MECHANICAL PROPERTIES AND OXIDATION OF NUCLEAR GRAPHITES

J.-M. Vidal and T.J. Mays
Department of Material Science and Engineering,
University of Bath, Bath, BA2 7AY, UK

Introduction

Moderator graphites in advanced gas-cooled reactors (AGRs) oxidise radiolytically in the carbon dioxide coolant, Best *et al.* [1], Kelly [2]. This leads to graphite weight loss, mainly in the form of a density reduction via porosity development. In turn, weight losses lead to mechanical properties such as Young's modulus and tensile strength. Reductions in mechanical properties can limit the safe operation and lifetime of moderators. At present the effects of radiolytic oxidation on moderator graphites are assessed from post-irradiation examination data from materials test reactors. Predictions of the safe operation of moderators are largely based on these data. They also involve simple models of radiolytic oxidation in graphites that, to give more confidence in predictions, need further development. This involves (i) measurement of mechanical properties, and characterisation of porosity, in thermally oxidised graphites, and (ii) refinement of models of oxidation weight loss, porosity development and mechanical property changes. This abstract focuses on (i), and indicates initial ideas for (ii).

Experimental

The material studied is an isotropic Gilsocarbon graphite, IM1-24 manufactured by Anglo Great Lakes Co Ltd. The samples supplied were taken from material archived from graphite bricks now used in reactors at Hunterston B AGR station. The microstructure of IM1-24 observed using optical microscopy is illustrated in Figure 1. Key features to note are the equiaxed Gilsocarbon filler phase A, containing lenticular 'onion skin' porosity B, and the matrix phase C containing rounder pores D. Both pore types arise in manufacture.

Bulk density, four-point bend flexural strength, and dynamic (sonic resonance) and static (tensile) Young's modulus, were measured on graphite bars, 100x15x15 mm, both as-received and after thermal oxidation in flowing, pure CO_2 at 900 °C for different times; samples were weighed and measured before and after oxidation to determine the fractional weight loss corrected for external burn-off.

After flexural testing, samples were retained for compression strength and He density measurements, and for microscopic examination and Hg porosimetry.

Results and Discussion

Table 1 lists property values for unoxidised IM1-24 graphite, which compare favourably with established values for this material. It may be noted that open porosity is mainly in the binder phase, while closed porosity is mainly in the filler.

Figure 2 is a plot of dynamic Young's modulus E for IM1-24 graphite as a function of corrected fractional weight loss w. Superimposed on Figure 2 are fits to the data using the Knudsen equation,

$$E = E_0 \exp(-bw),$$

where E_0 is the modulus at zero weight loss and b is a non-negative decay parameter (1/b is the fractional weight loss at which E_0 is reduced by a factor 1/e ~ 1/3). This equation is a popular model for the change in mechanical properties of graphites and other ceramics with porosity (or oxidation weight loss). The fits were obtained using either a non-linear, Simplex fitting routine or from linear regression of ln E as a function of w.

While both curves are good representations of the data, the non-linear fit passes though the zero weight loss data, unlike the ln-linear fit. Thus the former method - which implies proportionate rather than additive statistical errors - might be better model for these data. Estimated b values for the two curves are also different: the higher b for the non-linear fit means more rapid decay in E with increasing w. In Figure 3, the residuals of the non-linear and ln-linear fits are plotted as a function of weight loss. It can be seen at low weight losses that both fits overpredict the modulus while both fits underpredict the modulus at high weight losses. This indicates a degree of bias in both fits.

To explore this bias, non-linear Knudsen fits were carried out on low and high weight loss data ($w < 0.08$ and $w > 0.08$ respectively), Figure 4. Approximately the same number of data points are in each range.

Figure 4 shows that low weight loss data yield high estimated values of E_0 and b, while high weight loss data yield low values of these two parameters. This suggests that a single Knudsen fit is inappropriate for the whole dataset. If it is assumed as usual that Knudsen parameters are related to microstructure, especially pore structure, then the different fit ranges imply selective development of structure with weight loss. This is currently being explored using the techniques referred to in the experimental section, especially pore structure analysis. One idea being followed up is that closed pores in the filler particles, Figure 1, are opened at low weight losses and start to oxidise and change shape, resulting in a change in the way in which mechanical properties are reduced.

References

1. Best JV, Stephen WJ, Wickham AJ. Prog Nucl Energy 1985; 16(2):127-177.
2. Kelly BT. Prog Nucl Energy 1985; 16(2):73-96.
3. Kelly BT. Physics of Graphite. UK: Applied Science Publishers. 1981.

Acknowledgement

We thank British Energy Generation Ltd for financial support.

Table 1. Properties of unoxidised IM1-24 graphite.

Property	Mean value
dynamic Young's modulus, E / GPa	11.6
flexural strength, σ_F / MPa	32.8
compression strength, σ_C / MPa	62.5
bulk density, ρ / g cm^{-3}	1.82
open porosity, V_O / %	8.1
closed porosity, V_C / %	11.5

Figure 1. Optical microstructure of IM1-24 graphite.

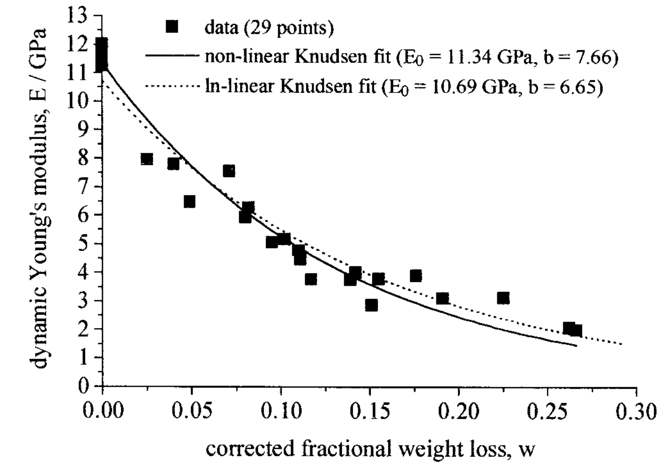

Figure 2. Dynamic Young's modulus as a function of corrected fractional weight loss for IM1-24 graphite. Fits are obtained using ln-linear and non-linear methods.

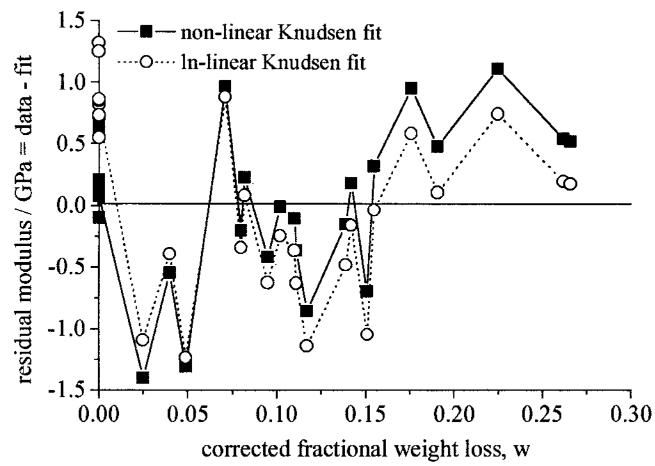

Figure 3. Residuals of the ln-linear and non-linear fits as a function of fractional weight loss.

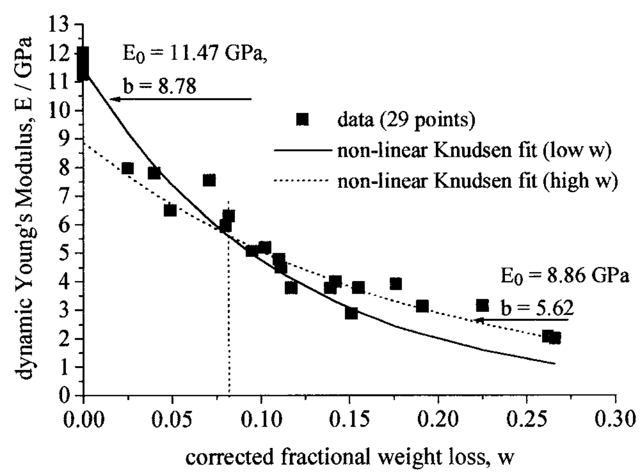

Figure 4. Dynamic Young's modulus as a function of corrected fractional weight loss for IM1-24 graphite. Fits are obtained using a non-linear method applied to low and high weight loss data.