



## Helium diffusivity in oxide nuclear fuel: Critical data analysis and new correlations



L. Luzzi<sup>a,\*</sup>, L. Cognini<sup>a,b</sup>, D. Pizzocri<sup>a</sup>, T. Barani<sup>a</sup>, G. Pastore<sup>c</sup>, A. Schubert<sup>b</sup>, T. Wiss<sup>b</sup>, P. Van Uffelen<sup>b</sup>

<sup>a</sup> Politecnico di Milano, Department of Energy, Nuclear Engineering Division, Via La Masa 34, 20156 Milan, Italy

<sup>b</sup> European Commission, Joint Research Centre, Directorate for Nuclear Safety and Security, P.O. Box 2340, 76125 Karlsruhe, Germany

<sup>c</sup> Idaho National Laboratory, Fuel Modeling and Simulation Department, 2525 Fremont Avenue, 83415 Idaho Falls, United States

### ARTICLE INFO

#### Keywords:

Inert gas behaviour  
Helium behaviour  
Diffusivity  
Oxide fuel

### ABSTRACT

Helium is relevant in determining nuclear fuel behaviour. It affects the performance of nuclear fuel both in reactor and in storage conditions. Helium becomes important in reactor conditions when high burnups are targeted or MOX fuel is used, whereas for storage conditions it can represent a threat to the fuel rods integrity. The accurate knowledge of helium behaviour combined with predictive model capabilities is fundamental for the safe management of nuclear fuel, with helium diffusivity being a critical property. For this reason, a considerable number of separate effect experiments in the last fifty years investigated helium diffusivity in nuclear fuel. The aim of this work is to critically review and assess the experimental results concerning the helium diffusivity. Experimental results are critically analysed in terms of the helium introduction technique used (either infusion, implantation or doping) and of sample characteristics (single crystal, poly-crystal or powder). Accordingly, we derived two different correlations for the diffusivity. Clearly, each of the new correlations corresponds to a limited range of application conditions, depending on the experimental data used to derive it. We provide recommendations regarding the proper application conditions for each correlation (e.g., in reactor or storage conditions).

### 1. Introduction

The knowledge of helium behaviour in nuclear fuel is of fundamental importance for its safe operation and storage (Olander, 1976; Rossiter, 2012). This is true irrespectively of the particular fuel cycle strategy adopted. In fact, both open and closed fuel cycles tend towards operating nuclear fuel to higher burnups (i.e., keeping the fuel in the reactor for a longer time to extract more specific energy from it), thus implying higher accumulation of helium in the fuel rods themselves (Rondinella et al., 2003). Moreover, considering open fuel cycles foreseeing the disposal of spent fuel, the helium production rate in the spent nuclear fuel is positively correlated with the burnup at discharge, and the production of helium (by  $\alpha$ -decay of minor actinides) progresses during storage of spent fuel (Crossland, 2012; Wiss et al., 2014). On the other hand, closed fuel cycles imply the use of fuels with higher concentrations of minor actinides (e.g., minor actinides bearing blankets, MABB), thus they are characterized by higher helium production rates during operation (Crossland, 2012).

Helium is produced in nuclear fuel by ternary fissions, ( $n,\alpha$ )-

reactions and  $\alpha$ -decay (Botazzoli, 2011; Ewing et al., 1995; Federici et al., 2007). After its production, helium precipitates into intra- and inter-granular bubbles and can be absorbed/released from/to the nuclear fuel rod free volume (Booth, 1957; Matzke, 1980). Helium can thus contribute to the fuel swelling (and eventually the stress in the cladding after mechanical contact is established), the pressure in the fuel rod free volume, and the gap conductance (giving feedback to the fuel temperature) (Piron et al., 2000).

Among the properties governing the behaviour of helium in nuclear fuel, its diffusivity and solubility govern the transport and absorption/release mechanisms (Maugeri et al., 2009; Nakajima et al., 2011; Talip et al., 2014a). Compared to xenon and krypton, helium presents both a higher solubility and diffusivity in oxide nuclear fuel (Belle, 1961; Petit et al., 2003; Rufeh et al., 1965). These high values of helium solubility and diffusivity are responsible for its peculiar behaviour, characterized by phenomena that are not observed for xenon and krypton (e.g., helium absorption, helium thermal re-solution from bubbles) (Donnelly and Evans, 1991).

A considerable amount of experiments has been performed with the

\* Corresponding author.

E-mail address: [lelio.luzzi@polimi.it](mailto:lelio.luzzi@polimi.it) (L. Luzzi).

**Table 1**  
Summary of the experimental works considered in this overview.

Ref.	Sample	Technique of He introduction	He release measurement method
(Belle, 1961)	UO <sub>2</sub> powder (0.16 μm)	Infusion	Dissolution and MS <sup>a</sup>
(Rufeh, 1964)	UO <sub>2</sub> powder (4 μm)	Infusion	Dissolution and MS
(Rufeh et al., 1965)			
(Sung, 1967)	UO <sub>2</sub> single-crystal (1 μm)	Infusion	Dissolution and MS
(Trocellier et al., 2003)	UO <sub>2</sub> poly-crystal	Ion Implantation	μNRA <sup>b</sup> <sup>3</sup> He(d,p)α
(Guilbert et al., 2004)	UO <sub>2</sub> poly-crystal (8 μm)	Ion Implantation	NRA <sup>3</sup> He(d,α)H
		Fluence <sup>3</sup> He (m <sup>-2</sup> ) = 10 <sup>20</sup>	
(Roudil et al., 2004)	UO <sub>2</sub> poly-crystal (10 μm)	Ion Implantation	NRA <sup>3</sup> He(d,p)α
		Fluence <sup>3</sup> He (m <sup>-2</sup> ) = 0.3·10 <sup>20</sup>	
		Fluence <sup>3</sup> He (m <sup>-2</sup> ) = 3·10 <sup>20</sup>	
(Ronchi and Hiernaut, 2004)	(U <sub>0.9</sub> , <sup>238</sup> Pu <sub>0.1</sub> ) O <sub>2</sub> poly-crystal	Doping	KEMS
(Martin et al., 2006)	UO <sub>2</sub> poly-crystal (24 μm)	Ion Implantation	NRA <sup>3</sup> He(d,α)H
		Fluence <sup>3</sup> He (m <sup>-2</sup> ) = (1.7 ± 0.06)·10 <sup>20</sup>	
(Pipon et al., 2009)	(U <sub>0.75</sub> , <sup>239</sup> Pu <sub>0.25</sub> ) O <sub>2</sub> poly-crystal	Ion Implantation	NRA <sup>3</sup> He(d,p)α
		Fluence <sup>3</sup> He (m <sup>-2</sup> ) = 5·10 <sup>19</sup>	
(Nakajima et al., 2011)	UO <sub>2</sub> single-crystal (18 μm)	Infusion	KEMS
(Garcia et al., 2012)	UO <sub>2</sub> poly-crystal	Ion Implantation	NRA <sup>3</sup> He(d,α)H
		Fluence <sup>3</sup> He (m <sup>-2</sup> ) = 10 <sup>20</sup>	
(Talip et al., 2014a)	(U <sub>0.999</sub> , <sup>238</sup> Pu <sub>0.001</sub> ) O <sub>2</sub> poly-crystal (10 μm)	Doping	KEMS

<sup>a</sup> Mass Spectrometry.

<sup>b</sup> NRA (Nuclear Reaction Analysis) is a nuclear method to obtain the profile of helium implanted in samples, using <sup>3</sup>He(d,p)α and <sup>3</sup>He(d,α)H reactions (Martin et al., 2006; Pipon et al., 2009).

goal of determining the diffusivity and solubility of helium in nuclear fuel (Belle, 1961; Garcia et al., 2012; Guilbert et al., 2004; Hasko and Swarc, 1963; Martin et al., 2006; Maugeri et al., 2009; Nakajima et al., 2011; Pipon et al., 2009; Ronchi and Hiernaut, 2004; Roudil et al., 2004; Rufeh, 1964; Sung, 1967; Talip et al., 2014a; Trocellier et al., 2003). In particular, several measurements have been made to determine the helium diffusivity as a function of temperature (Belle, 1961; Garcia et al., 2012; Guilbert et al., 2004; Martin et al., 2006; Nakajima et al., 2011; Pipon et al., 2009; Ronchi and Hiernaut, 2004; Roudil et al., 2004; Rufeh, 1964; Sung, 1967; Talip et al., 2014a; Trocellier et al., 2003), whereas few experiments are available to characterise Henry's constant,<sup>1</sup>(Belle, 1961; Blanpain et al., 2006; Hasko and Swarc, 1963; Maugeri et al., 2009; Nakajima et al., 2011; Rufeh, 1964; Sung, 1967; Talip et al., 2014a).

The experimental procedures available for measuring helium diffusivity differ mainly in the way in which the helium is introduced in the fuel samples. In particular, three introduction techniques are used: (i) infusion (Belle, 1961; Nakajima et al., 2011; Rufeh, 1964; Sung, 1967; Maugeri et al., 2009), in which the sample is kept in a pressurized helium atmosphere for a certain infusion time, (ii) ionic implantation (Garcia et al., 2012; Guilbert et al., 2004; Martin et al., 2006; Pipon et al., 2009; Roudil et al., 2004; Trocellier et al., 2003), in which a beam of <sup>3</sup>He<sup>+</sup> hits and penetrates the sample, and (iii) doping (Ronchi and Hiernaut, 2004; Talip et al., 2014a), in which α-decaying elements are introduced in the sample, resulting in an internal source of helium. These introduction techniques generate different helium distributions in the samples and induce different levels of damage to the crystal lattice of the sample (Labrim et al., 2007; Talip et al., 2014a). Depending on the introduction technique used, different measuring techniques are adopted to determine the concentration of helium introduced in the sample. A relation is then established between the helium concentration and the diffusivity (Rufeh, 1964; Sung, 1967).

Moreover, helium diffusivity has been measured for samples with different microstructures, i.e., single crystals, poly-crystals, and powders.

In the light of the profound differences in experimental techniques and in microstructure of the samples, the correlations derived from rough data fitting must be critically analysed. In fact, the spread of

available diffusivities is extremely large. Nevertheless, currently used correlations for the helium diffusivity are still derived from rough data fitting (Garcia et al., 2012; Nakajima et al., 2011; Ronchi and Hiernaut, 2004; Roudil et al., 2004; Talip et al., 2014a) or are intended to be upper/lower boundaries enveloping the data (Federici et al., 2007; Ronchi and Hiernaut, 2004).

In this work, we provide a complete overview of all the experimental results obtained for helium diffusivity in oxide nuclear fuel. The experimental results are classified according to the helium introduction technique used. At last, we derive empirical correlations and recommend the most suitable values of the helium diffusivity in the main cases of interest (e.g., in-pile, storage or annealing condition). The derivation of empirical correlations is complemented by an uncertainty analysis.

## 2. Review of experimental results

Early measurements of the helium diffusivity in oxide nuclear fuel have been performed since the 1960s. The growing interest in determining helium behaviour in nuclear fuel to assess its performance in storage conditions translated in several new experiments performed in the last twenty years. In this Section, we give an overview of all the experimental results available in the open literature, organized in chronological order, as reported in Table 1.

Helium can be introduced into oxide nuclear fuel samples by infusion (Nakajima et al., 2011; Rufeh et al., 1965; Sung, 1967), ion implantation (Garcia et al., 2012; Guilbert et al., 2004; Martin et al., 2006; Pipon et al., 2009; Roudil et al., 2004; Trocellier et al., 2003) or by doping the matrix with short-lived α-emitters (Ronchi and Hiernaut, 2004; Talip et al., 2014a). Fig. 1 shows a sketch of the different experimental techniques herein considered. Depending on the helium introduction technique, the crystalline lattice suffers different levels of damage. Crystalline lattices with different damage levels show different helium behaviour. Moreover, each technique used to introduce the helium in the sample has a corresponding specific technique to measure the amount of helium introduced.

Belle (1961) first studied the diffusivity of helium in a UO<sub>2</sub> powder. After his work, the helium diffusivity in oxide nuclear fuels was estimated by Rufeh (Rufeh et al., 1965; Rufeh, 1964) and Sung (1967) using UO<sub>2</sub> samples (some in powder form and some single crystal) with helium introduced through the infusion technique.

<sup>1</sup> Early work from (Rufeh, 1964; Sung, 1967) demonstrated the validity of Henry's law for the system helium/oxide fuel.

Download English Version:

<https://daneshyari.com/en/article/6759055>

Download Persian Version:

<https://daneshyari.com/article/6759055>

[Daneshyari.com](https://daneshyari.com)