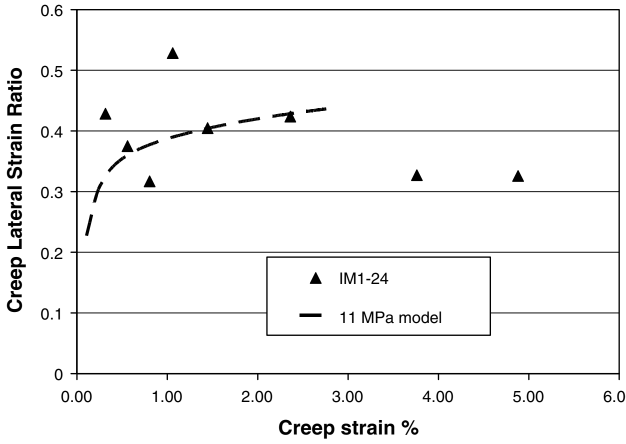

Fig. 15. Lateral creep strain ratios of IM1-24 specimens irradiated at 1050 °C.

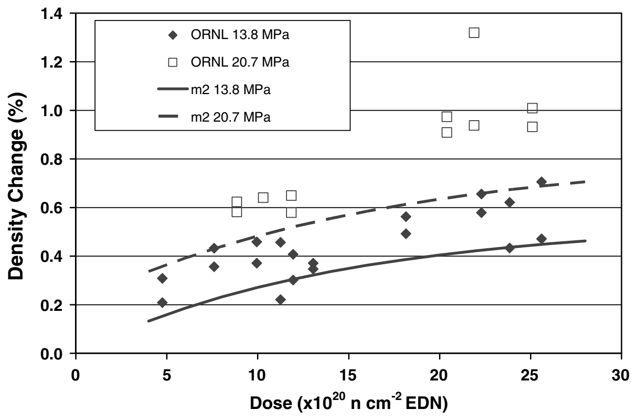

Fig. 16. H451 Graphite density change at 13.8 and 20.7 MPa irradiated at ORNL at 600 °C.

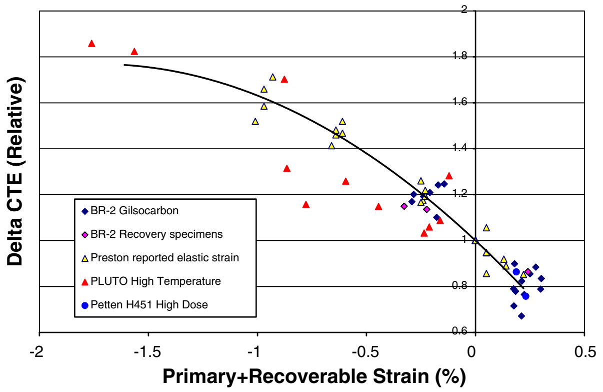

Fig. 17. Factorial change in CTE (20–120 °C) due to an externally applied strain.

between datasets, including UK experiments on the effect of elastic strain on unirradiated CTE [4].

This suggests that there is good evidence to suggest that the CTE would indeed recover if the load on the specimens was removed or reversed. This is an important observation arising from the new interpretation of the data, and is worthy of investigation as part of any future MTR creep programme.

4. Discussion

It has been demonstrated that a significant portion of irradiation induced total graphite creep strain is recoverable, substantially in excess of the 1 esu primary creep currently assumed. As a result, the corresponding rate of secondary creep is found to be substantially lower than hitherto assumed.

Using UK data obtained in BR-2, the recoverable strain has been determined to be 1 esu for primary creep and 3 esu for 'recoverable' creep. The secondary creep coefficient has been determined to be approximately constant at 0.15 per 10^{20} n/cm^2 EDN in the temperature range 350–600 °C. The alternative model has been applied to UK, US and German data and represents the creep strain data very well even up to high fluence, 160×10^{20} n/cm^2 EDN and does not require interaction strain.

It has been demonstrated that the creep strain induced changes to material properties, especially CTE, are not a function of secondary creep strain but, instead, appear to be proportional to the elastic, primary and recoverable strain components. Therefore the creep-induced change in material properties should also recover during an irradiation anneal, as can be considered to be taking place during stress reversal within an operating AGR moderator block.

At present, we are not aware of any controlled experimental data to confirm these implications of material property recovery. Therefore, any future graphite creep experiments should recognise the potential significance of property recovery and should ideally be designed to differentiate between the model proposed in this paper and any alternative methodology.

5. Conclusions

A re-evaluation of the information from UK, US and German creep experiments, has found that:

- There is evidence, from both thermal and irradiation annealing, for a recoverable strain several times that of primary creep, and a lower associated secondary creep coefficient than has been hitherto assumed.
- The strain induced change in CTE is not a function of secondary creep strain, but saturates after a threshold fluence.
- The fluence at which the recoverable strain saturates bears a striking similarity to that for the saturation of the CTE change.
- An alternative creep model has been developed and validated against low fluence creep data and predicts the high fluence creep behaviour of ATR-2E data reasonably well without the need for interaction strain.
- Some further investigation is required into lateral creep strain ratios and associated volume change.

Acknowledgement

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