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Simulation of heterogeneous atom probe tip shapes evolution during field evaporation using a level set method and different evaporation models

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ABSTRACT

In atom probe tomography (APT), accurate reconstruction of the spatial positions of field evaporated ions from measured detector patterns depends upon a correct understanding of the dynamic tip shape evolution and evaporation laws of component atoms. Artifacts in APT reconstructions of heterogeneous materials can be attributed to the assumption of homogeneous evaporation of all the elements in the material in addition to the assumption of a steady state hemispherical dynamic tip shape evolution. A level set method-based specimen shape evolution model is developed in this study to simulate the evaporation of synthetic layered-structured APT tips. The simulation results of the shape evolution by the level set model qualitatively agree with the finite element method and the literature data using the finite difference method. The asymmetric evolving shape predicted by the level set model demonstrates the complex evaporation behavior of heterogeneous tip and the interface curvature can potentially lead to the artifacts in the APT reconstruction of such materials. Compared with other APT simulation methods, the new method provides smoother interface representation with the aid of the intrinsic sub-grid accuracy. Two evaporation models (linear and exponential evaporation laws) are implemented in the level set simulations and the effect of evaporation laws on the tip shape evolution is also presented.

1. Introduction

Atom probe tomography (APT) is widely used for nanoscale 3D compositional characterization of a variety of materials ranging from metals and alloys [1–7], dielectrics [8,9], semiconductors [10,11] to biological [12,13] and geological materials [14]. The past decade has seen a dramatic development of APT instrumentation/data collection and sample preparation methods [15]. On the data collection front, local electrode and laser pulsing have been introduced widening the application of APT beyond conductive materials. On the sample preparation front, focused ion beam (FIB) based site-specific sample preparation methods have been introduced [16–18] permitting a better control of region selection for APT analysis as well as facilitating APT specimen preparation of nonconductive materials {Thompson, 2007 #44}. However a new challenge surfaces when the material systems analyzed by APT expand from the homogeneous metallic system to heterogeneous materials: the traditional APT data reconstruction protocol introduces artifacts due to the underlying assumption of uniform and isotropic evaporation.

The APT data reconstruction process is influenced by two distinct physical events that occur during APT analysis: field evaporation of the ions from the surface of an APT sample and projection of these evaporated ions to a position sensitive detector. This work will focus on the first step (evaporation process) which is a complex function of atom properties, atomic environment and emitter geometry. The traditional APT reconstruction algorithm for generating the 3D atomic position from the detector positions of ions is limited by the assumptions of homogeneous structure, uniform evaporation rate for different components and specific tip geometry [19]. The currently used reconstruction algorithm is modified from the method proposed by Bas et al. [19], and it is based on the assumption that the tip maintains a constant hemispherical shape during evaporation [20]. The apparent field strength F at the apex

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is determined by an approximation solution [21]:

$$F = \frac{U}{k \cdot r} \quad (1)$$

where U is the applied voltage, r is the radius of curvature and the empirical parameter k is the geometric factor/field factor accounting for the influence of the shaft and the geometry of the surrounding vacuum chamber [22]. During APT analysis to keep the ion detection rate constant, the applied voltage will be adjusted according to the increase in tip radius with evaporation. Using a simplified geometry model with the assumption of uniform hemispherical tip shape evolution, analytical solutions of the geometric factor has been derived in several models. These reconstruction protocols with analytical solutions of emitter evaporation and point projection have been applied in currently available data analysis tools. Generally, the analysis results agree well with simulation results and experimental data validated by other instrumentation. However, this is not always the case in a heterogeneous system consisting of phases with different evaporation behaviors. For most complex materials systems, the traditional reconstruction method will create global distortions in the final reconstructed data due to the magnification artifacts, which is mainly caused by the evaporation induced non-uniform apex shapes. Understanding such artifacts and improving the data analysis to account for such effects is very important to the further successful application of APT.

One exemplary illustration of the local magnification due to the heterogeneity is in the case of MgO embedded with gold particles [23,24]. Before evaporation, the APT needle sample has a smooth surface with gold particles embedded in the MgO matrix. After the evaporation, gold nanoparticles protrude out at the surface due to the slow evaporation rate, in contrast to the MgO matrix. In these experiments, the 3-D reconstruction analysis was carried out with the assumptions described above and the complex nature of the surface due to the Au particle protrusion from the surface contributes to trajectory aberrations. There has also been past experimental evidence provided by performing TEM imaging after interrupted APT analysis of multilayer samples highlighting the non-hemispherical tip geometry changes during the evaporation of layers with differing evaporation fields [25]. Recent APT-TEM correlation results on vertical Si-SiO_2 interfaces also highlight the non-hemispherical tip shape evolution during APT analysis of heterogeneous materials [26]. In addition to the experimental evidence, a number of simulation efforts has been focused on the evaporation of hemispherical APT tips consisting of layers or precipitates with varying evaporation fields [27–31].

To take full advantage of the high resolution inherited in atom probe tomography, a new efficient and accurate tip evaporation model is crucial for next-generation atomic structure reconstruction software [25,32–35]. In most tip evaporation models, finite difference or finite element models with atomic lattice structure is used to simulate the geometric tip evaporation and the following trajectory projection. Larson et al. [36] used a finite difference method to simulate the dynamic tip shape evolution during evaporation of a multilayer-structured tip, where each layer material had different evaporation field strength. Three different initial APT tip geometries were considered including two conditions where the interface between a high evaporation field layer material and low evaporation field material layer was considered perpendicular to the axis of the tip and one case where the interface between the same layers was considered to be parallel to the axis of the APT sample. In all three simulation cases, the interface structures in the reconstructed data were distorted due to non-hemispherical tip shape evolution during evaporation. This necessitated additional corrections to be imposed on the standard reconstruction protocol in order to obtain an accurate interface definition representative of the initial stage of the APT tip. There have also been finite element simulations aimed at understanding evaporation of heterogeneous composite materials consisting of nanoscale precipitates with widely different evaporation field compared to the matrix [37–39]. Pseudo 3D simulation models with revolution axisymmetric geometry have also been developed to reduce the computational expense [40–42]. Due to the large size of the sample compared with the atomic lattice and the size of APT chamber compared with the tip, the simulation can be extremely computationally expensive. To make it affordable in most models, the tip size is shrunk and the distance between the electrode and tip is shortened, where computationally efficient models are needed to overcome these difficulties.

To improve the computational efficiency, numerical simulations require more efficient approaches. In the last several decades, level set, phase field and many other analytical [43] and particle methods (DPD multiphase flow simulations [44]) that can be used for interface tracking have been widely applied to solve the dynamic moving interfaces. Examples are the application of the phase field and level set methods to simulate the solute precipitation/dissolution problem with evolving solid–liquid interfaces [45–48] and solid–solid interfaces [49–51]. Both level set and the phase field methods have subgrid scale accuracy through linear interpolation such that the interface can be more accurately represented while interface normals and curvature can be also conveniently calculated. The phase-field approach does not require explicit computing of the moving interface so that numerical difficulties associated with the moving interface are avoided and no explicit interface tracking is required. However, phase field approach requires a complex asymptotic analysis in order to find the relation between parameters of the sharp-interface model and the phase field model [52,53]. A level set approach does not require such an asymptotic analysis and can naturally handle the discontinuities at the interface through explicit interface tracking. Haley et al. [54] have applied the level set method for modeling homogeneous tip evaporation where curvature drive flow was used to mimic the tip evolution. In this study, the level set method is used to simulate the field evaporation of heterogeneous specimen coupling with a simultaneously updated full Poisson solution for an electric potential field.

2. Methods description

To validate the level set model developed for APT specimen evaporation, we compared the results with simulation results from the finite element and the finite difference method. Both methods solve the Poisson equation to retrieve the potential field and the local electric field, and then implement an evaporation process. The process is then iterated until stopped at a preset critical value of either tip height or evaporated ion volume/number.

2.1. Description of level set method

In our approach, a level set-based evaporation model is developed to simulate the evolution of tip surface morphology during the evaporation of multi-layered heterogeneous materials. Current evaporation models for APT utilize finite difference or finite element methods where the staircase representation is commonly used to describe the tip surface. The accuracy of tip geometry cannot be higher than the resolution of grid cell. Such methods are also not straightforward when the interface curvature and normals are involved in the calculation for atomic evaporation.

The level set method was originally introduced for solving multiphase flow problems involving dynamic and complex interface evolution between two phases. Historically these "moving boundary and/or interface" problems have been very challenging from a computational point of view. Level set methods [55] are based on the tracking or capturing of sharp interfaces, while complex

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