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Concurrent *in situ* ion irradiation transmission electron microscope

K. Hattar *, D.C. Bufford, D.L. Buller

Department of Radiation–Solid Interactions, Sandia National Laboratories, United States

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ABSTRACT

An *in situ* ion irradiation transmission electron microscope has been developed and is operational at Sandia National Laboratories. This facility permits high spatial resolution, real time observation of electron transparent samples under ion irradiation, implantation, mechanical loading, corrosive environments, and combinations thereof. This includes the simultaneous implantation of low-energy gas ions (0.8–30 keV) during high-energy heavy ion irradiation (0.8–48 MeV). Initial results in polycrystalline gold foils are provided to demonstrate the range of capabilities.

1. Introduction

The transmission electron microscope (TEM) has the well-known capability to observe specimens in real time at the nanoscale. Experiments performed *in situ* inside of the TEM provide the perfect platform for elucidating fundamental mechanisms governing material evolution in controlled environments. The knowledge gained from these experiments is essential to advancing predictive models of material response [1,2], which are important when traditional materials reliability testing cannot be performed either due to the remoteness of the application, harshness of the environment, longevity of service, or combinations thereof. Radiation environments pose substantial challenges due to obvious difficulties in performing experiments within high radiation areas, handling potentially radioactive materials, and with respect to the extended time often required for some effects to manifest.

In extreme circumstances, including space applications and nuclear reactors, the difficulties in predicting material response are exacerbated due to the often synergistic interactions that occur between various elements in the environment. This is seen first-hand in boiling water reactors, where radiation induced segregation can affect grain boundaries and interfaces in metal alloys, and shadow corrosion can affect dissimilar metals in close proximity [3,4]. Synergistic effects often manifest themselves when otherwise well-designed and controlled experiments exclude part of the radiation, stress, or corrosive environment. Tanaka et al. demonstrated minimal swelling in Fe–Cr alloys when irradiated with concurrent Fe/He and Fe/H beams, but more than one order of magnitude increase in swelling and void formation during irradiation with all three species at once [5]. In a similar fashion, due to the increased reliance on satellite based technology it has become essential to be able to predict device performance and thus materials properties in the radiation environment of space. Near-Earth space environments include a spectrum of energetic particles, which varies with distance and position relative to the planet. In addition to ions trapped by the magnetosphere, satellites are subjected to cosmic rays originating from the sun or other galactic sources. Microelectronic devices in these environments are not serviceable, and face degradation in performance and operation because of radiation damage events [6,7]. For all of these environments and many more, a fundamental experimental understanding of the evolution at the nanoscale, for which *in situ* TEM is ideally suited, is essential to validate and inform predictive models.

Not long after the invention of the TEM by Ruska in 1933 [8] and its commercialization in 1939 [9,10], several serendipitous observations of structural evolution dawned the beginning of *in situ* TEM experiments. These ranged from serendipitous ion implantation of the TEM foil in 1961 with oxygen from a contaminated tungsten filament [11], to observations of dislocation formation from displacement by energetic electrons [12]. Since these initial observations, a significant concerted effort has been made in the field to introduce a range of environments and radiation conditions, as well as thermal, electrical, and mechanical loading in a

* Corresponding author. Tel.: +1 (505) 845 9859.
E-mail address: khattar@sandia.gov (K. Hattar).

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controlled manner to the TEM sample. This has predominately been accomplished through the advancement of TEM holder technology applicable to the now standard side entry port design [13].

Throughout the last several decades, extensive research efforts have been undertaken at several laboratories around the world in an effort to introduce controlled ion beams into TEMs. An excellent overview of the development of these facilities and details of the ones operational in 2009 can be found in Ref. [14]. Each of these large, complex facilities operates under different experimental parameters dictated by the TEM utilized, the ion accelerator(s) attached, and the ion beamline specifics. These facilities have provided a wealth of fundamental insight into radiation–solid interactions over the last half century [15–21]. Over the same time period, *in situ* TEM deformation and failure studies have become increasingly refined. Initial controlled straining experiments with a simple applied external load have progressed to quantitative *in situ* tensile, compression, and indentation experiments where the associated stress and strain or load and displacement of the sample can be directly correlated to the evolution of the material observed in real time. The observations made using these evolving techniques have provided fundamental insight into the active mechanisms governing the plasticity of structural metals [22–28]. Finally, recent advancements in microfabrication tolerances have permitted rapid advancements in environmental TEM, from low vapor pressure conditions induced within the column or via dedicated facilities requiring significant amounts of differential pumping [29–32], to self-contained miniaturized environmental cells sealed by mechanically stable, electron transparent, amorphous membranes [33,34]. This recent advancement in vapor phase experiments now permits the incorporation of controlled liquid and gas environments into almost any TEM system.

Additionally, recent advancements in cathodoluminescence and electron tomography have been significant. *In situ* TEM cathodoluminescence is a technique that permits direct real time measurement of photons emitted from the material, as a result of the impinging electron beam. The collection efficiency of the technique has greatly advanced and now permits direct correlation between individual lattice defects and the resulting spectra [35]. Three-dimensional visualization techniques have seen rapid evolution due to significant computational advancements. In the past decade stereomicroscopy techniques have given way to full three-dimensional reconstructions that can be produced from automatically-collected tilting series [36], which permit a three-dimensional understanding of the material system. When performed sequentially with various *in situ* TEM techniques, a full four-dimensional understanding of the material's evolution is possible. Other advancements in traditional analytical TEM techniques outside of the scope of this manuscript have been recently reviewed here: [37,38]. The combination of these advancements permits the further development of complex *in situ* TEM experiments that can be run in overlapping extreme conditions not previously possible, with simultaneous potential for a greater wealth of experimental details to be obtained, analyzed, and incorporated into predictive models [39].

2. The instruments comprising the I^3TEM facility

In order to better understand the fundamental mechanisms governing microstructural evolution in a range and combination of extreme environments, Sandia National Laboratories has developed a concurrent *in situ* ion irradiation TEM (I^3TEM). This facility is housed within the Ion Beam Laboratory, a Sandia collaborative facility with a range of advanced ion beam capabilities [40,41]. The major components of the I^3TEM facility can be seen in Fig. 1, and include a 200 kV JEOL 2100(HT) TEM, a 6 MV EN Tandem Van de Graaff–Pelletron accelerator, and a 10 kV Colutron G-1 ion accelerator. The JEOL 2100(HT) LaB_6 microscope, Fig. 1A, was chosen as it provides a versatile platform with a high tilt pole piece best suited for the high contrast imaging that is most beneficial for resolving radiation damage. The microscope was located within 1 cm and 1° precision during installation, providing the Tandem beam a direct path to the sample with minimal steering. This TEM has been outfitted with a TVIPS 4k × 4k camera that permits high quality single electron sensitive images and a 1k × 1k retractable camera that permits acquisition of video at up to 30 frames per second (FPS) during the *in situ* TEM experiments.

To maximize the resolution and stability of the TEM, an effort was made to minimize noise from electrical, mechanical, and thermal sources surrounding the instrument before connecting the ion beamline. As such, the facility utilizes high quality electrical power and dedicated grounds. The portion of the building housing the TEM was built to meet the VC-E vibration criterion of 3.12 μm/s, and its foundation is separated from the rest of the building by an isolation joint. The TEM area is isolated from the rest of the Tandem hall by heavy coated fabric laser curtains, which provide both noise and light reduction needed during experiments, while allowing easy access to the enclosed TEM and beamline components for operation and maintenance. HVAC vents in this area were equipped with radial flow diffusers, which provide low velocity high volume airflow, minimizing vibration while still allowing consistent temperature control. In addition, the standard passive pneumatic dampeners for the JEOL 2100 column were replaced with active vibration isolators, which better dampen low frequency vibrations around 1 Hz inherently associated with the column. Finally, the goniometer with piezo-controlled elements provides movement precision of up to 0.4 Å/step and factory-specified drift rate of 0.2 nm/min, permitting direct, relatively seamless corrections for the small drift typically experienced during many *in situ* TEM experiments. The combination of these efforts resulted in a TEM that maintains a 2.5 Å point resolution during most *in situ* ion irradiation and implantation conditions.

The 6 MV Tandem and 10 kV Colutron are connected to the I^3TEM perpendicularly to the electron beam through a custom made electrically isolated and mechanically dampened port to ensure the highest resolution possible during *in situ* TEM experiments. The EN Tandem, Fig. 1B, originally manufactured in 1962, has been upgraded to use two Pelletron chains, Dowlish inclined field tubes, and a state of the art LabVIEW control system. The four ion sources available to the Tandem include SNICS (source of negative ions by cesium sputtering), Alphatross (rubidium exchange source for He^-), a duoplasmatron proton source (for negative ions of gas atoms), and an Hiconex 834 sputter source where negative ion species can be quickly changed. The combination of sources permits the acceleration of a wide range of ion species ranging between protons and Au with energies as low as 800 keV and as high as 88 MeV, respectively. Beams are produced with controlled ion fluxes ranging from 1.6×10^7 to 3.2×10^{13} ions cm^{-2} s^{-1} incident on the sample. Greater details of the ion beam conditions previously run in this Tandem accelerator can be found in Middleton's cookbook [42].

Similarly, the 10 kV Colutron G-1 (Fig. 1C) was produced in 1979 [43,44], but was extensively retrofitted with new power supplies, vacuum system, and LabVIEW based control system during installation in the I^3TEM facility. The Colutron can accelerate gas ions produced by a hot filament source to energies between 0.8 keV and 30 keV. Details on the various gas phases that have been accelerated out of similar Colutron guns can be found in these Refs. [45,46]. The upgrades to both accelerators have significantly increased the control and stability of the ion beams produced. In order to adequately manipulate the ion beams from both the Tandem and Colutron accelerators, a total of three quadrupole focusing magnets, two Einzel focusing lenses, five steering