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Status Summary of FY16 Atom Probe Tomography Studies on UCSB ATR-2 Irradiated RPV Steels

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SUMMARY

The University of California Santa Barbara ATR-2 RPV steel irradiation experiment was awarded in 2010 by the Nuclear Science User Facility (formerly ATR NSUF) through a competitive peer review proposal process. The experiment involved irradiation of nearly 1300 samples distributed over 13 capsules. The major objective of this experiment was to better understand embrittlement behavior of reactor pressure steels (RPV) at doses beyond which available data exists, yet may be achieved if reactor operating licenses are extended beyond 60 years. The experiment was instrumented during irradiation and active temperature control was used to maintain temperatures throughout the irradiation. Six samples were selected from the matrix of materials and prepared using the focused ion beam (FIB), located at the Center for Advanced Energy Studies (CAES), to perform atom probe tomography (APT) and examine the formation of high fluence embrittling phases. The nature of these phases is discussed and status of the overall experiment and modeling effort is provided.

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Status Summary of FY16 Atom Probe Tomography Studies on UCSB ATR-2 Irradiated RPV Steels

1. Introduction

The reactor pressure vessel (RPV) in a light-water reactor (LWR) represents the first line of defense against a release of radiation in case of an accident. Thus, regulations that govern the operation of commercial nuclear power plants require conservative margins of fracture toughness, both during normal operation and under accident scenarios. In the unirradiated condition, the RPV has sufficient fracture toughness such that failure is implausible under any postulated condition, including pressurized thermal shock (PTS) in pressurized water reactors (PWR). In the irradiated condition, however, the fracture toughness of the RPV may be severely degraded, with the degree of toughness loss dependent on the radiation sensitivity of the steel and the amount of neutron exposure.

Available embrittlement prediction models, quantified in terms of ductile to brittle transition temperature shifts (TTS), e.g. [1], and our present understanding of radiation damage are not fully quantitative, and do not treat all potentially significant variables and issues, particularly considering extension of operation to 80 years. Most important of these untreated issues is the formation of Mn-Ni-Si phases, which will likely not form until the later stages of reactor operation and thus have been referred to as "Late Blooming Phases" [2, 3]. There is now overwhelming evidence that these precipitates exist and will cause large amounts of embrittlement at extended life fluences, beyond that which would be predicted by the current models [4-6]. Thus, a primary objective of the LWRSP RPV task is to develop robust predictions of RPV embrittlement at extended life fluences of up 10^{20} n/cm^2 (>1 MeV), pertinent to plant operation of some pressurized water reactors (PWR) for 80 full power years. These models must include the effects of the Mn-Ni-Si precipitates, which have largely not yet formed in the current surveillance database. This report includes recent progress made on characterizing the irradiated microstructures of RPV model steels from the UCSB ATR-2 irradiation with a specific focus on Mn-Ni-Si (MNS) precipitation. While this large experiment includes collaboration with multiple universities and national labs, the primary focus here is on work done in the Center for Advanced Energy Studies (CAES) Microscopy and Characterization Suite (MaCS).

2. UCSB ATR-2 Irradiation

The main challenge associated with the development of a high fluence embrittlement prediction model is that there is very little surveillance data at extended life fluences. To address this issue, a large scale irradiation designed to reach a peak fluence of > $1\text{x}10^{20}$ n/cm^2 was recently completed in the Advanced Test Reactor (ATR) at the Idaho National Laboratory. The experiment, called UCSB ATR-2, was awarded to the University of California, Santa Barbara (UCSB) and its collaborator, Oak Ridge National Laboratory (ORNL), in 2010 with funding provided through DOE's Nuclear Science User Facilities. In collaboration with UCSB, the INL staff carried out conceptual design of the sophisticated instrumented irradiation test assembly. Following this, INL staff completed the construction and insertion of the test assembly into the reactor. A total of 172 alloys were included in the experiment and were acquired by UCSB and ORNL, including those contributed by Rolls Royce Marine (UK), Bettis Atomic Power Laboratory (US), and the Central Research Institute for the Electric Power Industry (Japan). Additionally, surveillance materials from various operating nuclear reactors are included to enable a direct comparison of results from a test reactor at high flux and a power reactor at low flux.

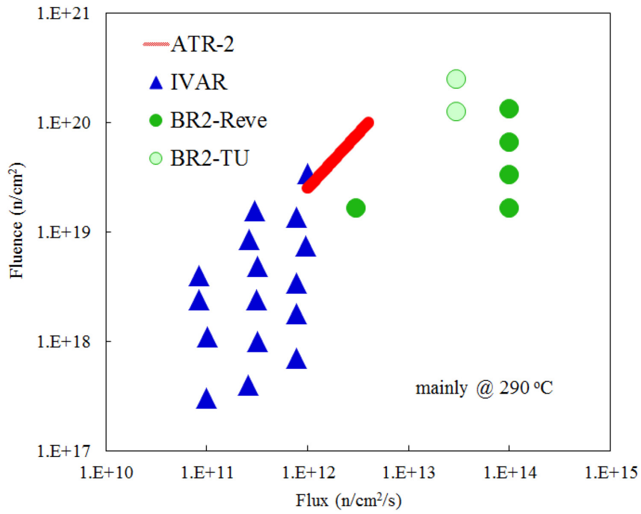

Figure 1.Fluence (n/cm^2) vs flux (n/cm^2/s) map for various irradiations completed in collaboration with UCSB.

The main objective of the experiment is to obtain a high f1uence, intermediate flux database to couple with a large body of existing data for a large set of common alloys (≥ 100) irradiated over a wide range of flux and fluence. Figure 1 shows the flux/fluence range for the ATR-2 experiment (red line). The results from the experiment will allow for direct comparisons with three existing test reactor databases (IVAR and two irradiations from the BR2 reactor, shown in triangles and circles, respectively) compiled by UCSB. Additionally, surveillance materials from various operating nuclear reactors are included in the ATR-2 experiment to enable a direct comparison of results from a test reactor at high flux and power reactors at low fluxes. In summary, a variety of relatively small specimens of many different RPV steels were irradiated in UCSB ATR-2, including many materials that have been irradiated and tested in previous test reactor and surveillance programs at different flux levels.

3. Experimental Setup

Six alloys from the UCSB ATR-2 irradiation were sent to the Center for Advanced Energy Studies (CAES) Materials and Characterization Suite (MaCS) for APT specimen fabrication. The composition of the alloys, which have systematic variations in Cu and Ni contents, are shown in Table 1. These samples were irradiated in Cup 7 of the UCSB ATR-2 irradiation to a fluence of 1.3×10^{20} n/cm^2 at a flux of 3.6×10^{12} n/cm^2/s at 290°C.

Table 1 Composition (wt.%) of the RPV model steels in the CAES APT matrix.

Alloy	Cu	Ni	Mn	Mo	P	C	Si
LC	0.41	0.86	1.44	0.55	0.005	0.14	0.23
LD	0.38	1.25	1.38	0.55	0.005	0.19	0.23
LG	0.01	0.74	1.37	0.55	0.005	0.16	0.22
LH	0.11	0.74	1.39	0.55	0.005	0.16	0.24
LI	0.20	0.74	1.37	0.55	0.005	0.16	0.24
CM6	0.02	1.68	1.50	0.54	0.007	0.15	0.17

Liftouts from all six alloys were prepared using the Quanta 3D FEG Focused Ion Beam (FIB) in the MaCS lab. The liftouts were then mounted on a sample coupon to create 7 Atom Probe Tomography (APT) specimens for each alloy. The individual APT needles were then partially sharpened to minimize the total activity and shipped to the University of California, Santa Barbara, where the final tip shaping, testing and analysis was performed. All samples were run in voltage mode in a LEAP 3000X HR at an analysis temperature < 50K, a pulse fraction of 20%, and an ion detection rate of 0.5-0.9%. Analysis was performed using the CAMECA Integrated Visualization and Analysis Software (IVAS).

4. Results

Atom probe tomography is currently ongoing on the alloy matrix with 2 alloys (LC and LH) completed thus far. An example atom map from a high Cu (0.4 wt.%), medium Ni (0.8 wt.%) steel is shown in Figure 2. A high density of precipitates, enriched in Cu, Ni, Mn and Si can clearly be seen. Further study of these precipitates shows that they are separated into a mostly Cu-enriched region and next to that, a mostly Mn-Ni-Si enriched region, seen in Figure 3, where regions rich in Cu are shown in the green surface and regions rich in Mn, Ni and Si are shown in the pink surface. In addition, a composition profile through a highlighted precipitate shows that the peak composition for Cu and Mn-Ni-Si are separated. This association between the two types of features has now been seen numerous times in both test reactor [4] and surveillance irradiated steels [6] and adds additional confirmation to the theory that the Cu precipitates act as nucleation sites for the separate Mn-Ni-Si phases. The MNS phases, which take much longer to form, will lead to significant, unaccounted for embrittlement at extended lifetimes.

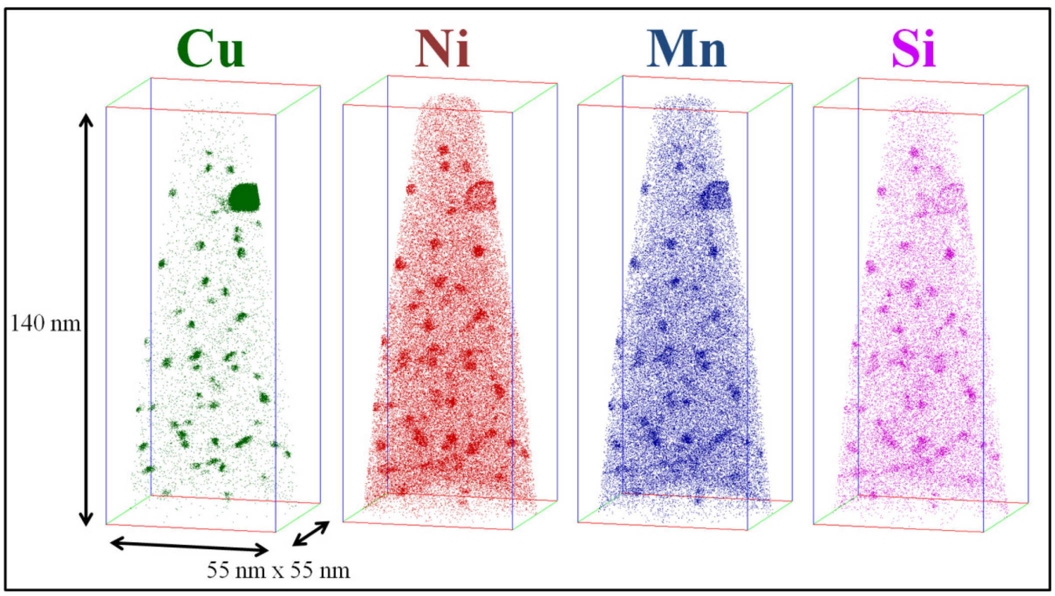

Figure 2. Elemental atom maps from a high Cu (0.4 wt.%), medium Ni (0.8wt.%) steel from the highest fluence (1.3x1020 n/cm2) condition.

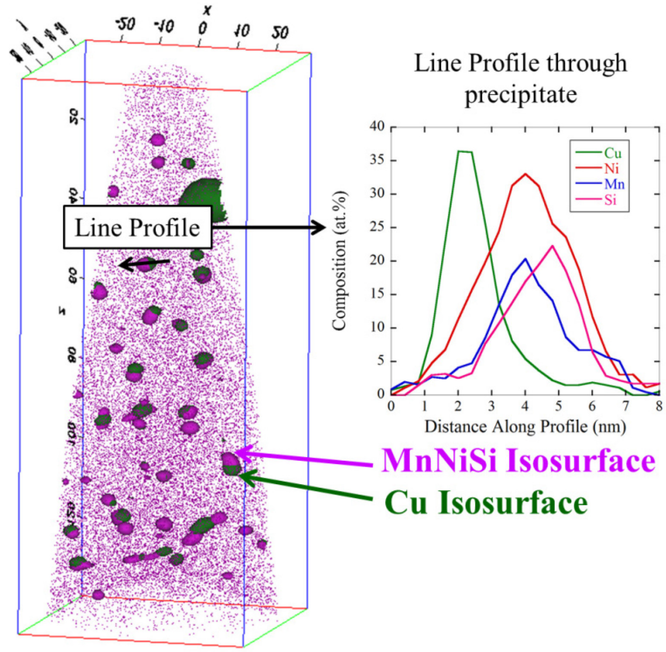

Figure 3. Atom map with Cu and MnNiSi isosurfaces and a line profile through one precipitate showing that the precipitates are a combination of Cu rich and Mn-Ni-Si rich regions.

5. Model Development

Pressure vessel embrittlement is governed by very complex interactions among environmental (flux, fluence, irradiation temperature) and compositional (Cu, Ni, Mn, Si, P) variables. UCSB has been working over the past several years to develop embrittlement prediction models for extended life fluences that include the effects of so called "Late Blooming Phases". The ATR-2 experiment will prove to be invaluable in further developing and calibrating these models. The current model is a microstructurally based Avrami model, which predicts the amount of precipitation, or volume fraction (f_v), in a given alloy as a function of fluence, then uses established correlations to convert these volume fractions to hardening and embrittlement. Ultimately, the goal is to create a model which can accurately predict embrittlement as a function of alloy chemistry and irradiation history to determine whether a specific RPV will maintain sufficient fracture toughness at extended lifetimes.

While the UCSB ATR-2 irradiation will ultimately be used in helping to fit this model, the initial results are being used to determine the accuracy of the preliminary model. The preliminary model, seen in Figure 4 was fit to the IVAR and BR2 experimental databases and was created prior to any ATR-2 data becoming available. The data points come irradiation conditions that span ≈ 3 orders of magnitude dose rate. The flux for each condition is given in the legend. The model shows remarkable agreement with the new ATR-2 data points, shown in large red circles in Figure 4. APT on the final 4 alloys will be completed shortly to compare their f_v with the Avrami predicted f_v.

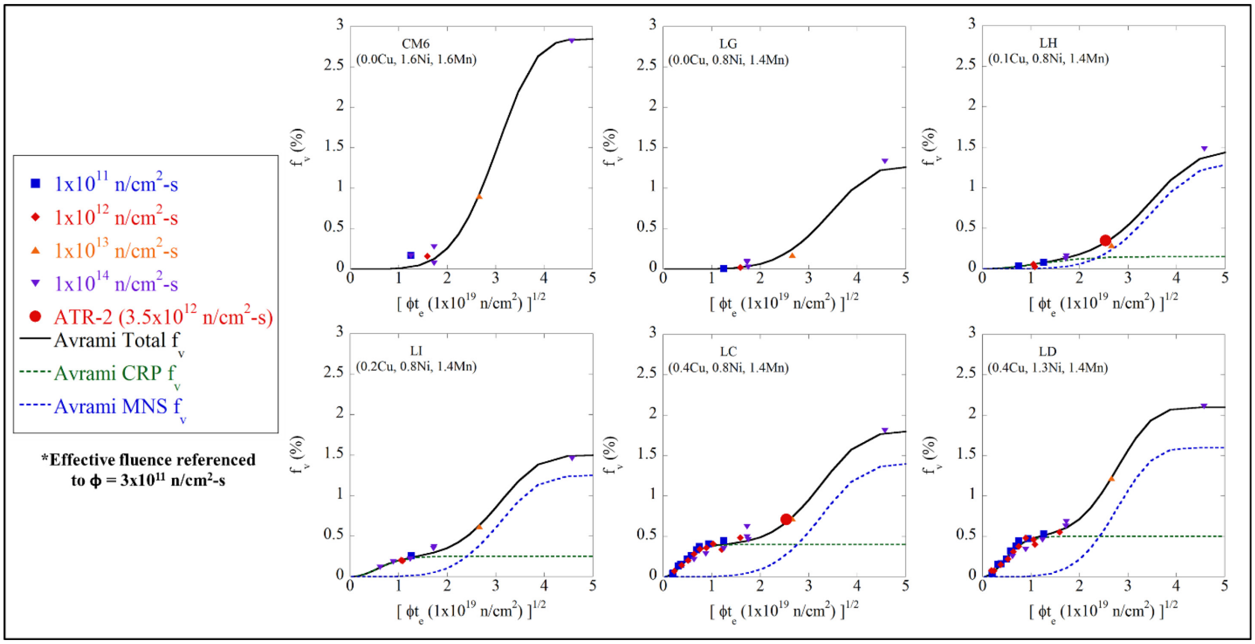

Figure 4.Preliminary UCSB Avrami model predicting fv(fluence) for the 6 alloys included in the CAES MaCS APT study. The ATR-2 data points (shown in large red circles) were not used in the fitting of the model.

6. Summary and Future Work

Extended life operation of our nation's light water reactors will require demonstrating that the reactor pressure vessels will operate within safety margins. The UCSB ATR-2 experiment is designed to investigate embrittlement at extended life fluences with two main goals. First, it will add to the general understanding of the underlying mechanisms leading to RPV embrittlement by generating large databases of both microstructural and mechanical property data. These databases will help to further refine correlations between microstructure and mechanical property changes under irradiation. In addition, the data generated in the experiment, for example the data presented above from the CAES MaCS lab, will be used to further refine and calibrate a reduced order embrittlement prediction model, which can be used by regulators to determine the amount of embrittlement a specific vessel can be expected to experience at extended lifetimes. The preliminary model created by UCSB accurately predicts the f_v from the first two alloys studied in APT from the ATR-2 experiment. Unexpected delays, primarily caused by issues with the LEAP at UCSB, have limited the total testing and analysis to only two alloys within the alloy matrix. It is expected that the other four alloys will be completed within the next several months.

7. References

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