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The design, setup and operational testing of the irradiation and corrosion experiment (ICE)

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Abstract

Developing and qualifying nuclear materials have traditionally been a very costly enterprise that often takes a decade or longer. The bottleneck has been the need to test materials under extreme conditions, where long irradiations times have been required. A irradiation and corrosion experiment (ICE) facility is introduced which uses low energy proton irradiation and enables testing of materials under a wide range of different conditions and environments with the benefit of relatively quick tests, low sample activation, efficiency and low cost, and the ability to obtain a wide range of data from one sample. These data can be compared to data gained from neutron irradiation experiments. Supplement to extreme conditions in nuclear reactors, this dedicated experiment will allow fast materials testing and basic studies in order to get a quicker and better understanding of the materials behavior in different environments under irradiation. We present the basic experimental design, experimental testing, and first operational experience.

1. Introduction

Developing, testing and qualifying nuclear materials have traditionally been a very costly enterprise that often takes a decade or longer. The bottleneck has been the need to test materials under extreme conditions, including irradiation and corrosion at high temperatures. Irradiation in test reactors can only accumulate a few to 30 dpa (displacement per atom) in a year [1], while advanced nuclear energy systems would benefit from the development of materials which withstand greater than a total dose of 20–30 dpa/year [2], and years of services in harsh environments (high temperature and corrosion). Integral-effect tests with irradiation and corrosion can only be conducted within large-scale national programs and in a limited number of test reactors around the world. As such there is a significant barrier to incorporate advances in modern materials into nuclear energy systems. The experiment introduced here allows performing fast, easy and relatively inexpensive material tests in a low energy proton and corrosion environment to evaluate the materials behavior. In the past it has been shown [3] that low energy proton irradiation damage in materials can be used to obtain comparable data to that obtained from neutron irradiation damage reactor experiments.

2. The irradiation and corrosion experimental (ICE) setup, pre experiment testing and calculations

The experimental configuration is presented in Fig. 1. The sample itself is the beam window that separates the corrosion medium from the vacuum (beam facing side). Therefore the backside of the sample is in contact with the corrosive medium while under irradiation. In order to be able to penetrate through the sample entirely with a low energy (4–6 MeV) proton beam, the sample HT-9 stainless steel has to be thinned down to 40–50 μm. The nominal composition of this material is 11.95% Cr, 1% Mo, 0.6% Mn, 0.57% Ni, 0.5% W, 0.4% Si, 0.33% V, and 0.2% C by weight with the balance in Fe. The sample

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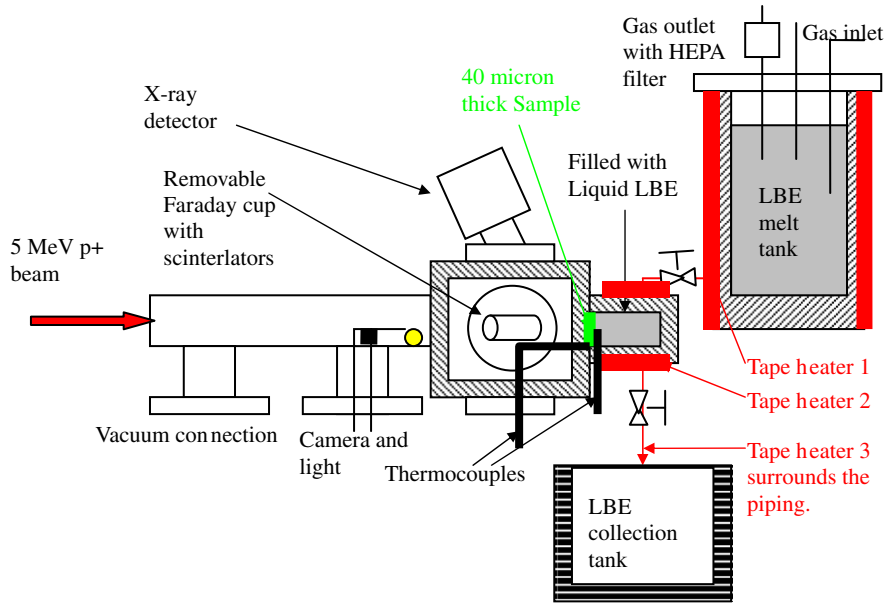

Fig. 1. Schematic illustration of the experimental setup on the platform of the experimental stand.

thinning was done with a large-scale TEM milling device (made at LANL). In this device, the sample rotates while a grinding wheel rotates perpendicular to the sample. This forms a concave sample (Fig. 2(a)). The grinding wheel diameter is ~200 mm. The concave shaped sample allows analysis after irradiation to different doses on the material over the radius of the sample back side (see Fig. 3). The calculations for a 5 MeV, 0.5 μA proton beam with a beam spot size of 8 × 8 mm using SRIM Monte Carlo simulation [4] show an accumulation of 0.1–1.4 dpa/100 h dose depending on location on the sample (see Fig. 3). An X-ray detector was used to quantify the total amount of protons hitting the sample by measuring the proton induced X-rays emission from major elements in the sample (such as Fe). The energy of the protons after penetrating through the 40–50 μm thick center of the sample is between 2 and 2.5 MeV. The beam stops rather quickly in the liquid lead bismuth eutectic (LBE) behind the sample. The total beam power deposited into the sample and liquid LBE is estimated to be 4.6 W/cm^2. The corrosion chamber was filled with LBE. The temperature of the medium plus the energy deposited in the sample by the beam determines the temperature of the sample during irradiation. In general, the temperature of our setup is limited mainly by sample itself and vacuum gasket material inside the chamber. K-type thermocouples close to the irradiation area on the front side of the sample and on the back side of the sample in the LBE were used to monitor the temperature (see Fig. 1).

Pre-test calculations (MCNPX and database-spread sheet calculations [5,6]) for the full available proton beam energy (~6 MeV) and an virtual beam current (~1 mA) show that the proton beam activated sample activity could be as high as ~14 mCi after 200 h of irradiation [7]. The main radioactive isotopes formed in steels are Co-56 and Co-57.

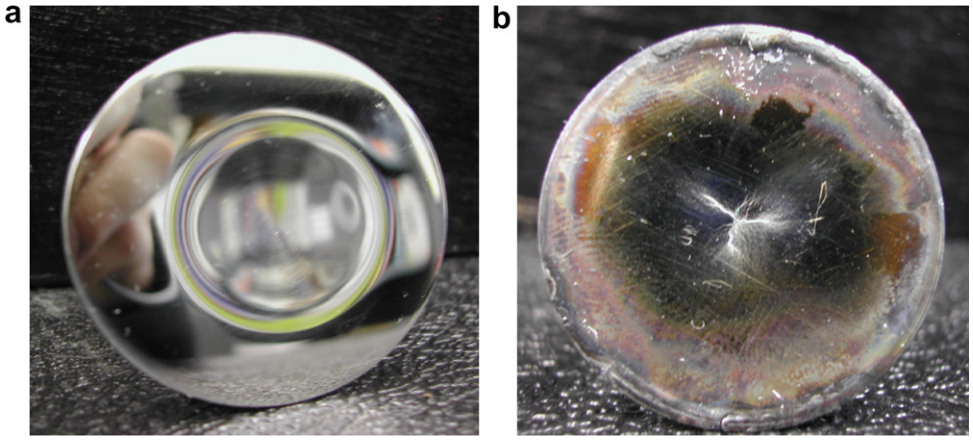

Fig. 2. The sample before the welding test (a) and after the welding test (b).

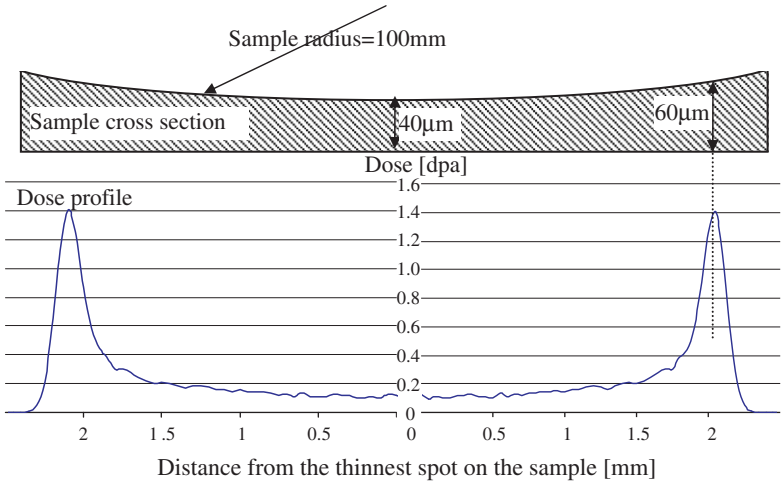

Fig. 3. The calculated dose the sample sample receives on its back side. The calculation parameters are: radius 100 mm, 0.5 μA beam current, 5 MeV beam power and irradiated area 8 mm × 8mm. No Gaussian beam shape was used.

The pressure leak test and beam line vacuum tests were performed prior to the experiment to ensure safe operation of the device during irradiation.

An initial weld test on the sample flange showed that the thermal gradient throughout the sample during welding ruptured the sample. An initial weld test on the sample flange showed that the thermal gradient throughout the sample during welding ruptured the sample (Fig. 2(b)). Before putting into the accelerator beam line, a liquid LBE experiment was conducted at 350 °C for 200 h. This experiment ensured the proper function of the valves, flanges, LBE container, fittings and the sample before the irradiation. Next, a pressure leak test of up to 65 psi was conducted at elevated temperature with compressed air (~650 °C) and LBE (~350 °C), respectively. The results showed sufficient sealing and no indication of the sample deformation. No pressure loss or LBE leaks were observed using the knife-edge seal. Upon connected in the beamline, the concave shape and thickness of the sample were confirmed using variable energy focused proton beam (~1 mm) and a carefully-designed Faraday cup located just behind the sample. Both calculations and measurements proved that a proton beam energy of 3.55 MeV was needed to penetrate through the center of the sample. By scanning the proton beam across the sample and increasing the energy the variation in thickness of the sample could also be detected. The dose vs. radius (thickness) calculated from the beam power penetration through the sample by using SRIM 2003 Monte Carlo simulation is shown in Fig. 3.

3. Conducting the irradiation and corrosion experiment

Fig. 4 shows radiation protection shielding and surrounding area of our experiment setup. The ICE was conducted in April 2007. The ICE setup was mounted on the L-30 beam line of IBML's 3 MV Pelletron Tandem Ion Accelerator. The experiment was conducted for 80 h continuously without any accidents or unexpected failures. The tape heaters were able to heat up the system to 350 °C for the entire duration. It was found that the beam increased the LBE temperature by an additional 50–60 °C. The oxygen level in the LBE was constant at its saturation point (~10^{-4} wt% O) at 350 °C (melt tank temperature). And the corrosion chamber was re-filled with fresh LBE every 5–10 h. During the experiment it was found that

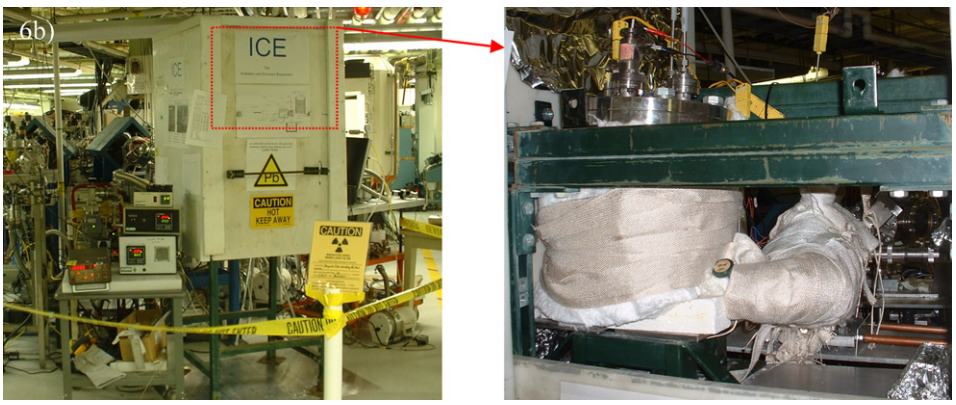

Fig. 4. An ICE setup in the beamline with the external shielding, control and data acquisition instruments.

Table 1
Summary of the limits of the ICE facility

ICE operation limits	Dose limit	Temperature limit	Medium limit	Beam power limit	Beam current limit
	Only the beam time limits the maximum dose	500 °C	The current design allows LBE, PB, Bi	6 MeV protons	$<1\ \mu A/cm^2$

the radiation level on the outside of the shielding was 36 mR/h. The sample activity right after the experiment was 200 000 dpm. Five months after conducting the experiment, the activity was found to be ~50 000 dpm. This confirms the calculations where Co^{56} was found to be the main isotope produced (half life = 77 days) during the experiment.

A summary of the operation limits of the ICE facility are presented in Table 1.

4. Outlook and conclusions

- Initial testing has shown that an irradiation and corrosion experiment (ICE) can be conducted at the Ion Beam Materials Laboratory (IBML) at LANL and hence, a dedicated beamline will be established.
- Testing shows that welding the specimen on the flanges creates stresses which fracture the sample but a conventional knife-edge seal sufficiently seals the chamber. The pressure tests conducted at elevated temperatures on the samples showed that a 40 μm thinned sample can withstand the pressure of 65 psi and therefore also the pressure caused by the liquid metal.
- The first ICE experiment was successfully conducted in IBML at LANL for 80 h without sample failure.
- The pre irradiation testing has been proven necessary and accurate.
- The sample analysis (SEM/WDX, AFM/MFM/C-AFM, Nanoindentation and TEM) will be performed as soon as the sample activity is below 20 000 dpm).

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