THERMAL HELIUM DESORPTION SPECTROMETRY OF HELIUM-IMPLANTED IRON–D. Xu, T. Bus, S. C. Glade, and B. D. Wirth (University of California, Berkeley)

OBJECTIVE

The objective of this work is to understand the kinetics and energetics of helium transport and clustering in iron implanted with He ions as a function of He ion energy and dose.

SUMMARY

Elemental iron implanted with He at different energies and doses is studied using thermal helium desorption spectrometry (THDS). Currently examined energies and doses include: 100 keV, and 1 x 10^{11}, 1 x 10^{13}, and 1 x 10^{15} He/cm^2, respectively. While no clear desorption signals have been observed for the two lower dose samples, the present results reveal that for the iron implanted to 1 x 10^{15} He/cm^2 the majority of the implanted He atoms desorb at ~ 1000ºC and at > 1100ºC. Both conventional reaction model and Johnson-Mehl-Avrami (JMA) transformation model kinetics were utilized to fit the lower temperature (~ 1000ºC) desorption event of the 1 x 10^{15} He/cm^2 dosed iron. Surprisingly, single (either 1st or higher) order fits can not adequately describe the event. Excellent fits are obtained when combining a lower (*n* ~ 1.1) order with a higher (*n* ~ 5.8) order JMA fit. Additionally, spurious desorption peaks and certain complex desorption features have been observed which may affect future THDS studies.

PROGRESS AND STATUS

Introduction

The development of fusion reactors requires knowledge of material behavior under fusion environments, in particular with regard to high levels of helium produced by (n,α) reactions. It has been established that implanted or internally produced He can cause significant mechanical property degradation [1-5]. A crucial aspect, therefore, is to understand how helium atoms migrate and are trapped by microstructural features in irradiated materials. While a large amount of theory, modeling and experimental research has been performed in the past years, the understanding of this problem is still far from complete. Thermal helium desorption spectrometry (THDS) has been employed to experimentally study irradiation-induced structural defects and their interactions with He atoms in a variety of materials. For example, nucleation and growth of He-vacancy clusters were reported in vanadium and vanadium alloys [6], and the sequential releases of interstitial He and He-Vacancy clusters were reported in SiC [7] based on the THDS spectra.

In iron and ferritic alloys, computer simulations have been performed on defect production in collision cascades caused by helium injection [8], effect of He-vacancy complexes on the mechanical properties [9], thermal stability of He-vacancy clusters in iron [10], and the He-grain boundary interaction [11]. Experimentally, nuclear reaction depth profiling [12], transmission electron microscopy [13], positron annihilation lifetime and coincidence Doppler broadening (CDB) techniques [14,15] have been used in addition to THDS [10,16–17] to study the He migration and He-induced defect clusters in iron.

In this work, we use THDS to study the kinetics and energetics of helium in iron implanted with 100 keV He to three different doses, 1 x 10^{11}, 1 x 10^{13}, and 1 x 10^{15} He/cm^2. Constant rate heating ramps were employed to thermally desorb the implanted He. The resulting desorption signals were fit to both conventional reaction model and Johnson-Mehl-Avrami (JMA) transformation model kinetics. Surprisingly, single (either 1st or higher) order fits can not adequately describe the signals in either model. Excellent fits were obtained when combining a lower (~ 1) order with a higher (~ 6) order in the JMA model. Spurious desorption peaks and complex desorption features observed are also presented.

Experimental Methods

Figure 1 is a sketch of our recently built THDS at UC Berkeley [18]. Both the sample chamber and the measurement (quadrupole mass spectrometer) chamber are maintained at ultra-high vacuum with a pressure of about 10^{-10} Torr (at room temperature). Both the sample holder and the resistive heating filament are made of tungsten. The He, as well as other species (N_2, H_2, etc.), is signaled by the mass spectrometer (maintained at room temperature) while the sample is being heated according to a desired temperature profile. The synchronized sample heating and mass spectrometer measurement are both controlled through a LABVIEW program which simultaneously records all relevant data. During an actual measurement, liquid N_2 is constantly flowing through a channel between the inner and outer walls of the sample chamber to prevent the temperature rise of the walls and subsequent heating of the gas species.

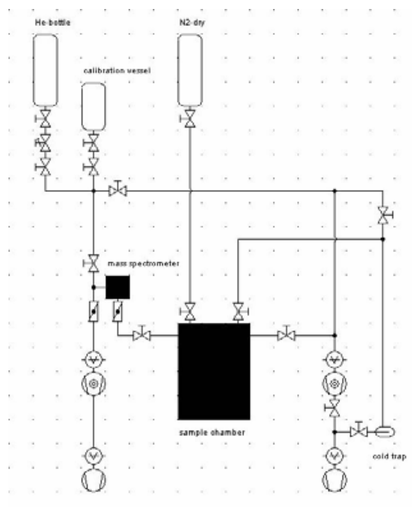

Fig. 1. Structural sketch of the Berkeley THDS instrument.

A THDS system can be operated in either static (no pumping during a measurement) or dynamic mode (gas being constantly pumped out during a measurement). In this study, dynamic mode was employed to prevent accumulation of desorbed He in the measurement chamber. In the dynamic mode with a fixed chamber volume *V*, assuming He at room temperature T_r, the He pressure inside the chamber *P*, as seen by the mass spectrometer, is governed by the differential equation:

$$VdP = K_B T_r d\overline{N} - \frac{PV}{\tau} dt \, , \tag{1}$$

in which τ is a pumping time constant, and $d\overline{N}$ is the number of desorbed He atoms from the sample in the time period of *dt*. The pumping constant τ is determined by the pumping speed and thus is adjustable. If τ is very small such that $dP/dt << P/\tau$ [1], then one obtains $d\overline{N}/dt \propto P$. However, as will be shown and discussed later, even when τ is indeed negligible, some non-negligible spurious signals (even peaks) which apparently do not result from the desorption of implanted He may still contribute to the measured pressure *P*. Therefore, a more careful expression should be: $d\overline{N}/dt \propto (P - P_{base})$ where P_{base} can be regarded as the signal measured from an otherwise-similar but non-implanted control sample. The control measurements were performed using the same settings (including the temperature control parameters,

[1]The accuracy of this assumption can be checked during data analysis by numerically comparing these two terms. τ can be found in calibration procedure. In this work, τ = 0.3s and the assumption is sufficiently satisfied.

the starting system pressure, the liquid N_2 flow rate, etc.) as for the corresponding actual desorption measurements.

A tube of 1 ml volume in connection with a 500 ml reservoir was used for calibration measurements. The calibration factor for the mass spectrometer was determined to be ~ 5.5 x 10^{-22}C/He-atom. Iron plates of 1mm thickness with a purity of 99.5% were purchased from Goodfellow and then commercially implanted with 100 keV helium to three different doses: 1 x 10^{11}, 1 x 10^{13}, and 1 x 10^{15} He/cm^2. Constant rate heating ramps at rates of 0.5 K/s and 1 K/s were used for both the control and the actual desorption measurements.

Results and Discussion

TRIM/SRIM calculations

The damage, He distribution, V/He (Vacancy/He) ratio and other factors related to He implantation in Fe were calculated with TRIM (SRIM 2003) software [19]. For 100 keV He implantation, the vacancy/He ratio is 87 and the peak He concentration at a dose of 1 x 10^{15}/cm^2 is about 700 appm, which appears at a depth of 340 nm.

Spurious peaks

A comparison between two samples: S10 (1 x 10^{15}/cm^2 dosed) and S12 (non-implanted control) is shown in Fig. 2a. The same heating control parameters were used for both samples which produced almost identical actual temperature profiles during the measurements. Fig. 2a shows that both samples exhibit a set of medium temperature He peaks in the range of ~ 600–880ºC, and, more importantly, the positions of these medium temperature peaks are almost identical for the two samples. However, the implanted sample S10 displays much stronger signals than the non-implanted S12 at temperatures higher than ~ 880ºC, including a fully developed peak at 1017ºC and a broad peak with an onset of ~ 1130ºC.

Apparently, the medium temperature peaks in the range of ~ 600–880ºC are not due to the desorption of implanted He, and thus are referred to as spurious peaks (signals) throughout this paper. The real identifiable desorption events start from above 880ºC in the 1 K/s ramping measurement of S10. Since the magnitudes of the spurious peaks are not negligible compared with the real desorption peaks, it is thus crucial to perform a control analysis before making peak assignments, particularly if the He desorption under consideration occurs in a relatively low temperature range (e.g., below 880ºC). Whether such spurious peaks also contributed to the observed signals in previous THDS studies is unclear.

While the exact origin of the spurious peaks is still under investigation, they appear to be related to the mutual desorption of several non-implanted species. As shown in Fig. 2b, other channels of the mass spectrometer, such as N_2, as well as the total system pressure, also exhibit peaks at basically the same temperatures.[2] Moreover, even a copper gasket was found to exhibit similar peaks on all channels of the mass spectrometer (including He) at relatively lower temperatures from 500 to 750ºC. It must also be noted that these spurious peaks do not appear when an empty-chamber (without any sample) is measured, indicating system cleanliness is not the problem. Rather it appears that surface contamination of the sample might be partly responsible for the spurious peaks.

[2]These other channels are distinguished from the He channel at temperatures higher than ~ 880 ºC where the implanted He starts to desorb. Note that a logarithm scale is used in Fig. 2b for comparison of different channels.