



Data Article

Data on Cu- and Ni-Si-Mn-rich solute clustering in a neutron irradiated austenitic stainless steel



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ABSTRACT

The data presented in this article is supplementary to the research article “Phase instabilities in austenitic steels during particle bombardment at high and low dose rates” (Levine et al.) [5]. Needle-shaped samples were prepared with focused ion beam milling from a 304L stainless steel that was irradiated with fast neutrons ($E > 0.1$ MeV) in the BOR-60 reactor at 318 °C to 47.5 dpa. Atom probe tomography (APT) experiments in voltage mode were then conducted on a Cameca LEAP 5000X HR. Atom position, range, and mass spectrum files after reconstruction with Cameca’s IVAS software are included. Cu- and Ni-Si-Mn-rich solute nanoclusters were identified and analyzed using the Open Source Characterization of APT Reconstructions (OSCAR) program. Python code for OSCAR [4], information on the program’s underlying algorithm, and sample output files are provided. A proximity histogram of a Ni-Si-Mn-rich cluster and a 1D density/solute concentration profile of a Cu-rich cluster are given

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to demonstrate OSCAR's analytical functionalities. The provided APT dataset is valuable for benchmarking phase instabilities in neutron-irradiated austenitic stainless steels that occur at high doses. The OSCAR program can be reused to process other APT data sets where solute nanoclustering is of interest.

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Specifications Table

Subject	Materials Science
Specific subject area	Characterization of an irradiated austenitic stainless steel
Type of data	Table, Graph, Code
How data were acquired	Atom probe tomography (APT) needles fabricated using FEI Quanta DualBeam and FEI Versa3D focused ion beams (FIBs). APT performed with Cameca Local Electrode Atom Probe (LEAP) 5000X HR. Reconstructions performed with CAMECA's Integrated Visualization and Analysis Software (IVAS). Solute clusters identified and analyzed with the Open Source Characterization of APT Reconstructions (OSCAR) Python code.
Data format	Raw, Analyzed
Parameters for data collection	APT- acquisition mode: voltage, temperature: 50 K, pulse fraction: 20%, pulse rate: 200 kHz, and detection rate: 0.50 Reconstruction- electric field factor: 4.5 and image compression factor: 1.65 Cu-rich cluster analysis- solute: Cu, threshold concentration: 5.0 at.%, linking distance: 1.07 nm, and minimum solute atoms: Ni-Si-Mn-rich cluster analysis- solute: Si, threshold concentration: 8.0 at.%, linking distance: 0.75 nm, and minimum solute atoms: 9
Description of data collection	APT needles were fabricated from a neutron irradiated 304L stainless steel using FIB. APT experiments were conducted and then volumes were reconstructed with IVAS software. Output files from IVAS (.pos and .rrng files) were used as inputs to OSCAR for the identification and analysis of Cu-rich and Ni-Si-Mn-rich solute clusters.
Data source location	Institution: University of Tennessee City/Town/Region: Knoxville, TN Country: USA
Data accessibility	With the article and in [4]; Repository name: Mendeley Dat Data identification number: 10.17632/375c38sf2h. Direct URL to data: https://doi.org/10.17632/375c38sf2h
Related research article	[5] S. M. Levine, C. Pareige, Z. Jiao, P. D. Edmondson, G. S. Was, S. J. Zinkle, A. Bhattacharya, Phase instabilities in austenitic steels during particle bombardment at high and low dose rates, Mater. Des. 217 (2022) 110588.

Value of the Data

- The data is useful for understanding radiation-enhanced precipitation (REP) and radiation-induced precipitation (RIP) in austenitic stainless steels (AuSSs), which are commonly used for light water reactor (LWR) core internals. This APT volume contains Cu-rich clusters in a neutron-irradiated AuSS, regarding which there are few available datasets in literature.
- REP and RIP information in AuSS samples neutron-irradiated to high doses at LWR-relevant temperatures (270 – 370 °C) is limited. Thus, the data given herein is valuable for benchmarking degradation in AuSSs. This information benefits regulators who must decide on license extensions for aging LWR reactors.
- The influence of radiation-enhanced Cu-rich clusters on the irradiated properties of AuSSs is unknown. As a result, the data herein can be used by future studies to assess the role of Cu-rich clusters in radiation hardening and embrittlement or cavity swelling in AuSSs.

- Methods for APT solute cluster analysis are continually evolving. Here, an open-source Python program, OSCAR, for APT cluster identification and analysis is provided. The given details on data treatment are important, since clusters detected in APT datasets can vary widely based on mass spectrum ranging or the method used for cluster identification [1,6].
- The maximum separation method built into IVAS tends to erroneously identify clusters when there is low contrast between matrix and cluster solute concentrations [1]. By comparison, OSCAR uses an iso-position method to improve cluster identification accuracy. The included program and information enable reproducible analysis of APT data sets with OSCAR.
- In irradiated AuSSs, Si-rich clusters may form by radiation induced segregation (RIS) or RIP. A sphericity filter is included in OSCAR, which can be used to systematically differentiate between RIS and RIP clusters based on morphology. This approach may be useful for characterizing other irradiated structural materials by APT, where distinguishing between RIS is RIP is important.

1. Data Description

The raw data for this article [4] was collected by APT from a 304L stainless steel irradiated by fast neutrons ($E > 0.1$ MeV) in the BOR-60 reactor at 318 °C to 47.5 dpa. The run number for the APT data set is R94_10497. The raw data files include R94_10497.pos, which gives the positions and mass to charge ratio of ions in the APT reconstruction, R94_10497.rnrg, which gives the ion assignments for mass spectrum ranges, and R94_10497_massSpec.csv, which contains the mass spectrum for the reconstruction. Average bulk composition measured from 20 APT needles after performing isotopic decomposition with IVAS is provided in Table 1.

The OSCAR program that was used for Cu and Ni-Si-Mn cluster analysis is included as a Jupyter notebook (OSCAR Iso-Position.ipynb) in [4]. The Python modules required to run the notebook (cluster_analysis.py, apt_utils.py, and visualization.py) are stored in the apt_pkg folder. Details on OSCAR are provided in the next section. Sample outputs for a Si cluster analysis are provided. These files include *posOSCAR*.csv, *clusters*.csv, *cores*.csv, *eroded clusters*.csv, and *summary*.txt (Note: * is a wildcard string in the file name). The *posOSCAR*.csv file includes the atom positions, mass to charge ratio, element IDs, and results from the local concentration analysis for the selected solute. The *clusters*.csv file includes information (atoms, mass, center of mass, and radius) on the clusters prior to erosion. The *cores*.csv file includes the core composition of each cluster and the calculated erosion distance. The *eroded clusters*.csv file includes information (atoms, mass, center of mass, and radius) on the clusters after erosion and results from the sphericity filter. Finally, the *summary*.txt provides a brief report on the cluster analysis results.

The graphs provided in Figs. 1 and 2 contain data generated when Ni-Si-Mn rich and Cu-rich clusters in data set R94_10497 were analyzed with OSCAR. The data in Fig. 1 is a proximity histogram (proxigram) measured for a Ni-Si-Mn-rich cluster. Concentrations for Fe, Cr, Ni, Si, and Mn are plotted as a function of distance from the initial cluster interface (the interface prior to erosion). Negative distances represent positions in the cluster, while positive distances represent positions outside the cluster (in the matrix). The erosion distance, $d_{erosion}$, was calculated by fitting a four parameter logistic function to the Fe curve. For this cluster, the calculated erosion distance (0.55 nm) is shown in gray. The data in Fig. 2 is a profile of Cu concentration and relative density measured across a Cu-rich cluster in the x-direction. The boundaries (minimum and maximum x-positions) of the cluster are denoted with dotted lines.

Table 1
Average bulk composition measured by APT after isotopic decomposition.

	Fe	Cr	Ni	C	Si	Mn	P	Mo	N	O	Co	Cu
at.%	62.02	21.84	11.57	0.08	1.26	1.85	0.06	0.35	0.31	0.09	0.25	0.23

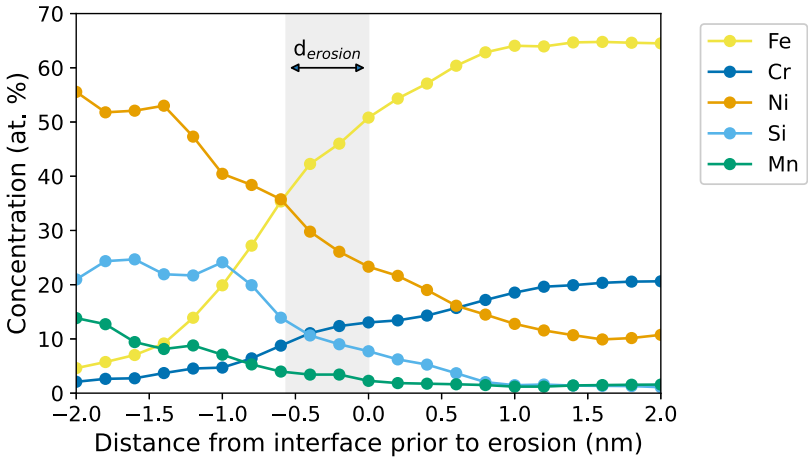


Fig. 1. A proximity histogram (proxigram) showing Fe, Cr, Ni, Si, and Mn concentrations for positions relative to the interface of an identified cluster. The portion of the cluster subject to erosion is shaded in gray.

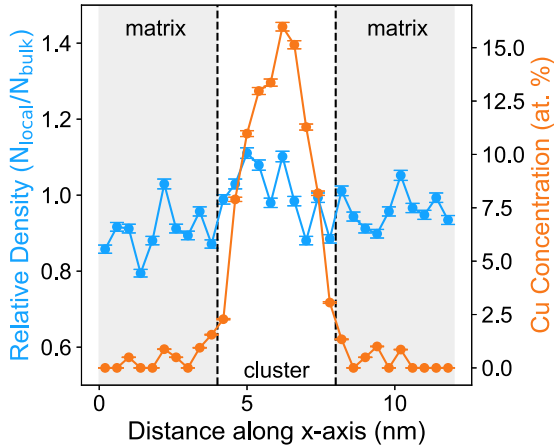


Fig. 2. The relative atomic density (left axis) and concentration of Cu (right axis) measured along the x-direction near a Cu-rich cluster. The dotted lines represent the bounds of the cluster.

2. Experimental Design, Materials and Methods

Full details on the examined 304L stainless steel and neutron irradiation conditions can be found in [4]. APT needles were fabricated at the Low Activation Materials Development and Analysis (LAMDA) laboratory at Oak Ridge National Laboratory (ORNL) using FIB milling. Initial cross-sectional lift-outs were made on a FEI Quanta DualBeam FIB using a 30 keV Ga ion beam. Needles were then sharpened using a FEI Versa3D FIB. To avoid milling induced artifacts, the energy and current of the Ga beam were progressively reduced during sharpening, with final tip polishing performed using 2 keV Ga ions.

Atom probe experiments were conducted on a Cameca LEAP 5000X HR at the Michigan Center for Materials Characterization (MC²). The run parameters for experiments consisted of voltage acquisition mode, 50 K temperature, 20 % pulse fraction, 200 kHz pulse rate, and 0.50 % detection rate. Volumes were reconstructed using Cameca's IVAS software with an electric field factor of 4.5 and an image compression factor of 1.65.

Both Cu- and Ni-Mn-Si-rich clusters were identified and analyzed in the given APT data set using OSCAR. OSCAR is an iso-position algorithm written in Python. The code can be run interactively using the “OSCAR Iso-Position.ipynb” Jupyter notebook in [4]. For successful operation, the apt_pkg folder must be saved to the same directory as the Jupyter notebook. Besides .pos and .rng files from IVAS, required user inputs are run number (run), the solute species for cluster identification (solute), bulk atomic density (bulkAtDens), detector efficiency (q), local concentration threshold (thres_conc), linking distance (dlink), minimum number of solute atoms allowed in a cluster (Nmin), ranged elements (elements_list), and whether to use the sphericity filter during analysis (separate).

The basic steps for cluster identification in the OSCAR program are as follows. First, the local solute concentration within a fixed radius around each atom is calculated. In the sample outputs provided, the radius was selected to ensure ~ 100 atoms were contained in each sphere. Next, atoms with a local solute concentration below a user defined threshold (thres_conc) are filtered out. The remaining atoms, which we shall call core atoms, are then formed into clusters based on a user defined linking distance (dlink). If the distance between two core atoms is less than or equal to the linking distance, those core atoms are merged into one cluster. After a cluster assignment is given to each core atom, clusters with less than a user defined number of solute atoms (Nmin) are excluded from further analysis.

To refine the size of each cluster, an erosion technique is used. As the width of the interface for each cluster may vary, a proxigram is first measured for each cluster. A proxigram is generated by calculating the composition of each element in shells within and surrounding the cluster. A minimum of 25 atoms is required in each shell so that percent error does not exceed 20%. A four parameter logistic function is then fitted to the profile of a user-defined element. For the analysis of Cu- and Ni-Si-Mn-rich clusters, we used the Fe profile to determine erosion distance. This is because Fe is the major element in 304L stainless steel, and therefore its profile has the lowest percent error. The erosion distance is taken as the absolute value of the logistic function x-midpoint (the x-value producing a response halfway between the minimum and maximum asymptote).

The core composition of each cluster is simultaneously calculated in OSCAR when the proxigram is measured during cluster erosion. The core composition is recorded as the asymptote of the logistic curve fit for each element. The minimum asymptote is used if the slope factor of the logistic curve fit is positive, while the maximum asymptote is used if the slope factor is negative. If a logistic curve cannot be fit to an element's profile, the mean value of the profile is recorded as the element's core concentration. It should be noted that cluster core compositions reported by OSCAR do not account for isotopic peak overlaps.

To analyze Ni-Si-Mn-rich clusters, the sphericity filter in OSCAR was used. The filter calculates for each cluster a sphericity parameter, ψ , defined here as R_{eq}/R_G . R_{eq} is the equivalent spherical radius of a cluster given by equation 1 where n is the number of atoms detected in the cluster, Ω is the atomic volume within the cluster, and Q is the detector efficiency.

$$R_{eq} = \sqrt[3]{\frac{3n\Omega}{4\pi Q}} \quad (1)$$

By contrast, a cluster's Guinier radius, R_G , is calculated with equation 2 based on the actual position of atoms within the cluster without assuming a geometry. Here m_j is the mass of atom j , r_j is the distance of atom j from the cluster's center of mass, and M is the total mass of the cluster.

$$R_G = \sqrt{\frac{5(m_1 r_1^2 + m_2 r_2^2 + \dots + m_j r_j^2)}{3M}} \quad (2)$$

For the data herein, equivalent spherical radius was calculated using a detector efficiency (q) of 0.52 and assuming a cluster atomic volume (pptatvol) of $1.2 \times 10^{-29} \text{ m}^3/\text{at}$, which is the atomic volume for G-phase ($M_6\text{Ni}_{16}\text{Si}_7$ with $M = \text{Ti, Nb, Mn, Fe and/or Cr}$). A user defined tolerance (tol) determines the minimum value required to pass the sphericity filter. For the

data herein, a tolerance of 0.25 was used and so the minimum passing value was 0.75. It should be noted that for a perfectly spherical cluster, ψ is expected to be unity.

After Cu-rich clusters were identified by OSCAR, clusters were examined for evidence of trajectory aberrations. This analysis was done by measuring the relative atomic density across clusters in the x-direction, similar to as in [2,3]. Here, relative atomic density is the ratio of the locally measured atomic density, N_{local} , to the measured bulk atomic density, N_{bulk} . When trajectory aberrations in APT data are significant, the relative atomic density spanning a cluster in the x-direction will deviate from unity. Local atomic density measurements were made for a series of rectangular volume slices. The thickness of each slice was 0.4 nm, and the y and z bounds for the volume were the same as the minimum and maximum y and z values bounding the cluster.

Ethics Statement

No ethical issues are associated with this work.

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability

[Data on Cu- and Ni-Si-Mn-rich solute clustering in a neutron irradiated austenitic stainless steel \(Original data\)](#) (Mendeley Data).

CRediT Author Statement

Samara M. Levine: Conceptualization, Software, Writing – review & editing, Visualization, Investigation; **Cristelle Pareige:** Conceptualization, Methodology; **Zhijie Jiao:** Conceptualization, Investigation, Funding acquisition; **Philip D. Edmondson:** Conceptualization, Supervision; **Gary S. Was:** Conceptualization, Supervision, Project administration; **Steven J. Zinkle:** Conceptualization, Supervision, Writing – review & editing; **Arunodaya Bhattacharya:** Conceptualization, Resources, Supervision, Project administration, Writing – review & editing.

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