Post-Irradiation Examination of Radioactive Materials at CANS

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Abstract

This paper is to present the experimental techniques available for post-irradiation examination at the Centre for Advanced Nuclear Systems (CANS). The McMaster CANS facility includes five hot cells with a variety of scientific equipment, including a computerized numerical control (CNC) mill, an MTS servo hydraulic testing unit, an opto-digital microscope, and Struers machines (Secotom-10, CitoPress-1, TegraPol-11) for cutting, mounting and polishing materialographic samples of radioactive materials for microscopic examination. A dual-beam microscope will provide additional laboratory support.

1. Introduction

The McMaster CANS facility [1], a hot cell laboratory for radioactive materials characterization research at the atomistic level, is scheduled to fully commission in 2017. The aim of the centre is to help scientists research the behaviors exhibited by various materials when they are exposed to radiation and extremely high temperatures over long periods of time. This will improve the maintenance, safety, and continued operation of nuclear power plants.

The facility includes five hot cells: receiving, CNC machining, sample preparation, mechanical testing, and optical testing. Each cell consists of a thick lead-infused shielded glass window, an in-cell crane for lifting equipment, and a pair of remote manipulators for handling material. The materials received can be processed through a sophisticated assembly line that logs samples, machines coupons for metallurgical testing with MTS equipment, polishes and etches, and views the microstructure evolution on each machining step with an Olympus DSX 500 opto-digital microscope. The polished specimens are then transferred to a FEI Versa 3D dual beam system for characterization and sample preparation.

The FEI Versa dual-beam system combines a Ga^+ focused ion beam (FIB) column and a field-emission scanning electron microscope (FE-SEM) column in one tool. It serves three basic functions: (1) imaging by e-beam / ion-beam. The SEM can reach a resolution of 1-2nm, while the FIB has a resolution of about 7-8nm; (2) at site-specific ion etching/polishing and cross sectioning; (3) at site-specific metal deposition. Furthermore, the SEM with energy and wavelength dispersive x-ray spectroscopy (EDS, WDS) and electro backscatter diffraction (EBSD) is used for topographic features, elemental analysis, grain microstructures and their crystallographic orientations. The first research conducted at CANS was the preparation of radioactive transmission electron microscope (TEM) samples using the FIB. The details of sample preparation, characterization and preparation-damage will be discussed in this paper.

2. Sample Preparation in Hot Cells

CANS provides a computerized numerical control (CNC) mill, Struers machines, an MTS servo hydraulic testing unit, and an opto-digital microscope. The Struers machines include the Secotom-10, CitoPress-1 and TegraPol-11 for precision cutting, mounting/pressing, and grinding/polishing, respectively. These machines are adapted to allow remote operation and remote handling of specimens using hot cell manipulators, as shown in Fig. 1a, for materialographic sample preparation of radioactive materials. For example, zirconium alloy tubes used to contain fuel bundles in CANDU nuclear reactors are cut into 25mm x 6mm x 4mm samples. The Secotom-10 then performs precise and deformation-free cutting of these larger samples into 6mm x 4mm x 2.5mm specimens. The specimens are then placed in a hot mounting press, and the conductive PolyFast mounting resin is added. A temperature of around 180°C and a force of about 250 bar is applied during the embedding of the specimen. A TegraPol-11 is used for the grinding/polishing process. The basic process is material removal using abrasive particles in successively finer steps to remove cutting damages from the surface until the required result is reached. The specific requirement of the prepared surface is determined by the particular type of analysis or examination, the preparation can be stopped when the surface is acceptable for a specific examination. Fig. 1b shows a zirconium specimen after mechanical polishing.

Fig. 1a: Hot cell facilities at CANS

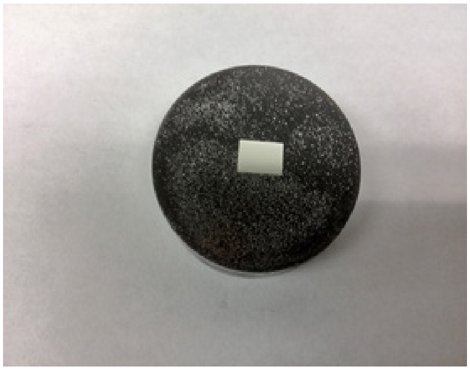

Fig. 1b: Zirconium specimen after mechanical polishing

The polished sample's microstructure will be checked under the Olympus DSX 500 opto-digital microscope in the hot cells and transferred to the FEI Versa dual-beam system.

3. Scanning Electron Microscope and X-ray Microanalysis

The energy dispersive x-ray spectroscopy (EDS) and wavelength dispersive X-ray spectrometers (WDS) are additional features of the FEI dual beam system that allows in-situ chemical analysis on the surface or at the cross-section of a sample. The analysis of the characteristic x radiation can yield both qualitative identification and quantitative compositional information from regions of a specimen as small as a micrometer in diameter. Another important feature is the capability of obtaining compositional mapping with characteristic x-rays. The x-ray images show the elemental distribution in the area of interest.

4. TEM sample Preparation Using Focused Ion Beam

The advantages of using FIB for radioactive sample preparation are as follows: (1) reduced personal radiation exposure and (2) the ability to prepare site-specific lift-outs. The electron beam is used to examine the sample surface and to select a specific region, i.e. a clean region with few cracks and scratches due to sample handling and sample preparation. Prior to ion milling, a platinum protection layer is deposited over the area of interest to protect the specimen surface from ion beam damage. Typically, a 30 keV Ga+ ion beam is used to sputter materials in such a way as to leave a thin wedge of material at the desired sample site, which must then be detached from the substrate and transferred using a micromanipulator arm to a specialized TEM grid for further thinning. The final thickness is typically less than 100 nm for high resolution TEM imaging. It should be noted that some degree of sample surface damage due the irradiation with Ga+ is inevitable [2]. The thickness of the damage layer formed on the FIB-prepared sample surface is roughly proportional to the primary energy of Ga+. Thus, the effect of radiation damage can be minimized at an accelerating voltage as low as 1-2kV. Fig. 2 shows the SEM images of Zirconium lamella prepared by FIB. It is interesting to observe the formation of Zirconium hydride [3]. The generation of local stress and introduction of hydrogen are the basic requirements for Zirconium hydrogenation. The hydrogen is generated from electron/ion beam stimulated decomposition of an organometallic precursor during the deposition of Pt. A method called gradient ion milling can be used to reduce the local stress, and reduce Zirconium hydrogenation. Care must be taken to consider the presence of such features and artifacts in order to avoid misinterpretation of the results.

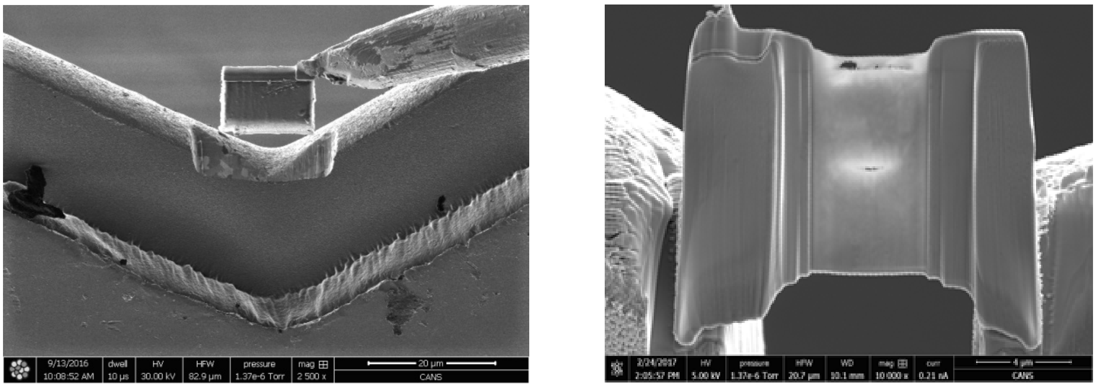

Fig. 2:   Preparation of Zirconium TEM sample by FIB

5. EBSD Crystallographic Analysis

The dual-beam system is also equipped with an electron backscatter diffraction detector (EBSD) that can be used to obtain crystallographic information about a sample by recording the diffraction patterns formed by electrons when they tunnel through a sample at glancing angles. EBSD is a powerful technique to characterize irradiated nuclear fuels to detect submicron-level damage. For EBSD, sample preparation is the key to fast and reliable analyses. Past experience has shown that diamond-polished samples are typically too rough to generate or reliably index Kikutchi patterns. Limited success has been reported for Zirconium samples carefully polished with colloidal silica. Conventionally, the best results are derived from Zirconium EBSD samples that have been electro-polished in an electrolyte consisting of 15-20% perchloric acid in methanol. However, this electro-polishing process should be done in a specially designed perchloric acid fume hood. Also, this technique requires close-in and hands-on manipulation of the samples for extended periods of time. This is not feasible with radioactive materials. Recently, FIB polishing has been developed to achieve a high quality surface for EBSD measurement. In CANS, we use the lift-out technique with a micromanipulator inside the FIB/SEM chamber for the preparation of EBSD samples. The procedure is similar to the preparation of TEM samples. The specimen from the substrate is removed and mounted to a Cu grid. The FIB polishing starts using 30kV Ga+ with 2nA current for coarse polishing, then 30pA current for fine polishing, and ends up at voltahe 5 kV or 2 kV with a low beam current ~16pA for final polishing to minimize the ion-induced damage. EBSD orientation mapping is then conducted with an EDAX Technology EBSD system on FEI dual beam system. Fig. 3 shows the EBSD sample prepared by FIB and its electron backscatter diffraction pattern.

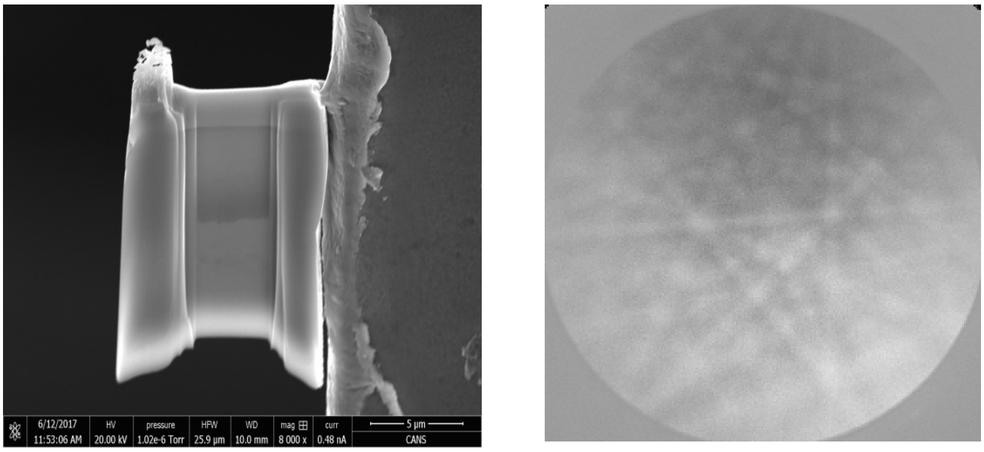

Fig. 3 (a) Zirconium EBSD sample prepared by FIB; (b) electron backscatter diffraction pattern

6. Other Subdivisions of CANS

CANS includes a nuclear thermal-hydraulic testing facility; a laboratory for studying material defects using positron annihilation spectroscopy. As well as the Nuclear Materials Characterization Facility, featuring a Three Dimensional Atom Probe located in the Brockhouse Institute for Materials Research.

7. Summary

This paper presents an overview of the post-irradiation examination services available at the Centre for Advanced Nuclear Systems (CANS), McMaster University. Easily shielded specimens from highly radioactive samples can be prepared inside the hot cell facility and subsequently be transferred to a FIB/SEM dual beam instrument for the microstructural characterization and the preparation of TEM samples. Careful attention to the details of the FIB processes can minimize artifacts such as re-deposition of sputtered material, ion implantation and microstructural modification.

Acknowledgment

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References

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