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Resolution of the carbon contamination problem in ion irradiation experiments

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ABSTRACT

The widely experienced problem of carbon uptake in samples during ion irradiation was systematically investigated to identify the source of carbon and to develop mitigation techniques. Possible sources of carbon included carbon ions or neutrals incorporated into the ion beam, hydrocarbons in the vacuum system, and carbon species on the sample and fixture surfaces. Secondary ion mass spectrometry, atom probe tomography, elastic backscattering spectrometry, and principally, nuclear reaction analysis, were used to profile carbon in a variety of substrates prior to and following irradiation with Fe^{2+} ions at high temperature. Ion irradiation of high purity Si and Ni, and also of alloy 800H coated with a thin film of alumina eliminated the ion beam as the source of carbon. Hydrocarbons in the vacuum and/or on the sample and fixtures was the source of the carbon that became incorporated into the samples during irradiation. Plasma cleaning of the sample and sample stage, and incorporation of a liquid nitrogen cold trap both individually and especially in combination, completely eliminated the uptake of carbon during heavy ion irradiation. While less convenient, coating the sample with a thin film of alumina was also effective in eliminating carbon incorporation.

1. Introduction

The application of ion irradiation to understand radiation effects in materials has been an active field of study since the 1950s. Numerous advances in our understanding of materials behavior have been made with ion irradiation including the discovery [1] and development [2] of radiation induced segregation, radiation-induced precipitation [3], void swelling [4], and radiation enhanced diffusion [5] among others. Recently, ion irradiation has gained increased attention in an effort to simulate the effects of radiation in a reactor environment. Various studies [6–8] have been conducted that show the capability of ion irradiation to qualitatively and quantitatively capture many, if not all, of the microstructure features created in reactor. The advantages of ion irradiation are many. Dose rates (typically 10^{-3} to 10^{-4} dpa/s) are much higher than under neutron irradiation (10^{-7} to 10^{-8} dpa/s) which means that 200 dpa[1] can be reached in days or weeks instead of decades. Because there is little activation, samples can be handled as if they were unirradiated, eliminating the need for the extremely high investment in time and cost connected with the use of hot cells and dedicated characterization instrumentation. Control of ion irradiation experiments is much better than experiments in reactor, and the result is that ion irradiation is 10–1000× less costly and 10–100x quicker than test reactor irradiation. Critical to the success of ion irradiation as a radiation damage simulation tool is that the ion irradiated microstructure reflects the damage created by the ions and is not influenced by external factors such as incorporation of impurities into the sample during irradiation.

Such is the case today in the radiation damage community that many laboratories are experiencing the pickup of carbon in their samples during ion irradiation. Carbon is incorporated into the irradiated microstructure, not just as a surface contaminant. The result is an alteration of the microstructure, most notably the formation of carbides, and modification of processes such as cavity evolution. Thus it is carbon incorporation into the sample over the depth of penetration of the ion beam (0.1 to several μm) that is important. Most reports come from self-ion irradiation of iron- and nickel-base alloys irradiated at high temperature. As this observation is clearly an unintended and unwanted effect, these

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[1] dpa is displacement per atom. For a value of one dpa, every atom will, on average, be displaced once.

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observations are seldom published in the open literature. However, there exists substantial earlier literature documenting this occurrence. Singer et al. [9,10] addressed observations of carbon uptake in Ti implanted steel and Ni-plated substrates. They found that irradiation with Ti resulted in incorporation of C into the solid surfaces. Carbon was distributed in a diffusion-like profile from the surface inwards. Similar observations were made for Cr and Ta-implanted steels. The authors speculated [9] and later provided evidence [10] that the uptake of carbon is due to vacuum carburization in which beam-enhanced or beam-induced adsorption and dissociation of residual CO and CO_2 molecules was responsible for the incorporation of carbon and the subsequent formation of carbides. Thomas and Bauer [11] observed carbide formation on Nb surfaces during proton irradiation at 1000 K. In fact, this process has been observed in many carbide-forming metals or alloys [12–17].

The problem has also been noted by the ion beam analysis community [18]. Healy [19] reported an extensive analysis of the factors causing the buildup of carbon on samples during ion beam analysis using a deuteron beam. They collected data on carbon contamination due to various factors, including vacuum pressure, beam area, beam contamination, beam current, temperature, and residual gas in the vacuum chamber. They concluded that the hydrocarbon component of the residual gas within the analysis chamber is the source of contamination, and that the hydrocarbons are cracked by the beam and attracted to the sample. They also noted that a cold trap near the sample minimizes contamination.

The transmission electron microscopy community has also observed the contamination of samples under the electron beam. In fact, it has been a common practice to use the buildup of carbon on the front and back surfaces of a sample to estimate sample thickness. Carbon on TEM and SEM samples was observed to occur only in the area under the beam. The TEM community has mitigated the buildup of carbon through the use of plasma cleaning of the sample prior to loading into the column, and by the use of a cold-finger near the sample during observation. Thus, the contamination of carbon can occur during heavy ion irradiation, ion beam analysis with light ions or under electron irradiation in the TEM or SEM. The common theme in all of these observations is that the contamination occurs only under the beam.

An example of carbon incorporation during ion irradiation include alloy 800H irradiated with 5 MeV Fe^{2+} at a temperature of 440 °C to a damage level of 20 dpa. Fig. 1 shows composition vs. depth profiles using three different techniques. Fig. 1a shows the carbon profile from nuclear reaction analysis utilizing the $^{12}C(d,p_0)^{13}C$ reaction with a deuteron energy of 1.5 MeV. The composition profile is characterized by a 1.7 nm thick surface layer of carbon, and enrichment below the surface, a sub-surface peak at about 1000 nm, and a depression below the bulk level starting at about 1300 nm and extending deeper into the sample. A SIMS

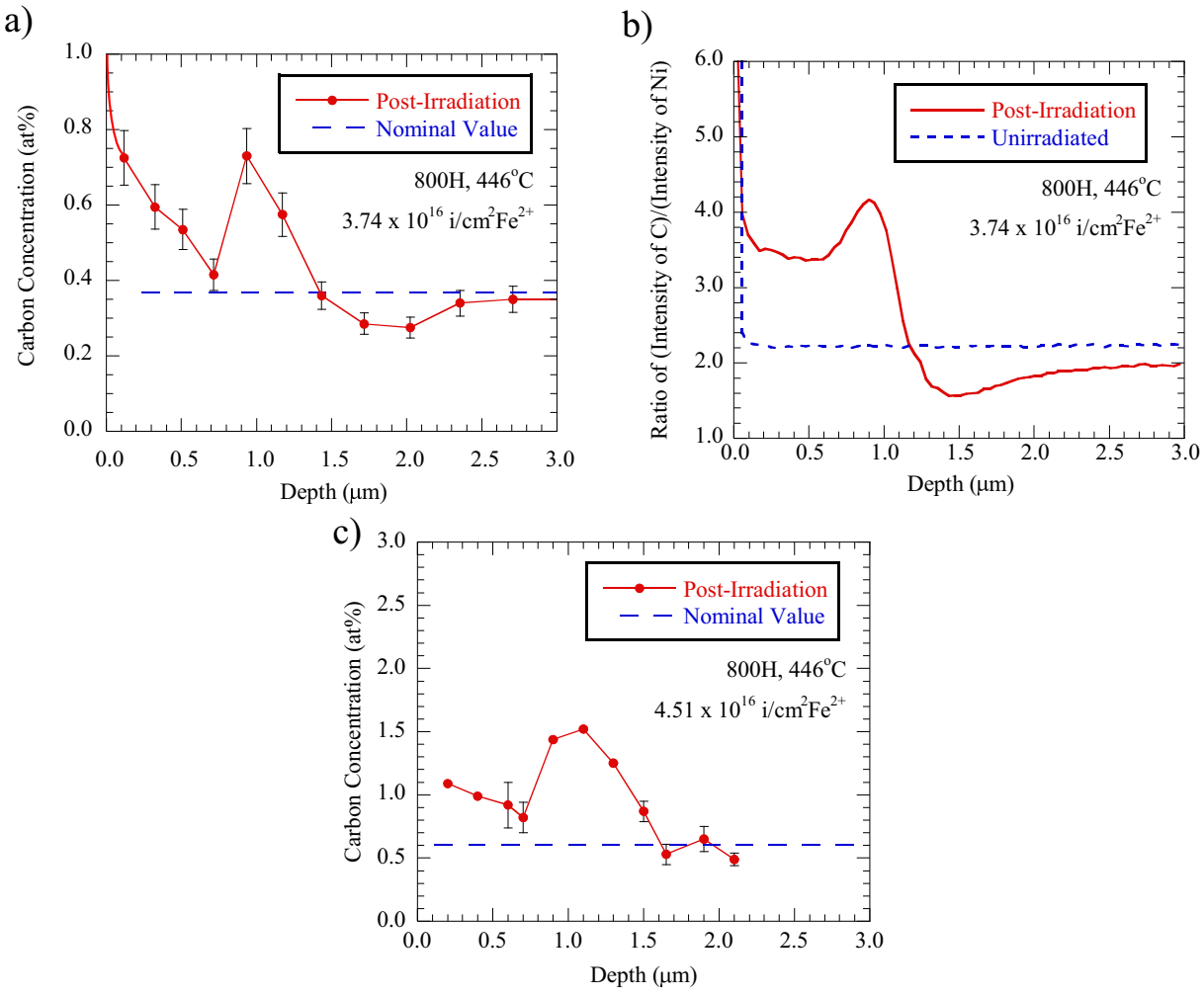

Fig. 1. Carbon concentration vs. depth profiles in alloy 800H following irradiation with 5 MeV Fe^{2+} at 446 °C to similar fluences by a) nuclear reaction analysis (4.51×10^{16} i/cm^2/17 dpa), b) secondary ion mass spectrometry (3.74×10^{16} i/cm^2/17 dpa plus He), and c) atom probe tomography (4.51×10^{16} i/cm^2/20 dpa).