

STUDY OF DIFFUSION MECHANISMS OF HELIUM ATOMS IN FACE-CENTERED
CUBIC METALS AFTER α -IMPLANTATION IN A CYCLOTRON

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ABSTRACT

Helium has been homogeneously introduced into gold foils at room temperature by α -implantation in a CV-28 cyclotron. After implantation the helium release was observed in isothermal and linear heating experiments.

The diffusion coefficient follows an Arrhenius behavior with $D_0 = 0.1 \text{ cm}^2/\text{sec}$ and $\Delta H = 1.7 \text{ eV}$. Possible diffusion mechanisms are discussed.

This work was performed at the Solid State Dept, of KFA - Jülich, West Germany.

INTRODUCTION

Helium in metals is produced by nuclear reactions of energetic particles. In nuclear reactors the interest in helium in metals is large, due to its production by (n, α) nuclear reactions in structural materials. Because helium has extremely low solubility in metals⁽¹⁾, the precipitation in the form of gas filled bubbles at elevated temperatures occurs⁽²⁾, where the helium atoms are mobile, which have detrimental effects on

mechanical properties. One typical example is the high temperature embrittlement⁽³⁾. To better understand this precipitation process, the diffusion of atomic helium in metals must be known. At present there are few experimental informations available on this properties⁽⁴⁾.

This work presents the study of the diffusion of helium in gold at temperatures above room temperature. The helium created by (n, α) reactions has been simulated by homogeneous α -implantation at a CV-28 cyclotron. The occurrence of free diffusion was proved comparing the dependence of release kinetics on helium concentration, sample thickness, time and heating rate to diffusion theory and clearly separated from agglomeration processes.

EXPERIMENTAL PROCEDURE

The experimental method has been described previously⁽⁵⁾.

The specimens were 99,999% Au-foils of thicknesses from 5 to 54 μm , which were annealed before implantation for 1 hour at 800°C in vacuum.

The helium was introduced homogeneously throughout the foil thickness by implanting α particles from a cyclotron at room temperature, in concentration between 10^{-9} to 10^{-5} atomic parts of helium.

The incoming 28 MeV α -particles passed through a 60 μm Al-window and a rotating degrader wheel before hitting the specimens. The Al-window separates the cyclotron vacuum from the helium gas in the chamber, which the specimens were cooled. The beam current density was limited to about 0.15 $\mu\text{A}/\text{cm}^2$.

In order to obtain a variable energy, a rotating degrader wheel was used, which supported 51 Al-foils of various thicknesses which results α -ranges in Au between 0 and 83 μm , and gives steps in the implantation depths of 1.7 μm .

After implantation the specimens were dropped in a furnace in a UHV-chamber. This furnace consisted of an alumina tube and a tantalum heater. The specimens were dropped onto an alumina plate in the middle of the alumina tube to which two W-Re thermocouples were attached. The accumulation of the released helium was followed by a quadrupole mass spectrometer during isothermal and linear annealings, with heating rate of 50 K/min.

RESULTS

In fig. 1 helium desorption curves, for linear anneal, from 18 μm Au foils of different He-concentrations are shown as a function of reciprocal temperature.

The ordinate gives the released fraction of the initial He-concentration c_0 , which was determined by melting the specimens. For concentration of $c_0 \leq 2.10^{-8}$ atomic parts of helium, for Au-18 μm , the curves fall together within the experimental error (dotted line). At higher concentrations deviation from this behavior occur at decreasing temperatures. It may be observed from another thicknesses that the point of deviation is further shifted to lower temperatures with increasing foil thickness.

The free diffusion problem of a gas release from a foil can be solved analytically^(5,6) and represented within 0.1% error by the equation:

$$\frac{c_0 - c}{c_0} = 2 \left(\frac{4Dt}{\pi d^2} \right)^{\frac{1}{2}} \quad \text{for} \quad \frac{c_0 - c}{c_0} \leq 0.5 \quad (1)$$

where: D - diffusion coefficient

d - specimen thickness

t - time

Considering only small helium concentrations, fig. 2 shows experimental $(c_0 - c)/c_0$ curves. These curves varied reciprocally

with thickness, as relation (1), and correspond to free diffusion. The details of the deviations from the free diffusion behavior can be qualitatively explained by immobilization of helium by clustering. In the case of free diffusion, helium diffuses freely, unaffected by clustering, in Au specimens, for which the product of the thickness d [m] and initial concentration c_0 [atomic parts] is less than $4.6 \cdot 10^{-13}$ m.

Isothermal experiments are showed in fig. 3. The straight (dashed) lines are predicted by diffusion theory, according eq.(1) that shows that free diffusion can only be assumed if $(c_0 - c)/c_0$ is independent of c_0 and proportional to \sqrt{t} and d^{-1} .

One problem in these experiments is that the zero point of the time axis is not exactly known, since it takes some time for the specimen to attain temperature after being dropped into the furnace. Therefore a constante value Δt was subtracted from the time passed after dropping the specimen. These values are choosen such that the release curves fit the straight lines at small times as closely as possible. As long as the \sqrt{t} -law in the isothermal release experiment is obeyed over a time regime much larger than Δt , the uncertainty introduced by the choise of Δt seems tolerable⁽⁷⁾.

Diffusion constants as a function of temperature are derived from the curves in fig. 2 and 3 by applying equation (1), and are showed in fig. 4. The results are compared to self-diffusion⁽⁸⁾ and to diffusion of hydrogen⁽⁹⁾ in gold. The isothermal results agree with the linear heating results, giving on the average an Arrhenius behavior of the diffusion constant:

$$D = 10^{-1.0 \pm 0.4} [\text{cm}^2/\text{s}] \exp (-1.7 \pm 0.1 [\text{eV}]/kT) \quad (2)$$

A single D -value reported in the literature⁽¹⁰⁾ is also in

fair agreement.

DISCUSSION

It is assumed that after room temperature implantation, all helium atoms are trapped in vacancies, that means in substitutional positions. Mobility may be then regained by two diffusion mechanisms:

- a) The first is the usual vacancy mechanism which is assumed to operate in most substitutional alloys. In this case the activation energy, ΔH_V , is in the following range:

$$Q_{2V}^{sd} - E_V^F \leq \Delta H_V \leq Q_V^{sd} \quad (3)$$

where: Q_{2V}^{sd} - activation energy for self-diffusion by divacancies

E_V^F - formation energy of a vacancy

Q_V^{sd} - activation energy for self-diffusion

- b) helium occupies also substitutional positions and moves by jumping out of this site, diffusing interstitially until it is trapped again in a vacancy. In this case of dissociation one obtain the following activation energy, ΔH_D

$$\Delta H_D = E_{HeV}^B + E_{He}^M - E_V^F \quad (4)$$

where: E_{HeV}^B - binding energy between He and a vacancy

E_{He}^M - migration energy of a He atom

In the table 1 activation energies calculated from eq. (3) and (4) are compared to the experimental data. Included is self-diffusion data for D_{10} , the preexponential factor for tracer, which may apply for the vacancy mechanism. It may be estimated that the preexponential factor for the dissociative mechanism is smaller than for the vacancy mechanism. But this estimate, as well as the experimental preexponentials, contain sufficient errors to prevent a detailed comparison. The activation energy clearly favour the

Table 1: Comparison of preexponential factor D_0 [cm^2/s] and activation energy, ΔH [eV] to estimates for the vacancy diffusion mechanism and dissociation mechanism.

Vacancy ^{a)}			Dissociation ^{b)}	Experiment	
D_{10}	Q_V^{sd}	$Q_{2V}^{\text{sd}} - E_V^{\text{F}}$	ΔH_D	D_0	ΔH
$10^{-1.40}$	1.76	1.42	1.35	$10^{-1.0}$	1.70

a) ref. (8)

b) $E_{\text{HeV}}^{\text{B}}$, E_{He}^{M} from ref. (11), E_V^{F} from ref. (12)

the vacancy mechanism. The activation energy indicates that the substitutional helium atom is better represented as a host atom than as vacant lattice site. This is the case for Ag and Al⁽⁷⁾, but in the case of Ni the activation energy favours the dissociative mechanism⁽¹³⁾.

CONCLUSIONS

1. Measurement of helium release in isothermal and linear-heating experiments as a function of time (or heating rate), thickness and initial concentration made it possible to determine diffusion constants of isolated helium atoms.
2. Comparison with theoretical estimates favour the vacancy mechanism for helium diffusion in Au, Al and Ag and dissociative mechanism in the case of Ni.

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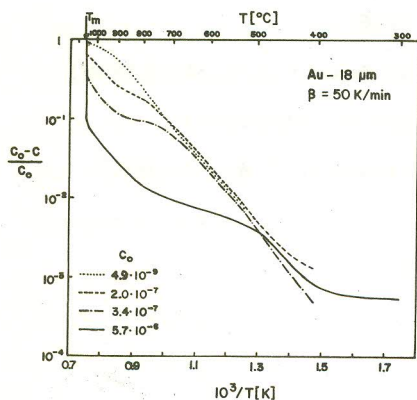


Fig.1: Released fraction of helium from 18 μm -Au foils, as a function of reciprocal temperature during linearly heating at a rate of 50 K/min.

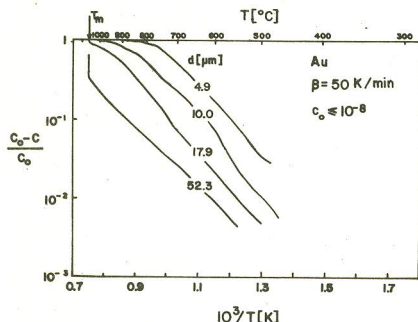


Fig.2: Released fraction of helium in Au, of various thicknesses and for $c_0 \leq 10^{-8}$ atomic parts, as a function of reciprocal temperature during linearly heating at a rate of 50 K/min.

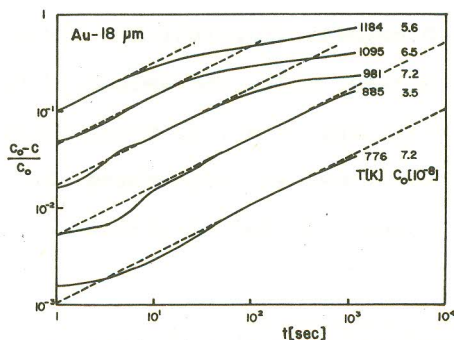


Fig.3: Fraction of helium released isothermally from 18 μm -Au foils.

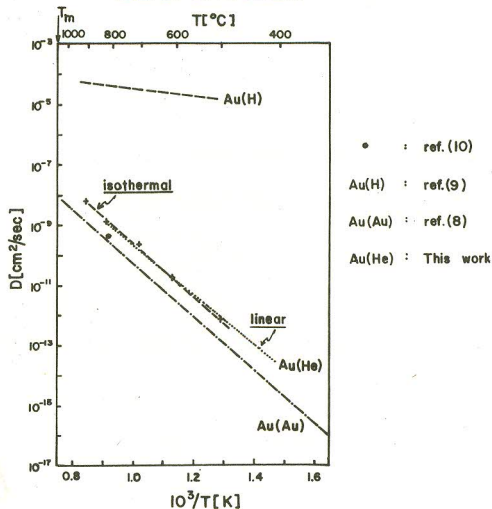


Fig.4: Diffusion constants of helium in Au derived from the measurements in fig. 2 and 3.