Modelling Property Changes in Graphite Irradiated at Changing Irradiation Temperature

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Abstract

A new method is proposed to predict the irradiation induced property changes in nuclear graphite, including the effect of a change in irradiation temperature. The currently used method to account for changes in irradiation temperature, the scaled fluence method, is demonstrated. The proposed method, which makes use of evolution equations to describe the rate of change of the property of interest, is described in detail. The property value is obtained by integrating the evolution equation. A sudden change in irradiation temperature instantaneously affects the rate of change, but the property value itself remains continuous. Numerical examples are used to demonstrate the increased accuracy of the proposed method.

1   Introduction

Nuclear graphite refers to graphite that is manufactured for the specific use as a moderator in a nuclear reactor. The material properties of this graphite are required for safe mechanical design. However, due to irradiation these mechanical properties change. These property changes are usually available as isothermal property tables or curves e.g. [1]. Curve fits to this data provide closed-form expressions used to model property changes during isothermal operation of a nuclear reactor.

However, during start-up and shutdown it is inevitable that some property changes will occur during non-isothermal conditions. Also, during the lifetime of a nuclear reactor it is common that the operating temperature will change periodically, primarily due to operational and licensing requirements. These scenarios make it necessary to be able to predict property changes due to changes in irradiation temperature. The currently accepted method to predict these property changes if a change in irradiation temperature occurs is the KTA 3232 rule [2], which recommends the scaled fluence rule proposed by Price and Haag [3].

2   The scaled fluence method

Consider the isothermal property curves depicted in Figure 1 (a), at temperatures T_a and T_b. If at some fluence γ_1 the irradiation temperature suddenly changes from T_b (before) to T_a (after),
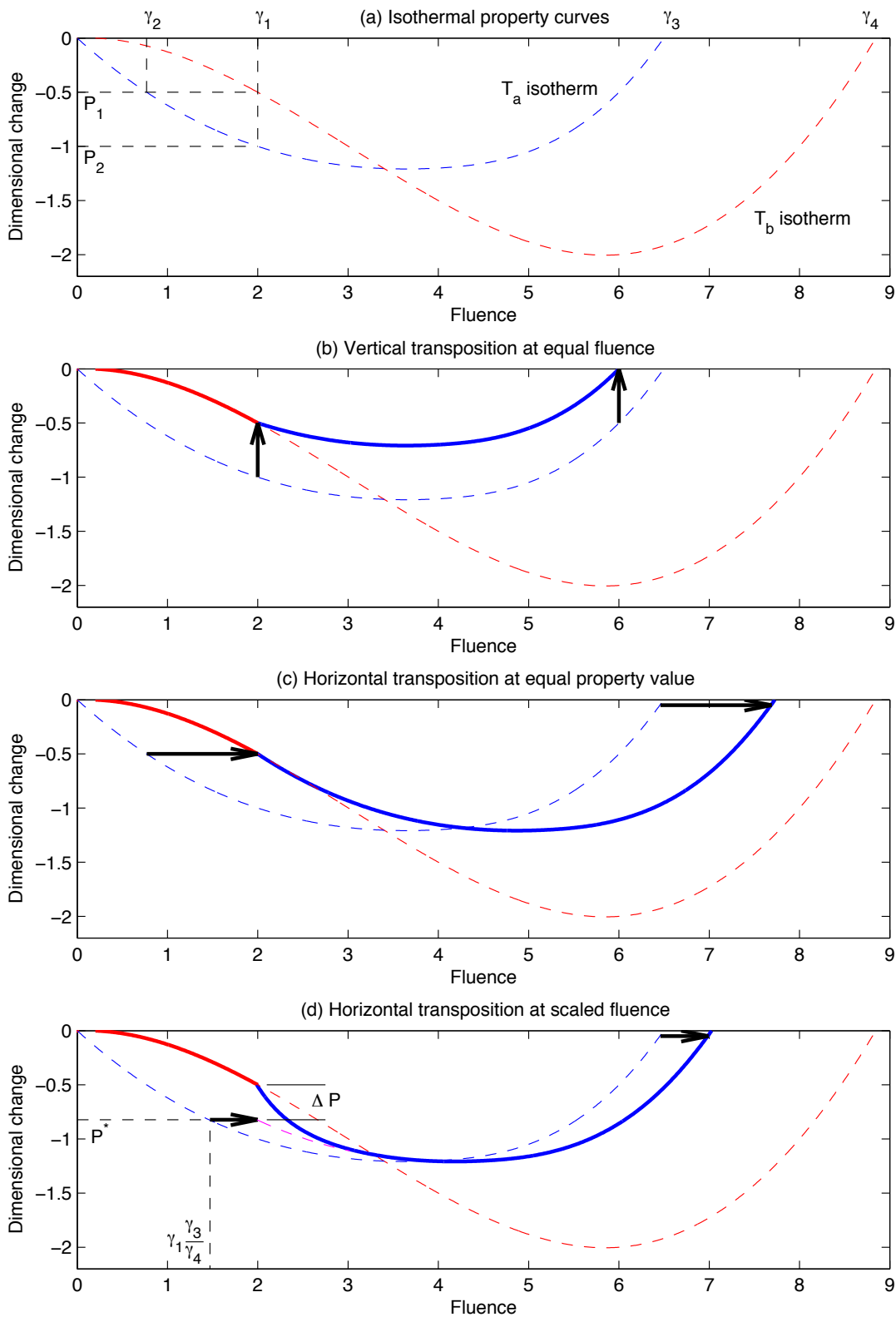
Figure 1: Illustration of the three rules investigated by Price and Haag.

it is desirable to predict the resulting property value. Price and Haag [3] investigated three methods to predict this required property change. The three methods are:

1. Vertical transposition at equal fluence (Figure 1 (b)): This rule states that the T_a proporty isotherm at γ_1 is shifted vertically to join the T_b property isotherm at γ_1. The vertical shift is $P_1 - P_2$. No theoretical justification for this rule has been proposed.

2. Horizontal transposition at equal property value (Figure 1 (c)): This rule states that the T_a isotherm at property value P_1 is shifted horizontally to join the T_b propert isotherm at γ_1. The horizontal shift is $\gamma_1 - \gamma_2$. This rule has some theoretical justification, if it is assumed that a given property (dimensional change in this case) corresponds to a given state of total irradiation.

3. Horizontal transposition at a scaled fluence (Figure 1 (d)): This rule requires some specific knowledge on the behaviour of nuclear graphite at high fluence. Of particular interest are irradiation induced dimensional changes. Irradiated graphite gradually shrinks (called Wigner shrinkage strain), reaches a maximum shrinkage and then the shrinkage becomes smaller to eventually reach a point of expansion. The fluence at which the dimensional change curve transitions from shrinkage to expansion (i.e. dimensional change equals zero) is called the lifetime fluence, since it can be regarded as the "usable lifetime" of the graphite at the particular temperature. This rule scales the fluence axes for all the isothermal curves by their respective lifetime fluences i.e. all the curves now cross the zero dimensional change line at a scaled fluence of one. Horizontal transposition now occurs at equal scaled fluence. For the specific example, the horizontal shift is $\gamma_1(1 - \frac{\gamma_3}{\gamma_4})$. This procedure usually results in a gap (ΔP) between the two isothermal curves. This gap is assumed to decrease exponentially according to the expression

$$P = P^* + \Delta P e^{-\gamma/\tau} \qquad (1)$$

where P is the predicted property, P^* is the property value of the transposed isothermal curve, γ is the fluence measured from the temperature change point and τ is a time constant.

Price and Haag found that the scaled fluence method is more accurate than the other two rules. They applied their technique to predict the changes in linear dimensional change, Young's modulus, thermal conductivity and thermal expansivity after a change in irradiation temperature.

3   Evolution equation method

In this paper an alternative method is proposed to model material properties. Similar to state variable based hardening laws for use in plasticity (such as the mechanical threshold stress model [4]), the approach is to rather evolve the particular property from some known initial condition. Specifically, an evolution equation of the form

$$\frac{dP}{d\gamma} = f(P, T) \qquad (2)$$

is proposed, where P is the material property of interest, f is some evolution equation to be determined for this property, T is the temperature and γ is the irradiation fluence. The motivation

for such an evolution equation based model is the assumption that if the operating conditions suddenly change, the *rate* of change of the particular property will change instantaneously, but the property value itself remains continuous. This requirement is statisfied for all three rules studied by Price and Haag. In the case of rule three, the continuity of the property value itself is enforced after the fact by assuming that the gap ΔP decreases exponentially.

The potential of this approach is demonstrated by applying it to the linear dimensional change (Wigner strain) in irradiated graphite. Isothermal curves for gilsocarbon-based graphite (Dragon grade 95) are presented in Figure 2. Notice that the 800°C curve contains three regions: initial hardening, then softening and finally hardening again. A versatile description would therefore be to model dimensional change as a combination of i) a curve that gradually saturates to a large negative value, ii) a curve that quickly saturates to a small positive value and iii) a curve that increases monotonically. The dimensional change $\frac{\Delta L}{L}$ is thus considered to be the sum of three mechanisms i.e.

$$\frac{\Delta L}{L} = \left(\frac{\Delta L}{L}\right)_1 + \left(\frac{\Delta L}{L}\right)_2 + \left(\frac{\Delta L}{L}\right)_3. \tag{3}$$

In such a description it is implicitly assumed that three independant mechanisms are responsible for the observed dimensional change. These three curves can be obtained by integrating the differential equations:

$$\frac{d(\frac{\Delta L}{L})_1}{d\gamma} = \theta_1 \left(1 - \frac{\left(\frac{\Delta L}{L}\right)_1}{S_1}\right) \tag{4}$$

$$\frac{d(\frac{\Delta L}{L})_2}{d\gamma} = \theta_2 \left(1 - \frac{\left(\frac{\Delta L}{L}\right)_2}{S_2}\right) \tag{5}$$

$$\frac{d(\frac{\Delta L}{L})_3}{d\gamma} = \theta_3 \left(1 + \frac{\left(\frac{\Delta L}{L}\right)_3}{S_3}\right) \tag{6}$$

where $\theta_1, \theta_2, \theta_3, S_1, S_2$ and S_3 are temperature dependent material parameters that are obtained by solving an inverse problem to best fit the experimental data. For this particular property the initial conditions $\left(\frac{\Delta L}{L}\right)_i = 0$ for $i = 1, 2, 3$ are used. Although analytical solutions exist to these differential equations, it is common practise to implement material models of this type into finite element codes, where the differential equations are integrated numerically. It is however noteworthy that the analytical solution to each of these differential equations is an exponential function, demonstrating similarity to the exponential function used in the scaled fluence method.

For this particular problem, the material parameters for each temperature are treated independently. Hence, a total of 24 parameters (6 parameters for each of the 4 temperatures) are required to model the isothermal curves in Figure 2. These 24 parameters are stored in the single design vector $\boldsymbol{x} = [\boldsymbol{x}_{600°C} \; \boldsymbol{x}_{800°C} \; \boldsymbol{x}_{900°C} \; \boldsymbol{x}_{1200°C}]$, where the design vector for each of the temperatures contain the six parameters $\boldsymbol{x}_{T°C} = [\theta_1 \; S_1 \; \theta_2 \; S_2 \; \theta_3 \; S_3]_{T°C}$. The inverse problem determines the material parameters that minimize the least square error between the experimental property data and the numerically computed data. Specifically it was found that the formulation

$$\text{Minimize w.r.t. } \boldsymbol{x} \quad f(\boldsymbol{x}) = \alpha_1 f_1(\boldsymbol{x}) + \alpha_2 f_2(\boldsymbol{x}) + \alpha_3 f_3(\boldsymbol{x}) \tag{7}$$

provides good quality fits, where f_1 is the least square error for the isothermal data in Figure 2, f_2 is the least square error error for the irradiation temperature change data in Figures 4 and 5,

and f_3 is the norm of $\boldsymbol{x}$. The scaling factors α_1, α_2 and α_3 can be adjusted to weight the importance of the three functions. Since much more isothermal data is available than irradiation temperature change data, the choice of $\alpha_1 = 1$ and $\alpha_2 = 10$ ensures that the irradiation temperature change data is fitted well. Function f_3 is necessary due to the particular choice of the model. Since $\left(\frac{\Delta L}{L}\right)_1$ is always negative and $\left(\frac{\Delta L}{L}\right)_2$ and $\left(\frac{\Delta L}{L}\right)_3$ are always positive, many different combinations exist where the sum of all three components appear more or less the same. In order to reduce this effect, the norm of $\boldsymbol{x}$ is added to the cost function. A value of $\alpha_3 = 0.01$ proved to be sufficient to prevent unnecessarily large values for S_1, S_2 and S_3. The optimal material parameters that accurately predict the isothermal and irradiation temperature change data are summarized in Table 1. Figure 2 illustrates the agreement between the isothermal data and the evolution equation based model. The relative contributions of $(\Delta L/L)_1$, $(\Delta L/L)_2$ and $(\Delta L/L)_3$ to $(\Delta L/L)$, for the four isothermal curves, are illustrated in Figure 3. Notice that each of the three mechanisms have a non-trivial contribution to each of the four isothermal curves.

Parameter	600°C	800°C	900°C	1200°C
θ_1	-0.7010	-0.8495	-2.1868	-2.7504
S_1	-11.1390	-6.0009	-16.0001	-5.8600
θ_2	0.6860	1.0363	1.9216	1.3373
S_2	1.9870	2.2443	5.4285	2.6262
θ_3	0.0394	0.0090	0.2461	0.2366
S_3	0.1596	0.0160	0.8092	2.1393

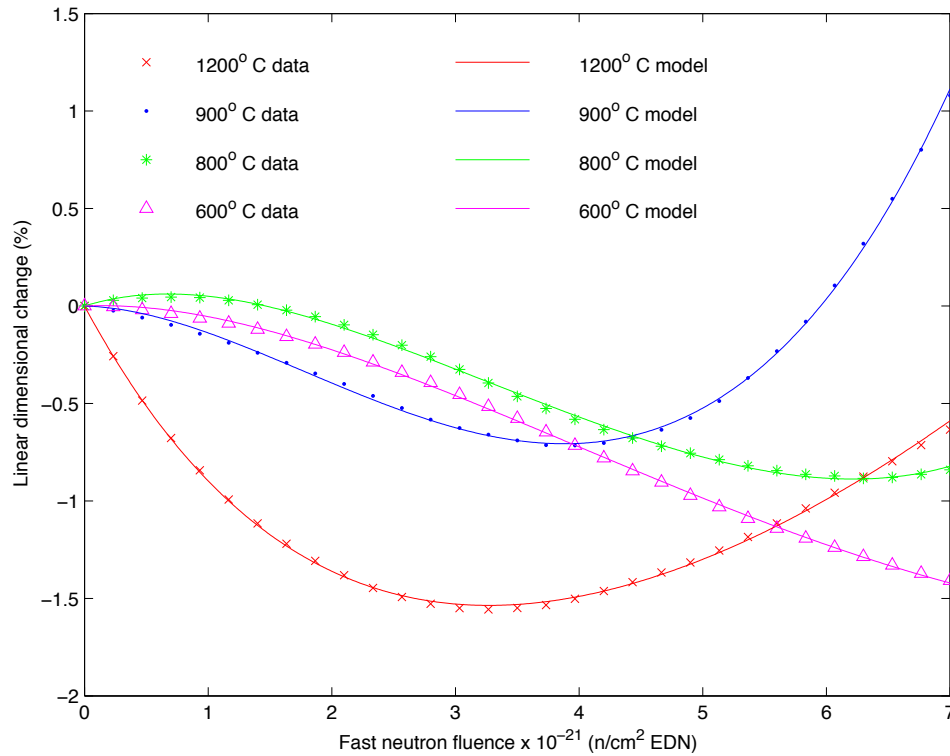**Table 1:** Optimal material parameters obtained by solving an inverse identification problem.

Figure 2: Ability of the proposed evolution equation model to reproduce the isothermal linear dimensional change curves for Dragon grade 95 graphite. Data from [3].
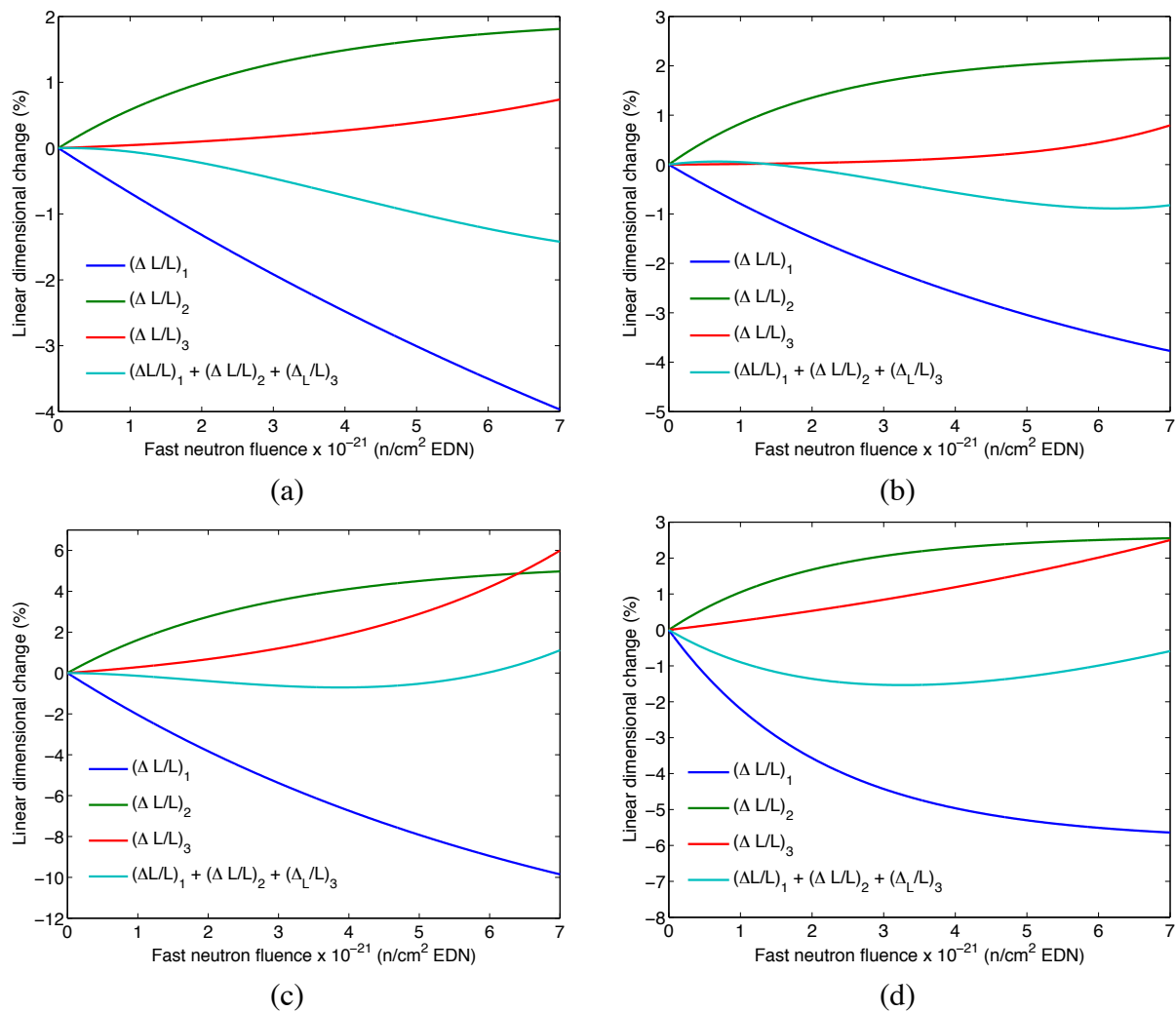
Figure 3: $(\Delta L/L)$ as a sum of $(\Delta L/L)_1$, $(\Delta L/L)_2$ and $(\Delta L/L)_3$ for (a) 600°C, (b) 800°C, (c) 900°C and (d) 1200°C.

Figures 4 and 5 illustrate the ability of the proposed evolution equation based model to predict property values after a sudden changes in the irradiation temperature. Notice the superior accuracy of the proposed method as compared to the scaled fluence method, especially for the case where the irradiation temperature changes from 900°C to 1200°C in Figure 4.

One drawback of the proposed method is the large number of material parameters. This is due to the fact that 6 material parameter values are associated with each temperature. Since the approach followed here is purely phenomenological, problem specific knowledge on how some of these parameters can be expected to vary as a function of temperature is not exploited. If this is done, it can lead to a reduction in the total number of parameters. The main purpose of this work is however to illustrate that an evolution equation based model can be used to predict nuclear graphite property changes, and not that it can do so with a minimal set of parameters. Specifically, the model has the ability to take irradiation temperature change into account.

The ability of the proposed method to take irradiation temperature change into account is directly due to the evolution equation form as stated in Eq. (2). Note that the fluence γ does not appear explicitly on the right hand side. The history of how the particular property evolved
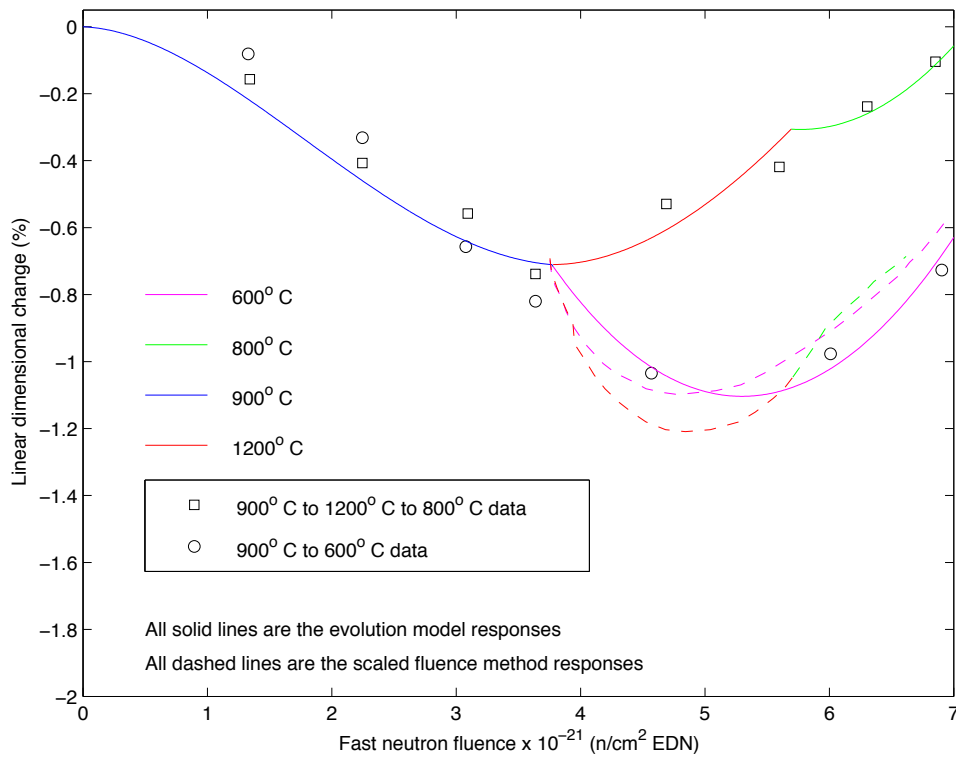
Figure 4: Ability of the proposed evolution equation model to account for changes in irradiation temperature from 900°C, compared to the scaled fluence method. Data from [3].
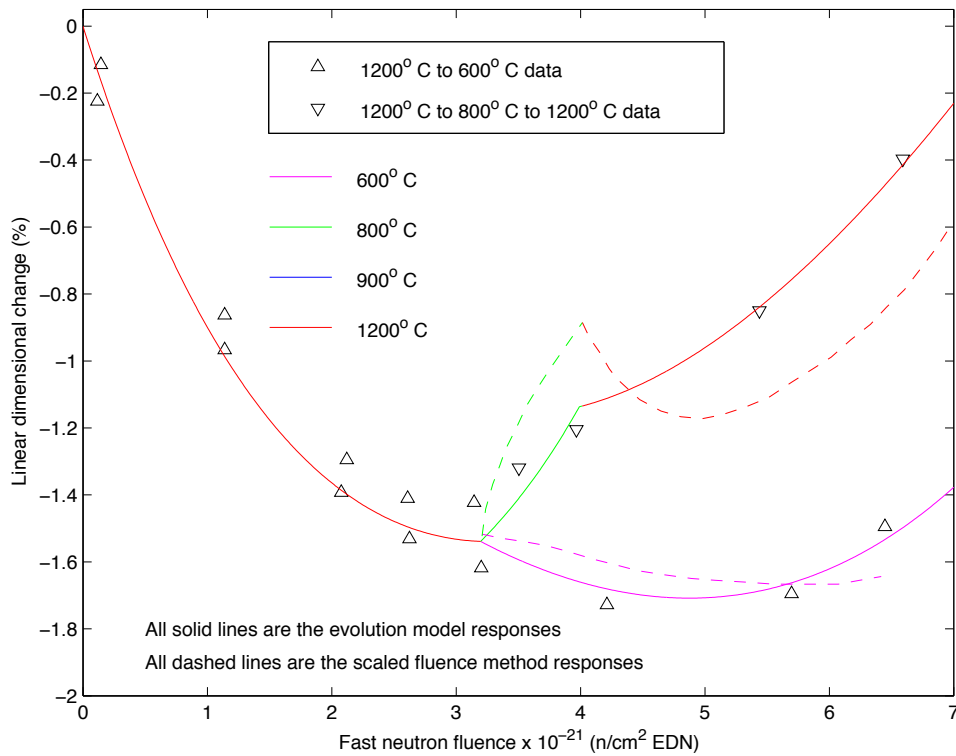

Figure 5: Ability of the proposed evolution equation model to account for changes in irradiation temperature from 1200°C, compared to the scaled fluence method. Data from [3].

to date is implicitly contained within the property itself. Conventional models attempt to describe the Wigner strain by some explicit function of temperature and fluence, usually making use of polynomials, exponentials and power laws. The model by Tsang and Marsden [5] is an example of such a model. It is determined by making use of proper statistical analysis and optimization. However, the goal of their technique is to determine a closed form expression for the Wigner strain in terms of fluence and temperature. Therefore this model cannot be used to accurately predict property changes due to a sudden change in irradiation temperature. Closed form models of this type simply jump instanteneously from one isothermal curve to the other.

4 Conclusion

An novel state variable based evolution equation was proposed to model material property changes due to irradiation. In particular, the method can also predict property changes after a sudden change in irradiation temperature. The proposed method is much more accurate that the currently used scaled fluence method. The form of the model is also well suited for implementing it into a finite element code, where it is common practise to evolve state variables in every time step by numerically integrating the differential equations that describe state variable evolution.

References

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