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EPRI Project Manager

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ABSTRACT

A number of studies in the literature have concluded that dislocation loops, irradiation-induced hardening, and radiation-induced segregation at the grain boundary are not sufficient indices to assess susceptibility to irradiation assisted stress corrosion cracking (IASCC). Irradiation induced hardening and dislocation loop density and size, as determined by transmission electron microscopy (TEM), no longer change significantly past 5 dpa. However, studies of IASCC crack initiation of field-retrieved materials suggest fluence is an important factor up to 20-25 dpa. It is therefore not possible to diagnose relative degrees of IASCC susceptibility based on these parameters. This update reviews the data in EPRI's *Program on Technology Innovation: Effects of Post Irradiation Annealing on IASCC* (3002005475) along with other studies that apply TEM to quantify radiation-induced dislocation loops *and* atom probe tomography to quantify radiation-induced solute clusters.

The aggregate data suggest that proton-irradiated and light water reactor (LWR)-neutron-irradiated stainless steels have different values of the following ratio: solute cluster number density/Frank loop number density. The data also suggest that the ratio continues to increase beyond 5 dpa when the Frank loop number density and size saturate, driven by the increase in solute cluster number density. It is therefore important to investigate and establish whether clusters affect irradiated materials behavior, particularly past 5 dpa. If they do, then changes in solute cluster number density, size, volume fraction, or cluster/Frank loop ratio may serve as an index of susceptibility that can guide IASCC risk assessment.

Keywords

Atom probe tomography
Irradiated stainless steel
Irradiated microstructure
Solute clusters
Frank loops
Irradiation assisted stress corrosion cracking (IASCC)

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1 REVIEW OF AVAILABLE DATA IN THE LITERATURE

Several studies in the literature, including the work documented in *Program on Technology Innovation: Effects of Post Irradiation Annealing on IASCC.* EPRI, Palo Alto, CA: 2015. 3002005475, have now applied transmission electron microscopy (TEM) to quantify dislocation loops and atom probe tomography (APT) to quantify solute clusters [1, 3, 4, 6, 7] in the irradiated microstructure of stainless steels. It is of interest to review, in this brief update, the observations and the reported cluster/loop ratios, which also touch upon differences that may exist between proton-irradiated and LWR-irradiated microstructures.

Currently, different researchers can apply different parameters to quantify APT data within the IVAS analysis software provided by the manufacturer, Cameca[1], and a few groups use proprietary algorithms. Therefore, data from different researchers cannot be simply collected together and analyzed, except with caution and caveats. For this reason, EPRI and EDF have sponsored an APT Round Robin, involving 10 international organizations, to achieve better agreement on how APT data should be collected and analyzed; one of the co-PI's of EPRI 3002005475, Emmanuelle Marquis, is the technical advisor to the Round Robin, and the other co-PIs are participants.

However, several BWR-irradiated stainless steels in the range of 3.5-13 dpa have been analyzed by Marquis and coworkers [7], according to the same protocol as this study[2] [6]. Therefore, these data (both neutron-irradiated and proton-irradiated) can be considered together with assurance and are plotted in Figure 1-1. Only Ni-Si clusters are considered because they are strongly associated with dislocation loops, whereas Al-enriched clusters and Cu-enriched clusters are not [7]. Their number densities are compared to the Frank loop number densities via a ratio.

The (BWR) neutron-irradiated cluster/loop ratio vary from 0.1 at 3.5 dpa to 1.7 at 13 dpa. APT analysis indicates that Ni-Si-rich clusters are observed primarily on Si-segregated dislocation loops and fragments [7]. This is not surprising, since dislocation loops can serve as sites of heterogeneous nucleation.

In contrast, the proton-irradiated CP304 of EPRI 3002005475 has a much higher cluster/loop ratio of 14.6, suggesting that a substantial number of "free clusters" is present. The linear density of Ni-Si rich clusters on dislocations, as determined by APT, is 12 clusters per 100 nm. The average dislocation loop diameter according to APT is 9.4 nm (Table 3-4 of EPRI 3002005475) [or 7.4 nm according to TEM (Table 3-3 of EPRI 3002005475)]. Therefore, there are ~3.5 clusters are associated with the average dislocation loop. Consequently, approximately (3.5/14.6)x100%=24% of the clusters are associated with dislocation loops while the majority ~76% should be "free clusters."

[1] Cameca became the only manufacturer of atom probe tomographs, after its acquisition of Imago in 2010.

[2] The raw APT datasets were collected by CRIEPI (Japan) within a CRIEPI-EPRI collaboration using voltage-only mode (no laser assist), as are the data of this study.

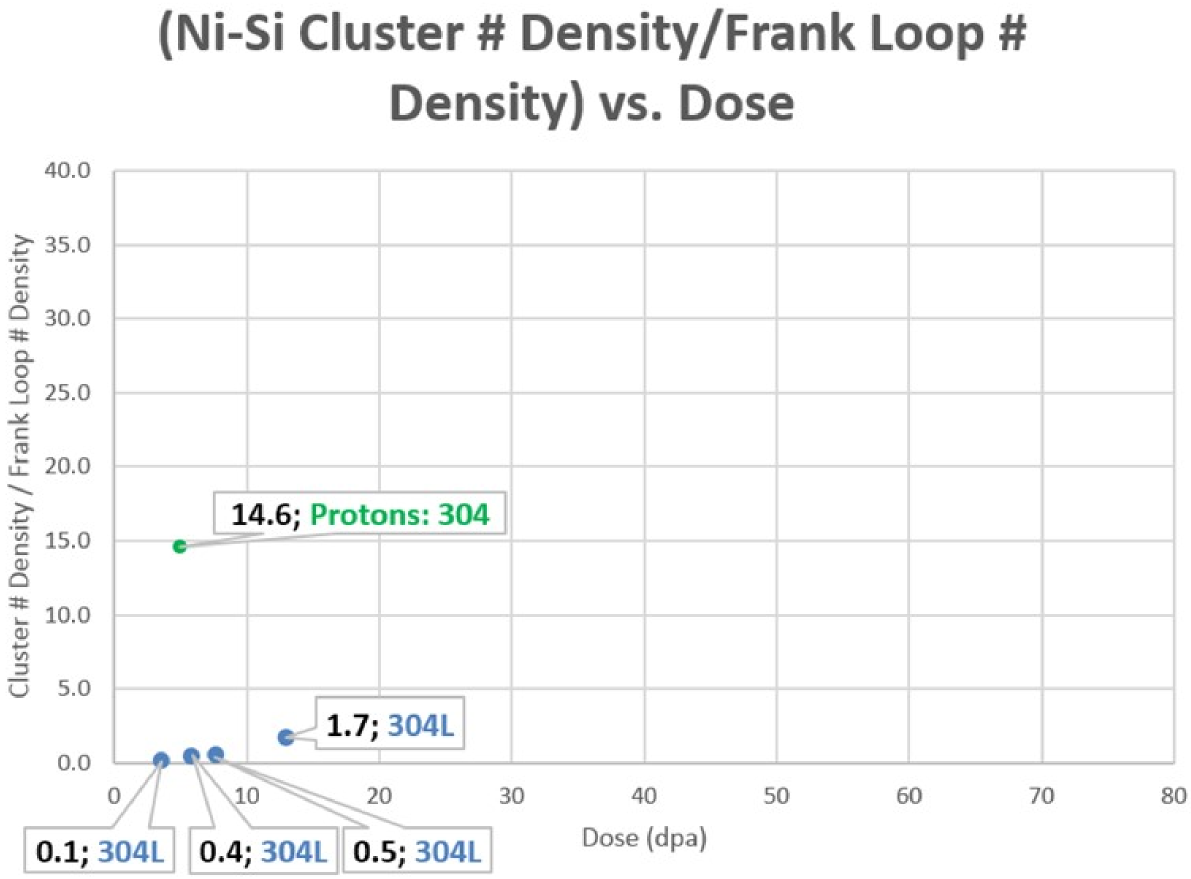

Figure 1-1
The ratio of (cluster number density/Frank loop number density) has been plotted versus dose. These datasets have been analyzed according to the same protocol. The datapoint in green (proton-irradiated 304SS) comes from EPRI 3002005475. The datapoints in blue [(BWR) neutron-irradiated 304L SS] comes from the CRIEPI-EPRI-UMich study [7]. Note that the proton-irradiated dose of 10 dpa "full-cascade" has been divided by 2 to convert it to 5 dpa "Khinchin-Pease" to facilitate comparison to (BWR) neutron-irradiated data [9]. Only Ni/Si clusters have been plotted because the CRIEPI-EPRI-UMich study indicate that they are strongly associated with dislocation loops, whereas Al-enriched clusters and Cu-enriched clusters are not.

The existence of "free clusters" might be somewhat puzzling, since homogeneous nucleation is difficult. One possible explanation is that as cluster decoration of dislocation loops progress past, say, 4 clusters, the dislocation loops are consumed and becomes a larger cluster. Dislocation loop density saturates around 2 dpa and their size saturate around 5 dpa. [10] Clusters might therefore have a role in maintaining the steady-state dislocation loop density, balancing their creation and annihilation.

Four additional data points are added to the same plot in Figure 1-2. *The APT data analysis was conducted according to different protocols, so they must be considered with caution and, at most, we can only interpret trends.* But it is interesting to note that:

- The two additional proton-irradiated data points [3, 5] also indicate high cluster/loop ratios relative to neutron-irradiated data points at comparable dose.

- The (PWR) neutron-irradiated data point at 24 dpa (cluster/loop ratio of 6.9) [4], when considered together with the BWR neutron-irradiated data points, suggests that the cluster/loop ratio can continue to increase with dose. Since dislocation loop density saturates at relatively low doses (although there is scatter) [10], the increase in the cluster/loop ratio is driven by the increase in the cluster number density.
- The (PWR) neutron-irradiated data point from CW 316SS at 74 dpa (cluster/loop ratio of 38.5) [8], may suggest that the cluster/loop ratio can continue to increase with dose, but may also suggest that network dislocations associated with cold work act as nucleation sites that enhance the formation of solute clusters.[3] In this context, it should be noted that there is only one data point from CW 316SS in Figure 1-2. More data is clearly necessary.

Although Figure 1-2 can be interpreted to suggest that the cluster/loop ratio can continue to increase with dose, the increase cannot, of course, persist indefinitely, since Si is the "limiting reactant" and will run out earlier or later, depending on the amount of Si from heat to heat. For example, in their study, Jiao and Was [3, 5] indicated that at 4.5% volume fraction of Ni-Si clusters, 19-34% of the available Si in their material were consumed by Ni-Si clusters. (The data points from their study are included in Figure 1-2.)

Broader assessment of the details discussed here requires more consistent, accurate quantification among different studies.

The reason such questions are of practical interest is that dislocation loop density and size generally saturates (no longer changes significantly) past ~5 dpa [10], but studies of IASCC crack initiation of field-retrieved materials suggest fluence is an important factor up to 20-25 dpa. Therefore, it has not been possible to "diagnose" relative degrees of IASCC susceptibility based on past microstructural examinations that focused on TEM quantification of dislocation loops.

It is therefore important to investigate and establish whether clusters affect irradiated materials behavior, particularly past 5 dpa. If they do, then changes in cluster number density, size, volume fraction, or cluster/loop ratio may serve as an index of susceptibility that can guide IASCC risk assessment.

[3] The network dislocations associated with cold-worked stainless steels are typically removed by ~20 dpa under neutron irradiation, but prior to their disappearance, they may influence the formation of solute clusters.

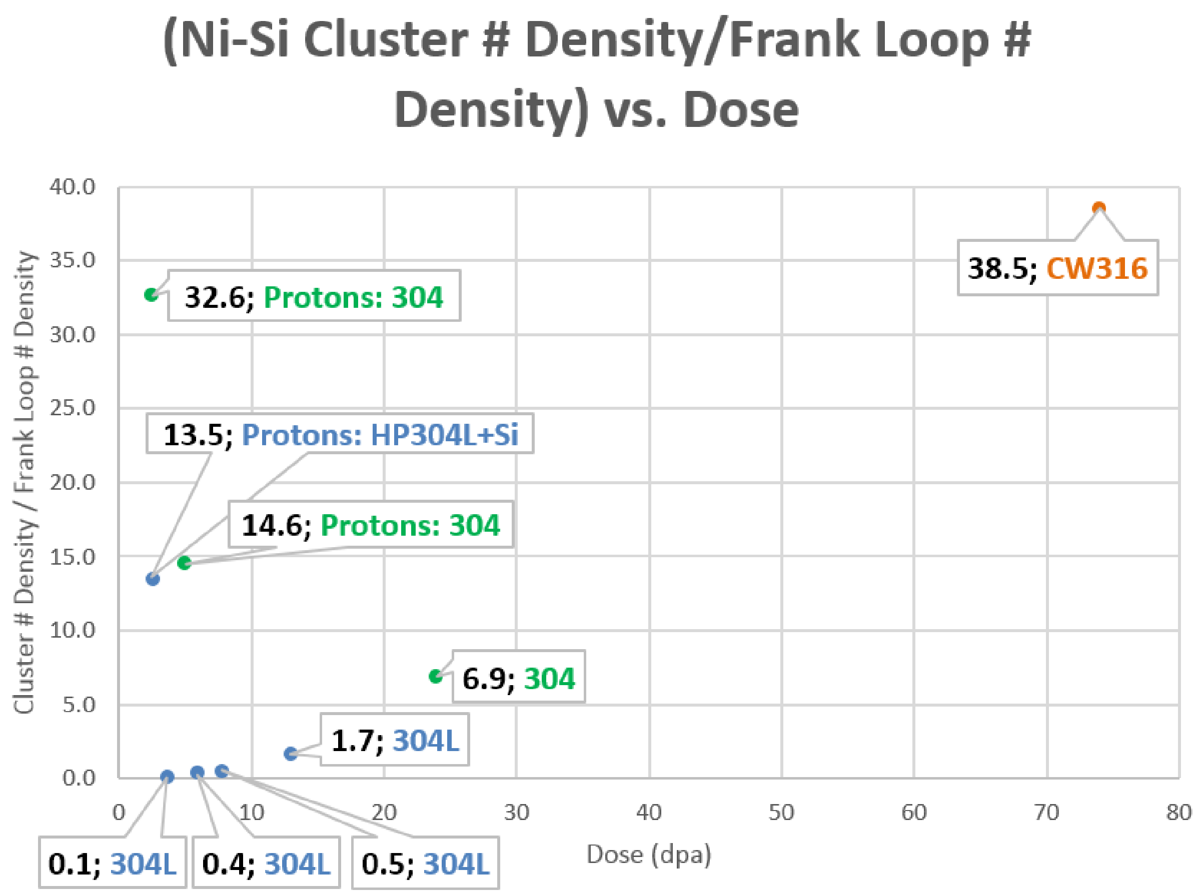

Figure 1-2
The ratio of (cluster number density/Frank loop number density) plotted versus dose, including four data points not in Figure 1-1, whose APT data not analyzed by the same protocol. Two additional data points from proton-irradiated stainless steels, at 2.5 dpa came from [3, 5]; one additional data point from PWR-irradiated 304SS came from [4]; and one additional data from PWR-irradiated CW 316SS came from [8]. For proton irradiation, "full-cascade" dose values calculated by SRIM has been divided by 2 to convert them to "Khinchin-Pease" dose values to facilitate comparison to neutron-irradiated data [9]. Only Ni/Si clusters have been plotted because the CRIEPI-EPRI-UMich study [7] indicate that they are strongly associated with dislocation loops, whereas Al-enriched clusters and Cu-enriched clusters are not.

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