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A graph-theoretic approach for characterization of precipitates from atom probe tomography data

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ABSTRACT

Atom Probe Tomography (APT) represents a revolutionary characterization tool that allows direct-space three-dimensional, atomic-scale resolution imaging along with the chemical identities of each detected atom. Quantitative analysis of APT data to perform characterization of precipitates in alloys gives clear insights into the structure–property relationships and helps in achieving the larger goal of materials-by-design. Most techniques currently used to extract precipitate topology and interface information from APT data are efficient; however, they are based on homogenization of the rich point cloud data which is inherently lossy. Furthermore, these methods require a specified, usually heuristic, concentration-level to draw iso-contours in order to extract characteristics of the precipitate topology. These twin issues of homogenization and heuristics are compelling rationale for the development of a robust, scalable, heuristic-free, graph-based framework, which we call **Gra**ph methods for **P**recipitate **Top**ology Characterization (GraPTop). This framework is motivated by the equivalence between a 3D point cloud data of atoms and an undirected, weighted, labeled graph. By considering the 3D point cloud data as an undirected, weighted, labeled graph, we leverage powerful graph-based algorithms to identify the local topology of precipitates without the necessity of any heuristics. Since GraPTop is based on nearly linear-complexity graph-algorithms, it is scalable to extremely large datasets. Furthermore, the performance of this framework is insensitive to the complexity of the geometry or the number of the precipitates in the point cloud data. We showcase this framework by analyzing several regions of interest in a point cloud Al–Mg–Sc (Aluminium–Magnesium–Scandium) specimen APT data and extract several interesting measures describing the precipitate topology like area, volume, and nonconvexity.

1. Introduction

Atom Probe Tomography (APT) [18,16] represents a revolutionary characterization tool for material scientists by providing direct-space three-dimensional, atomic-scale resolution with chemical identities of all the detected atoms. It involves controlled removal of atoms from a specimen's surface by field evaporation and then sequentially imaging and analyzing them with a TOF (Time of Flight) mass spectrometer. This technique currently provides the highest spatial resolution of any microanalysis technique. This capability provides a unique opportunity to experimentally study – with atomic resolution – chemical clustering and 3-D distributions of atoms, and directly test and refine atomic and molecular based modeling studies. While APT is a powerful technique with the capacity to gather information containing hundreds of millions of atoms from a single specimen, the ability to effectively use this information creates significant challenges. The main technological bottleneck lies in handling the extraordinarily large amounts of data in a reasonable amount of time [20]. This imposes a constraint for any quantitative technique to analyze the data to be both scalable and efficient.

One key material-science problem that can be analyzed using the APT is the characterization of precipitates in multi-component systems [20]. Of particular interest is the analysis and classification of precipitate topology, shape, size distributions as well as their interfacial properties [12,22,23,25,13,24]. Addressing this problem can give clear insights into structure–property relationships (especially in the context of energy storage devices) and, thus, help in achieving a larger goal of accelerated materials-by-design [33]. Recent work in detecting nano-scale bio-geo-chemical interfaces [21] also show the increasing relevance and applicability of atom probe tomography in fields outside materials science.

There exist several contemporary techniques for analyzing precipitates and clusters in 3D atom probe data. These chemical clusters are normally defined by calculating the concentration of

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different elements in the sample across a chosen bin size [8,30], through the use of nearest neighbor approaches [27,7], and cluster finding approaches [15,28,2]. Specifically, some of the approaches for precipitate analysis include: (i) proximity histograms, (ii) Fourier analysis, and (iii) friends-of-friends analysis. While chemical analysis of a chosen bin size through proximity histograms is a convenient technique for linking spatial features with chemistry and has shown good results [5,19], the definition of the region analyzed is mathematically arbitrary. The region is defined by a user-defined chemical threshold value, and therefore a precipitate is defined largely through assumption. Additionally, the concentration is calculated as an averaging of the voxels comprising the region, thereby reducing the information resolution. Fourier analysis of APT data has previously been performed for analyzing regions of interest with the objective of identifying crystallographic structure [29,31]. Among the outputs of this approach are mean precipitate size, shape and composition. While this approach is well suited for crystallographic analysis, the extensive computer memory requirements and limited resolution away from the poles prevents the ability to define the detailed shape and interface of precipitates [6]. Finally, a friends-of-friends approach for analyzing phases has shown promise [10,17]. This approach is based on the finding solute atoms that are nearer to each other in the solute phase as opposed to the matrix. However, this approach is inefficient for high solute concentrations, and also has been found to falsely identify clusters due to bridging effects and insensitivities of parameters [26].

The standard approaches for defining precipitates, such as through proximity histograms or cluster analyses, result in precipitates defined with largely convex surfaces. For example, defining precipitate regions based on atomic clustering requires inputting a parameter defining the maximum distance allowed between atoms within the same cluster and an additional envelope parameter which effectively serves as defining the convex hull of the precipitate. However, even a small number of atoms within the cutoff distance can extend the envelope region to incorporate a region of limited solute concentration. By defining convex volumes, either regions of low solute concentration are included in the defined precipitate, or conversely high solute regions are omitted. The ability to define a non-convex surface for precipitates is necessary because the morphology of a cluster within this convex hull can be defined only by capturing the non-convex surfaces of these precipitates. Parameters traditionally measured from convex surfaces include volume and surface area, while non-convex parameters capture information including degree of kinetic coagulation. For example, clusters that are at initial stages of kinetic coagulation will have large non-convexity.

Most current approaches to performing precipitate analysis on APT point cloud data are either based on homogenization, or are dependent on heuristics to characterize the precipitates. These issues motivate the development of an efficient and heuristic-free method for performing the characterization of precipitates that can directly work with the point cloud (APT) data without homogenizing it. In this work, we detail a method of performing cluster selection and surface construction using a graph-based formalism that is heuristic-free, works directly with the point cloud data without homogenization into concentration fields, is very scalable to analyze very large data-sets, and is applicable to a wide range of chemistries, environments, and geometries. We call this framework, **Gra**ph based methods for **P**recipitate **Top**ology Characterization (GraPTop). The following observations motivated our choice of a graph-based approach:

- The APT point-cloud data of atomic positions and their chemistry can be equivalently represented using an undirected, weighted, labeled graph. Each atom becomes a graph vertex with a label denoting its chemistry. Each vertex is connected to its neighboring vertices through edges whose weight is proportional to the distance between the vertices (atoms). Fig. 1 shows a simple example of this concept.
- Most precipitate characterization properties (like size, shape, number of atoms, bounding shape, etc.) can be naturally recast as estimating properties of the equivalent graph.

Furthermore, a graph-based formalism is ideally suited for large-scale APT datasets, particularly due to the fact that:

(1) Graph based methods are well-studied and have fast and efficient algorithms – for computing neighborhood and distance information – that are important for precipitate characterization. Furthermore, a graph approach directly works on the point cloud data without homogenizing it.
(2) Graph based methods are easily scalable and, hence, can be easily extended to larger problem sizes. Given the fact that APT deals with atomic scales, even for a moderately dense material specimen a minute increase in the dimensions of the region of interest can cause an exponential increase in the size of the dataset. In such cases, scalability of the method becomes a critical factor in qualifying the applicability of a technique.
(3) A graph-based approach is generic. That is, by making modifications to the definitions of parameters like edges, weights, and labels, different problems relating to the physical process can be solved. For example, while we focus on extracting precipitate shapes in this work, by replacing the Euclidean distance with radial distance (as weight definition) one can study the radial distribution of atoms in a precipitate. We have recently used such analysis to characterize the morphology of thin film organic photovoltaic [33,32].

The outline of this paper is as follows: Section 2 gives an overview of the framework. Section 3 details the methodology and

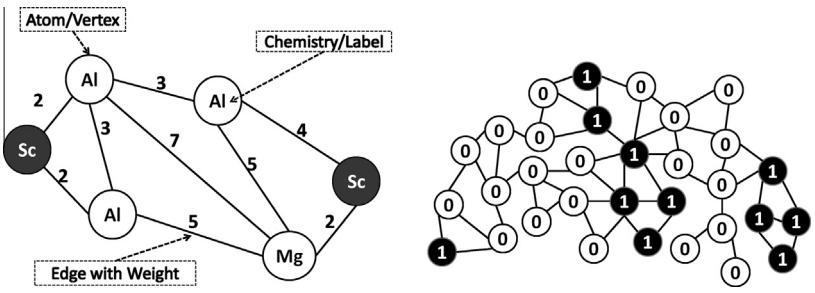

Fig. 1. (Left) Simple example of an Al–Mg–Sc alloy illustrating the equivalence between a graph and point cloud data. (Right) A larger example where the precipitate is labeled black and the solvent is labeled white.