

A Volumetric Framework for Registration, Analysis and Visualization of Nanostructured Materials

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Abstract. Data from atomistic simulations of nanostructured materials pose challenges to conventional scientific visualization and analysis pipelines. At the nanoscale, definition of surfaces is only approximate, with multiple possible physical models and high uncertainty due to the underlying discrete geometry. Scientists often sidestep defining the surface entirely, and rely on the relative positions of atom nuclei to conduct analysis. For visualization, however, modalities relying on nucleus positions (ball-and-stick or extruded surfaces) provide limited understanding of the underlying model. To better represent material boundaries, it is desirable to use a volumetric charge density model for both analysis and visualization of the material interface. In this paper, we demonstrate a volumetric pipeline for nanoscale materials analysis, which employs the same data structures for efficient volume rendering, registration and geometric analysis (surface area, volume, curvature) of nanostructured materials. We apply our framework to analyze simulated atomic diffusion in amorphous aluminum oxide nanobowl structures, aimed at improving catalysis processes.

1. Introduction

Innovation in renewable energy technologies demands advanced catalytic and biomass conversion technologies, and drives ongoing research initiatives at the US Department of Energy. Optimizing catalysis with atomic-scale nanostructures is central to these efforts. Complimenting experimental synthesis, computational chemistry provides insight into the atomic-scale behavior of materials. Understanding and validating these simulations requires domain-specific analysis and visualization techniques.

Most atomistic nanoscale simulations involve tens or hundreds of thousands of atoms. Although the simulations themselves are computationally demanding, they are not large by the standards of traditional visualization analyses. Nonetheless, semantic interpretation of this data is not always straightforward in general-purpose scientific visualization software [2, 4]. Chemistry-specific packages [1, 5, 6] are designed primarily for ball-and-stick modeling, and often rely on separate software to plot and analyze charge density or electrostatic potential fields. Where it exists, visualization of these volumetric data typically consists of precomputed isosurface meshes, limiting possibilities for interactive classification. However, classification is particularly pertinent to materials data, where boundaries possess inherently high uncertainty. Comparative and uncertainty volume visualization techniques have been studied in other contexts [9]; we propose applying them to nanoscale computational chemistry simulations.

2. Volumetric Framework for Nanostructure Analysis

Our work focuses on molecular models that are cumbersome to visualize as ball-and-stick and static isosurfaces. The ability to characterize a molecular system with specific shape and structure is especially desirable. Using novel amorphous aluminum oxide (Al_2O_3) nanostructures, “nanobowls”, as catalyst supports has been proposed to engineer catalysts with more than one functionality, which can then be tailored to mimic natural catalysis [3,8,12]. The stability of synthesized nanostructures under reaction conditions is a key for the applications. One underlying factor affecting the stability of such nanostructures is the atomic diffusions of aluminum and oxygen in the bulk and on the surface, which are temperature dependent. The rate of volume decrease of the nanostructure (e.g. nanobowl) as a function of temperature can be used as a qualitative indicator of the structural stability. We model the diffusion process in nanobowl models consisting of 10,000 - 20,000 atoms (depending on the nanobowl diameter), using DL_POLY [11], a molecular dynamics (MD) code. The MD simulations of pre-equilibrated system were conducted at temperatures in the range of 1000 - 1500 K. The MD time step is one femtosecond (10^{-15} second). The intermediate structures of the nanobowl are saved every 10 picoseconds (10^{-12} second) during simulations. Analysis and visualization is conducted every 5 of these steps, or 50 picoseconds. The simulations discussed here are run for 1.5 nanosecond (1.5×10^{-9} second).

The goal is then to determine which structures are stable, given different bowl radii and temperatures. This is determined by the atom geometry as the simulation progresses. As the structure disintegrates, both surface area and void space volume decrease. Geometric analysis can be conducted from atom positions, even though they do not directly define a surface. Since structural stability can be measured relatively, this is sufficient for most analysis. Ultimately, we seek visualization and analysis that correlate to experimental results using electron microscopy, which requires modeling the physical surface of the compound. As full density functional theory (DFT) computation would be prohibitively costly, we use an atom-decomposed charge density model to approximate the charge density field as a structured volume. We conduct analysis on this model and use volume rendering to visualize our results. Lastly, we correlate these results with those of the raw positional data.

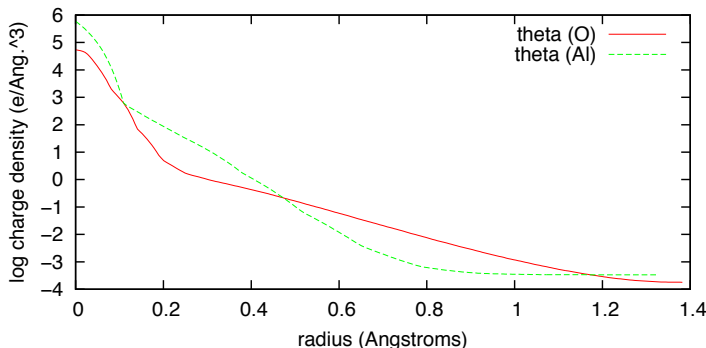


Figure 1. Charge density from Al-Al and O-O bonds plotted as functions $\theta_{Al}(r)$ and $\theta_O(r)$ of radial distance r . The charge density along the Al-O bond is equivalent to the superposition of those along Al-Al and O-O. Therefore we use θ_{Al} and θ_O to model the charge density around Al and O, respectively.

2.1. Generating the Charge Density Field

Numerous models exist for defining surfaces at the atomic scale, depending on chemicals and their bonding behavior [10]. Because of the strong ionic nature of bonding in alumina, we can model the charge density with a superposition of static ionic charge densities. Specifically, for atom positions $\{\vec{p}_i\}$, we have

$$r_i(\vec{x}) = |\vec{x} - \vec{p}_i|$$

$$\rho(\vec{x}) = \sum_i \theta(r_i(\vec{x})).$$

We evaluate θ via a lookup table from the calculated all-electron charge density of a DFT computation on bulk $\alpha - Al_2O_3$, as shown in Figure 1. We then generate a structured volume by summing total charge density from each contributing atom, for each voxel in the volume. We use a scale factor of 4 voxels per \AA , yielding roughly $300 \times 300 \times 170$ (58 MB) single-precision float volume data per geometry step.

2.2. Bowl Classification and Registration

Having constructed the charge density field ρ , we wish to extract the bowl by examining the range (classification) and spatial domain (registration) of ρ . For the former, we can identify segments of the charge density distribution corresponding to void space, compound, and boundary regions. Because the charge density is extracted from a bulk calculation, the values of $\theta(r)$ at large r , appropriate for defining the boundary between surface of the nanostructure and vacuum, is uncertain. Nonetheless, our classification enables us to visualize this interface. We use the minimum measured charge for θ_{Al} , ($3 \times 10^{-2} \text{ e}/\text{\AA}^3$) as an inner isovalue bound for our bowl surface, and a small nonzero $\epsilon = 10^{-4} \text{ e}/\text{\AA}^3$ as an outer bound. In our transfer functions, we have color coded charge densities corresponding roughly to void (blue), oxygen (red) and aluminum (green) thresholds. Since we use the same charge density model for all time steps, we only need to perform classification once for all simulation runs. From this we segment the nanobowl (Figure 2).

In registration, we seek the spatial extents of the nanobowl prior to analysis. Through the diffusion process, atoms can move, significantly altering the height of the bowl and the structure near the rim. We wish to take accurate measurements of the bowl structure, changes in geometry notwithstanding. To accomplish this, we measure the bowl as a height function $Z(x,y)$, and determine $\bar{Z}_\rho = \sup\{z \mid \rho(z) > 0\}$, the maximum non-void height via our classification. We then compute a distribution of \bar{Z}_ρ for each time step in the simulation. The mode of this distribution, or most common maximum height, is a good indicator of the upper plane of the structure, thus the spatial top of the bowl.

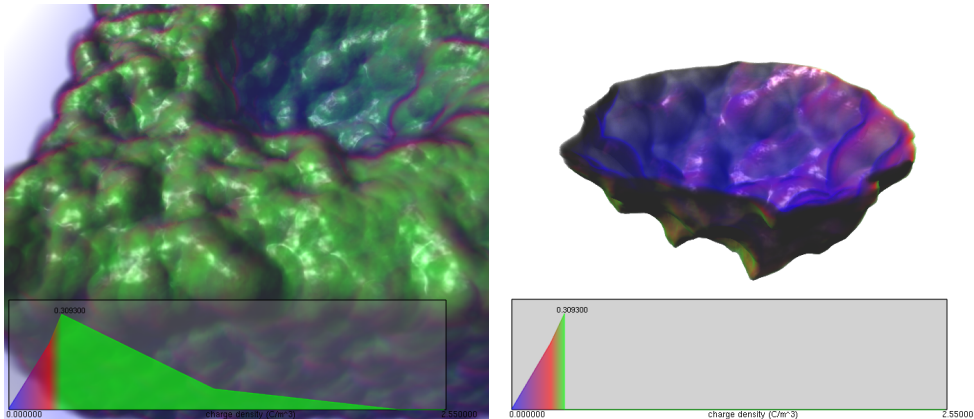


Figure 2. Classification of the charge density volume, showing uncertainty in the material interface via a red-blue colormap in the lower range of the transfer function.

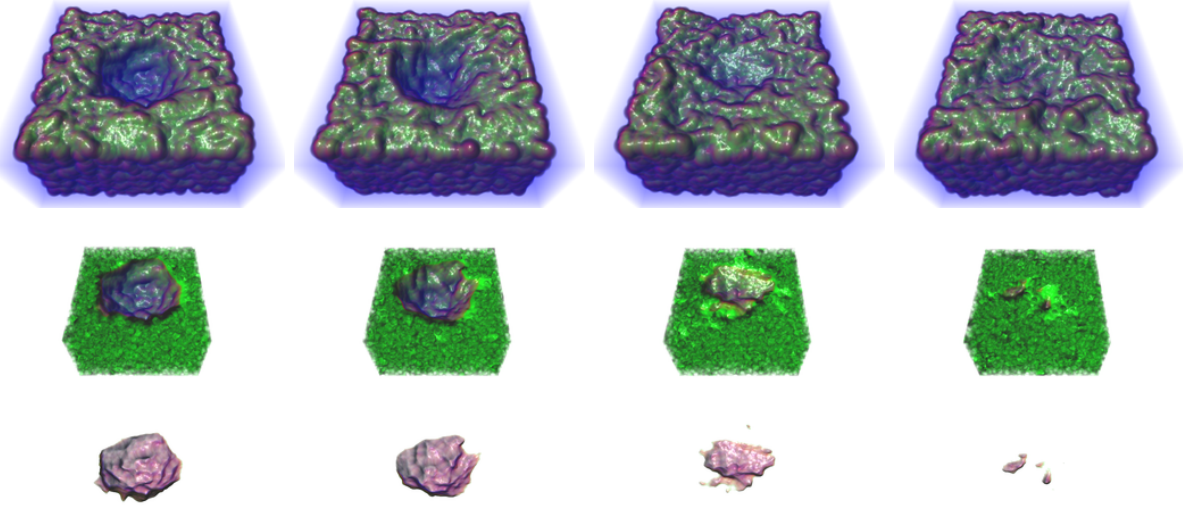


Figure 3. Comparative visualization at 800 picoseconds of nanobowls at four temperatures (from left to right: 1000K, 1200K, 1300K, and 1350K). Rows show the full potentials volume, registered bowl window, and segmented bowl void space.

2.3. Combining Data Structures for Analysis and Visualization

Volume and surface area can be estimated by cell count at the same time as registration. Units are then normalized by the volume dimensions into cubic Angstroms. For each geometry step,

$$\begin{aligned}
 V(i, j) &= \bar{Z}_\rho - Z(i, j) \\
 V &= \sum_{i, j} \max(V(i, j), 0) \\
 A &= \sum_{i, j} \{1 \text{ if } V(i, j) > 0, \text{ else } 0\}
 \end{aligned}$$

Any volume renderer with 1D transfer function capabilities suffices for visualization. Seeking portability and interactivity on laptops with weak or no GPU resources, we chose an efficient CPU-based solution [7]. This approach has the advantages of not relying on progressive LOD simplification for interactive performance on modest hardware, and having access to full CPU main memory when switching between geometry steps. This renderer requires precomputation of a min-max bounding volume hierarchy (BVH). While the time to compute this is not significant (300 ms per instance), it is an added postprocess step. However, this structure proves useful in optimizing additional analysis. We have performed more accurate surface area computation (via marching tetrahedra and marching cubes) and Gaussian curvature to identify the bowl rim. By omitting regions outside our classification, the BVH provides order-of-magnitude better performance than computation over the full volume.

3. Results and Discussion

In Figure 4, we compare volume in \AA^3 from our charge density field model with the original positional analysis. The original positional analysis consisted of a gridded heightfield interpolating nucleus positions, with a Gaussian smoothing filter to adjust for temporal noise. This provides a good picture of overall trends, and is sufficient to determine whether a structure is stable or not. With our charge density model, we note greater fluctuation in volume measurements. Nonetheless, the same overall trends remain. We note that the uncertainty in the exact surface location (inner and outer thresholds) does not overly affect geometry, although it is useful as a visual indicator. Clearly, it would be desirable to validate visual and statistical results of our model with real-world microscopy data from experimental groups.

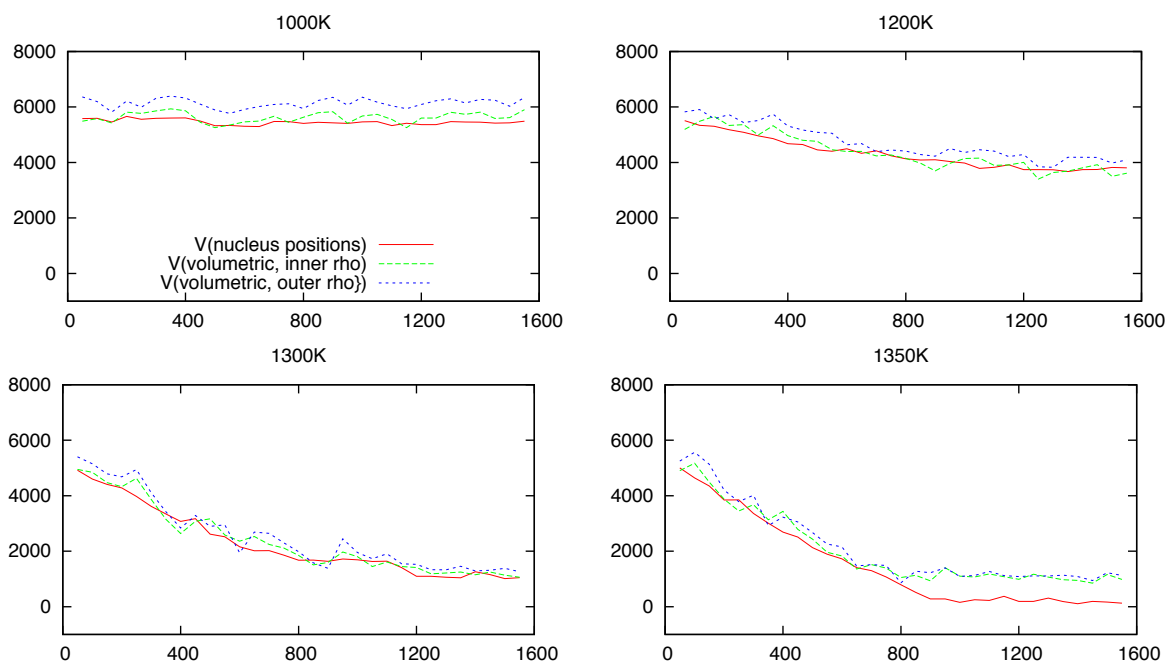


Figure 4. Analysis of bowl volume (\AA^3), over the simulation time (picoseconds), for the runs shown in Figure 3. We compare bowl volumes computed from a heightfield interpolated nucleus positions, with our volumetric classification approach, showing lower and upper possible bounding regions of the surface (Figure 2).

We find our spatial discretization is sufficient to preserve geometric quality, without generating an overly large data footprint. Nonetheless, the selected steps of five simulation runs generated 8 GB of data on disk, as opposed to 110 MB of particle data. Generating volume data for each iteration (1 fs) of the MD simulation would require roughly 400 TB per run. While this may be impractical, greater temporal refinement and more comparative analysis could shed light on variation in geometric structure through the diffusion process. Moving forward, we would like to expand analysis of approximate charge density fields to larger-scale problems, and leverage them towards computational steering or ensemble computation.

Acknowledgments

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