New Ion Beam Materials Laboratory for Materials Modification and Irradiation Effects Research

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Abstract

A new multifunctional ion beam materials laboratory (IBML) has been established at the University of Tennessee, in partnership with Oak Ridge National Laboratory. The IBML is currently equipped with two ion sources, a 3 MV tandem accelerator, three beamlines and three endstations. The IBML is primarily dedicated to fundamental research on ion-solid interaction, ion beam analysis, ion beam modification, and other basic and applied research on irradiation effects in a wide range of materials. An overview of the IBML facility is provided, and experimental results are reported to demonstrate the specific capabilities.

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Keywords: tandem accelerator; ion beam analysis; ion-solid interaction; irradiation effects

1. Introduction

Ions with keV to MeV energies can be used to controllably change electrical, optical, structural, mechanical and chemical properties of materials in the surface region up to a few micrometers. Ion irradiation has the advantage of having well-controlled conditions that allow for fundamental studies on ion-solid interaction and material response to extreme radiation environments. Applications of such energetic ions include advanced electro-optical device fabrication, nanostructure engineering, and radiation effect studies for nuclear materials and space exploration [1 - 10]. For nuclear energy applications, controlled ion irradiation approaches can be utilized to explore a wide range of irradiation conditions (e.g., dose, dose rate, damage energy density, and temperature) in nuclear materials and achieve better understanding from separate effects evaluation and testing, as well as combined or simultaneous effects of integrated irradiation studies. Advances in understanding the response of materials to energetic ions have significantly contributed to the knowledge of irradiation damage from high-energy neutrons on microstructure evolution, and changes in chemical and physical properties in advanced structural steels, fuels, and cladding materials. Progress made in ion-solid interactions, including defect production, microstructure evolution and damage recovery, has led to many technological advances to create new functionalities in metals, semiconductors, ceramics and polymers [1-10].

Diminishing research and training capabilities in the USA for ion beam analysis (IBA), ion beam modification, fundamental understanding of ion-solid interaction, and irradiation effects research in materials have prompted the University of Tennessee (UT), in partnership with Oak Ridge National Laboratory (ORNL), to develop a new multifunctional Ion Beam Materials Laboratory (IBML, <http://ibml.utk.edu/>) located on the UT campus. The primary mission of the UT-ORNL IBML is to address the scientific challenges associated with both fundamental and applied programs, as well as educational and training needs. Since operation of this laboratory began in the fall of 2012, it has been utilized to support various research needs for national energy and security missions. The purpose of this

paper is to describe the capabilities in this new facility with some recent studies on defect formation and damage accumulation.

2. Overview of IBML facility

The IBML, with laboratory space of $\sim 300 \text{ m}^2$ (45 ft $\times$ 70 ft), is equipped with two ion sources, an injection magnet, an electrostatic tandem ion accelerator, an analyzing magnet, three high-energy beamlines, and three endstations, as shown **Fig. 1**. In addition, one low-energy beamline and its endstation after the injection magnet are in the development stage. The IBML has a separate sound-proof control room with a glass wall overlooking the accelerator, beamline and endstation areas. This facility is capable of accelerating a variety of ion species over a range of energies from a few hundreds of keV to a few tens of MeV to meet the needs for education, fundamental research and applied programs.

One ion source is a radio frequency (RF) charge exchange ion source (Alphatross) for producing gas ions. At the IBML, this ion source is used to produce helium ions. The other ion source is a Source of Negative Ions by Cesium Sputtering (SNICS) II. Most heavy ions, as well as hydrogen ions, are produced from a solid cathode by Cs^+ sputtering. Elemental or compound cathode material containing the required elements is compacted inside a cathode rod, normally made out of Cu due to its high thermal and electrical conductivity. For Ni or Fe ions, pure Ni or Fe cathodes are used to avoid difficulties of mass separation from Cu. The negative ion beam, produced from either the Alphatross or the SNICS source with pre-acceleration energies of a few tens of keV, is bent at a 30° angle (**Fig. 1**) by the injection magnet into the central beamline of the accelerator. The accelerator is a 3.0 MV Pelletron (model 9SDH-2) tandem electrostatic accelerator, manufactured by National Electrostatics Corporation (NEC, WI, USA, <http://www.pelletron.com>). Calibration of ion energy was carried out in generating voltmeter (GVM) mode using a resonant reaction $^{16}\text{O}(\alpha, \alpha)^{16}\text{O}$ [11]. The resultant peak energy is well within the error for the reported resonant peak energy with an overall error of $\pm 10 \text{ keV}$.

A unique design of the injection magnet at the low-energy end of the accelerator is the three bending ports, which makes low-energy ion beam capabilities available at an insignificant additional cost.

A 0° port is positioned along the central line where the ions travel to the accelerator (**Fig. 1**). The other two ports are at $\pm 50^\circ$. One low-energy (LE) beamline is shown in **Fig. 1** downstream from the injection magnet along the $+50^\circ$ port away from the central beamline of the accelerator. Such a design enables 100° bending of light ions (He at this IBML) from the Alphasross ion source, and modest bending (20°) of heavy ions from the SNICS source. Based on the voltage (up to 80 keV) and current capabilities (up to a few tens μA) of both ion sources, this LE beamline allows applications, such as low-energy high-current implantation, shallow doping, and surface modification. Specifically, the penetration depth from H and/or He implantation from this low-energy capability can be controlled to match that from heavy ion irradiation produced from the high-energy end of this accelerator. Such overlap of the ion ranges meets the requirements to investigate cavity formation under heavy ion irradiation with and without H and He present in various nuclear materials for reactor applications. Moreover, low-energy implantation of certain fission products into the surface region from this low-energy capability will also enable migration studies of fission products to the surface and to the bulk under subsequent irradiation conditions at elevated temperature. As a preliminary test, 50 keV He ion implantation into a glass sample was performed, and the beam shape and uniformity was monitored by the luminescence from the sample. Further development of this LE beamline will include adding an electrostatic steerer to deflect the beam, an einzel lens to focus the beam, a beam profile monitor to observe beam shape, a Faraday cup for current measurement, double slits or collimator to define beam size. A customized position- and temperature-controlled target chamber will allow H and He implantation or other surface modification by heavy ions to be performed at temperature ranging from 150 to 1000 K.

The analyzing (switching) magnet at the high-energy end of the endstation has seven ports at -45° (L1), -30° (L2), -15° (L3), 0° (L4), $+15^\circ$ (L5), $+30^\circ$ (L6), and $+45^\circ$ (L7), with respect to the accelerator. Three high-energy beamlines are located at the L3, L5, and L6 ports downstream from the analyzing magnet, as shown in **Figs. 1** and **2**. The energetic positive ions after the accelerator are focused using a magnetic quadrupole and a Y-axis electrostatic steerer into the analyzing magnet and bent into one of the

three research beamlines. The L6 beamline and its endstation are devoted to rapid routine ion beam analysis at room temperature. Two other beamlines (L3 and L5) with temperature-controlled endstations are equipped with some unique capabilities for ion beam analysis, modification, and radiation effect studies that will be described in the following section.

3. Endstations and capabilities

There are three operational endstations. The manipulators or goniometers in these endstations are computer-controlled, and the working pressure is in the low 10^{-5} Pa range. Higher vacuum conditions are possible, if needed.

3.1 Endstation at beamline +30° (L6)

The physical and chemical properties of many materials are dominated by the characteristics of the surface, interface, and near-surface composition and structure. Understanding how the surface region affects material properties depends on the availability of surface analysis techniques. Over the last few decades, demands on developing and improving these analysis techniques have been strengthened by demands from the electronics manufacturing and device industry. Ion beam analysis for thin film characterization has, therefore, developed into a mature field of applied science and today comprises numerous techniques. The endstation at the +30° beamline (L6) is equipped with most of the standard ion beam analytical capabilities, including Rutherford backscattering spectroscopy (RBS [12-14], Non-Rutherford backscattering spectroscopy (NRBS), conventional elastic recoil detection analysis (ERDA) [15-18], and nuclear reaction analysis (NRA) [19, 20].

This room-temperature ion beam analysis endstation is equipped with a computer controlled motorized five-axis target manipulator attached through the bottom port of a vacuum chamber. The well collimated parallel beam, such as H, He and C ions, passes through the beam profile monitor, X-Y steerer, double slits, and a cylindrical aperture to impinge on a sample. The large versatile sample holder (15 mm × 90 mm or 15 mm × 45 mm) can load multiple samples with different size and shape, as shown in **Fig. 3**.

1 This combination of the multiple-sample handling capability, together with the loading lock system,
2 enables rapid sample exchange within a relatively short period of time. There are two solid-state detectors
3 (ORTEC, USA) with configurations shown in **Fig. 4**, one fixed Si detector with a scattering angle (θ) of
4 168° and solid angle of 2.3×10^{-3} sr, and one movable Si detector with a scattering angle ranging from
5 30° to 170° and solid angle of 1.2×10^{-3} sr . A rotatable foil holder mounted in front of the movable
6 detector (**Fig. 3**) allows up to six foils with different thicknesses/composition for conventional ERDA
7 measurement. Foil position is controlled from outside the vacuum chamber. While the fixed detector
8 located underneath the probing beam at the backscattering angle of 168° has an advantage of combining a
9 large scattering angle (for optimized mass resolution), as well as grazing incident (α) and exit (β) angles
10 (for optimized depth resolution) in the RBS and NRBS measurements, the movable detector allows NRA
11 and conventional ERDA measurements performed at required detection angles. An equipped
12 microcomputer with multichannel analyzer card and software are designed for automated data collection
13 and data analysis. Individual users may use this endstation independently for their research, following the
14 necessary training.

15 All the silicon detectors (ORTEC, USA) used in L6, L5 and L3 endstations for RBS
16 measurements are calibrated routinely, or mandatorily when different settings of the associated
17 electronics are chosen. For energy calibration, standard calibration samples, consisting of a thin mixed
18 AgCuAu layer with thickness varying from 5 to 20 nm deposited on a carbon substrate, are used. During
19 the storage in air, the metal layers are oxidized, which provides an extra calibration point from the surface
20 O peak. In addition, other calibration samples with thin layer of Pt or Ag on Si substrate were also used
21 occasionally for additional calibration points or confirmation. The scattering angle is also confirmed using
22 the standard calibration samples. The complication associated with Si detector energy calibration is
23 discussed elsewhere [21-25].

24 Ion beam analysis is utilized to probe the composition of and to obtain elemental depth profiles in
25 the near-surface region of solids. The depth resolution is typically from several nanometers to a few tens

of nanometers, depending on the nature of samples and measurement geometry. Atomic depth resolution may be achieved with advanced detection systems. While the elemental composition and film thickness are characterized by RBS at random directions, irradiation-induced damage in crystalline samples can be determined by RBS along one or more major channeling directions. RBS measurements are normally performed using a 2.0 MeV He^+ beam with Si detectors located at scattering angles of 168° and 150° relative to the incoming beam. NRBS measurements may be carried out at higher He beam energies [2,11], such as at 3.5 MeV to enhance the detection sensitivity for O and C atoms. Such NRBS measurements enhance scattering cross sections for lighter elements, can probe deeper and provide better signal separation. For example, NRBS has been utilized to better determine possible changes in O/Ce ratio in nanocrystalline CeO_2 films due to ion irradiation [26] or changes in disorder level on both the Si and C sublattices in SiC crystals from a single backscattering measurement [27].

Irradiation-induced interface mixing has been investigated at this endstation based on RBS results. Nanocrystalline zirconia films with thickness of ~ 400 nm were deposited on a (001) silicon substrate by ion-beam assisted deposition (IBAD) [26, 28]. The zirconia films were irradiated at endstation L3 with 12 MeV Au^{4+} ions at ion fluences ranging from 1×10^{14} to $2 \times 10^{16} \text{ cm}^{-2}$. RBS measurements were carried out at endstation L6 using 2.0 MeV He^+ to probe the entire irradiated zirconia film and the interface region. For clarity purpose, only the spectra from the as-deposited film and the films subjected to high-fluence irradiation are shown in **Fig. 5**. While possible composition change (the O/Zr ratio) is not obvious within the detection uncertainty, irradiation-induced interface mixing can be observed with increasing Au fluences, starting at $2 \times 10^{15} \text{ cm}^{-2}$ and becoming evident at $6 \times 10^{15} \text{ cm}^{-2}$. After a fluence of $2 \times 10^{16} \text{ cm}^{-2}$, a wide mixed region over a 100 nm thick is formed, as shown in **Fig. 5**.

To demonstrate the conventional ERDA capability at the L6 endstation, a measurement was conducted to determine an implanted He profile in SiC. A single crystal 3C SiC was implanted with 65 keV He^+ ions to $1 \times 10^{16} \text{ cm}^{-2}$ at 7° off the surface normal to avoid channeling implantation. The ERDA experimental configuration is shown in **Fig. 6(a)**. A beam of 11 MeV O^{4+} ions was directed onto the specimen at 75° off the surface normal, and a Mylar foil ($\text{C}_{10}\text{H}_8\text{O}_4$) with a density of 1.4 g cm^{-3} and

thickness of 9.6×10^{19} atom/cm² (10 μ m) was placed on the rotatable foil holder in front of the movable Si detector (**Fig. 3**). This Mylar foil can stop oxygen ions with energy up to 13.5 MeV. Therefore, it completely blocks the forward scattered beam and recoiled heavy particles, except for H and He from the surface region as shown **Fig. 6(b)**. The He depth profile was estimated using the Stopping and Range of Ions in Matter (SRIM) 2012 code [29], and the peak of the He concentration was predicted to be at a depth of 350 nm from the surface, as shown in the inset of **Fig. 6(c)**. The depth profile in nanometers was acquired by converting the channel numbers based on a linear energy calibration of electronics. The helium concentration profile determined from the conventional ERDA spectrum is shown in **Fig. 6(c)**. The peak He concentration is about 0.8% (8000 appm) at ~ 350 nm from the sample surface predicted using SRIM2012 [29]. The difference in He distribution between the SRIM prediction and the measured data may be attributed to the underestimation of the Mylar foil thickness.

3.2 Endstation at beamline +15° (L5)

The endstation L5 has a custom designed ultra high vacuum (UHV) target chamber (stainless steel 355.6 mm in diameter and 660.4 mm in height) with several ports located at three horizontal levels for vacuum control and analytical capabilities. The ports at the bottom level are designed for vacuum control (connection to pumps and vacuum gauges). The middle level is the beamline level that contains ports for beam controls, temperature measurements, ion beam analysis, and viewports. The top level opens the possibility for future additions, such as adding an ion gun or electron gun for multiple beam irradiation capabilities. This target chamber accommodates a customized liquid-nitrogen cooled manipulator from the top opening flange. The manipulator (Thermionics, NW, WA, USA) has 3 axes of rotations and 3 axes of translation and is ideal for effective *in-situ* channeling measurements to study defect production/annihilation and damage recovery. The motorized polar, azimuthal and flip rotation axes range of $\pm 180^\circ$, $\pm 180^\circ$, $\pm 25^\circ$ with the corresponding resolutions better than 0.01° , 0.001° , and 0.025° per step, respectively. In the routine channeling measurement program, the finest steps of the polar

1 and flip rotation are typically set at 0.018° , and 0.02° , respectively. The motorized X, Y and Z axes have
2 ranges of ± 25.40 , ± 25.40 and ± 203.20 mm, respectively, with a resolution of 0.03 mm. The samples can
3 be mechanically mounted on a 17 mm diameter circular molybdenum plate (0.5 mm in thickness) by
4 molybdenum spring-loaded clips, with a chromel–alumel thermocouple clamped to the sample surface or
5 the molybdenum plate. The samples can be heated up or cooled down within a temperature range of -120
6 to 800°C . The manipulator, with an internal liquid-nitrogen circuit, allows sample cooling from room
7 temperature down to -120°C in 100 min. Samples can be heated from the rear using an electron
8 bombardment heater. It consists of a low power tungsten filament, which is capable of heating most
9 sample materials to over 300°C . An adjustable bias supply of up to -500 V provides an acceleration
10 potential up to 100 mA emission. Samples can be efficiently heated from room temperature to $800 \pm 5^\circ\text{C}$.
11 A fixed surface barrier detector (Ortec PIPS) is mounted on the middle level (the same horizontal plane as
12 the ion beam) and has a fixed scattering angle of 155° for RBS and channeling measurements with a solid
13 angle of 1.1×10^{-3} sr. The scattering geometry is the same as shown in **Fig. 4** (left).

14 One essential prerequisite for ion implantation is the correct determination of the ion fluence. At
15 both L3 and L5 endstations, a Faraday cup, located below the sample holder, is employed to measure the
16 beam current. A current monitor and digital counter (CMDC) was developed in order to monitor the
17 current on the samples during irradiations or ion beam analysis. The system is constructed using an
18 inexpensive open-source microcontroller board, Arduino (<http://www.arduino.cc/>). Two configurations
19 are developed: a single channel CMDC based on an Arduino Uno, and a four-channel CMDC based on an
20 Arduino Micro. For the one-channel version, the input signal goes directly into a digital Pin that is
21 connected to a hardware 16-bit Timer/Counter. This unit is able to count standard TTL pulses with a
22 minimum length of $0.5\ \mu\text{s}$ and a minimum time of $0.2\ \mu\text{s}$ between pulses. The four-channel version is a
23 more sophisticated system, including a logic buffer (SN74HCT125), two dual 16-bit counters
24 (SN74LV8154), and an Arduino Micro. The buffer decouples and protects the following Integrated
25 Circuits and the Arduino Micro itself. The two counters are controlled by the Arduino Micro, which

1 activates the counting and the communications. The data are fed to the Arduino through an 8-bit parallel
2 digital bus. The minimum length of a standard TTL pulse is 40 ns. This four-channel CMDC
3 configuration can be interfaced with a Brookhaven Instr. Corp. 1000C Current Integrator, or an ORTEC
4 439 Digital Current Integrator, or used to count standard TTL pulses. Each channel is capable of
5 monitoring a current meter or pulse generator, independently from the other channels. The boards are
6 controlled and powered through a USB cable connected to a computer that acts as a data acquisition and
7 logging system. A portable Graphical User Interface (GUI) software program (**Fig. 7**) was written with
8 Python (<https://www.python.org/>), and was tested both on Windows and on Mac OS X. The software is
9 capable of displaying the acquired data in real time and calculates the collected charge during the
10 experiment, as well as storing and loading data from a file. This GUI can also display the value of the
11 total collected charge, the average of the current, and its standard deviation.

12 The ion fluence can be calculated from the beam spot size that is normally regulated by the
13 double slits on the beamline close to the target chamber. Uncertainty in beam current measurements exists
14 due to the offsets of the slits, incomplete secondary electronic suppression of the Faraday cup, as well as
15 limited resolution of the current meter (higher uncertainty at low current close to 1 nA), current integrator
16 and other associated electronics. Fluence calibration is, therefore, necessary. Several fluence calibrations
17 have been performed for both L3 and L5 beamlines and endstations. Au ions were employed in the
18 calibrations because of the achievable high beam current and high backscattering cross section of He on
19 Au (for RBS measurements). One example of fluence calibration is illustrated in **Fig. 8**, where a Si
20 sample was implanted with 2.25 MeV Au²⁺ ions. After the Au implantation to an ion fluence of 6.1×10^{15}
21 cm⁻², random RBS was performed using a 2.0 MeV He⁺ beam. As shown in **Fig. 8**, the signal from the
22 implanted Au ions appears in a background free region (channels from 800 to 1150), easily separable
23 from the signal of the light Si substrate (channels 750 and below). Moreover, a large scattering cross
24 section leads to a reasonable backscattering yield thereby minimizing the measurement and analysis
25 uncertainty of RBS. The concentration of Au shown in the RBS spectrum is determined by fitting the data
26 to a simulation using SIMNRA software [30]. The diamonds in **Fig. 8** are the data from the RBS

measurement, and the dashed line is the fit from SIMNRA. In this example, while the intended fluence is $6.1 \times 10^{15} \text{ cm}^{-2}$, the fluence determined by SIMNRA simulation is $6.3 \times 10^{15} \text{ cm}^{-2}$. Such fluence calibration has been performed several times since the fall of 2012 using Au ion with different energies in different targets. Some of the Si samples implanted to different ion fluences were analyzed using Time-of-Flight secondary ion mass spectrometry (SIMS), and the intended ion fluences were confirmed using the SIMS technique. A summary of the calibration measurements is provided in **Table 1**, showing that the uncertainty of our irradiation fluence is within $\pm 10\%$.

The main purpose of the L5 endstation is to efficiently study defect production and damage accumulation using ion channeling techniques (RBS/C). With this in mind, the beam optics, the manipulator and sample holder are designed to allow irradiation studies to be performed at well controlled areas or spots on one crystalline sample, as demonstrated in the following experiment. Ion irradiations of 900 keV Si^+ with fluences ranging from 7.0×10^{13} to $1.4 \times 10^{15} \text{ cm}^{-2}$ were carried out at room temperature on a single crystalline 3C SiC sample (NOVA, 3.8 μm film on Si substrate). The Si irradiation was performed at 60° off the surface normal to produce a shallow damage profile that can be readily measured by the channeling technique. The average dose in dpa (displacements per atom) was estimated under full-cascade simulations using the SRIM code (2008 version) [29] under the assumptions of a sample density of 3.21 g cm^{-3} and threshold displacement energies of 20 and 35 eV for the C and Si sublattices, respectively [31]. The conversion factor at the damage peak from ion fluence (10^{14} cm^{-2}) to dose (dpa) is 0.072 under the 900 keV Si^+ irradiation at 60° off the surface normal. The corresponding damage accumulation was investigated using both RBS and NRBS (**Fig. 9(a)** and **Fig. 9(b)**, respectively) techniques along the $\langle 111 \rangle$ -axial channeling direction. As shown in **Fig. 9(b)**, eleven spots with size of $3 \times 3 \text{ mm}^2$ were irradiated on the same sample with a 0.5 mm gap between the spots. Smaller spots can be irradiated if needed. To ensure uniform irradiations, the ion beam was rastered over the designated irradiated area with fixed horizontal and vertical scan frequencies of 517 and 64 Hz, respectively. The crystal was aligned with the He beam of a probe size of $1.5 \times 1.5 \text{ mm}^2$. With the precise computer-controlled manipulator, all the irradiated regions can be measured at their corresponding XZ positions

along the same channeling orientation. Such an approach/procedure significantly reduces the measurement time, as compared to irradiating different samples and finding the channel direction on each of the samples. In the SiC irradiation study, both 2.0 MeV He⁺ and 3.5 MeV He⁺ were performed, as shown in **Fig. 9**. Comparing the spectra from both sets of the measurements, damage production under 900 keV Si⁺ irradiation is clearly observable on the Si sublattice from both measurement conditions with a similar trend of damage accumulation responses. While the 2.0 MeV He⁺ RBS measurements provide good depth resolution of the damage profile of the Si sublattice (channels from ~300 to ~580) due to the higher energy loss of 2.0 MeV He beam penetrating the sample (as comparing with 3.5 MeV He beam), no useful information can be extracted for the C sublattice (below ~ channel 300), as shown in **Fig. 9(a)**. On the other hand, the collision between a C atom and a 3.5 MeV He⁺ is non-Rutherford, the backscattering cross section is ~ 6 times higher than that for the 2.0 MeV He⁺ beam. This is clearly demonstrated by the sharp edges of both Si (channel 1030) and C (channel 460), as well as the enhanced backscattered yield from the C sublattice in **Fig. 9(b)** versus that in **Fig. 9(a)**.

Another example demonstrating the capabilities of this endstation in studying defect production and accumulation is shown in **Fig. 10**. RBS channeling spectra were measured from SrTiO₃ irradiated with 900 keV Au⁺ at fluences ranging from 1.3×10^{13} to 2.1×10^{14} cm⁻² at room temperature. Damage profile in dpa and Au distribution were determined by the SRIM full-cascade simulations (2008 version) [29] under the assumptions of a sample density of 5.118 g cm⁻³ and displacement energies of 45, 70, and 80 eV for O, Ti and Sr, respectively [32]. The irradiated doses at the damage peak range from 0.05 dpa to 0.8 dpa. Subsequent *in-situ* investigation of the damage was carried out to demonstrate the high efficiency of the irradiation and analysis capabilities. The RBS measurements were performed using a 3.5 MeV He⁺ beam along the <100>-axial direction with a Si detector located at a scattering angle of 155° relative to the incoming beam, as shown in **Fig. 10 (a)**. The beam energy of 3.5 MeV was chosen due to the lower electronic energy loss in the surface region (as compared with a 2.0 MeV He beam), and, therefore, to have a wide spread of the damage profile and reasonable separation of the damage yield between the Sr and Ti signals for dechanneling analysis. The RBS spectra were analyzed using an iterative procedure to

1 achieve the relative disorder level [32]. The damage accumulation curve determined from the Sr
2 sublattice as a function of irradiation dose at the damage peak is shown in **Fig. 10(b)**, where a sigmoidal
3 dependence is observed. The whole accumulation curve is based on ion irradiation and ion channeling
4 measurements from more than 10 irradiation fluences that can be accomplished within one day with a
5 good experimental plan.

6 The high-resolution goniometer allows fine angular scans and image scans, which are unique
7 capabilities of this endstation. A sample ($\text{CeO}_2/\text{SrTiO}_3$) with an epitaxial CeO_2 thin film grown on a
8 SrTiO_3 substrate was measured in the angular scan mode to study interface quality and possible lattice
9 mismatch at the interface. A 70 nA beam of 3.5 MeV He^+ ions was collimated through the double slits
10 (set at $1.5 \text{ mm} \times 1.5 \text{ mm}$) onto the sample. Backscattered He was detected by the Si surface barrier
11 detector located at a backscattering angle of 155° from the incoming beam. The channeling spectra taken
12 along the [100] direction of the $\text{CeO}_2/\text{SrTiO}_3$ sample are shown in **Fig. 11(a)**. From the RBS
13 measurements, the film thickness of 160 nm can be estimated by assuming a theoretical density of 7.65 g
14 cm^{-3} . The spectrum in the region from channel 1500 to 1650 is the yield from the Ce film, where both the
15 surface peak and increased disorder at the interface are clearly evident. Angular scans around the [100]
16 and [113] channeling directions were carried out, as shown in **Figs. 11(b) and (c)**. The angle between [100]
17 and [113] is calculated to be 25.24° . The rocking curves, shown in **Figs. 11(b) and (c)**, were obtained by
18 integrating the regions of interest (ROI) on the CeO_2 film (channel from 1510 to 1647) and the SrTiO_3
19 substrate (channel from 650 to 1447), respectively. The dashed-dot lines are marked at or near the
20 minimum yield to guide the eyes. Detailed analysis shows that the angle shift between the film and the
21 substrate along the [100] channel varies from 0.02° to 0.06° , the shifts along the [113] channeling
22 direction are around 0.05° , and the uncertainty in the angle scans is on the order of 0.05° . The results in
23 **Fig. 11** indicate that, within the measurement uncertainty, the interface of the $\text{CeO}_2/\text{SrTiO}_3$ sample is
24 mainly coherent.

25 An image scan technique has also been tested at this endstation. The results in **Fig. 12** are the
26 spectral maps acquired from a 2D scanning mode of RBS measurements. A single crystal Fe-15%Cr-

15%Ni sample with fcc structure was analyzed using 3.5 MeV He⁺. The scan range was $\pm 1.44^\circ$ with a scan step of 0.072° . Finer scan steps may be used at the cost of a longer measurement time and if the sample is insensitive to helium ion damage. After performing several image scans at different locations (involving more than 10000 rotations), the crystal can be re-aligned using the pre-determined values of the channel direction, demonstrating high reproducibility and reliability of this customized manipulator.

3.3 Endstation at beamline -15° (L3)

This L3 endstation is also a custom designed UHV endstation. It has been recently added to extend the high-temperature irradiation capability and is shown in **Fig. 13**. This high-temperature irradiation endstation consists of a target chamber (**Fig. 13(a)**) using the same design as the target chamber at beamline L5. A custom-built temperature-controlled manipulator (Thermionics, NW, WA, USA) has one rotatable axis with a polar rotation range of $\pm 180^\circ$ and no azimuthal and flip rotations (**Fig. 13(b)**). There are 3 translation axes of X, Y and Z with ranges of ± 12.7 , ± 12.7 , and ± 203.2 mm, respectively. Due to the relatively simplified design, filament radiant heating or liquid-nitrogen cooling is conducted more efficiently, and a wider temperature range, from -150 to 1200 °C can be reached. Besides the high-temperature and uniform irradiation capabilities, this endstation is also designed to perform *in-situ* ion beam analysis.

In order to achieve uniform irradiation over a relatively large area, a raster scanner is installed on the beamline for the purpose of providing uniform beam intensities and wide irradiation areas at the target position. Identical to that for beamline L5, the fixed scanning frequencies are 517 and 64 Hz for the horizontal and vertical direction, respectively, with a maximum scanning voltage of 2 kV. The sample holder (44.5 mm in diameter) is larger than the one in L5, but has a similar design. Such a configuration can produce relatively uniform beam flux at the target position over the area up to 17 mm long and 17 mm wide at elevated or cryogenic temperatures. The samples can be mechanically mounted on a circular molybdenum plate (25.4 mm in diameter and 0.5 mm in thickness) by molybdenum spring-loaded clips,

1 with a chromel–alumel thermocouple clamped to the sample surface to monitor sample temperature. For
2 room-temperature irradiations, larger irradiation areas are possible on a simplified sample holder as no
3 heating or cooling is required. The ion fluence at the L3 endstation was calibrated in a similar manner as
4 was done for the endstation L5 (Table 1). Uniform ion irradiations have been performed by rastering the
5 ion beam over a large irradiated area. Beam uniformity can be monitored by measuring and balancing the
6 current on the double slits (four electrically isolated plates). These slit sets, placed directly upstream from
7 the endstation, define the irradiation area. The double slit currents can be monitored from the accelerator
8 control panel allowing dynamic control of the beam steerers to minimize axial drift. For long irradiations,
9 the Faraday cups located on the beamline and below the sample holder (**Fig. 13(b)**) in the target chamber
10 can be inserted into the beam to measure the ion current. Uniformity of irradiations has been verified by
11 ion-induced luminescence from quartz targets and by large-area Au implantations in Si samples, where
12 implanted fluences and the uniformity of different irradiated regions from the same implantation are
13 confirmed by RBS and secondary ion mass spectroscopy measurements. The largest deviation is within
14 $\pm 10\%$ (Table 1).

15 The main purpose of this endstation is to study radiation damage mechanisms and their effects on
16 microstructural evolution and physical property changes in nuclear materials for advanced fission and
17 fusion reactor designs. Ion irradiation allows well-controlled and tunable conditions (as compared to
18 neutron irradiation), thereby providing a broad range of damage production and ionization rates to study
19 separate and combined effects of cascade damage and ionization, and create uniform or non-uniform
20 damaged regions and local stress fields. Ion irradiation has, therefore, been utilized as a useful tool for
21 separate and integrated irradiation studies [27]. At this endstation, an irradiation study can be performed
22 over a wide range of ion energies, masses, fluences, dose rates, temperatures, all of which can provide
23 data on the kinetics of radiation effects. In several ongoing research projects, ion irradiation is used to
24 study the separate effects of cascade damage from fission products (irradiation dose, stopping powers,
25 ratio of nuclear to electronic energy deposition, dose rate, temperature, etc), and combined effects on
26 microstructure evolution to high doses (up to 100 dpa) over a wide temperature range [26-28,33]. The

irradiation studies of nanocrystalline ZrO_2 under high-dose Au irradiation shown in **Fig. 5** were performed at this endstation. From systematic ion irradiation studies, it is possible to build physics-based constitutive models to provide insights on defect kinetics and, ultimately, enable the prediction of fuel behavior at the lower dose rates of fission reactor environments [34]. As mentioned earlier, ion irradiations can be carried out in this endstation from cryogenic temperature -150 to 1200 °C to study and evaluate the impact of both thermal and irradiation effects. Photos of the high-temperature stage configuration are shown in **Figs. 13(c) and (d)**. The design of the manipulator allows for efficient cooling of samples from room temperature down to -150 °C in 50 min. Samples mounted on the high temperature stage can be heated radiantly from the rear of the platen using a tungsten filament (for resistive heating). Sample temperatures can be rapidly raised from room temperature to 1200 ± 2 °C controlled by an automated program. The heating rate can be set to a few tens of °C per min up to 230 °C per min. It is worth noting that accurate measurement of sample temperature depends on careful mounting of the thermocouple, caution should be taken into the sample holder design. Depending on the target temperature ranges, the sample temperature can be monitored during the irradiation using two thermocouples (one determines the temperature on the backside of the holder and the other clamps to the sample surface) and one additional infrared pyrometer.

Besides ion irradiation, the L3 endstation is equipped with IBA capabilities. Time-of-flight elastic recoil detection analysis (TOF ERDA), complementary to RBS and conventional ERDA demonstrated in Section 3.1, is a unique ion beam analysis technique available at this endstation. For MeV helium ions commonly used in RBS, the primary ions backscattered from the heaviest elements have the highest energy, and the ratio of the cross sections can be approximated by $d\sigma_{\text{light}} / d\sigma_{\text{heavy}} \approx (Z_{\text{light}} / Z_{\text{heavy}})^2$ for $M_1 \ll M_2$. While RBS is sensitive to heavy elements in a light target and conventional ERDA is normally limited to detect very light elements (H and He), as illustrated in the examples shown in **Figs. 5, 6 and 8-11**, TOF ERDA detects both light and heavy elements in a target, with superior mass resolution for light isotopes [15-18, 35-41]. In an ERDA configuration with a detector in a forward scattering geometry, one

1 can detect forward-scattered recoiling atoms from a single collision. With heavy ions ($M_1 \gg M_2$), the cross
2 section for recoils is approximately proportional to Z_2/M_2 and, therefore, the recoil cross section does not
3 decrease dramatically for light elements. It is possible for TOF ERDA to separate isotopes of hydrogen,
4 helium, lithium, boron, carbon and oxygen.

5 Several TOF ERDA tests have been carried out at the L3 endstation, as shown in **Fig. 14**. The
6 time-of-flight and Si energy detector (TOF-E) telescope consists of two carbon-foil time detectors [18,42]
7 separated by a flight length ($L=647.5$ mm) followed by a silicon energy detector placed 80 mm after the
8 second time detector. The carbon foils are 5 and 10 $\mu\text{g cm}^{-2}$ thick (2.5×10^{17} and 5×10^{17} atoms cm^{-2} ,
9 respectively) for the first and second time detectors, respectively. The energy detector, placed after the
10 second time detector, is a silicon charged particle detector with a 50 mm^2 diameter active detection area.
11 A collimator is placed immediately in front of the detector. The recoiling target atoms were detected at an
12 angle $\phi = 30^\circ$ relative to the incoming beam direction. The incident angle θ_1 of primary ions and exit
13 angle θ_2 of recoils were both at 75° to the sample surface normal. The incident ions were 20 MeV $^{197}\text{Au}^{7+}$
14 ions. The output signal from the first time detector discriminator was delayed and fed together with the
15 signal from the second time detector to a time-to-amplitude converter in an inverse start-stop mode in
16 order to minimize dead time. A conventional preamplifier-main-amplifier spectroscopy chain was used
17 for signals from the energy detector. The coincident TOF and energy signals from each recoil were stored
18 in list mode by the FAST ComTec MPANT software [43]. Two-dimensional energy-time histograms of
19 MgO and SiC samples are shown in **Fig. 15**. In the MgO sample, both H and C contaminations are clearly
20 detected. In the SiC sample, beside H and O signals from surface contamination, projectile Au^{7+} ions are
21 also detected, as shown in **Fig. 14(b)**.

22 Ion beam induced luminescence (IBIL) and cathodoluminescence (CL) are two newly added
23 capabilities at the multipurpose endstation (L3) to provide insights to radiation damage mechanisms.
24 These techniques can be utilized to acquire information on intrinsic defects, and to investigate the
25 formation of color centers, the kinetic evolution with increase of irradiation fluence, and microscopic

processes resulting from damage accumulation. Since visible light results from the outer shell transitions, it provides information on the nature of chemical bonds in materials, as well as the local chemical environment of compounds, trace substitutes, and microstructures of the network. Both IBIL and CL techniques can excite deeper-level electrons due to large energy transfer to target electrons, is therefore considered to be a monitoring tool for material modification during irradiation. These two luminescence techniques are tested and validated using known scintillation materials at room temperature, including model oxides of α -SiO₂, quartz, SrTiO₃, and MgO, and other complex pyrochlores and silicates.

A schematic diagram of the *in-situ* luminescence experimental setup is shown in **Fig. 15**. The light emitted from the samples was transmitted through a silica window port placed at 150° with respect to the ion beam direction, and collected using a 25-mm-diameter, 4-cm-focal-length silica lens into a silica optical fiber of 1 mm diameter located outside of the vacuum chamber. The light was guided to an imaging spectrometer, consisting of a monochromator (Acton Research Corp. SpectraPro-2556, 600 lines/mm, 500-nm blazed grating) and a liquid nitrogen-cooled back illuminated UV enhanced CCD camera multichannel array detector (Princeton Instruments, Spec-10: 2KBUV). The evolution of the IBIL spectrum was monitored in spectroscopic mode with Full Vertical Binning (FVB) pixels addition for wavelengths from 250 to 1000 nm. The spectral resolution is better than 0.05 nm. During test runs, the low-current ion beam was used to avoid overheating and electric arcs on the samples.

To demonstrate the newly operational luminescence capability, a comparative study of the ionoluminescence is carried out on crystalline quartz and amorphous silica using 3.5 MeV He and 8 MeV O ions. The irradiation conditions are summarized in **Table 2**. It has been reported [44-46] that each swift ion impact generates a nanometric amorphous track when electronic stopping power is above the threshold value of ~2 keV/nm [47]. Moreover, the electronic damage is cumulative, amorphization can occur through overlapping ion tracks even below the threshold [48]. Some representative IBIL emission spectra are shown in **Fig. 16**. Helium ions were selected due to their low stopping powers, and thus few vacancies were produced relative to those by oxygen ions peaked at the end of ion range (see **Table 2**). In other words, He beam may be considered as a non-damage ion probe. The spectra are deconvoluted into

four different emission bands. All of these bands have been previously reported in the literature and ascribed to substitutional impurities of Fe^{3+} (band at 1.65 eV) [49], non-bridging oxygen hole color centers (red band at 1.9 eV) [50], radiative recombination of the self-trapped exciton with an E' center (band at 2.26 eV) [49] and the blue band associated with the radiative emission from self-trapped excitons (STE, band at 2.7 eV) [50]. The agreement of our results in **Fig. 16** with the literature results validates our luminescence setup.

The two broad emission bands lying at 1.9 eV (652 nm) and 2.7 eV (459 nm) are referred here as red (1.9 eV) and blue band (2.7 eV), respectively. In the case of silica, the blue band is clearly dominant over the red one under both He and O irradiation. For quartz, while the red band dominates under He irradiation, the blue one becomes more intense under O irradiation. It should be noted that, in spite of the apparent difference for the two materials at the initial damage stage (**Fig. 16** (a) and (b)), they become quite similar at the final damage stage after O irradiation (**Fig. 16** (c) and (d)). Moreover, with continuing irradiation, decrease in luminescence yields of both bands is observed with the accumulation of ion damage in silica, but the blue band is clearly predominant over the red one with a constant ratio of ~ 5 . For quartz, the red emission is initially dominant but gradually taken over by blue emission with further damage accumulation. The blue emission becomes overwhelmingly dominant in the highly damaged quartz. After a high enough irradiation fluence, both materials reach a similar damaged phase. All those observations are consistent with the assignment of the red band to color (NBOHC) centers generated during the recombination of STE [50], whereas the blue band is associated with the radiative emission, exciton self-trapping and recombination, occurring preferentially at heavily strained Si-O bonds, such as those present in silica or in a damaged quartz crystal [51]. In silica, the strong reduction of the yield for the two emissions after the ion beam damage could be attributed to the compaction of the Si-O-Si network [52]. For quartz, the irradiation response indicates a competition between recombination at NBOHC centers and STE sites. In summary, while the experiments demonstrate the luminescence capability, the results have demonstrated the analytical power to provide insights on the evolution of the

1 recombination sites (strained bonds) with irradiation fluence, as well as on the processes of structural
2 damage during ion irradiation.

3 Accurate information on energy loss when energetic ions go through materials is essential for
4 fundamental research, electronic and optical device fabrication, and prediction of the performance of
5 materials subject to high irradiation environments. Despite intense research on energy loss of charged
6 particles in matter for over almost a century, the electronic stopping power is not adequately described,
7 particularly in the case of heavy ions in compound targets [53-55]. Only a few ion-target combinations
8 have been studied in sufficient detail to allow interpolation and extrapolation on purely experimental
9 grounds. At the same time, the range of validity of existing scaling relations is largely unknown, and a
10 comprehensive theory allowing predictions is not available. To address this long-term challenge, one task
11 at this endstation is to determine electronic energy loss in matter. By utilizing the TOF-E telescope with
12 unique data analysis approaches, energy loss in self-supporting foils can be determined [53,56]. As shown
13 in **Fig. 17**, the material of interest in a thin foil form can be reproducibly moved into and out of the ion
14 path between the TOF spectrometer and the Si detector. A monoenergetic beam (ions of interests) from
15 the accelerator is forward scattered into the TOF-Si detector telescope by a scatter target to produce
16 particles with continuous energies. The TOF and energy signals of the ions are recorded in a coincident
17 mode with and without the alloy foil (or any self-supporting foils), from which the electronic energy loss
18 in the foil will be determined with high accuracy [53,56]. This approach presents a significant advantage
19 where the stopping powers can be measured over a wide energy region in a similar time frame as the
20 conventional approaches for one single data point. This method has been applied to determine the
21 electronic stopping power for very heavy ions (e.g. I and Au) in materials containing light elements (e.g.
22 SiC and SiO₂). We have found that the SRIM2012 code [29] significantly overestimates the electronic
23 stopping power of very heavy ions (e.g. Pt and Au) in light targets (e.g. SiC, GaN, and SiO₂) [53, 55].

24 **4. Conclusion**

1 The UT-ORNL Ion Beam Materials Laboratory has been developed on the campus of the
2 University of Tennessee. This facility is well suited for ion beam analysis, ion beam modification of
3 materials, and research on ion-solid interaction and irradiation effects in materials. Most of the routine
4 and some unique irradiation and ion beam based analysis capabilities are developed and tested. Since it
5 began operation in the fall of 2012, irradiations and measurements are conducted to meet increasing
6 demands from universities, national laboratories, and industry.

7 **Acknowledgements**

8 The UT-ORNL Ion Beam Materials Laboratory (IBML) was established with support from the
9 UT-ORNL Governor's Chair program. The development of the high temperature endstation (L3) was
10 supported by an infrastructure grant from the U.S. Department of Energy (DOE), Nuclear Energy
11 University Programs. Y. Zhang is grateful to the Presidential Early Career Award for Scientists and
12 Engineers (PECASE) support through DOE Office of Basic Energy Sciences (BES), Materials Sciences
13 & Engineering Division (MSED). Y. Zhang, C. L. Fontana and W. J. Weber were partially supported by
14 DOE, BES, MSED in the development and demonstration of unique capabilities for damage accumulation
15 studies. K. Jin and Y. Zhang acknowledge the support on energy loss determination from Department of
16 Defense (DOD) Defense Threat Reduction Agency (DTRA), Research and Development (R&D)
17 Enterprise, Basic and Applied Sciences Directorate. C. L. Fontana was partially supported by ORNL
18 LDRD for the TOF ERDA development. Y. Zhang is also grateful to the assistance and support from
19 Gregory A. Norton, Jeremy Kasmarek, and Dan Bernhardt at National Electrostatics Corp.

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Table 1 Calibration of ion fluence at endstations L3 and L5.

Endstation	Irradiation conditions	Theoretical ($\times 10^{15} \text{ cm}^{-2}$)	Measured ($\times 10^{15} \text{ cm}^{-2}$)	Error (%)
L5	1 MeV Au in Si	5.0	5.5	+10
	1 MeV Au in SiO ₂	5.0	5.2	+4
	3 MeV Au in SiO ₂	5.0	4.7	-6
L3	2.25 MeV Au in Si	6.1	6.3	+3
	5 MeV Au in Si	5.3	5.5	+4
	7 MeV Au in Si	6.1	6.4	+5
	10 MeV Au in Si	5.3	4.8	-9

Table 2. Irradiation parameters for the ions and materials used in this work, calculated using the SRIM code (2008 version) [29], where S_e is the electronic stopping power at the surface, S_n is the maximum value of the nuclear stopping power and R_p is the projected ion range. The average dose in dpa (displacements per atom) was estimated under full-cascade simulations for the ion range avoiding the damage peak region (90% R_p) for the specific irradiation fluences used.

Silica					
Ion	Energy (MeV)	S_e (keV/nm)	S_n (keV/nm)	R_p (μm)	<dpa>
He	3.5	0.19	0.002	14.0	1×10^{-3}
O	8	1.78	0.04	6.2	5.4×10^{-2}
Quartz					
Ion	Energy (MeV)	S_e (keV/nm)	S_n (keV/nm)	R_p (μm)	<dpa>
He	3.5	0.23	0.003	12.0	5.7×10^{-4}
O	8	2.14	0.05	5.1	5.3×10^{-2}

Figure Captions

- Fig. 1.** Schematic layout of the UT-ORNL IBML, including a separated control room, the two ion sources, a 3 MV tandem accelerator, beamlines and end-stations.
- Fig. 2.** A photograph of the three high-energy research beamlines with L3 on the left, L5 in the middle, and L6 on the right.
- Fig. 3.** A photograph of the inside of the L6 endstation showing the internal configuration.
- Fig 4.** Schematic drawing of the possible RBS and NRBS scattering geometries for the movable (left) and fixed (right) Si detectors located in L6 endstation, respectively. The thick straight arrow denotes the incoming beam, and the thin arrow is the backscattered beam, where α is the incident angle, β the exit angle, and θ the scattering angle.
- Fig. 5.** RBS spectra of ZrO_2/Si samples irradiated with 12 MeV Au^{4+} ions to different fluences up to $2 \times 10^{16} \text{ cm}^{-2}$. The RBS measurements were conducted using 2.0 MeV He^+ beam. For clarity purpose, every fifth data point is plotted.
- Fig. 6.** Conventional ERDA measurement of a 3C SiC sample implanted with 65 keV He^+ ions to $1 \times 10^{16} \text{ cm}^{-2}$ at 7° off the surface normal: (a) schematic drawing of the experimental configuration, (b) the ERDA spectrum of the sample where surface H is clearly detected, and (c) He depth profile in the sample determined from the ERDA measurement (triangles) with a smooth line fit to guide the eye. The energy calibration is $E \text{ (keV)} = 3.1178 \text{ (keV/ch)} \times \text{Channel} + 85.51 \text{ (keV)}$. Inset is the SRIM2012 prediction (solid line) of the He profile.
- Fig. 7.** Schematic diagram of the four-channel CMDC. A logical buffer (SN74HCT125) is used to decouple and protect the circuit, and two dual 16-bit counters (SN74LV8154) are then used to count the pulses. The results are sent to an Arduino Micro in a parallel 8-bit bus.
- Fig. 8.** RBS spectrum of Si implanted using 2.25 MeV Au^{2+} ions to an intended fluence of $6.1 \times 10^{15} \text{ cm}^{-2}$. The diamonds are the data from the RBS measurement using 2.0 MeV He^+ beam, and the dash line is fit using SIMNRA based on a fluence of $6.3 \times 10^{15} \text{ Au cm}^{-2}$.
- Fig. 9.** (a) Rutherford backscattering employs 2.0 MeV He^+ along the $\langle 111 \rangle$ channeling direction of SiC. The corresponding dose at the damage peak ($\sim$ channel 460) is noted on the plot. Both random and virgin spectra are included for comparison. (b) 3.5 MeV He^+ Non-Rutherford backscattering spectrum, the enhanced carbon yield is due to the enhanced scattering cross section. The inset shows the eleven spots with size of $3 \times 3 \text{ mm}^2$ irradiated on the same sample with a 0.5 mm gap between the spots. For clarity, every ninth or sixteenth data point is plotted in (a) or (b), respectively.
- Fig. 10.** (a) RBS spectra of SrTiO_3 irradiated with 900 keV Au^+ to different ion fluences. Both Random and virgin spectra are included as references. The corresponding dose at the damage peak is marked on the plot. For clarity, every eighth data point is plotted. (b) Relative disorder as a function of local dose at the damage peak in SrTiO_3 , the disorder level is determined using the signals from the Sr sublattice.
- Fig. 11.** (a) Random and channeling RBS spectra along the $[100]$ direction of a CeO_2 film epitaxially grown on a SrTiO_3 substrate. For clarity, every ninth data point is plotted. Angular scans of the

sample with ROI from the film (channels from 1510 to 1647) and substrate (channels from 650 to 1447) around the [100] direction (b) and [113] direction (c), respectively.

Fig. 12. RBS image scans of Fe-15%Cr-15%Ni (100) single crystal using 3.5 MeV He⁺ beam; (a) 3D view and (b) 2D view. The scan range is $\pm 1.44^\circ$ and scanning step is 0.072° . High quality of the crystal is evident.

Fig. 13. L3 endstation: (a) photo of the target chamber, (b) inside of the chamber, (c) heated sample holder at 827 °C with a large one-side polished Cu sample loaded for temperature calibration (two thermocouples are mounted), and (d) heated sample holder at 700 °C with a few 3C SiC samples loaded.

Fig. 14. TOF ERDA energy-time histograms for (a) MgO and (b) SiC. The projectile beam is 20 MeV Au⁷⁺ ions.

Fig. 15. Schematic experimental setup for online IBIL measurements.

Fig. 16. IBIL spectra and the corresponding component bands from silica (a, c) and quartz (b, d) under 3.5 MeV He and 8 MeV O ions. The dash curves represent the emission bands used to deconvolute the spectra. The intensities are normalized based on ion beam flux and energy.

Fig. 17. Single ion technique utilizing a TOF-Si detector telescope for electronic energy loss measurement. Data analysis procedure is described.

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