Image-driven machine learning for microstructure characterization: A case study in the uranium-molybdenum system

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Abstract

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1. Introduction

No standard, general, and widely applicable method of quantifying microstructure image data currently exists in the field of materials science. While American Society for Testing and Materials (ASTM) standards exist for quantifying some microstructural features, for example average grain size measurement and nodule count in ductile iron, these standards are limited in terms of applicability to a wide range of material systems with varying features of interest. Existing standards are insufficient for scientists looking to develop more quantitative, less biased, and more generalizable microstructure-processing-properties relationships. More complex mi-

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crostructural features, such as impurity phase precipitates, inclusions, voids, dendrites, and lamellae are typically quantitatively described in a subjective manner, using methods specifically developed for that particular material system of interest (methods selected by expert, after expert identifies microstructural features of interest). Low-level pixel processing is commonly used (thresholding) using open-source software such as ImageJ. Image processing can be automated or semi-automated using tools such as the image processing toolbox in MATLAB or sci-kit image library in Python, however automating low-level pixel processing routine does not allow for these methods to be more broadly applicable, it simply makes image analysis more efficient. In order to reduce bias introduced into the materials characterization process (from subjective analysis of image data), more widely applicable methods of quantitatively describing microstructures must be used.

In this work, a deep neural network is developed for the task of quantifying image data for the specific characterization task of kinetic modeling in a case study on the uranium-molybdenum system.

Uranium-Molybdenum (U-Mo) alloys are under investigated as a candidate low enriched uranium (LEU) fuel system in order to reduce proliferation concerns associated with continued use of highly enriched uranium (HEU) fuel systems in research and radioisotope production facilities worldwide [1, 2, 3, 4, 5, 6, 7]. U-Mo is a promising LEU fuel system due to high U density achievable [], and acceptable fuel performance []. Both dispersion and monolithic designs have been previously studied. The monolithic design, detailed in References [8, 9], and is the focus of the work presented here. LEU is alloyed with 8-10 weight percent (wt%) Mo in order to stabilize the body centered cubic (BCC) gamma phase (γ-U) which has acceptable swelling and mechanical stability when subjected to neutron irradiation [8].

U-10Mo experiences a eutectoid reaction at approx. 555-565C where the γ-U matrix phase forms α-U and U2Mo(γ') as decomposition products: γ-U $\rightarrow$ α-U + U2Mo γ'. Other methods have been used to quantitatively analyze the eutectoid decomposition in U-Mo alloys during sub-eutectoid annealing, including: X-ray diffraction, differential scanning calorimetry. Image analysis is the only technique that can detect (and quantify) the extent of this phase transformation when it is less than 20% [10].

1.1. Related work

This work builds upon work previously reported in Reference [11], which concluded that convolutional neural networks used for feature extraction in

micrograph classification represented image data well. The work presented here uses CNN's for a higher-level task of kinetic modeling, versus the simpler task of image recognition (classification). The methods used in this work are well established in the computer vision and machine learning disciplines, and are gaining popularity for use in materials science and engineering applications.

1. DeCost and Holm - multi-class microstructure image classification using VBoW and SVM [12]
2. DeCost and Holm - characterization of powder micrograph images using VBoW, compared to segmentation-based particle size analysis [13]
3. DeCost and Holm - characterization of powder feedstock materials for metal additive manufacturing using SIFT-VLAD system [**?**]
4. DeCost and Holm - image segmentation using an off-the-shelf deep neural network architecture [14]

1.2. Contribution

This work contributes to both computational materials science and microstructure characterization fields. Here, we investigate the applicability of feature extraction (via deep learning methods, specifically neural networks) for quantitatively describing microstructure image data, in order to relate to kinetic parameters (k and n in the KJMA equation), which has not yet been explored. The methods and approach utilized in this work can be generalized and applied to kinetic modeling in a variety of other material systems, particularly where image data is heavily used for relating microstructure to processing parameters or material properties and performance.

2. Methods

2.1. Experimental Materials and Methods

Depleted U alloyed with 10 wt% Mo (referred to here as DU-10Mo) alloys were studied here in order to perform characterization tasks on U-Mo alloys with low radioactivity in comparison to prototypic fuel materials which contain between 19 and 20% ^{235}U.

The U-10Mo samples analyzed in this work were fabricated at the Y-12 National Security Complex at Oak Ridge National Laboratory by melting depleted U, adding Mo, and casting in graphite molds. After casting, samples were subjected to a homogenization anneal (900°C for 48 hours),

followed by a sub-eutectoid annealing treatment at 500°C. The time of this sub-eutectoid annealing ranged from 1 to 100 hours. A DU-10Mo alloy cast and homogenized was also studied analyzed here to understand the starting microstructure prior to sub-eutectoid annealing.

The homogenization annealing treatment was performed in an inert gas atmosphere (Argon) using a high-temperature furnace (MTI Model VBF-1200X). The argon gas flow rate was $11.7\times10^{-6}m^3/s$. The furnace was heated at a rate of 10°C per minute to 350°C, held at 350°C for 15 minutes, then temperature was increased to the homogenization annealing temperature (900°C) then held for 48 hours. A furnace cool was performed (furnace turned off), and argon gas was forced into the chamber to assist with sample cooling. The cooling rate from high temperature to 650°C was 25°C/minute, then from 500°C to 350°C, the cooling rate dropped to 3°C/minute [10**? ? ?**].

Sub-eutectoid annealing treatments were also performed in a furnace with an Argon atmosphere. The homogenized samples were placed in a furnace which was heated to 500°C at a rate of 7°C per minute. After the annealing treatment was completed for a given time step (1-100 hours), the furnace was turned off and the samples were cooled to room temperature at a rate of 3°C per minute [10].

It is noted here that although there are several other steps in the fuel fabrication process, the focus of this work was to investigate the impact of annealing time (at 500°C) on the formation of the α-U phase, the presence of which is known to be detrimental to fuel performance [].

All image data was obtained from samples prepared from sections of DU-10Mo that were mounted in epoxy and polished using standard metallographic techniques and equipment. In order to obtain an acceptable surface finish needed for imaging, samples were polished according to the procedure detailed in Reference [15].

2.2. Image Data

Image data used in this work was collected using the following two scanning electron microscopes (SEMs): a FEI Quanta dual beam Focused Ion Beam/Scanning Electron Microscope (FIB/SEM), and a JEOL JSM-7600F Field Emission SEM. The backscatter electron detector was used for improved atomic number (Z) contrast. Images were taken over a range of magnifications from 250x to 1,000x. The data set for training, testing, and validation was comprised of a XX original images for each time step at the

sub-eutectoid annealing temperature of 500°C. Images of microstructures produced after four different times (1, 10, 50, and 100 hours) were analyzed, in addition to one sample that was just cast and homogenzized, and thus corresponds to an annealing time of 0 hours.

2.3. Data Analysis Approach

2.4. Low-Level Pixel Processing

2.5. Data Generation

2.6. Machine Learning Model

2.7. Model Validation

2.8. Software Specifications and Selection of Model Parameters

All of our experimentation was carried out with Python version 2.7 with the help of various open-source libraries. The opencv, scipy, skimage, numpy, and sklearn packages (compatible with the Python version used in this work) were used for training, testing, and validation.

3. Results and Discussion

3.1. Results from Model Training and Testing

3.2. Kinetic Modeling

Suggested Figures for publication

1. Introduction: Series of micrographs showing phase transformation that occurs during sub-eutectoid annealing - **Liz**
2. Data Analysis Approach section: flowchart of approach **Liz**
3. Machine Learning Model section
   (a) Schematic of network architecture
   (b) Summary table of model parameters
4. Results Section
   (a) Table of - sample ID, annealing time at 500C, ground truth, results from segmentation using the neural net - **Liz, Mofii**
   (b) Example of how ground truth was determined (segmented image using multi-level thresholding in matlab) - **Liz**
   (c) Example of segmented images using neural net - **Mofii**
   (d) MSE and MAPE versus Epoch for training and testing - **Mofii**
   (e) Plot of quantification of microstructure versus time at 500C (S-curve) - **Liz, Mofii**
   (f) k,n parameters in KJMA equation - **Liz**

4. Limitations

Limitations associated with the data analysis approach presented here are generally related to reliance on domain knowledge and the amount of original image data available.

Image data labeling was performed using based on domain knowledge of SEM imaging and the U-10Mo material system. Images were labeled according to the sub-eutectoid annealing time at 500°C. Low-level pixel processing was performed in order to determine the ground truth of area fraction measurements used to compare to results from deep learning methods. Additionally, 2D image data is used in this work to quantify amount of phase transformation product, which is an inherently 3D object, thus 2D image data is an inherently limited representation of material microstructures. Obtaining 3D information on microstructures is possible via serial sectioning and data reconstruction methods, however these methods are much more time consuming in comparison to simpler imaging only. It is also of primary interest in this work to investigate microstructure quantification/characterization methods that can be applied to existing image data sets in order to leverage the wealth of image data in materials science communities. Thus, the data analysis approach presented here is limited to using area fraction as a proxy for volume fraction.

5. Conclusions

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Add acknowledgement for Aritra if he is not listed as author

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Contributions

D.J.L. and B.Y. conceptualized the data analysis approach presented here. E.J.K. and A. C. performed all data analysis. E.J.K. performed all analysis via low-level pixel processing, and contributed to analysis using a machine learning model originally developed by A.C. E.J.K led analysis of results and manuscript writing. V.J. coordinated the fabrication and metallographic preparation of UMo samples used in this work. A.D. contributed to discussions about results and its significance to phase transformations in metal alloys and specifically the uranium-molybdenum system studied here. All authors contributed to manuscript preparation.

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