Deep learning-enabled probing of irradiation-induced defects in time-series micrographs

Kory Burns; Kayvon Tadj; Tarun Allaparti; Liliana Arias; Nan Li; Assel Aitkaliyeva; Amit Misra; Mary C. Scott; Khalid Hattar

APL Mach. Learn. 2, 016117 (2024)
https://doi.org/10.1063/5.0186046
Deep learning-enabled probing of irradiation-induced defects in time-series micrographs

Kory Burns,1,a) Kayvon Tadj,3 Tarun Allaparti,2,3 Liliana Arias,3,4 Nan Li,5 Assel Aitkaliyeva,6 Amit Misra,7 Mary C. Scott2,8,9 and Khalid Hattar10,a)

AFFILIATIONS
1Department of Materials Science and Engineering, University of Virginia, Charlottesville, Virginia 22904, USA
2Department of Materials Science and Engineering, University of California-Berkeley, Berkeley, California 94720, USA
3Sandia National Laboratories, P.O. Box 5800, Albuquerque, New Mexico 87185, USA
4Department of Nuclear Science and Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA
5Center for Integrated Nanotechnologies, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA
6Department of Materials Science and Engineering, University of Florida, Gainesville, Florida 32611, USA
7Department of Materials Science and Engineering, Department of Mechanical Engineering, Ann Arbor, Michigan 48109, USA
8Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA
9National Center for Electron Microscopy, Molecular Foundry, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA
10Department of Nuclear Engineering, University of Tennessee-Knoxville, Knoxville, Tennessee 37901, USA

a)Authors to whom correspondence should be addressed: koryburns@virginia.edu and khattar@utk.edu

ABSTRACT
Modeling time-series data with convolutional neural networks (CNNs) requires building a model to learn in batches as opposed to training sequentially. Coupling CNNs with in situ or operando techniques opens the possibility of accurately segmenting dynamic reactions and mass transport phenomena to understand how materials behave under the conditions in which they are used. In this article, in situ ion irradiation transmission electron microscopy (TEM) images are used as inputs into the CNN to assess the defect generation rate, defect cluster density, and saturation of defects. We then use the output segmentation maps to correlate with conventional TEM micrographs to assess the model’s ability to detail nanoscale interactions. Next, we discuss the implications of preprocessing and hyperparameters on model variability, accuracy when expanded to other datasets, and the role of regularization when controlling model variance. Ultimately, we eliminate human bias when extrapolating physical metrics, speed up analysis time, decouple reactions that happen at 100 ms intervals, and deploy models that are both accurate and transferable to similar experiments.

© 2024 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/). https://doi.org/10.1063/5.0186046

I. INTRODUCTION
In situ transmission electron microscopy (TEM) is a powerful technique that allows us to observe high resolution phenomena at the nanoscale while simultaneously analyzing the dynamics of an undergoing process.1–4 Accordingly, with the developments in in situ TEM over the past few years and the dynamic frame rates of modern cameras,5,6 experimentalists often have hundreds, sometimes thousands, of micrographs to process to understand an individual physical process at the appropriate time scale. Automated analysis tools available now should not only increase efficiency but also decouple subtle interactions that humans miss during data
analysis. However, there is an added layer of complexity when trying to model microstructural changes occurring during radiation damage to structural materials.

In radiation materials science, \textit{in situ} TEM is used as a unique tool to analyze complex defect structures produced in response to extreme environments. Of practical importance, materials that can survive high doses of radiation are needed as we explore the dimensional stability of Generation IV reactor materials. Fundamental studies encompass the impact high energy particles have on the microstructure of a variety of materials, citing the interaction of point defects leading to dislocation networks, vacancy clusters, interstitial clusters, and inert gas bubbles. By analyzing the number of defects and their size and shape, we gain insight into the irradiation doses that begin to fundamentally alter materials’ properties and cause long timescale performance degradation, often limiting the performance of engineering systems. However, microstructural damage varies widely in different materials, even changing with the number of preexisting defects prior to irradiation. Accordingly, a large framework of data and experimentation is needed to explore as many enumerations as possible to design radiation tolerant materials for advanced nuclear energy technology.

In response, the nuclear materials community has made valiant efforts to automate defect characterization and extract the most information from large datasets. A faster regional-based convolutional neural network (R–CNN) was previously used to identify information from large datasets. A faster regional-based convolutional neural network (R–CNN) was previously used to identify defects from the images, and post-processing is used on the output segmentation maps. The multi-step process here was designed to recognize salient features of an image with a higher degree of accuracy. The input data then goes through linear layers with \textasciitilde \textasciitilde reduced by a factor of 2, making it easier for UNet++ layers to handle. A patch-based learning strategy is then implemented to recognize salient features of an image with a higher degree of accuracy. The input data then goes through linear stretching to reduce the intensity ratio between the defects and the background, making it seamless to filter out artifacts or unwanted characteristics from the image, as shown in Fig. S3. Finally, an 80/20 random split between the training/test set was implemented.

II. MATERIALS AND METHODS

A. Experimental methods

Experiments were performed using \textit{in situ} ion irradiation at a 200-kV JEOL-JEM 2100 TEM. A TVIPS 1k camera enabled \textit{in situ} imaging, capturing 5 fps. A 6 MV EN tandem ion accelerator was used to generate a 3 MeV Cu\textsuperscript{6+} broad beam. The stage was tilted 30° toward the beamline to form a 60° angle between the ion beam and the sample normal so that the sample faced the ion beam. The temporally uniform but spatially graded Cu beam spot size current was measured using a Faraday cup located upstream of the TEM on the beamline. For further details on the sample preparation, the method used to form a two-beam image, and calculating the thickness of the TEM foil, refer to our previous work by Li \textit{et al.}\textsuperscript{23} Accordingly, to further diversify the training set, we include an additional \textit{in situ} dataset from a self-ion irradiation of Au, with a similar snapshot shown in Fig. S2(a). Additionally, the hand-labeled masks for the Au “still” frames are displayed in Fig. S2(b). Details on the Au irradiation can be found in our previous work.\textsuperscript{24} There were two separate sequences of videos collected in this experiment to collect the data used for the Cu irradiation and the Au irradiation, respectively.

B. Computational methods

1. Acquisition of dataset and labeling

Over 200 000 defects were hand-labeled to establish the ground-truth data. \textit{In situ} observation yielded insight into the defect generation rate and interaction of defects. All defects in the micrographs were hand-labeled pixel-by-pixel by domain experts to establish the ground-truth data. In Fig. S1(a), we give a snapshot of the training dataset for the Cu foils, which is an extrapolation of “still” frames from the video captured in Movie S1. In Fig. S1(b), the corresponding hand-labeled masks are shown to display a comprehensive labeling process from the still frames. This \textit{in situ} dataset from the Cu foils had similar microstructures throughout the experiment, which increases the probability of overfitting the model during training.

2. Image analysis process flow

Extrapolating features from images requires an end-to-end process from image filtering prior to training to extracting features from the output segmentation maps. The multi-step process here was built using a combination of previously studied steps,\textsuperscript{25} with some new processes integrated. Figure 1 provides a synopsis of this process, where we break each section down into the following subparts:

1. Data normalization: The input image is analogous to the “still” frame introduced in Fig. 1(a). The size of the image is then reduced by a factor of 2, making it easier for UNet++ layers to handle. A patch-based learning strategy is then implemented to recognize salient features of an image with a higher degree of accuracy. The input data then goes through linear stretching to reduce the intensity ratio between the defects and the background, making it seamless to filter out artifacts or unwanted characteristics from the image, as shown in Fig. S3. Finally, an 80/20 random split between the training/test set was implemented.
FIG. 1. Diagram of the workflow dedicated to the detection and segmentation of defects in Cu metal during in situ ion irradiation of TEM images. The full analysis of the workflow is described in four sections: (1) Data Normalization: Every input image is rescaled and reduced from a $1024 \times 1024$ to a $512 \times 512$ pixels image and split into patches of the same size. (2) UNet++ training: Every patch corresponding to the images in the dataset from (1) is transformed to augment the training size, and UNet++ is trained. (3) Probability map post-processing: All the patches in the test set are processed with the trained UNet++. A probability map is obtained after the reconstruction of the patches. A binary mask is obtained on the trained image by thresholding the probability map. Finally, the mask is processed using a transform, binary closing, and filtering of small objects to facilitate the separation of touching and overlapping defects to label unique identifiers for each object. (4) Feature Extraction: contour detection is employed to detect the borders of defects, and then the images are carefully calibrated to measure size and shape features based on contour detection.

2. UNet++ training: 80% of the entire dataset analyzed was implemented into the training set. To increase the number of analyzed micrographs and add complexity to the dataset, the micrographs were augmented with three modulations. They were rotated in iterations of $90^\circ$, zoomed in at five different magnifications, and introduced with noise through a low-filter Gaussian blur. Visual representations of the augmentation are found in Fig. S4. Next, training and passing of the images through the UNet++ architecture were performed.

3. Probability map post-processing: The probability maps of the patches are a segmentation of the features of the patches generated in Sec. I. A binary classification was adopted to yield absolute probability values, where each pixel in the image was given a value based on the intensity of the original image.
Pixels below a particular threshold were given a value of 0 (copper metal), and pixels above a certain threshold were given a value of 1 (defect clusters). The threshold value (set to 0) is used to classify the pixel values. The maximum value (255) is assigned to pixel values exceeding the threshold. We then used Otsu’s binary threshold since this approach determines the optimal global thresholding values for the user based on the intensity distribution in the image histogram. The complete image was then reconstructed by stitching the patches of the threshold back together. The pixel values of 1 were then given a label, thereby giving every defect cluster a unique label in the returned array. Finally, we get the full output segmentation map, which can distinguish between overlapping defects, touching defects, and defects that coalesced.

4. Feature extraction: To detect the contours around the defects, a loop in the range of contours is run and iterated through. An outline of the shape was then drawn to determine the centroid of the boundary. The shape is then detected based on the number of contour points within the outline, the detected shape is characterized, and the center point is given a shape. Finally, we can extract the size and shape metrics from each of the labels (defects) from the output segmentation map.

This sequence of steps was used for all the frames in the dataset for consistency in analysis. Note that some steps (i.e., data augmentation in the section of UNet++ training as opposed to Data Normalization) could be applied to a different section; however, emphasis is placed on the sequence in which each process is performed.

III. RESULTS
A. Model performance

Model performance was based on metrics we can quantify by calculating the intersection over union (IoU) and computing the Dice score. First, we provide a comparative visual of the input (raw) images, the ground-truth dataset, and the predicted images from the output of the process flow, as shown in Fig. 2(a). Accordingly, we take two samples from the training set and two from the test set and calculate the defect count from the ground truth and predicted dataset, displayed as a bar plot in Fig. 2(b). The ground truth dataset for Cu irradiation (Training Set 1) and Au irradiation (Training Set 2) had a defect count of 143 and 23, respectively. The corresponding predicted masks had a defect count of 144 and 24 defects, respectively. It should be noted that both experiments characterized defects produced during self-ion irradiation experiments. However, to determine the robustness of this approach to any defect observed in a TEM, we applied this algorithm to a dataset of implanted He bubbles in Pd metal to see if the segmentation algorithm was transferable to a defect class that has an intensity gradient across its domain. The He was implanted into a thin Pd slab at $1 \times 10^{17}$ cm$^{-2}$ and thermally annealed at 400, 450, 500, and 550 °C, respectively. For this, the ground truth had a defect count of 317 defects, while the predicted had 241 defects. The large variance is a good
B. Quantifying defect interactions

As mentioned previously, one of the main benefits of in situ TEM is the ability to perform real-time experiments while analyzing local regions of a material under an external stimulus and simultaneously probing the dynamics of a reaction. In Fig. 3, we highlight the defect interactions commonly observed in our experiments by indicating regions of interest with arrows. First, defect generation is shown in Fig. 3(a), as impinging ions generate vacancies at the site of impact. The energy of the ions is sufficiently high to overcome the threshold energy of Cu atoms in the target material.10 Next in Fig. 3(b) is defect coalescence, in which we discuss the interplay between dislocations and stacking fault tetrahedra (SFT). These defects interact aggressively with each other at high strain rates,12 which we impart on the Cu slab from prolonged exposure to the ion beam stimulus. Third is defect fragmentation in Fig. 3(c), which occurs when an impinging ion interacts with a preexisting defect in the target material. Fragmentation rates increase with the number of dislocations in a close vicinity,13 so their occurrence is expected to increase with fluence.

In Fig. 3(d), defect annihilation is highlighted, which depends strongly on the separation between dislocations. In general, there is more free energy for dislocations in a closer vicinity to each other,14 making these dislocations easier to annihilate in the presence of impinging ions. A defect is annihilated within 0.4 s, which is quantitatively visualized from the intensity decay of the cluster [visualized in the line profile in Fig. 3(e)]. It should be noted that defect clusters leave debris behind after annihilation because the annihilation is triggered by a dipole transformation; additionally, dislocations rarely have a fixed orientation. The center of mass remains stationary relative to the surrounding defects, as shown in the line profile, and the intensity variance reduces in contrast relative to the Cu grain. Accordingly, even with the small debris left behind, the model does not pick up the defect after it is annihilated, as the contrast is closer to that of the Cu grain than that of the defect. The fluences in Fig. 3(f) were selected based on the inflection points indicated by the red X annotations in the plot, which are used to describe the stages of defect evolution in Cu.23,35 Here, when the density of defects is proportional to the irradiation dose (i.e., n = 1, where one impinging ion generates 1 defect), the defects are displaced directly in the displacement cascades. The incubation zone represents the onset of Cu being displaced from equilibrium with a slope greater than 1. As the fluence increases from prolonged exposure to the ion beam, the slope decreases, indicating the interaction of adjoining defects.

Figure 4(a) provides a visual of how defects are tracked using the OpenCV object tracking software. A tile stack of output segmentation maps was converted to a NumPy array in their respective time series. Each defect was given a unique label based on its (x, y) coordinate list in the image and its occurrence in time. Accordingly, this allows one to visualize how a particular defect changes its displacement through irradiation. Figure 4(b) provides a breakdown of the count of defects present pre-irradiation and arising from irradiation. The defect clusters that were present pre-irradiation decrease with increasing fluence, while the irradiation-induced defects oscillate between increasing and decreasing in density with increasing fluence. We attribute this to a combination of the defect interaction events discussed in Fig. 3 since the ions impinging on the Cu slab have a steady dose, and without interactions, a progressive increase in the number of defects would be observed. Figure 4(c) displays the TEM images and corresponding masks that complement Fig. 4(b) and the annotation in Fig. 3(f). The preexisting defects are shown in black, while the irradiation-induced defects are displayed in gold. Irradiation-induced defects accumulate aggressively both within the Cu grain and along the grain boundary.

C. Correlative data analysis

In Figs. 5(a)–5(c), a density plot of the defect density, eccentricity, and area are given relative to each metric for the irradiation of Cu. The plots represent the mean value of each variable across the entire experiment. Each plot provides a distribution of a single variable vs the relationship of two variables in a dataset and is used here as an effective method to identify trends between independent variables. Plots 5a-c have the same density spread throughout the

<p>| Table I. Important hyperparameters involved in the selection process during hyperparameter optimization. |</p>
<table>
<thead>
<tr>
<th>Optimizer</th>
<th>Loss</th>
<th>Learning rate, Lr (min Lr)</th>
<th>Scheduler</th>
<th>Weight decay</th>
</tr>
</thead>
<tbody>
<tr>
<td>Adam</td>
<td>BCEDiceLoss 0.001 (0.000 01)</td>
<td>CosineAnnealingLR</td>
<td>0.0001</td>
<td></td>
</tr>
</tbody>
</table>
FIG. 3. Defect interactions during in situ ion beam irradiation of Cu. (a) Defect generation displays how impinging ions create defects in Cu. (b) Defect coalescence of neighboring defects conjoining to form one. (c) Fragmentation of a larger defect to smaller ones. (d) Series steps of defect annihilation over a timeframe of 0.4 s. Colored lines represent where the line scan was taken to construct the plot in (e), where the defect of interest is annihilated. (f) Plot of fluence vs defect density, where red X annotations highlight inflection points in the graph.
FIG. 4. Tracking of defects through experimentation. (a) Process detailing how time series output segmentation masks are stacked, each defect identified, and movement tracked. (b) Histogram highlighting the proportion of fabrication-induced defects to irradiation-induced defects as a function of increasing fluence. (c) Images and corresponding masks complimenting histogram shown in (b) to display how fabrication vs irradiation induced defects progress with increasing fluence. Units for numerical values in (c) are in cm$^{-2}$.

In Fig. 5(d), the median, mean, and max area are shown throughout the experiment. The median value of the defect sizes remains the same, a good indication that irradiation-induced defects generated are the same size as fabrication-induced defects, while the slight fluctuations in size can be attributed to nanoscale interactions and defects interacting. In Fig. 5(e), a plot of the fluence vs the eccentricity is displayed. The eccentricity increases at the initial onset of the irradiation, indicating a distortion in the morphology of the defects becoming more elongated and a larger variance in the major and minor axis lengths. This onset occurs when the Faraday cup is removed to initiate the irradiation. There is a slight thermal shock on the metal that causes a small amount of stage drift. This creates false (ghosting) on the defects, making the size read higher than what is real. In both plots, the max and mean have an unusually higher value for the first data point, which we attribute to a false identifier at the onset of irradiation. The plot in Fig. 5(d) shows a poor correlation of the fluence vs area, as the size of the defects oscillates around a mean through the experiment. The plot in Fig. 5(e) is a plot of the fluence vs eccentricity, showing a steady increase during the first half of the experiment, followed by the values fluctuating around a mean. The outliers in Figs. 5(a)–5(e) are explained by fighting drift as a microscopist at the beginning of an experiment. This is when a small area and low defect density (and, therefore, low fluence) are present simultaneously. Figure S5 is included to show how the standard deviation of area and eccentricity both evolve over time. Notably, the standard deviations of the area assist in highlighting an outlier at the beginning, as the “ghosting” around the defects during drift will make the defects appear larger in a non-uniform manner.

Correlative methods of tying together in situ TEM and data science serve as a platform for rapid, accurate, precise, and non-biased identification of defects. We correlated the properties of defects based on output segmentation maps to understand the physics at multiple time scales by stitching back together micrographs in their temporal sequence. With this approach, it becomes possible to sift through massive amounts of time-series data and decouple the kinetics of a given reaction, only limited by the specifications of the electron microscope used. Uniquely, we emphasize a transferable approach by diversifying our micrographs through various in situ experiments and augmentation. We also show how many features of defects can be compared to each other to globally understand an irradiation response. However, we have also seen that one training regime is not universal to all types of defect characterization, so future work will be geared toward developing a customized process flow for the identification of He and Xe bubbles in similar materials systems.
IV. CONCLUSIONS

We have developed an end-to-end process that allows one to plug in time-series TEM images and extrapolate feature characteristics to probe the physics of any nanoscale reaction that can be captured within the capabilities of the camera frame rate and the resolution of the microscope. To date, we have assembled the largest labeled in situ ion irradiation dataset in two-beam (diffraction contrast) conditions, which includes over 1000 images and over 250,000 defects, making for one of the most comprehensive data-driven studies in this field. Notably, we found that

i. hyperparameter optimization, including multiple experiments in the training set and increasing variability in the input, was paramount for preventing the overfitting of the model,

ii. defect classification does not scale 1:1 from dislocation loops/SFTs to helium bubbles. The primary driving force behind this issue is the opposite contrast produced by the electron beam between the intrinsic defects in Cu and Au and the extrinsic defects in Pd. Accordingly, a separate model, or a vastly expanded model with thousands of micrographs with a high density of He bubbles in the training set, would have to be trained for an algorithm to classify broader datasets in radiation materials science,

iii. in situ datasets can often battle drift when the onset of an external stimulus is introduced to the materials. The noise aberrations can create “false” metrics in the images and generate inaccuracies in the data. It is important to selectively go through a dataset and understand where this happens (as seen in this work) or develop an approach that removes drift-related noise (i.e., a denoising autoencoder), and,

iv. the end-to-end process developed in this work was suitable for characterizing irradiation-induced defects in face centered cubic metals such as Cu and Pd, with rapid analysis and good accuracy.

Last, this approach can be adopted to quantify the characteristics of defects in similar materials’ systems and understand the dynamics of processes occurring at multiple time scales. Further work is needed to separate dislocation loops and SFTs in the micrographs.

SUPPLEMENTARY MATERIAL

See the supplementary material for additional details: the Cu dataset and the hand labeled masks (S1); the Au dataset and the hand labeled masks (S2); unsupervised image segmentation on regions
of interest (S3); data augmentation (S4); and the effect of the LIF neuron’s threshold (S5).

ACKNOWLEDGMENTS

This work was primarily funded by the US Department of Energy in the program “4D Camera Distillery: From Massive Electron Microscopy Scattering Data to Useful Information with AI/ML.” This work was performed, in part, at the Center for Integrated Nanotechnologies, an Office of Science User Facility operated for the U.S. Department of Energy (DOE) Office of Science. Sandia National Laboratories is a multimillion laboratory managed and operated by National Technology & Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International, Inc., for the U.S. DOE’s National Nuclear Security Administration under contract DE-NA-0003525. The views expressed in the article do not necessarily represent the views of the U.S. DOE or the United States Government. Los Alamos National Laboratory, an affirmative action equal opportunity employer, is managed by Triad National Security, LLC, for the U.S. Department of Energy’s NNSA under Contract No. 89233218CNA000001.

AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Kory Burns: Conceptualization (equal); Data curation (equal); Formal analysis (equal); Investigation (equal); Methodology (equal); Visualization (equal); Writing – original draft (equal); Writing – review & editing (equal). Kayvon Tadi: Conceptualization (supporting); Data curation (supporting); Investigation (supporting). Tarun Allaparti: Data curation (supporting). Liliana Arias: Data curation (supporting). Nan Li: Conceptualization (supporting); Investigation (supporting); Writing – review & editing (supporting). Assel Aitkaliyeva: Writing – review & editing (supporting). Amit Misra: Investigation (supporting); Project administration (supporting); Supervision (supporting); Writing – review & editing (supporting). Mary C. Scott: Funding acquisition (equal); Project administration (equal); Supervision (equal); Writing – review & editing (equal). Khalid Hattar: Formal analysis (equal); Funding acquisition (equal); Project administration (equal); Supervision (equal); Writing – review & editing (equal).

DATA AVAILABILITY

All the code supporting this work is available via GitHub. All images are available via a downloadable link on The Box.

REFERENCES


E. Lang, T. Clark, R. Schoell, K. Hattar, and D. P. Adams, “In situ investigation of ion irradiation-induced amorphization of (Ge0.5Sb0.5)2−xCex [0 ≤ x ≤ 0.12],” J. Appl. Phys. 133, 135302 (2023).


37. See https://github.com/KoryBurns/Defects-APL for codes used to reproduce the findings in this work.

38. See https://virginia.box.com/s/6yh13zvrlcip85kqoos2a8kndq7d2vk for curated data, raw data, ground truth labels, and our output results from this work.