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Helium Infusion In Nuclear Materials

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ABSTRACT

Increasing the burnup limits for economic reasons and preferring MOX for the possibility of recycling spent material are among the two main reasons that lead to a greater production of helium in fuel and a consequent higher fission gas release in nuclear fuel. These are a major concern for the performance of the fuel rods in operation but also their safe management during disposal to avoid any potential radioactive release to the environment. Generally, these classes of problems could be handled through progressively more comprehensive and predictive models. However, these require an increased understanding of the physical properties and transport mechanisms for helium behaviour in nuclear fuels and materials. For this, helium has been infused in glasses used for the vitrification of radioactive waste, paving the way for further investigation in materials considered for nuclear waste disposal, besides nuclear fuel.

1 INTRODUCTION

Helium is produced in reactor by ternary fissions (with a helium fission yield equal to 0.22%[1] [1]), (n, α) reactions (mainly $^{16}O(n, \alpha)^{13}C$) and α-decays. Since helium production increases exponentially as a function of the burnup and since almost all the actinides are α-emitters, helium behaviour becomes more relevant in oxide fuel at high burnup, in $(U,Pu)O_2$ mix-oxides (MOX) and in particular in storage/disposal conditions. In detail, after the production, helium can be either released from the fuel, increasing the fuel rod internal pressure, or retained in the fuel (dissolved in the lattice or precipitated into intra- and/or inter-granular bubbles), contributing both to the degradation of the thermal conductivity of the fuel and to its gaseous swelling[2]. In addition, helium is generally used as filling gas in the fuel rods of current Light Water Reactors (LWRs which represent nowadays 82.5% of the reactors in operation worldwide), and of Gen IV nuclear reactors as well. During the first several months of operation, helium initially loaded in the free volume of the fuel rod can be absorbed into the

[1] The mean value of about 0.22% per fission is the helium ternary fission yield calculated for standard MOX fuels and it results slightly higher compared with the one for UO_2 fuels. This value is consistent with previously reported values of 0.2 - 0.3% per fission in both U and Pu [2].
[2] Gaseous swelling is defined as the isotropic increment of nuclear fuel volume caused by the precipitation of gas into both intra- and inter-granular bubbles.

fuel pellets [3]. All these phenomena can lead to safety and performance issues during the operation of fuel rods. Furthermore, beyond the potential impact of helium behaviour on both the fuel performance and the operation of a fuel rod, it is even more relevant in spent fuel, since it affects the long-term storage of nuclear fuel. In fact, when the rods are stored after irradiation in the reactor, helium is continuously produced in the spent fuel matrix due to α-decaying actinides. Hence, the accumulation of radiogenic helium next to α-damage creates bubbles at grain boundaries, which may change the spent fuel mechanical properties and could possibly cause loss of grain cohesion, with the ultimate risk of reducing the spent fuel pellet to powder [4 - 6]. On the other hand, if helium is released from the spent fuel matrix, it could increase the internal pressure on the cladding (representing the first confinement barrier) and lead to important consequences for the safety and, at worst, to its rupture.

In case of a fuel cycle backend including re-processing and storage of minor actinides in high level waste (HLW) glass, the helium behaviour is a key aspect to consider to predict its long term evolution [7]. The mechanical integrity could also be impacted by over-pressurized helium bubbles even if compared to crystalline fuel the open volumes in the glassy structure are more important. To this purpose, to enhance the prediction of helium behaviour in storage conditions, in this work, helium infusions have been carried out in materials other than the ceramic nuclear fuels, such as amorphous confinement matrices used for the immobilisation of α-active waste. All the infusions occurred at the same temperature (i.e., 300°C), but in different conditions of pressure and the experiments have been performed by means of a resistance-heated autoclave. After the infusions, the samples have been annealed by means of the Thermal Desorption Mass Spectrometer (TDMS) device and the helium released has been measured as a function of temperature, thanks to the Quantitative GAs MEasurement System (Q-GAMES) [8]. These preliminary experiments have been performed on simple model glass. Low temperatures are sufficient to promote diffusion in a glassy structure and free volumes are numerous so that the conditions are optimum to test the set-up. Previous studies were carried out in simple glass [9] but not at such high pressures, which constitutes the novelty here. In addition, a first infusion in a resistive heated autoclave in UO_2 is reported and the helium desorption compared to the one of a glass sample.

2 EXPERIMENTAL SET-UP

All the helium infusions have been performed in APIS (All-Purpose Infusion Set-up), which is a high-pressure system installed at Joint Research Centre of the European Commission in Karlsruhe, Germany (JRC-KRU) for infusing gases in nuclear materials. In detail, APIS consists of two autoclaves which can operate separately or in parallel: a newly designed laser-heated autoclave used for infusing gas in uranium dioxide pellets and a resistance-heated autoclave shown in Fig. 1 and used in the herein presented experiments for infusing gas in transparent amorphous nuclear materials. Specifically, the latter device is a commercial autoclave marketed by Sitec - Sieber Engineering AG with a chamber volume of 0.8 mL and it can reach 2000 bar and 600°C. Moreover, the temperature can be easily managed by means of a controller connected to four heaters which being positioned equidistant inside the cylindric body of the autoclave ensure uniform heating of the sample. The sample-holder consists of a zirconia cylinder of 1 cm of diameter and 1 cm high. Thanks to the temperature controller, this resistance-heated autoclave is suitable also for experiments that requires long time of infusion since it can safely operate even for several consecutive days. In addition, being heated by heaters instead of a laser, its applicability is extended also to transparent materials, allowing the infusion of helium in glasses. Furthermore, the peculiar conical shape of the support on which the sample-holder is placed guarantees a perfect adherence between the fixed and the mobile parts of the autoclave which indeed results extremely tight. However, during the

opening/closing of the autoclave, since there are no windows, the movable part must be handled with extreme care to avoid damages in the point of contact, which would compromise the tightness.
In addition, a limitation of this autoclave lies in the long times it takes to clean it. In fact, to seal and protect the metal parts, a thin layer of anti-seize and lubricating paste is spread on the screw which allows fixing the mobile part to the fixed one. This nickel-based paste, resistant to high temperatures and pressures, must however be carefully removed after each experiment and for this purpose, the various removable components of the autoclave are cleaned in an ultrasonic bath.

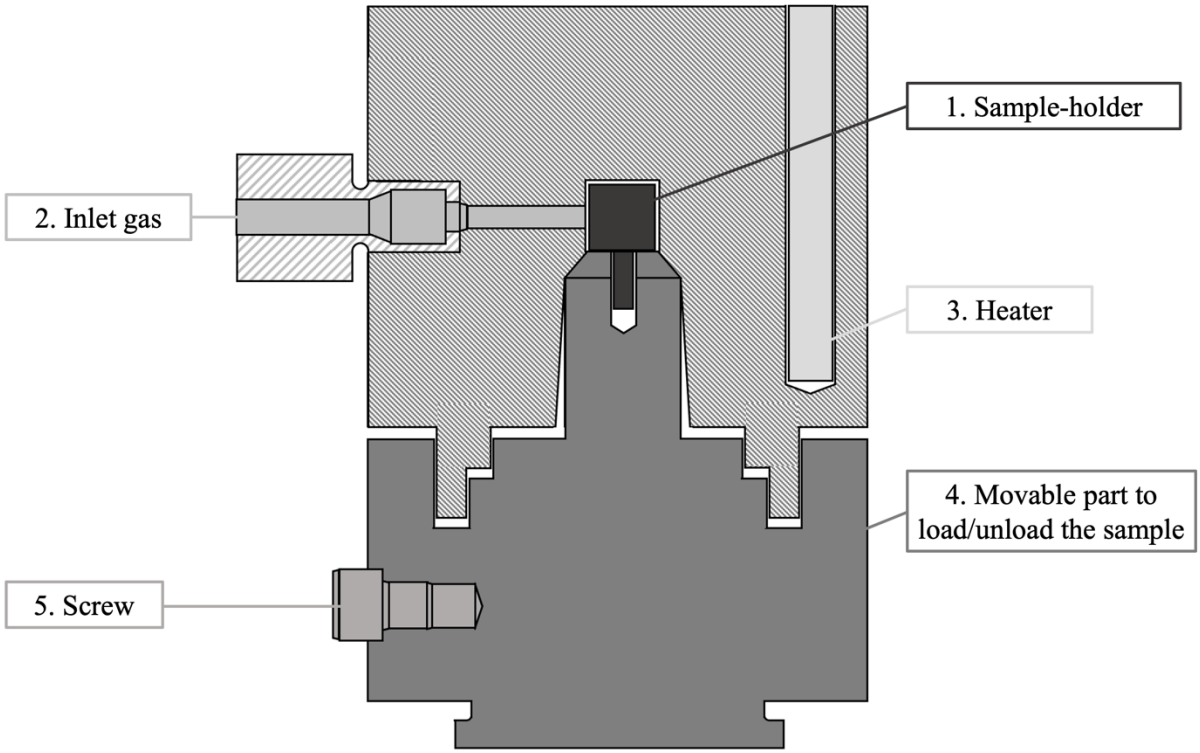

Figure 1: Schematic representation of the resistance-heated autoclave used to infuse helium in the glasses, with focus on the main elements which characterize it.

3 SAMPLES DESCRIPTION

The infused samples were squares 5x5 mm of CJ1, a transparent glass studied in the nuclear field for the vitrification of radioactive waste and whose composition is detailed in Table 1. Each sample, to be distinguished, was named with the glass name (CJ1) followed by a number (i.e., CJ1.1, CJ1.2, CJ1.3 and CJ1.4).

Table 1: Composition of the glass CJ1 investigated in the nuclear field as potential material for the vitrification of radioactive waste.

Glass CJ1	SiO_2	Na_2O	B_2O_3
mol %	67.7	14.2	18.1

4 EXPERIMENTAL RESULTS

All the infusions occurred at 300°C for 3 hours, but at different pressure values (ranging from 100 to 1000 bar), while all the samples underwent a similar annealing treatment (i.e.,

temperature ramp of 30°C min^{-1} from T_{room} up to 600°C, as shown in Fig. 2). After the infusions performed in APIS, the high-pressure system described in Section 2, the samples have been moved to a Thermal Desorption Mass Spectrometer (TDMS), in which they have been annealed in vacuum conditions. After the thermal treatment, the helium released from the samples have been discharged and by pumping, it has been collected in a high-pressure chamber and quantitatively measured by means of a Quantitative GAs MEasurement System (Q-GAMES) [8]. In detail, in Fig. 3 are reported the experimental curves of helium released from the infused CJ1 glasses as a function of the temperature. Given the high solubility of helium in amorphous materials, to avoid any helium leaking at room temperature, as soon as infused the glass samples have to be transferred immediately to TDMS. Moreover, to detect any macroscopic change, a picture of each sample has been taken by means of a camera installed in an optical microscope at every single step of the whole experiment (i.e., before the infusion, after the infusion and after the annealing). It is worth mentioning that already after the infusion performed at 1000 bar and 300°C for 3 hours, some unexpected helium bubbles appeared, as observable from the left panel of Fig. 4. In addition, to verify the reproducibility of these experiments and test the reliability of the set-up, each measurement has been performed twice, obtaining similar results. For the sake of brevity, are herein shown in Fig. 5 only the results obtained for the samples CJ1.2 and CJ1.4, two identical glasses which have been infused and annealed at the same conditions (i.e., Infusion Conditions – IC = 500 bar, 300°C, 3 h and Annealing Conditions – AC = temperature ramp of 30°C min^{-1} from T_{room} up to 600°C).

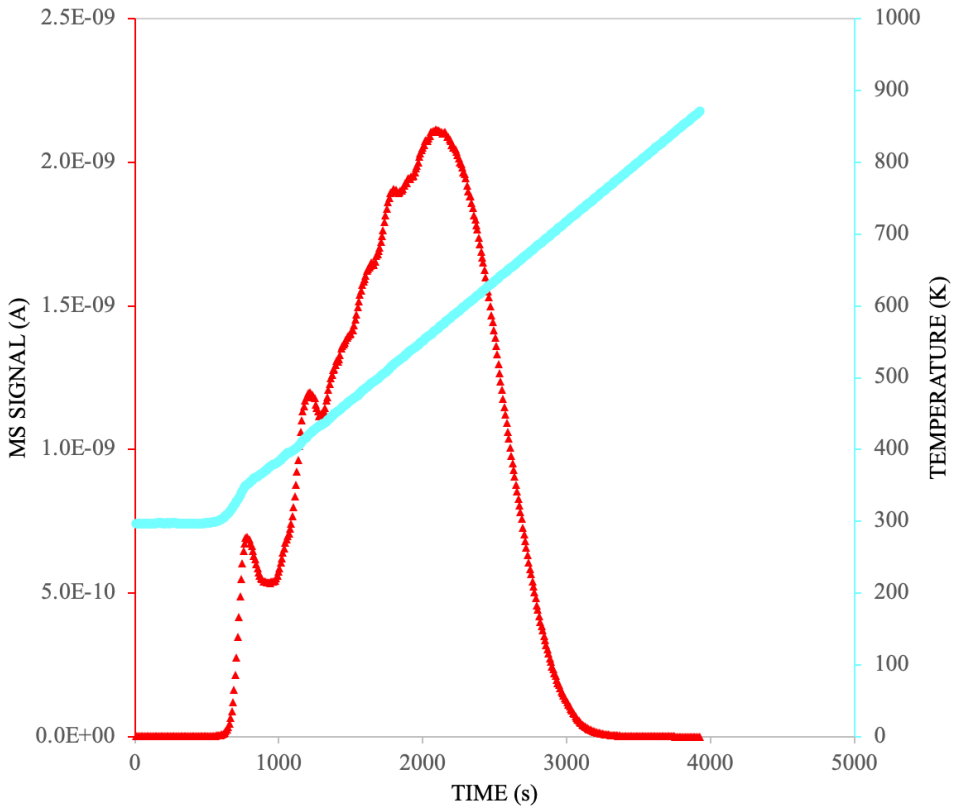

Figure 2: In red, experimental curve of helium released for thermal desorption from infused CJ1.2 glass. In light blue, temperature history of the thermal treatment to which the CJ1.2 sample was subjected.

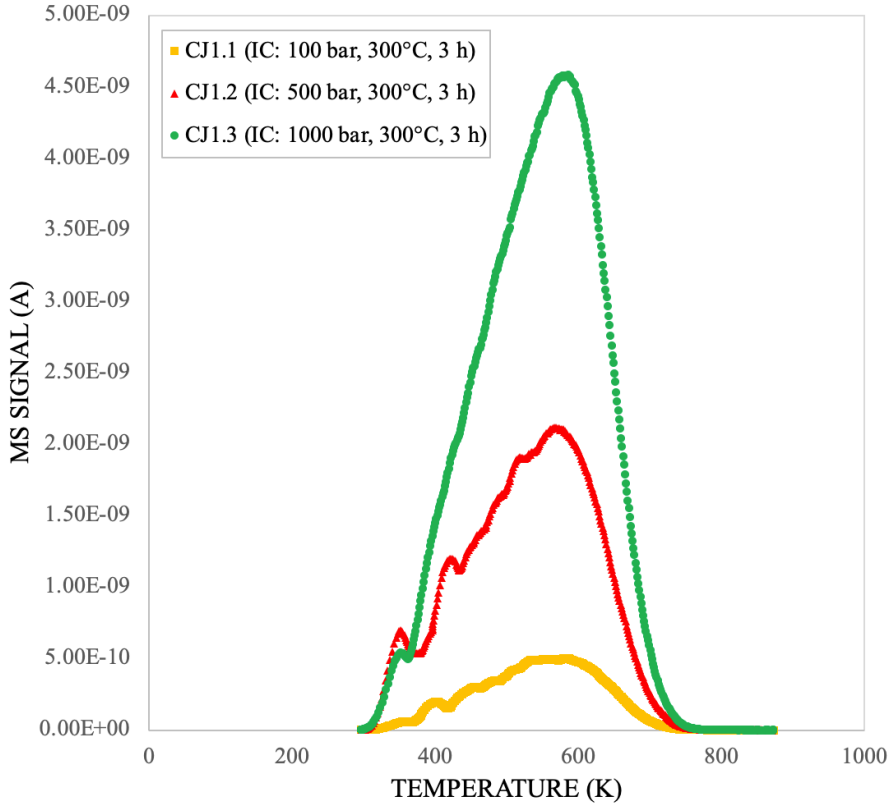

Figure 3: Comparison of the signals measured by means of thermal desorption mass spectrometer (TDMS) coming from glasses infused at the same temperature and for the same time, but in a different condition of pressure as reported in the legend (IC = Infusion Conditions). After the infusion, all the samples have been submitted to the same annealing treatment (i.e., temperature ramp of 30°C min^{-1} from T_{room} up to 600°C).

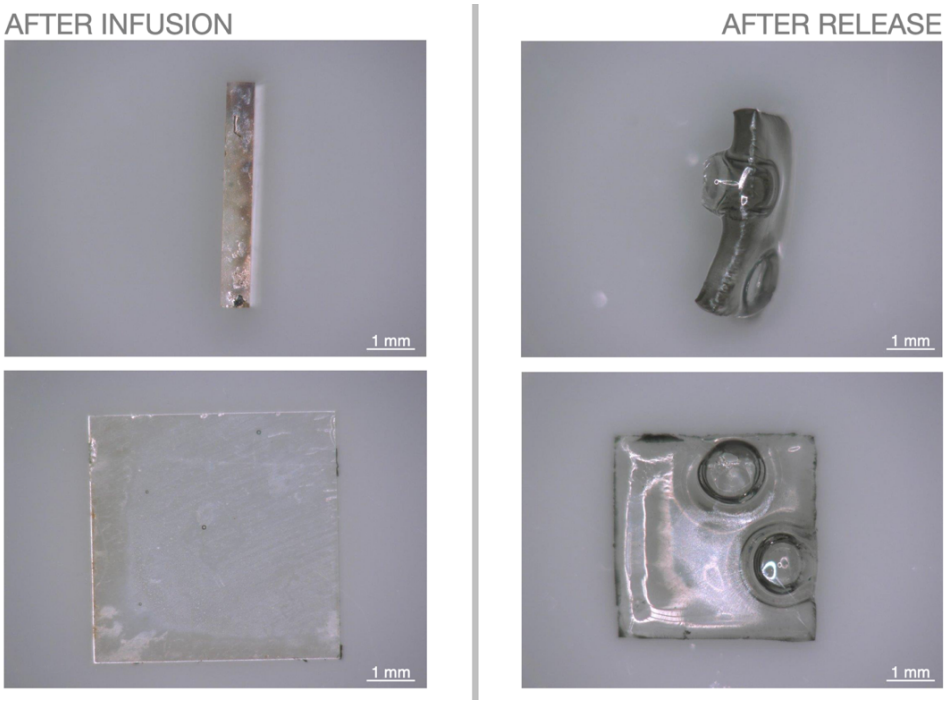

Figure 4: Picture of the same glass CJ1.3 after an infusion at 1000 bar and 300°C for 3 hours (left panel) and after an annealing from T_{room} up to 600°C with a ramp of 30°C min^{-1}.

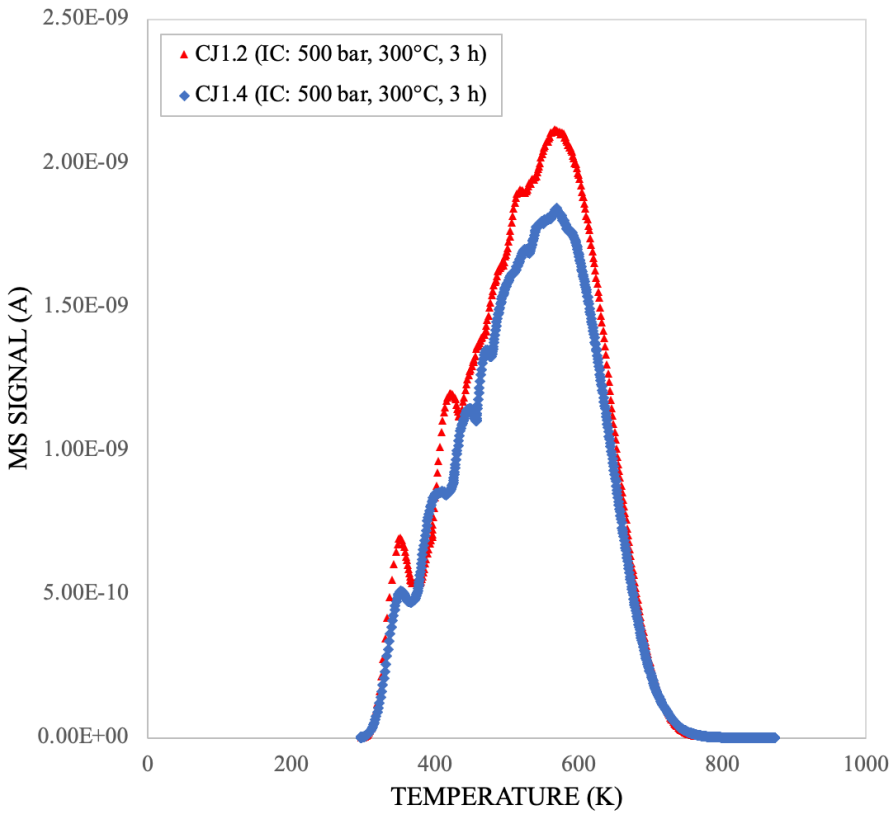

Figure 5: Comparison of the signals measured by means of thermal desorption mass spectrometer (TDMS) coming from two identical glasses, infused and annealed under the same conditions (Infusion Conditions – IC = 500 bar, 300°C, 3 h and Annealing Conditions – AC = temperature ramp of 30°C min^{-1} from T_{room} up to 600°C).

The use of the infusion has been initally guided by the study of nuclear fuel and in particular to fuels having cumulated some substantial amount of helium either by their composition and/or by waiting long enough time for radiogenic helium to build-up. In Fig. 6 is reported the helium that desorbed from a polycrystalline UO_2 sample (figure 6 – left panel – shows its microstructure at nanometric scale) infused at 1000 bars and 300°C for 30 hours. It is known that diffusion in the crystalline structure of UO_2 is extremely slow as is also its solubility [10 – 12]. However, even at low temperature some helium has infused in UO_2 and its release starts as for the CJ1 glass at the beginning of the thermal treatment and continues at much higher temperature (up to 400°C for the CJ1 glass and up to 1000°C for the UO_2 sample, respectively). For the UO_2, the peak of desorption is at slightly lower temperature compared to that for the glass, which coincides with the infusion temperature (i.e., 300°C). Moreover, looking at Fig. 6, it emerges that, unlike the green curve, in the orange one, which corresponds to the desorption measurement of helium from UO_2, there are numerous burst release which are probably attributable to the instantaneous helium release from the several pores which characterize the microstructure of the ceramic sample and are absent in the amorphous one.

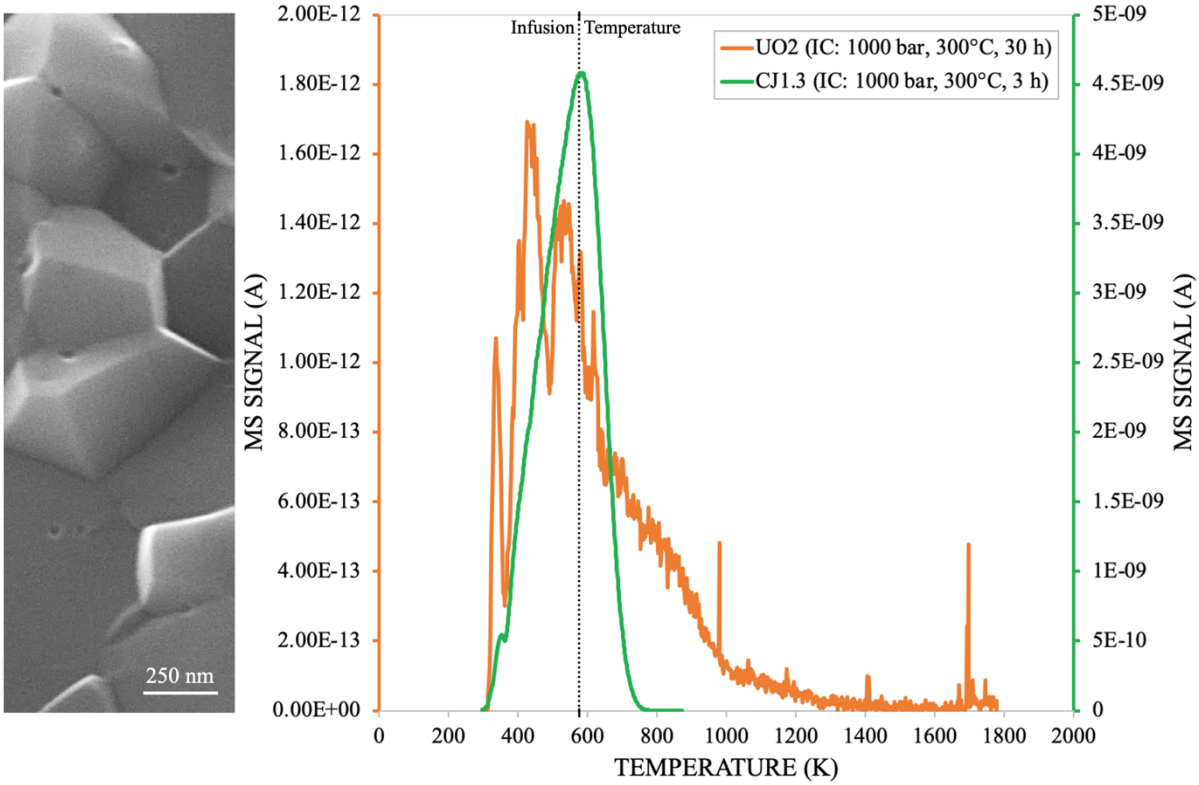

Figure 6: Comparison of the signals measured by means of thermal desorption mass spectrometer (TDMS) coming from a CJ1 glass sample and a UO_2 sample infused at the same temperature and pressure but for a much longer time.

5 CONCLUSIONS

Helium behaviour has been investigated in CJ1 glass samples infused in a resistance-heated autoclave. Besides the progress in the understanding how this glass behaves under different conditions of pressure, the new data herein reported represent a significant step towards the assessment of the transport mechanism of helium in amorphous materials. These measurements are also important to support the development of new models with enhanced capabilities for a safer management of spent fuel in storage conditions.

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