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In-situ high temperature irradiation setup for temperature dependent structural studies of materials under swift heavy ion irradiation

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ABSTRACT

An *in-situ* high temperature (1000 K) setup is designed and installed in the materials science beam line of superconducting linear accelerator at the Inter-University Accelerator Centre (IUAC) for temperature dependent ion irradiation studies on the materials exposed with swift heavy ion (SHI) irradiation. The $Gd_2Ti_2O_7$ pyrochlore is irradiated using 120 MeV Au ion at 1000 K using the high temperature irradiation facility and characterized by *ex-situ* X-ray diffraction (XRD). Another set of $Gd_2Ti_2O_7$ samples are irradiated with the same ion beam parameter at 300 K and simultaneously characterized using *in-situ* XRD available in same beam line. The XRD studies along with the Raman spectroscopic investigations reveal that the structural modification induced by the ion irradiation is strongly dependent on the temperature of the sample. The $Gd_2Ti_2O_7$ is readily amorphized at an ion fluence 6×10^{12} ions/cm^2 on irradiation at 300 K, whereas it is transformed to a radiation-resistant anion-deficient fluorite structure on high temperature irradiation, that amorphized at ion fluence higher than 1×10^{13} ions/cm^2. The temperature dependent ion irradiation studies showed that the ion fluence required to cause amorphization at 1000 K irradiation is significantly higher than that required at room temperature irradiation. In addition to testing the efficiency of the *in-situ* high temperature irradiation facility, the present study establishes that the radiation stability of the pyrochlore is enhanced at higher temperatures.

1. Introduction

Investigations of the properties of nuclear materials at extreme conditions such as high temperatures (~1000 K), exposure to radiation doses of around 200 displacements per atom (dpa), and severe environments like contact with salt or liquid metal [1], are important for their applications in Gen IV fast reactors and safe disposition of nuclear wastes as well as for the wide acceptability of nuclear energy. This explains the quest for the advanced materials which are stable towards above mentioned extreme conditions. The $A_2B_2O_7$ (pyrochlores) is one such class of compounds which shows high melting point, good thermal conductivity, non toxic and very high radiation resistance [2], and are possible candidates for such applications. There are various reports available in the literature related to the effect of the low and high energy ion irradiation on these materials. Energy dependent ion irradiation studies on the $Gd_2Ti_2O_7$ single crystals showed that ion fluence required for amorphization is much higher at low energy (5×10^{13} ion/cm^2 for 5.2 keV/nm) ion irradiation in comparison to high energy (5×10^{12} ion/cm^2 for 29 keV/nm) irradiation [3]. Temperature dependent *in-situ* microscopy studies on the single crystals of rare-earth titanate pyrochlores using 1 MeV Kr^+ ion irradiation showed that radiation resistance of the pyrochlore structure to ion beam-induced amorphization is not only affected by the relative sizes of the *A* and *B* site cations, but also the cation electronic configuration and the structural disorder [4]. Formation of strain-free, buried and disordered defect-fluorite layer within an ordered $Gd_2Ti_2O_7$ pyrochlore matrix is reported [5] on ion irradiation using 600 keV Ar^+ ions at the fluence of 6.5×10^{14} ions/cm^2. Ti-rich pyrochlores $Gd_2Ti_2O_7$ are initially transformed to a disordered, defect-fluorite structure by irradiation [6], but finally, become fully amorphous on irradiation using 1.0 MeV Kr^+ at an ion fluence of 7.6×10^{14} ions/cm^2. Radiation stability of the $Gd_2(Ti_{2-x}Zr_x)O_7$ is also found to increase by the substitution of Zr for Ti [7]. We have reported ion irradiation studies on the pyrochlores ($Gd_2Zr_2O_7$, $Nd_2Zr_2O_7$ and $Gd_2Ti_2O_7$ of varying r_a/r_b value 1.46, 1.54, and 1.75, respectively) with different swift heavy ion (SHI) of varying S_e. It was shown that $Gd_2Zr_2O_7$ and $Nd_2Zr_2O_7$ transform to anion-deficient fluorite and $Gd_2Ti_2O_7$ does not amorphize even with ion of highest S_e and fluence [8–10]. In summary, ion

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irradiation can induce amorphization, damage accumulation, plastic flow, implant precipitation, re-crystallization by thermal annealing, swelling and precipitation of new crystalline phases in the $A_2B_2O_7$ pyrochlore [11]. Most of the work reported in the literature is available on the ion irradiation studies performed at the 300 K temperature. Since, the temperature of the core of the reactor is about to 1000 K, therefore, it is important to investigate the effect of temperature on the radiation stability of the pyrochlore. In view of this, a high temperature irradiation facility has been designed, installed and tested in beamline of superconducting linear accelerator (LINAC) of IUAC.

The present work reports the design, fabrication and installation of high temperature ion irradiation facility in the materials science beamline. The newly installed facility is used for the study of radiation stability of the $Gd_2Ti_2O_7$ pyrochlore on irradiating at high temperature (1000 K). The structural modifications induced by the high temperature ion irradiation are explained on the basis of the competing processes of defect annealing by high temperature and amorphization by the electronic energy loss.

2. Design, fabrication and installation of high vacuum based high temperature ion irradiation setup

The *in-situ*/online facilities [12–14] is advantageous in view of better time economy and reproducibility of the data. Such an in situ facility becomes furthermore important for particle accelerators. In order to investigate the effect of temperature on the irradiation stability of materials, a high temperature set up is installed in the high vacuum chamber of the materials science beamline of the LINAC at the IUAC Delhi. For minimum conduction and transmission of heat from the UHV heater to vacuum chamber and beamline components, a custom-designed target ladder is fabricated. The UHV button heater procured from Heat Wave Labs Inc, USA is installed on this specially made target ladder using insulating machinable glass ceramic. The commercial machinable glass ceramic (Macor, supplied by Corning Incorporate) used in high temperature setup, is made up of fluorphlogopite mica embedded in a borosilicate glass matrix. It is a good thermal insulator, and is stable up to temperatures of 1273 K, without out gassing. The coefficient of thermal expansion (300–1073 K) is 1.3×10^{-5}/K. Fig. 1 shows the schematic diagram of the high temperature target ladder (Fig. 1a), the photographs of high temperature set up (Fig. 1b), high temperature set up installed in the high vacuum chamber (Fig. 1c), and illuminated view of target ladder at 1000 K in vacuum (Fig. 1d). The temperature of the sample is controlled using programmable Watlow EZ-Zone PM temperature controller. The maximum set temperature in the controller is 1643 K. The commercial UHV heater (Model #101491-01) is procured from Heat Wave Lab, USA. It consists of a heating filament which is fixed in a cylindrical shaped radiation shield. The heater is able to attain a temperature of about to 1500 K. The temperatures are measured by a chromel–alumel thermocouple mounted at the centre of the heater (TM1) and connected to a temperature controller. The thermocouple, which has a thin flat tip, is inserted through a small hole in the backside of the radiation shield chamber and perfectly contacted with the heater filament for accurate measurement. Fig. 2 shows the heating and cooling curves for the high temperature setup. During test experiment, variation in the temperature is found to be ±2 K, which is acceptable for structural modification studies. To measure the real temperature of the target surface, another K-type thermocouple (TM2) is mounted on the surface of the target. The temperature value of the TM2 is lower than the TM1 and the temperature difference is about 40 K. This difference arises due to poor heat conduction between heating surface of heater and target because target is mounted with help of clips. For ion irradiation studies, the ion beam has to be aligned on the quartz sample so that it can be viewed before targeting it on the sample. Therefore, a quartz sample is also mounted on the target ladder in the same vertical plane as that of the sample and heater. The vacuum in the chamber is created using turbo molecular pump backed with a rotary pump.

3. Preliminary results: *in-situ* temperature dependent ion irradiation studies on $Gd_2Ti_2O_7$

The experimental results of temperature dependent 120 MeV Au^{+9} ion irradiation effects on the structural properties of polycrystalline $Gd_2Ti_2O_7$ and its damages created by electronic excitation are presented in this section. Polycrystalline $Gd_2Ti_2O_7$ samples were synthesized by standard multi-step solid-state route at the Bhabha Atomic Research Centre, Mumbai (BARC), India. Gd_2O_3 and TiO_2 (AR grade) were taken as reactants. The stoichiometric amounts (1:1) of reactants were weighed, ground and pelletized. The samples were heated at 1200 °C and 1300 °C with intermittent grinding and repelletizing. The single-phase $Gd_2Ti_2O_7$ was observed at 1300 °C. To achieve highly sintered pellets and to annihilate the defects, final annealing was done at 1450 °C for 48 h under slow-cooled conditions (2 °C/min). Pellets with greater than 90% theoretical density were obtained which were further used for irradiation studies. The crystallographic phase of the as-prepared pyrochlores samples in powder form is confirmed using XRD studies. The pyrochlore structure ($A_2^{3+}B_2^{4+}O_6O'$), is the superstructure of fluorite structure (MX_2; $Fm\bar{3}m$ space group) with two cations and one-eighth fewer oxygen anions. In this structure, the A^{3+} cations are generally rare-earth elements occupying 16d Wyckoff positions (1/2,1/2,1/2) whereas smaller B^{4+} are transition metal elements that lie on 16c Wyckoff positions (0,0,0). There are two different oxygen sites, one is the 48f oxygen coordinated to two B^{4+} and two A^{3+} cations, while 8b oxygen is coordinated to four A^{3+} cations. The ion irradiation is carried out using the 15UD Pelletron accelerator facility to investigate the irradiation induced effects on the structure stability of the pyrochlore structure. The value of the ion current is measured using a secondary electron suppressed Faraday cup and measured ion flux was 6.25×10^9 ions/sec-cm^2. The power deposited by the ions is about 0.12 W/cm^2. There is no observable increase in the temperature of the sample on irradiation. Table 1 lists the energy losses and projected ion ranges [15] in $Gd_2Ti_2O_7$ having density [16] $\rho = 6.567$ g/cm^3. In the present cases, the electronic energy loss for Au ions is significantly higher than the corresponding nuclear energy loss, and thus electronic energy loss dominates the interaction process. In order to characterize the radiation damage and structural modifications, the samples were characterized using *in-situ* XRD for room temperature irradiated sample and *ex-situ* XRD for high temperature irradiated sample. The XRD measurements were performed with the Bruker D8-Advance diffractometer [12], using Cu Kα (0.15406 nm) and operating at 40 kV and 40 mA. The X-ray patterns were recorded in standard θ–2θ geometry in a range of $2\theta = 20$–$80°$ with a step size of 0.02°. The penetration depth of X-rays in the present is about 1.73 μm, as calculated for main peak (222). The ratio S_e/S_n is vary from 58 (surface) to 40 (depth ~ 1.73 μm) which is almost constant up to probing depth of the X-ray diffraction. All irradiations were carried out at room temperatures under a vacuum of 10^{-5} mbar.

Here, two experiments have been performed simultaneously (a) irradiation at 300 K where sequential *in-situ* diffraction (Irradiation-Analysis-Irradiation and so on...) is performed and (b) irradiation at 1000 K where subsequent *ex-situ* diffraction is performed.

For 300 K irradiation–simultaneous *in-situ* diffraction, structural studies of the pristine and ion irradiated $Gd_2Ti_2O_7$ are carried