Measurement of Helium Diffusion in Metals

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40th Tritium Focus Group, Albuquerque, NM
October 23-25, 2018
Overview

- Helium is generated in tritium exposed materials by beta decay.
- It is insoluble in metals and diffuses to the surface or collects in high-pressure bubbles, eventually affecting the materials structural integrity.
- Modeling this process requires knowledge of the effective He diffusion and trapping parameters.
- Previous efforts with He implantation have focused on bubble growth and the onset of blistering.
- Here, a new, “gentle” implantation technique is described where He clustering is reduced. It involves
  - a low energy, short He implantation pulse at low temperature
  - followed by a rapid thermal desorption ramp.
- An analytic expression of desorption behavior yields both diffusion and trapping data.
  - Fickian diffusion from slab (short time limit)
  - SRIM implantation profile
- Results: first experimental data on He diffusion in Ni, Cu, and Pd.
Experimental System

- Samples, 100 µm foils annealed: large grain
- He energy (0.5-5.0 keV) range = 4-20 nm
- Spot size, 4-6 mm
- Implant pulse (1-100 nC) .1-10 appm He
- Sample evacuation (20 s)
- Rapid He-TDS (1-2 K/s)
The thermal desorption spectrum shows both diffusive behavior and He release from traps.
Analytic expression for diffusive desorption

- Diffusion from a plane sheet of half-thickness \( R \) (Crank, per Kass*):

\[
M_t = 2 \left( \frac{D_t}{\pi R^2} \right)^{1/2} \left\{ 1 + \sum \exp\left(\frac{-nR^2}{D_t}\right) \& \text{erfc}\left(\frac{nR}{\sqrt{D_t}}\right) \right\}
\]

For small times, the release from one side, \( M_t = \left( \frac{D_t}{\pi R^2} \right)^{1/2} \)

- Crank also shows this solution for constant diffusivity \( D \) may be transformed for \( D(t) \) by replacing \( D t \) by

\[
\tau = \int_0^t D(t') \, dt'
\]

- With a linear ramp \( T = T_0 + \beta t \) and \( D = D_0 \exp\left(-\frac{E_D}{kT}\right) \),

\[
\tau = \left( D_0 kT^2/\beta E_D \right) \exp\left(-\frac{E_D}{kT}\right)
\]

- Substituting, for large \( E_D/kT \)

\[
M_t = \left( \frac{kT^2 D_0}{\pi \beta R^2 E_D} \right)^{1/2} \exp\left(-\frac{E_D}{2kT}\right), \quad \text{proportional to } 1/R
\]

or

\[
\ln(M_t/T) = \frac{1}{2} \ln \left( \frac{kD_0}{\pi \beta R^2 E_D} \right) + \frac{E_D}{2kT}, \quad \text{linear with } 1/T
\]

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R is determined from Implantation Profile

SRIM: 2 keV He implantation into Pd

ION RANGES

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<tr>
<th>Ion Range</th>
<th>108 Å</th>
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<td>Straggle</td>
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• For non-interacting He, the cumulative release is the sum of release from layers with R determined by right side of implant profile.

• Since $M_t \propto \frac{1}{R}$, the effective thickness is

$$\frac{1}{R_{\text{eff}}} = \frac{1}{n} \sum_n \left( \frac{1}{R_n} \right)$$

2 keV He on Pd: $R_{\text{eff}} = 169$ Å

• For a 500 s spectrum, errors from the absence of release from the left side of the profile ($R < \frac{R_{\text{eff}}}{10}$) occur during the 20 s evacuation.
Typical fit to plot of $\ln \left( \frac{M_t}{T} \right)$ vs $1/T$

2 keV He on Pd, $\beta = 2$ K/s

- Plot is linear over several orders.
- From slope $S$, $E_D = 2kS = 0.092$ eV
- From $y$-intercept $I$, $D_0 = 2\pi\beta R^2S \exp (2I) = 2.6 \times 10^{-12}$ cm$^2$/s

- Rapid rise yields $E_T = 0.32$ eV

(Pd interstitial-vacancy recombination will eject He from vacancy)
# Table of results

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<tr>
<th>Run</th>
<th>E(keV)</th>
<th>β(K/s)</th>
<th>E_D(eV)</th>
<th>D_0(cm²/s)</th>
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Arrhenius plot comparing He diffusivities

- Uncertainty: shift is due to normalization
- Release from traps obscures end of diffusive spectrum.
- Correct by stopping ramp before release from trap.
Summary

An experimental technique is being developed that appears capable of measuring He diffusivities in metals.

- $10^{10}$ to $10^{11}$ He atoms are implanted by a short, few keV He ion pulse at low temperature
- The sample chamber is quickly evacuated, then opened to a getter-pumped gas analyzer, with some additional He pumping for fidelity of the He desorption behavior
- The sample temperature is rapidly ramped to 400 K, producing
  (i) interstitial He diffusion from the sample and
  (ii) escape of He from trapping sites at higher temperatures.
- The desorption spectrum is analyzed with a linear expression describing Fickian diffusion, under short time and $E_D/kT > 10$ approximations.
- The analysis gives both $D_0$ and $E_D$.
- He diffusion in Cu, Ni, and Pd is found to be around $10^{-13}$ cm$^2$/s at room temperature, but He trapping differs significantly.
Next Steps

• Reduce uncertainties
  - Energy (background correction)
  - Normalization (stop ramp)

• Lower implant temperature

• Vary implant fluence (clustering, trapping)

• Examine diffusivity in other materials
  - NG, fusion, alloys