Available online at www.sciencedirect.com

ScienceDirect

Geochimica et Cosmochimica Acta 192 (2016) 248–257

Geochimica et Cosmochimica Acta

www.elsevier.com/locate/gca

Diffusion of helium in SiC and implications for retention of cosmogenic He

D.J. Cherniak [a,*], E.B. Watson [a], R. Trappisch [b], J.B. Thomas [c], D. Chaussende [d]

[a] *Department of Earth and Environmental Sciences, Rensselaer Polytechnic Institute, Troy, NY 12180, USA*
[b] *Department of the Geophysical Sciences, The University of Chicago, and Chicago Center for Cosmochemistry, Chicago, IL 60637, USA*
[c] *Department of Earth Sciences, Syracuse University, Syracuse, NY 13244, USA*
[d] *Laboratoire des Matériaux et du Génie Physique, CNRS – Grenoble INP, 3 parvis Louis Néel, BP, 257, 38016 Grenoble, France*

Received 10 February 2016; accepted in revised form 4 August 2016; available online 11 August 2016

Abstract

Diffusion of helium has been characterized in silicon carbide of cubic and hexagonal (4H and 6H) forms. Polished sections of SiC were implanted with ^{3}He at 100 keV at a dose of $1 \times 10^{15}/\text{cm}^2$. The implanted SiC samples were sealed under vacuum in silica glass ampoules, and annealed in 1-atm furnaces. ^{3}He distributions following all experiments were measured with Nuclear Reaction Analysis using the reaction $^3\text{He}(\text{d},\text{p})^4\text{He}$. For He diffusion in cubic SiC and 4H hexagonal SiC we obtain the following Arrhenius relations:

$D_{\text{cubic}} = 1.83 \times 10^{-6} \exp(-254 \pm 10\ \text{kJ mol}^{-1}/RT)\ \text{m}^2\ \text{s}^{-1}$.

$D_{4H} = 4.78 \times 10^{-7} \exp(-255 \pm 29\ \text{kJ mol}^{-1}/RT)\ \text{m}^2\ \text{s}^{-1}$.

While He diffusion is considerably slower in SiC than in many silicate phases, He retentivity may be limited under some conditions. For example, helium will be lost from SiC grains over much shorter timescales than potential survival times of SiC presolar grains in the solar nebula. When exposed to impact heating followed by slow cooling, nearly complete loss of He from SiC grains near the site of impact will occur within several hours to a few days. For SiC grains at greater distance from impact sites, He would be better retained, depending on the rapidity of cooling. At tens of km away from a large impactor, where peak T would be ~800 K, SiC grains would lose about 50% of their He if the grains cooled within a few thousand years, and 5% if they cooled within a few tens of years. At greater distances where heating is more modest (500 K and lower), SiC grains would be quite retentive of He even for cases of very slow cooling. Helium would also be retained in cases of impact heating followed by very rapid cooling. For these short heating pulses, 10 μm diameter SiC grains would retain more than 50% of their He for peak heating temperatures of 2173, 1973 and 1773 K for durations of 3, 10 and 60 s, respectively.

Keywords: SiC; Helium; Diffusion; Nuclear reaction analysis; Presolar grains

1. INTRODUCTION

Silicon carbide is a major constituent of interstellar dust grains (e.g., Bernatowicz et al., 1987; Zinner et al., 1987; Zinner, 2013; Amari et al., 1994). In addition, it has important technological applications in nuclear reactors as cladding or containment materials (e.g., Riccardi et al., 2004; Nozawa et al., 2009; Yvon and Carré, 2009; Snead et al., 2007; Katoh et al., 2012), as well in electronic devices for

---

[*] Corresponding author at: Department of Earth and Environmental Sciences, Science Center 1W19, Rensselaer Polytechnic Institute, 110 8th St., Troy, NY 12180, USA. Fax: +1 518 276 2012.

E-mail address: chernd@rpi.edu (D.J. Cherniak).

http://dx.doi.org/10.1016/j.gca.2016.08.007

high temperature, high frequency and high power applications (e.g., Raynaud, 2001).

Because of its importance as a material in nuclear reactor and nuclear waste applications and for electronic devices, the effects of He implantation and subsequent annealing on SiC have been studied extensively (e.g., Chen et al., 2000; Oliviero et al., 2002, 2003; Jiang et al., 2002; Zhang et al., 2003; Vincent et al., 2009; Miro et al., 2011; Barbot et al., 2011; Linez et al., 2013). He migration and defects in SiC structures have also been explored theoretically with density functional theory calculations (e.g., Van Ginhoven et al., 2006; Cheng et al., 2011). Direct profiling to characterize diffusion of implanted ^{3}He and ^{4}He in SiC has been undertaken using the techniques of nuclear reaction analysis (NRA) and Elastic Recoil Detection (ERD) (e.g., Jiang et al., 2002; Smith et al., 2004; Sauvage et al., 2007; Miro et al. 2011). He diffusion in SiC has also been characterized through bulk degassing of He-implanted materials (Jung, 1992). However, there are wide variations in He diffusivities determined in the existing experimental studies, and observations have been made of complex behaviors of He in SiC, which have been attributed to trapping-detrapping phenomena and may depend on He implant dose and energy as well as annealing conditions.

In this work, we attempt to further quantify helium diffusion in SiC by examining different SiC structural forms, both cubic and hexagonal (4H and 6H). We conduct experiments over a range of time–temperature conditions to evaluate temperature dependence of He transport, and assess whether measured diffusivities exhibit time invariance at constant T, which could be an indicator of the presence or absence of trapping effects. A similar approach to that used in our earlier investigations (Cherniak et al., 2009; Cherniak and Watson, 2011, 2012, 2013) of He diffusion is taken here, with ion implantation used to introduce ^{3}He, and ^{3}He concentrations measured by nuclear reaction analysis (NRA) using the reaction ^{3}He(d,p)^{4}He. Relatively low doses of implanted He are used to minimize effects of implantation-induced defects, and to avoid the possibility of He aggregation, which has been associated with high He implant doses. Materials were also characterized with IR reflectance spectroscopy prior to and following ion implantation to assess the effects of He implantation on crystal integrity.

2. EXPERIMENTAL PROCEDURE

2.1. Sample preparation

The silicon carbide samples were single-crystal gem moissanite purchased from a commercial jewelry supplier, SiC in cubic from, grown by the continuous feed physical vapor transport (CF-PVT) method (Chaussende et al., 2005, 2009), and commercially-produced polished slabs of single-crystal hexagonal SiC (both 6H and 4H forms) obtained from the MTI Corporation. The hexagonal and cubic structure SiC samples were cut into pieces 3–4 mm on a side using a low-speed saw with a diamond blade. The gem moissanite was polished to 0.3 μm alumina, followed by a chemical polish with colloidal silica to create a flat surface about 3 mm on a side. After polishing and cutting, samples were cleaned ultrasonically in distilled water and ethanol.

2.2. Ion implantation and diffusion experiments

The prepared SiC samples were mounted for ion implantation on an aluminum plate using carbon paint. The samples were implanted at room temperature with 100 keV ^{3}He ions produced in the Extrion ion implanter at the Ion Beam Laboratory at the University at Albany, with a dose of 1×10^{15} ^{3}He/cm^2. All of the samples were implanted simultaneously, so they received the same implant dose.

Experimental assemblies for diffusion experiments were prepared by sealing the implanted SiC samples in silica glass ampoules under vacuum, along with some Si powder to act as a scavenger for residual oxygen and nitrogen. Experiments were run in 1-atm furnaces at temperatures from 749–1150 °C, for times from 30 min to 9 days (Table 1). Experiments at temperatures under 1100 °C were run in Kanthal-wire wound vertical tube furnaces, monitored with chromel–alumel (type K) thermocouples; those at higher temperatures were run in horizontal tube furnaces with $MoSi_2$ heating elements, monitored with Pt-Pt10%Rh (type S) thermocouples. In all cases, temperature uncertainties were ~±2 °C. Following diffusion anneals, samples were removed from the furnaces and allowed to cool in air. They were then removed from capsules and cleaned in baths of ethanol.

2.3. NRA analysis

^{3}He distributions in the samples were measured with nuclear reaction analysis using the ^{3}He(d,p)^{4}He reaction (e.g., Pronko and Pronko, 1974; Dieumegard et al., 1979; Payne et al., 1989; Paszti, 1992). Analyses were performed at the 4 MeV Dynamitron accelerator at the University at Albany. This nuclear reaction has been used to measure ^{3}He in a variety of minerals and ceramic materials, including apatite (e.g., Miro et al., 2006; Cherniak et al., 2009), zircon (Cherniak et al., 2009), zirconia (Gosset et al., 2002; Costantini et al., 2003; Trocellier et al., 2003a,b), titanite (Cherniak and Watson, 2011), rutile (Cherniak and Watson, 2011), olivine (Cherniak and Watson, 2012) and REE phosphates (Cherniak and Watson, 2013). The protons produced in the reaction, along with backscattered deuterons and products of various (d,p) and (d,α) reactions induced with major elements constituting in the sample, were detected with a solid state surface barrier detector with 1500 μm depletion depth and 100 mm^2 area positioned at 167.5° with respect to the incident beam. The beamspot incident on sample surfaces was ~1 to 1.5 mm on a side. A 7.5 μm thick Kapton foil was placed in front of the detector to stop some of the backscattered deuterons.

For these analyses, we take the approach used in Cherniak et al. (2009), performing analyses over a range of energies to better define the profile. Analyses at 4 or 5 different incident deuteron energies were performed, using energies ranging from 0.5 to 0.9 MeV, with energy steps