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Development of a device for helium thermal diffusion investigations by IBA in self-irradiated nuclear glass

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ABSTRACT

To minimize the amount of nuclear waste issuing from the nuclear power plants, the solution adopted in France consists in the reprocessing of spent fuel to isolate long lived and high level radioactive waste (minor actinides and fission products). They are incorporated into a glassy matrix in order to be placed in dedicated long-term disposal repository. The confinement of the radioelements depends strongly on the integrity of the glassy matrix which could be damaged by the radiations and the generation of helium produced by α-decays of the minor actinides. In the past few years, several studies were conducted in order to understand the behaviour of helium, especially its thermal diffusion into the glassy matrix [1–3]. However none were conducted on self-irradiated samples and a validation on radioactive glasses and in the temperature range of the repository conditions is still needed.

For this purpose, a specific setup was developed on the analysis chamber of the nuclear microprobe dedicated to radioactive samples in Saclay [4]. The temperature of the sample is controlled during all the experiment, in the range from 143 to 323 K; ^{3}He ions are implanted at low temperature. Helium profiles are measured at low temperature using the ^{3}He(d,p)^{4}He reaction, as-implanted and after several stages of annealing. We will present the developed setup and show the preliminary results of the measurements made on non-active samples.

1. Introduction

A consequence of the production of electricity by the mean of nuclear power plants is the generation of nuclear waste. In order to limit their volume, the strategy chosen in France consists in reprocessing spent fuel, i.e. operating a selective chemical separation in order to recycle U and Pu. Quantification of the level of hazard towards the environment and the population in the next hundreds of centuries is made according to the activity and half-life of produced isotopes. The most hazardous category, the High Level Waste (HLW) represented in France at the end of 2010 only 0.2% of the global volume of the nuclear wastes, but 96% of their radioactivity [5]. To ensure a safe confinement in long term disposal, HLW, composed of Long Lived Fission Products (LLFP) (e.g. ^{99}Tc, ^{135}Cs) and Minor Actinides (MAs, e.g. Am, Cm, Np), was vitrified and placed in dedicated stainless steel containers.

In France, the selected conditioning matrix is a complex borosilicate glass, known as R7T7 [6], designed to hold up to 18.5% of radioactive waste oxides. During the long term disposal period, it has to keep its rule of confinement. It could however be altered by internal radiation damage. Disintegration of MAs produces α particles and recoil nuclei (RNs). The heavy RN induces on a short distance a large number of atomic displacements that could modify the glassy structure while the alpha particle ends as a helium atom incorporated in the structure. The effect of alpha decay radiation damage on R7T7 type glass is discussed in [7]. Given the accumulation of helium in long run, its behaviour in the glass has to be understood and predicted to guarantee the confinement performances of the disposal device. In the past few years, Ion Beam Analysis (IBA) was widely used to investigate the helium thermal diffusion in various nuclear matrices [1,2,8–10]. ^{3}He ion is implanted in samples and profiles are measured as-implanted and after annealing using the ^{3}He(d,p)^{4}He reaction. Depth distribution modifications allow deriving diffusion coefficient and activation energy parameters.

Helium diffusion in oxide glasses [1–3] is higher than in oxides ceramics [8–10] which could lead to important migration at room temperature. In order to prevent this migration, the sample has to be maintained to a low temperature during implantation and profile measurements, and controlled during the annealing phase.

None of these studies examined the influence of self-irradiation. The aim of this paper is to present the special device developed on

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CASIMIR [4] in order to regulate the temperature of radioactive samples in the range [143 K, 323 K] into the analysis chamber. We will afterwards illustrate the performances of the developed facility describing the first measurements of non-radioactive samples.

2. Setup description

2.1. Nuclear environment of the device

The nuclear microprobe of LEEL [11], CEA Saclay, France, is equipped with a 3.75 MV HVEC Van de Graaff accelerator. The selecting magnet redirects the beam to one of the two microfocusing beam lines. The first one is used for the analysis of "classical" samples. The analysis chamber of second beamline is implanted in a radioactive controlled area. Received in the lab in shipping casks, radioactive samples are inserted in hot cells and handled with slaved arms. Controls are made to ensure that they are non-contaminant before transferring them into the analysis cell. In the absence of radioactivity, this cell can be accessed with the agreement of the radiosafety officer in order to allow maintenance or IBA measurements preparation. Before transferring the sample, it is closed by a shielding door and access is strictly forbidden.

Developing a device dedicated to control the sample temperature in such an environment represented a real challenge. The "active" part of the device is confined in a shielded closed cell. All the controlling parts which require direct intervention must be installed in an accessible location, but still in the restricted area, therefore subject to constraining rules and procedures.

2.2. Organisation of the setup

Fig. 1 shows a schematic view of the setup organisation. Heating or cooling of the sample is obtained by circulation of gaseous nitrogen N_2. The gas bottles are placed outside of the building and N_2 is piped into the controlled area at a pressure of 4 bars. According to the temperature controller requirement, N_2 can be either cooled or heated. Gas cooling is obtained by circulation of the gaseous nitrogen in a liquid nitrogen Dewar located in the controlled area in order to ensure periodic refills. Heating is generated by annular resistances fixed around an exchange reservoir crossed by the gas in the analysis cell, as close as possible from the analysis chamber. Both gas lines are merged before entering the analysis chamber.

In the analysis chamber, nitrogen flows through a small copper tank, heating or cooling the sample, stuck on a thin Cu plate, tightly pressed on the surface of the Cu tank. The gas is evacuated through very high efficiency filters after returning to room temperature.

Monitoring of the temperature is implemented by a PID controller. It commands the opening/closing of two electrovalves allowing the circulation of the gas in the heating or cooling devices. It controls also the power supply of the resistances. The input temperature of the PID controller is measured by a thermocouple (TC) placed on the entrance pipe of the gas in the analysis chamber in order to obtain a very stable regulation. The sample is fixed with a thin layer of epoxy resin on a copper plate and sample temperature is measured by a second TC set on the plate. Comparisons were made to verify that there is no difference between the sample temperatures and its Cu support. The sample temperature history is recorded on a computer during the whole experiment. Both TC chains were calibrated by simultaneous measurements with a certified thermometer in liquid nitrogen and in water at various temperatures from 273 to 353 K.

A Teflon block is used to interface the goniometer to the sample holder in order to guarantee the thermal insulation of the sample and Cu tank. The Cu plate with a special design for handling with slaved arms is pressed on the Cu thermal exchanger by a couple of springs to improve the thermal contact. Reference samples, used for beam refinement and energy calibration, are fixed in a special compartment.

The sample holder and the Cu tank are presented in Fig. 2.

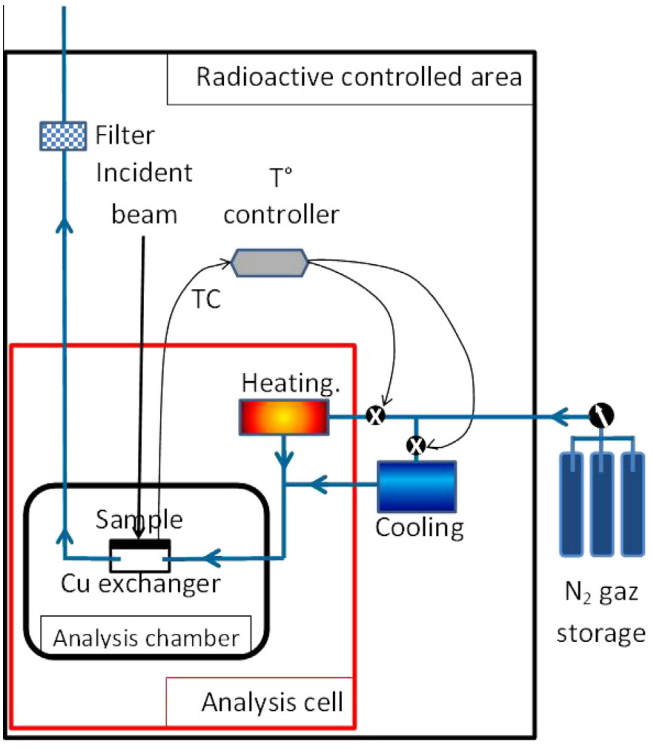

Fig. 1. Schematic representation of the heating/cooling device.

3. Measurement of non-radioactive samples

First measurements with the new setup were performed with non-radioactive samples. It was necessary to test each step of the

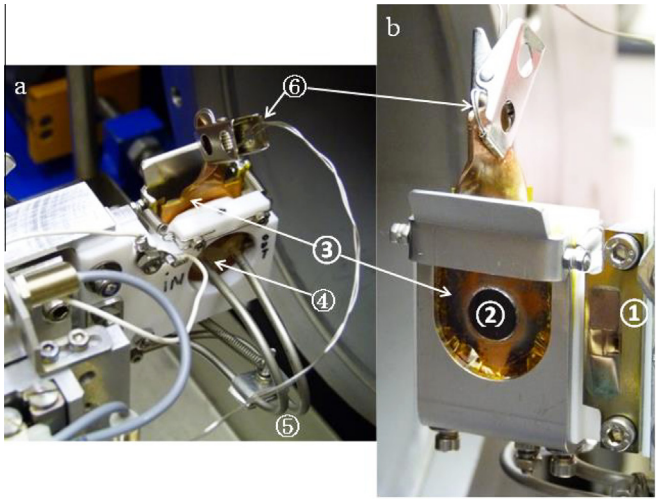

Fig. 2. Rear (part a) and front (part b) views of the sample holder, containing the reference samples (1). The sample (2) is glued on a Cu plate (3), pressed on the Cu tank (4) in which hot or cold N_2 circulates (5 and 6). Thermocouples (7) allow the temperature measurement on the Cu plate.