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High-temperature annealing of graphite: A molecular dynamics study

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HIGHLIGHTS

- Changes to the AIREBO reactive potential, show progress towards simulating thermal annealing of damaged graphite.
- The reactive potential used in the Carbon model was re-parameterized according to DFT interstitial results.
- The potential for reactive potentials to be accurate and feasible enough, to simulate annealing events in graphite is shown.
- It is shown that the body of experimental work on graphite irradiation damage can be accessed computationally.

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ABSTRACT

A modified AIREBO potential was developed to simulate the effects of thermal annealing on the structure and physical properties of damaged graphite. AIREBO parameter modifications were made to reproduce Density Functional Theory interstitial results. These changes to the potential resulted in high-temperature annealing of the model, as measured by stored-energy reduction. These results show some resemblance to experimental high-temperature annealing results, and show promise that annealing effects in graphite are accessible with molecular dynamics and reactive potentials.

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1. Introduction

The cause of the Windscale nuclear reactor accident (1957) was the release of stored-energy from the graphite moderator that was subject to low temperature radiation damage (the Wigner Effect [1]). As a result, there is much interest in controlling this release by annealing the graphite during reactor operation. As a result of this interest, some computational work has been done in this area [3–6]. This work shows that the AIREBO [7] graphite computational model can produce annealing effects: Specifically, how at 750 K (477 °C), the stored-energy of damaged graphite is almost two orders of magnitude less than the stored-energy of heavily damaged graphite at 300 K (27 °C). The AIREBO carbon potential used in this work has modified parameters which better fit DFT self-interstitial results. This approach of combining DFT interstitial results with empirical methods has become well-established (e.g. Meslin et al. [8]). The annealing results of this modified potential show resemblance to experimental annealing results - which also show a significant drop of stored-energy in graphite at 723 K (450 °C) vs. 300 K (30 °C) - see Fig. 1 below. However, a direct claim of reproducing experimental annealing is not made because of the small cell size used (1000 atoms), which was necessary to counter the long simulation times. Although the modified potential shows significant improvement towards reproducing graphite annealing (compared to the original potential), more work his needed to understand the effects of periodicity in the annealing dynamics of the model.

Fig. 1 below (R.E. Nightingale 1962 [2]) shows the experimental results of the accumulation of stored-energy in graphite at different temperatures, when it is exposed to increasing dosages of damaging neutrons. The dosage is indicated on the x-axis, in units of $\mathrm{M\,w\,d/(A\,t)} \times 10^{+3}$ (not the erroneous $\mathrm{M\,w\,d/(A\,t)} \times 10^{-3}$

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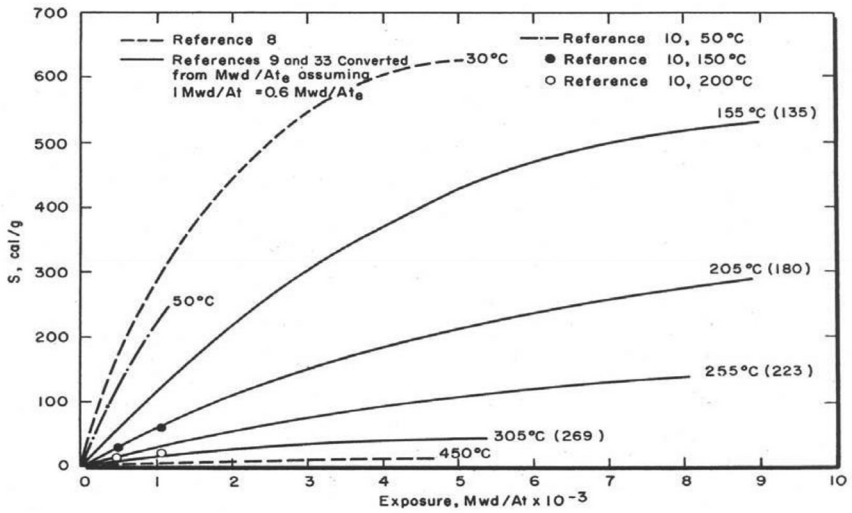

Fig. 1. This figure is taken from "Nuclear Graphite" R.E. Nightingale, Figure 12.2, page 329, 1962 [2]. It shows the experimental accumulation of total stored-energy as a function of dose in graphite at various irradiation temperatures.

shown on the axis in the original Nightingale Figure). M w d means "Mega watt days", and A t means "adjacent ton of Uranium fuel". These exposure units are specific to the measurement setup [2]. The topmost plot shows that at 30 °C(303 K, room temperature), the stored-energy monotonically increases with dosage to an asymptote, where the stored-energy reaches a maximum of 630 cals/g. The accumulation of stored-energy is caused by increasing interstitial defects (or damage) in the graphite, as the damage is worsened by neutron dosage. Dosages of over 5000 M w d/(A t) fail to accumulate any further damage because the graphite material is already saturated with damage/defects.

However, notice what happens at 450 °C (or 723 K). The thermal energy of the carbon atoms in the graphite is sufficient enough to overcome the interstitial defect energy barriers, so there is significantly less stored-energy accumulated (only about 15 cal/g).

2. Airebo parameterization using density functional theory (DFT) interstitial energies

Fig. 2 below was taken from Andris Gulans' work (Gulans 2011) on interstitials in graphite [9]. (a), (b) and (c) show the top view, the side views and the potential energy surface respectively. The most stable defects are the S type interstitial defects (S″, S′ and S), followed by the G type (G′, G) and D. (a) Also shows the reaction pathways between the defects that give rise to the potential energy barriers shown in (c).

In our work, we focus only on the pathway minima that define the defects; where the interstitial is in a local minimum with respect to the reaction coordinate. Potential energy calculations of equilibrium positions (S″, S′, S, G′, G, D) are straightforward because the simulations/calculations are convergent. However potential energy calculations for non-equilibrium parts of the pathway are notoriously ill-defined. Fortunately, simply focusing on correcting these local minima of the AIREBO potential is sufficient for the model to succeed in improving its thermal-annealing properties.

In the plot below (Fig. 3), Gulans' DFT minima results (also shown in Fig. 2c) are shown again; they are the thick black horizontal bars, with the defect type labels just below (S″ S′, S, G′, G and D). To calculate AIREBO results for these minima, the same system configurations were used (as detailed in Gulans 2011), and the

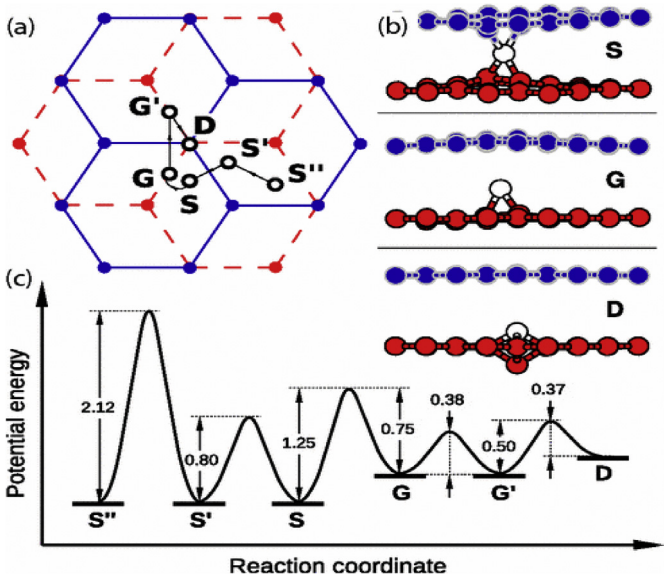

Fig. 2. This figure is from Andris Gulans' 2011 DFT work on interstitials in graphite [9]. (a) and (b) show the top view and the side views of the single interstitials respectively. (c) Shows the potential energy surface; the S type defects (S″, S′, and S) are the most stable, followed by the G type interstitial defects (G′ and G), followed by the D interstitial defect. The potential energy differences are in eV units.

systems were minimized against the original AIREBO potential. The original AIREBO potential (labeled with the AIREBO symbol) is a reasonably good match for the G and D type interstitials.

However, for the S type interstitials, they are significantly too stable relative to the DFT result. This may indicate why the original AIREBO potential can't reproduce thermal annealing at 723 K. In Fig. 3, the absolute potential energy values of the defects are missing from the y-axis; but they are shown (in eV) in Table 1 below.

The modified potential (M) and original AIREBO potential give the same result for D. The philosophy behind the modified potential is to make minimal and theoretically justified changes to the force field. As a result, the priority is to keep the hierarchy of effects the

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