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Application of backscatter electrons for large area imaging of cavities produced by neutron irradiation

V.I. Pastukhov [a, b, c], S.A. Averin [a, c], V.L. Panchenko [a, c], I.A. Portnykh [a], P.D. Freyer [d], L.A. Giannuzzi [e], F.A. Garner [c, f, g, *]

[a] Joint Stock Company "Institute of Nuclear Materials" (JSC "INM"), Zarechny, Sverdlovsk Region, Russia
[b] Ural Federal University Named After the First President of Russia, B. N. Yeltsyn, Ekaterinburg, Russia
[c] National Research Nuclear University MEPhI (Moscow Engineering Physics Institute), Moscow, Russia
[d] Westinghouse Electric Company, Pittsburgh, PA, USA
[e] L.A. Giannuzzi & Associates LLC, Fort Myers, FL, USA
[f] Radiation Effects Consulting LLC, Richland, WA, USA
[g] Texas A&M University, College Station, TX, USA

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ABSTRACT

It is shown that with proper optimization, backscattered electrons in a scanning electron microscope can produce images of cavity distribution in austenitic steels over a large specimen surface for a depth of ~500–700 nm, eliminating the need for electropolishing or multiple specimen production. This technique is especially useful for quantifying cavity structures when the specimen is known or suspected to contain very heterogeneous distributions of cavities. Examples are shown for cold-worked EK-164, a very heterogeneously-swelling Russian fast reactor fuel cladding steel and also for AISI 304, a homogeneously-swelling Western steel used for major structural components of light water cooled reactors. This non-destructive overview method of quantifying cavity distribution can be used to direct the location and number of required focused ion beam prepared transmission electron microscopy specimens for examination of either neutron or ion-irradiated specimens. This technique can also be applied in stereo mode to quantify the depth dependence of cavity distributions.

1. Introduction

Cavities in the form of voids or gas-pressurized bubbles are often formed during displacive irradiation of metals and can lead to changes in metal volume and also to changes in various physical and mechanical properties [1–4]. While changes in density or physical dimension can be used to estimate the aggregate cavity volume or swelling, transmission electron microscopy (TEM) is most often used to image and quantify the density and size distribution of radiation-induced cavities. Microscopy of components such as fuel pin cladding and reactor structural components has historically required cutting the radioactive component into pieces of manageable size, followed by the preparation of a thin electron-transparent foil specimen via electropolishing, a process which adds complexity and difficulty in material handling and also involves the production of additional liquid radioactive waste.

More recently, focused ion beam (FIB) production of very small electron-transparent lamellae has been used to prepare specimens for imaging cavity microstructure, eliminating production of additional liquid radioactive waste, but possibly producing radioactive contamination in the FIB chamber. When an irradiated specimen is known or suspected to contain very heterogeneous distributions of cavities, however, the thin area surrounding the perforation in TEM analysis presents only a very small sub-region of the specimen, and therefore it may be difficult to obtain statistically representative results of microstructure and cavity morphology that are typical of the bulk specimen. In some cases the very heterogeneity of void or bubble ensembles, as well as the heterogeneity of other accompanying microstructural and microchemical features coexisting in the same volume, may determine where perforation occurs during electropolishing, yielding a selective and potentially misleading picture of the overall cavity and precipitate distribution.

* Corresponding author. Radiation Effects Consulting LLC, Richland, WA, USA.
E-mail address: frank.garner@dslextreme.com (F.A. Garner).

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Conversely, state-of-the art scanning electron microscopes (SEM) may be used to image a large field of view (e.g., up to hundreds of micrometers or more) with ultimate resolution on the order of ~1 nm [5–9]. Sample preparation for SEM is also much easier than for TEM examination. Therefore, employing SEM for imaging radiation-induced cavities is desirable if it can be demonstrated that proper imaging techniques can be developed. Obtaining SEM large-area overview images can also identify regions for subsequent and more detailed TEM work using site-specific FIB specimen preparation methods. Such a technique would also be very useful for examination of ion-irradiated specimens that are not necessarily radioactive, but that have cavities that are distributed in a layer that is only a micrometer or less from the ion-incident specimen surface.

Secondary electrons are usually collected in an SEM when surface information is desired since they possess low energy (<~50 eV) and correspondingly, their escape depth from the target surface is only ~1 nm for metals and up to 10 nm for insulators [7,8]. Thus, secondary electrons are suitable for surface imaging, but are not suitable for sub-surface cavity imaging in metals. Conversely, back-scattered electrons (BSE) have energies up to that of the primary incident electron, and thus, may originate from a depth of micrometers from the sample surface, depending on the incident electron beam energy and the composition of the target [7–9]. The depth of BSE emission depends on the average atomic number within the interaction volume and the accelerating voltage.

It should be noted that the average width of the volume producing back-scattered electrons is significantly larger than the diameter of the primary electron beam and that cavities near or within the interaction volume can alter the escape probability of the BSE. Fig. 1 shows that, depending on the relative depths and sizes of the cavity and the interaction volume, the number of BSEs escaping the surface can be modified and under some conditions can produce an image of the cavity. A more detailed finite element modeling of this phenomenon and its tendency to provide a cavity image for a given cavity size and depth will be discussed later in the paper.

BSEs have been previously used to characterize porosity in many materials including nuclear fuel, shale, ceramic coatings on dental implants, and integrated circuits [8,10–15]. In most previous applications of BSE imaging the porosity was either connected to the surface [10–14] or was covered by a thin deposited layer [15]. In this paper we explore the use of SEM BSE imaging to characterize radiation-induced sub-surface cavity formation in irradiated metals where the porosity is not connected to the surface. BSE analyses are presented on three specimens of EK-164 stainless steel examined in Russia and on a fourth specimen of AISI 304 stainless steel examined in the USA. These two alloys are representative of complex non-uniform microstructures and simple uniform microstructures, respectively.

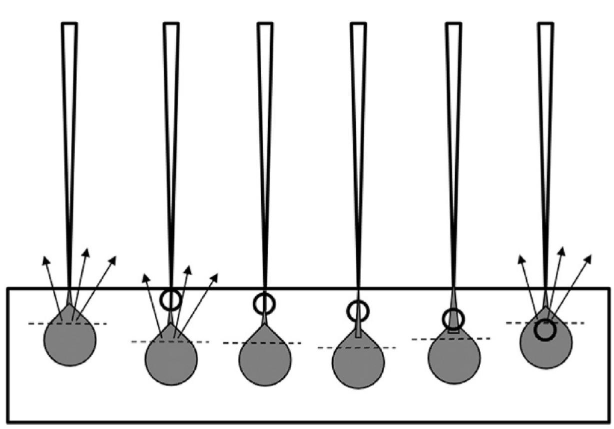

Fig. 1. Schematic illustration showing how cavities can alter the number of BSE that exit the beam-incident surface, depending on the relative sizes and depths of the cavity and the interaction volume.

2. Materials and procedure

2.1. Russian EK-164 cladding steel from BN-600

The first specimen set was derived from a thin (~0.4 mm) fuel cladding tube constructed from 20% cold-worked EK-164 austenitic steel (16Cr-19Ni-2Mo-2Mn-Si-Ti-Nb-V-B) after irradiation in a fuel assembly in the Russian BN-600 fast reactor. Irradiated specimens were examined from several elevations on the cladding tube that possessed various temperature and dose combinations. The Russian code GEFEST developed by the Institute of Physics and Power Engineering for this reactor were used to calculate the accumulated displacement dose, using the standard NRT dpa model, as well as the time-dependent temperature history for any position on the cladding tube. Time-averaged temperatures are reported for the inside face of the cladding tube. Time-averaged temperatures are necessary because burn-up of fuel produces a gradual decrease in the local cladding temperature, although deliberate periodic increases in neutron flux are sometimes performed, a process that raises the burn-up rate and therefore raises the temperature abruptly back toward its initial value, before it again declines with additional burn-up.

After cutting of small sections from the fuel pin, followed by removal of the fuel and cleaning to remove fuel and sodium residue, specimens were prepared for SEM analysis by mounting in a standard Lucite mold employing a conductive resin, followed by conventional metallographic grinding and final polishing using a suspension of colloidal silica to remove residual surface damage. A Struers Tegra System was used for grinding and polishing, employing standard methods developed for stainless steel. It was determined in exploratory studies that a necessary precondition for effective SEM-BSE cavity imaging requires that the final surface preparation must yield a very flat, featureless surface that is free of residual cold-work or significant roughness. Standard polishing with colloidal silica was found to meet this requirement. In general it was found that relative changes in local surface topography of <100 nm were sufficient to produce good SEM images of cavities.

The mounted specimens were examined using a Tescan Mira3 LMU SEM with a Schottky field emission electron source and a backscatter ring detector scintillator positioned above the sample. The SEM was operated at various electron beam energies in the range of 15–30 keV at a working distance that varied from 7 to 13 mm, depending on the specimen examined.

Some portions of the cladding were also examined by TEM analysis using specimens prepared by electrochemical-thinning. Examination was performed on a JEOL 2000EX to allow direct comparison of TEM and BSE images of the cavities in the same electron-transparent volume. TEM specimens were prepared by mechanically grinding 3 mm diameter disks to a thickness of ~130 μm, followed by twin-jet electropolishing using a 10% HCl – 90% acetic acid solution at 10 °C.

2.2. Western AISI 304 structural steel from EBR-II

To assess the general applicability of this technique to other alloys and irradiation conditions, a previously TEM-examined specimen derived from thick hexagonal blocks of ~5% cold-worked 304 stainless steel was reexamined using the BSE technique. This specimen was part of a larger experimental series and