8.8 Analytical Model of Homogeneous Helium Trap-Mutation Evolution in Tungsten —

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OBJECTIVE

The objective of this research is to support the prediction of irradiation damage properties of bulk tungsten-based materials using computational methods.

SUMMARY

Homogeneous trap-mutation events in which helium (He) clusters above a minimum size displace tungsten (W) atoms from their lattice sites are simulated with molecular dynamics. Using a He concentration of up to 4600 appm and temperature up to 2050 K (half the absolute melting temperature), analytical models are formulated to describe the evolution of the number of trap-mutation nucleation sites (N_C) and the number of created self-interstitial atoms (N_I) as a function of He concentration and temperature. Consistent time scaling with respect to concentration (linear scaling) and temperature (Arrhenius scaling) is observed in the case of N_C curves. Therefore, a single analytical model can be obtained. On the other hand, while the linear time scaling still applies among the N_I curves, the Arrhenius time scaling holds only during early stages of the evolution due to the temperature-dependence of the saturation value (larger at higher temperatures). Hence, for N_I, a specific model is formulated for each temperature.

PROGRESS AND STATUS

Introduction

Fusion devices employing the deuterium+tritium reaction will produce helium (He) and neutrons with characteristic kinetic energies of 3.5 and 14.1 MeV, respectively. These neutrons will cause two major challenges in the design of plasma-facing materials and first-wall materials by enabling solid and gaseous transmutation reactions as well as by inducing atomic displacement damage. Transmuation produced helium enhances displacement damage by forming bubbles that potentially facilitates void formation in the materials. On the grain boundaries, helium atoms cause significant interface decohesion. Another detrimental effect of helium is the formation of dendritic structures known as “fuzz” of the tungsten (W) surface [1]. When helium is formed, it is initially at interstitial sites. Subsequently, clustering occurs and large enough clusters (typically of size 7 at 1000 K) start to displace W atoms from their lattice sites, a process known as trap-mutation. The purpose of the current work is to develop an analytical curve to describe the time-evolution of the number of nucleation sites and the number of created W self-interstitial atoms (SIAs) due to a homogeneous trap-mutation process as a function of He concentration and temperature.

Simulation details

The molecular dynamics (MD) technique was employed to simulate trap-mutation. For the W-W interaction, the Finnis-Sinclair type potential developed by Ackland and Thetford [2] was taken, in which the short-range part was then modified to harden the repulsion [3]. Modification was also done at distances relevant to self-interstitial configurations to improve defect formation energies. The simulations were performed using LAMMPS software [4]. The simulations were

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performed in the *NPT* ensemble at zero pressure and with a Nosé-Hoover thermostat. Periodic boundary conditions were applied in all axis coordinates. The simulation cell contains 524,288 W atoms. Helium atoms were introduced randomly at interstitial sites with no initial clustering, i.e. all He atoms were isolated interstitial atoms from each other.

RESULTS

Four different He concentrations at three different temperatures were simulated mostly up to 2 ns. The exact conditions are summarized in Table 1. At 300 K, no trap-mutation events occurred within 2 ns, therefore the data at this temperature will not be analyzed further. We first investigate the number of nucleation sites of a trap-mutation event. As time goes on, a nucleation site may contain more than one vacancy; however it will still be counted as one site. The number of nucleation sites and He atoms is denoted by N_C and N_{He} respectively. Figure 1a. and 1b show the evolution of N_C/N_{He}. Within 2 ns, saturation is reached for the 2050 K curves particularly the 2300 and 4600 appm data, meanwhile for the 1025 K curve only the 4600 appm data reaches saturation. From the 2300 and 4600 appm data, N_C/N_{He} saturates at approximately 0.12 regardless of temperature. A more accurate reading is 0.123 for the 2300 and 4600 appm data at 2050 K.

Table 1. Time and conditions for helium trap-mutation simulations in tungsten. The 1025 K represents 0.25 of the melting temperature (T_m) from the interatomic potential. The simulation cell contains 524,288 tungsten atoms.

	300 K	1025 K (0.25 T_m)	2050 K
134 appm (70 atoms)	2 ns	2 ns	2 ns
460 appm (241 atoms)	2 ns	10 ns	10 ns
2300 appm (1205 atoms)	2 ns	2 ns	2 ns
4600 appm (2410 atoms)	2 ns	2 ns	2 ns

Scaling of time, t, with respect to He concentration, x, is shown in Figure 2a. at 1025 K. In this figure, the proper time corresponds to that of the 4600 appm curve. It appears that the 2300 and 460 appm curves superimpose the 4600 appm curves if their respective timetag is scaled by a factor of 1/2 and1/10. This indicates that time scales linearly with concentration, i.e., data with twice concentration saturates twice faster ($t \rightarrow xt$). This simple scaling is well reproduced between the 4600 and 2300 appm data at 2050 K as shown in Figure 2b. We note that the 460 appm data at 2050 K does not follow this scaling that well, this particular curve saturates at 0.07 instead of 0.12. If this curve is multiplied by a factor of 0.12/0.07 (denoted as red dashed line and labeled as “460 appm adjusted” in Figure 2b), it appears to follow the scaling as well.

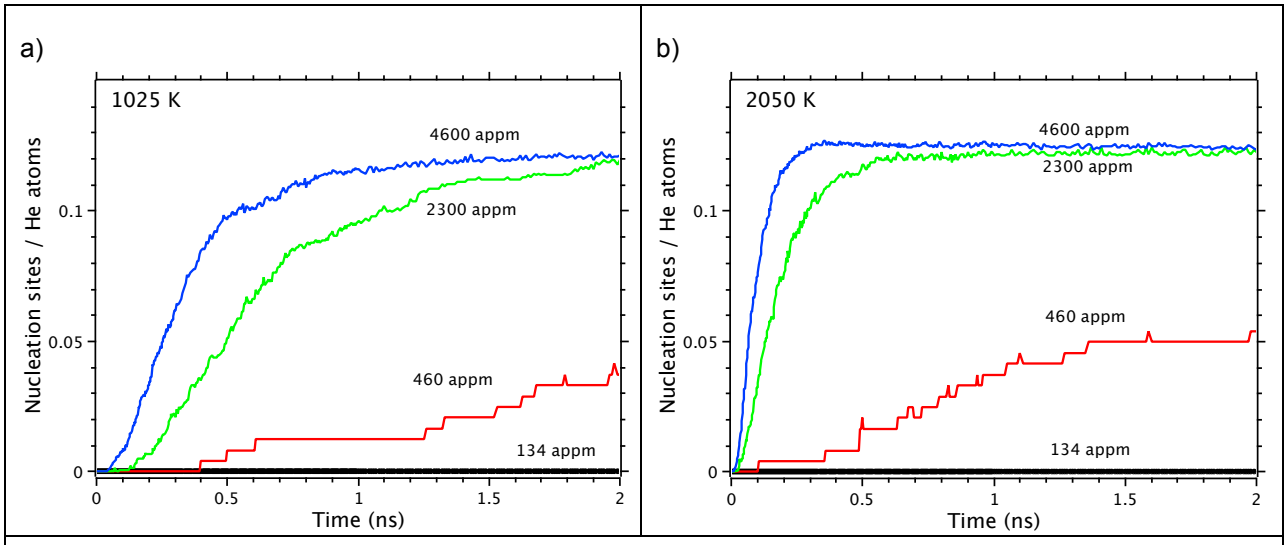

Figure 1. Evolution of the number of nucleation sites for the trap-mutation process due to homogenous He interstitial atoms in bulk W. No trap-mutation events are observed for the 134 appm data within 2 ns of MD simulation.

Scaling of time with respect to temperature, *T*, is shown in Figure 2c for the 2300 appm data. In this figure, the time of the 2050 K data is taken as the proper time. From Figure 2c, it is evident that Arrhenius scaling with effective migration energy of 0.23 eV fits the 1025 K curve to the 2050 K curve remarkably well ($t \rightarrow t \exp(-\mu\beta_T)$, where μ = 0.23 eV and $\beta_T = 1/(kT)$). This scale is also reproduced between the 4600 appm data at 1025 K and 2050 K as shown in Figure 2d. The value of μ = 0.23 eV is slightly larger than the migration energy barrier (at 0 K) of a single He (0.21 eV) from the interatomic potential [3]. In a recent work of Reference [5], it was shown that the migration energy of small He clusters increases with temperature (non-Arrhenius diffusion at high temperatures) due to the increased number of super-basins. For example, the effective migration energy of He_2 cluster is 0.26, 0.41 and 0.69 eV at 0, 1025 and 2050 K respectively. Therefore, the value of μ = 0.23 eV suggests that the trap-mutation nucleation kinetics is governed predominantly by the migration of single He atoms.

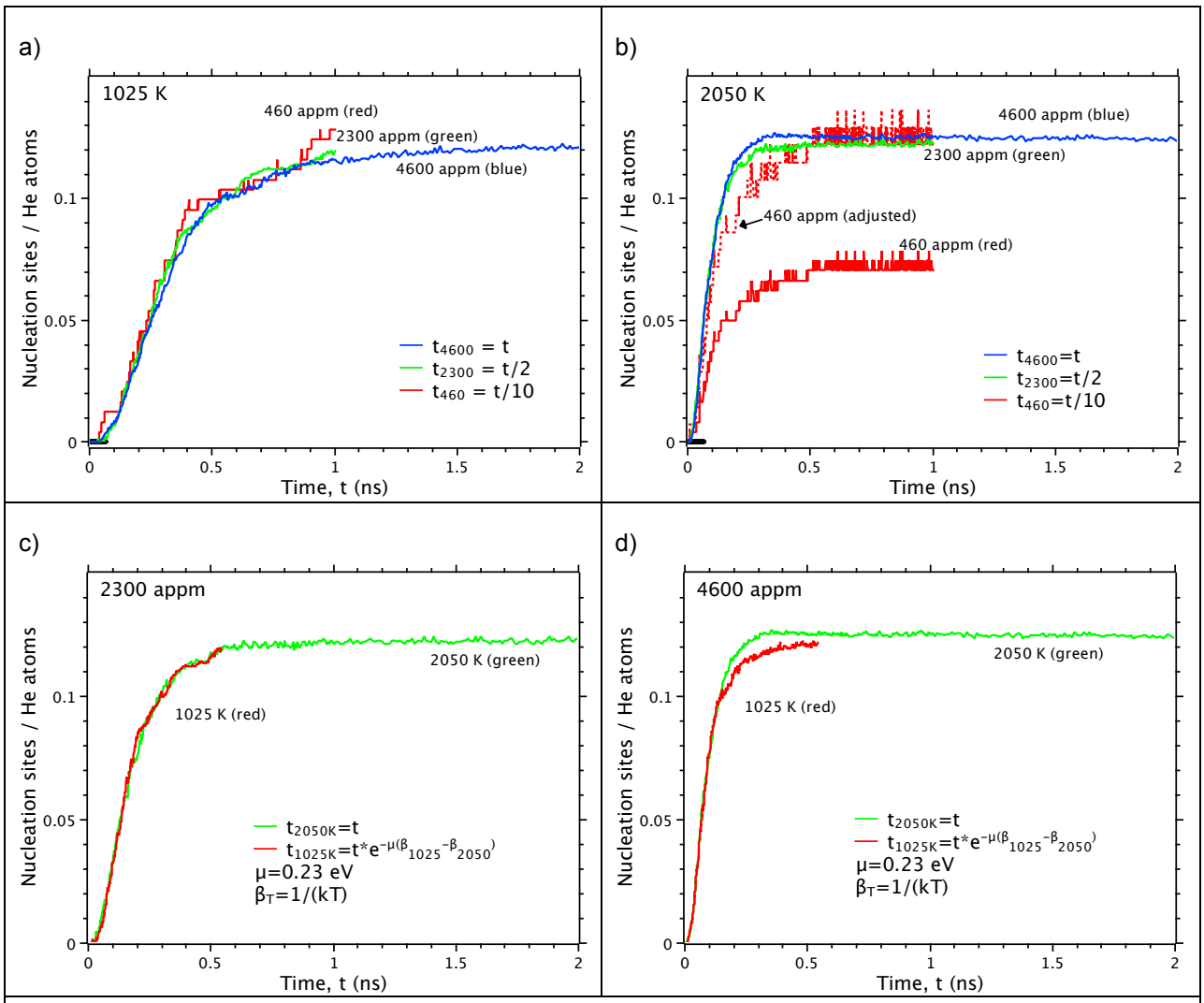

Figure 2. Panels a) and b) show MD time scaling with respect to He concentration *x*, (*t* → *xt*). Panels c) and d) show the time scaling with respect to temperature *T*, (*t* → *t* exp(-μβ$_T$) where μ = 0.23 eV.

The time scaling analyses suggest that a single empirical curve exists to model the evolution of the number of trap-mutation nucleation sites as a function of He concentration and temperature. In particular, the following model applies well in the vicinity of 4600 appm and 1025 K conditions. Motivated by the saturation behavior at $t \rightarrow \infty$ and that the curves increase superlinearly at $t \rightarrow 0$, the curves are fitted to the following functional:

$$a[1 - \exp(-bt^n)] \tag{1}$$

Where the saturation value is taken to be a = 0.12. Figure 3 shows the curve fitting result for the 4600 appm and 1025 K data, i.e. b = 4.69 and n = 1.60. Using this data, the empirical model for estimating the number of nucleation sites N_C can be written as

$$\frac{N_C}{N_{He}} = 0.12[1 - \exp(-4.69\tau^{1.60})] \quad (2)$$

$$\tau = \frac{x}{4600} t e^{-0.23\left(\frac{1}{T} - \frac{1}{1025}\right)\left(\frac{300}{0.0258}\right)} \quad (3)$$

where the He concentration x is in appm, temperature T is in K and time t is in ns. The performance of Equations 2 and 3 in approximating N_C/N_{He} curves for 2300 and 4600 appm at 1025 and 2050 K is demonstrated in Figure 3b. For conditions near 1025 K, Equations 2 and 3 can be used as is. For conditions near 2050 K, a better prediction is obtained by using the above Equations with a saturation value of a = 0.123 instead of 0.12.

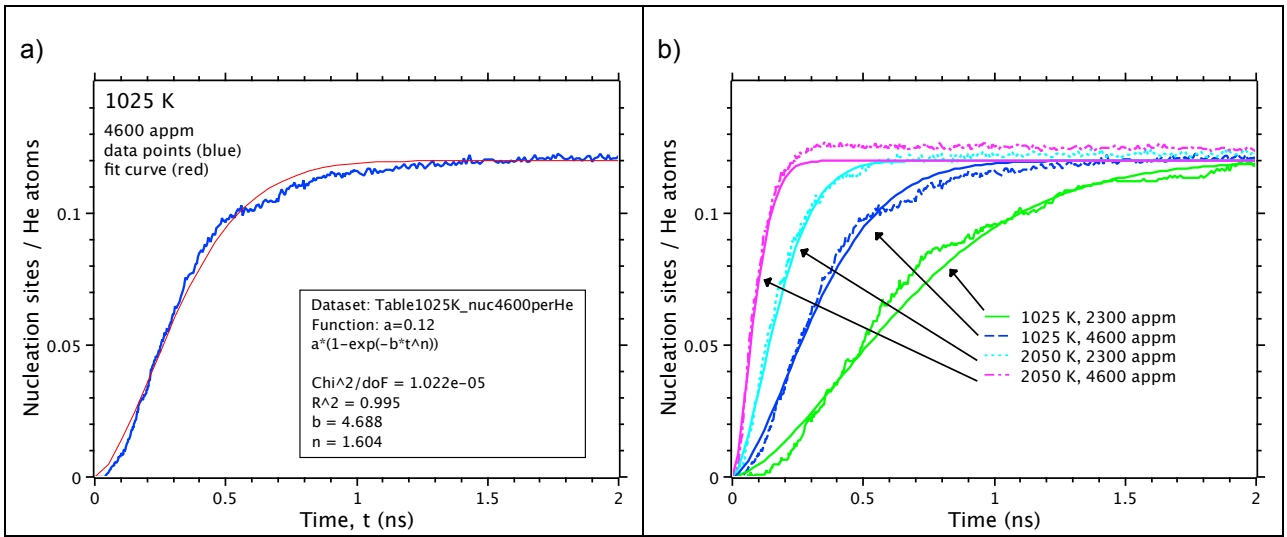

Figure 3. a) Curve fitting of the 4600 appm and 1025 K data. b) The fit curve from a) is used to estimate the curves for other conditions simply by using a time scaling as given in Equation 3 (see text).

Next we discuss the evolution of the number of SIAs, N_I. Similar to the case of nucleation sites, the curves of N_I/N_{He} also exhibit the same time scaling with respect to He concentration as shown in Figures 4a and 4b. Excellent scaling is observed for the 1025 K data. At 2050 K, the 2300 and 4600 appm data also scales well, however the 460 appm data at this temperature shows anomalous behavior as in the case of the nucleation sites curve. At 2050 K, the N_I/N_{He} curve of the 460 appm rises more slowly than predicted by the time scaling. Nevertheless, it appears to saturate to the same value as the other curves. The scaling with respect to temperature is displayed in Figures 4c and 4d using the same effective migration energy μ = 0.23 eV. The scaling works well to describe the early part of the curves (up to ~50% saturation). However, a perfect fit cannot be obtained due to t apparent differences in the saturation value, which are considerably larger at higher temperatures. At 1025 K, the saturation value is about 0.15, while at 2050 K it is 0.20. One could probably introduce another scaling parameter related to the saturation value as a function of temperature. However, the exact dependence cannot be determined with only two temperatures as it is clearly nonlinear.

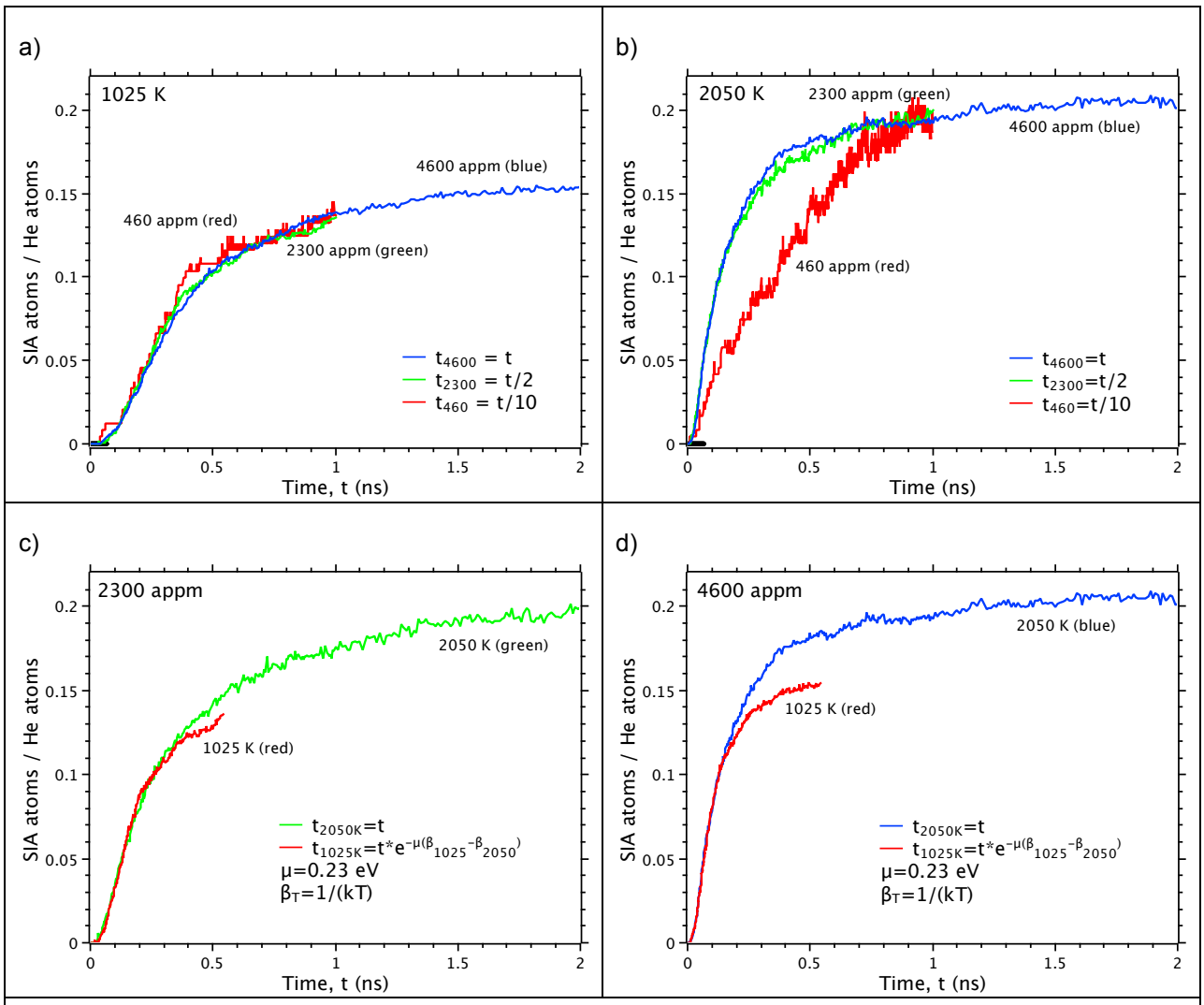

Figure 4. Evolution of the number of SIAs created by He trap-mutation. Panels a) and b) show MD time scaling with respect to He concentration *x*, (*t* → *xt*). Panels c) and d) show the time scaling with respect to temperature *T*, (*t* → *t* exp(-μβ_T) where μ = 0.23 eV.

Consequently, two separate curve fits were calculated for the 1025 and 2050 K results. For both temperatures, the 4600 appm data was used for the fitting. The resulting empirical models as a function of He concentration for 1025 and 2050 K conditions are respectively:

$$\frac{N_I}{N_{He}} = 0.15[1 - \exp(-2.89\tau^{1.43})], \quad \tau = \frac{x}{4600}t \qquad \text{(for 1025 K)} \tag{4}$$

$$\frac{N_I}{N_{He}} = 0.20[1 - \exp(-5.34\tau^{1.03})], \quad \tau = \frac{x}{4600}t \qquad \text{(for 2050 K} \tag{5}$$

Where time *t* is in ns and He concentration *x* is in appm. The performance of the models for describing other He concentrations is depicted in Figure 5. The 1025 K model describes the evolution well even for the 460 appm. While the 2050 K model sufficiently fits the 2300 and 4600 appm conditions.

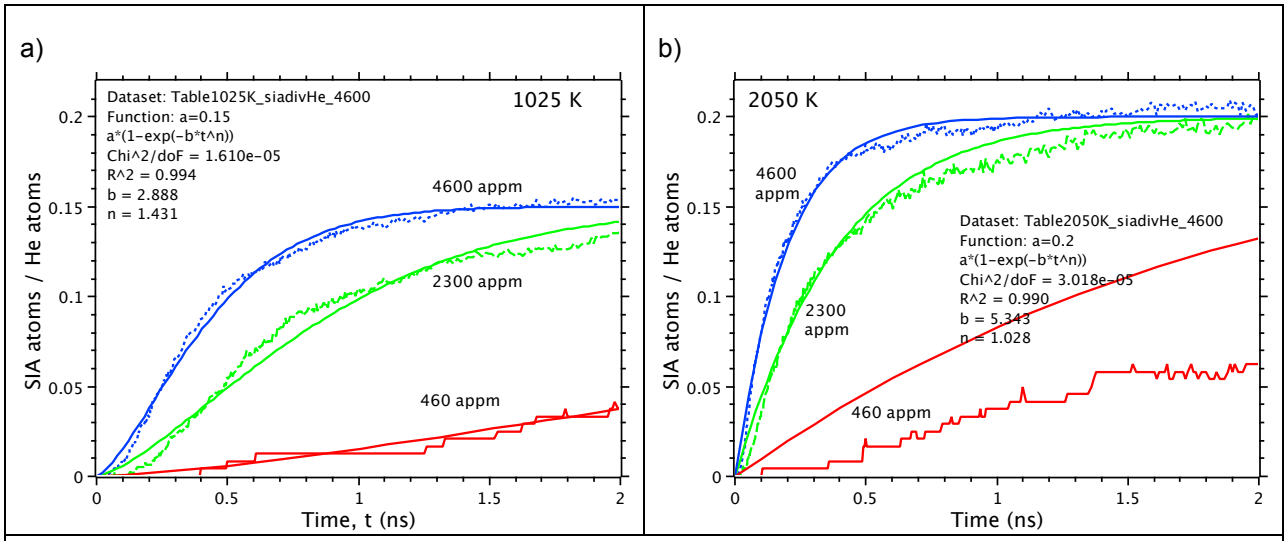

Figure 5. A curve of the 4600 appm is used to estimate the curves for other concentrations simply by using the time scaling given by Equations 4 and 5 (see text).

ACKNOWLEDGEMENT

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