

Supporting Information: The Birth of NaCl Crystals: Insights from Molecular Simulations

G. Lanaro and G. N. Patey

*Department of Chemistry, University of British Columbia,
Vancouver, British Columbia, Canada V6T 1Z1*

(Dated: July 7, 2016)

I. DISTRIBUTION OF OTHER PROPERTIES

Figures S1 and S2 show distributions of various properties for clusters of 10 and 30 ions, respectively. The properties considered and the calculation methods are given below.

- **Average neighbor count** is calculated by counting for each ion of the cluster the number of neighbors within 0.6 nm. The counts are then averaged over all ions in the cluster.
- **Volume** is approximated by encasing a cluster in a cuboid box, subdivided into $32 \times 32 \times 32$ grid points, and counting the number of grid points that are inside of the cluster, the volume is obtained by multiplying this number by the volume of the grid element (Figure S3). The radius that each ion covers is chosen to be the ionic radius plus the solvent radius, according to the definition of excluded volume¹.
- **Surface area** is approximated by extracting the surface from the volumetric representation described above using the marching cube algorithm², a procedure inspired by the work of Xu et al³.
- **Sphericity** is the ratio of the surface area of a sphere with equivalent volume and the surface area of the cluster. It is defined as

$$\pi^{1/3} \frac{(6V)^{2/3}}{A} ,$$

where V is the cluster volume and A is the cluster surface area. Defined this way, the sphericity parameter is 1 when the object is perfectly spherical and assumes lower values for less spherical objects. This parameter is often used to represent the compactness of a three dimensional object.

- **Hydration** is a measure of water content in the cluster and is calculated by counting the number of water molecules within 0.3 nm for each ion in the cluster, then averaging the counts over all the ions in the clusters.
- **Radius of gyration** is defined by

$$R_g^2 = \frac{1}{N} \sum_a (\mathbf{x}_a - \bar{\mathbf{x}}_a)^2 ,$$

where the sum is over the N ions in the cluster, \mathbf{x}_a is the position of the ion and $\bar{\mathbf{x}}_a$ is the geometric center of the cluster. It is commonly used as a measure of compactness.

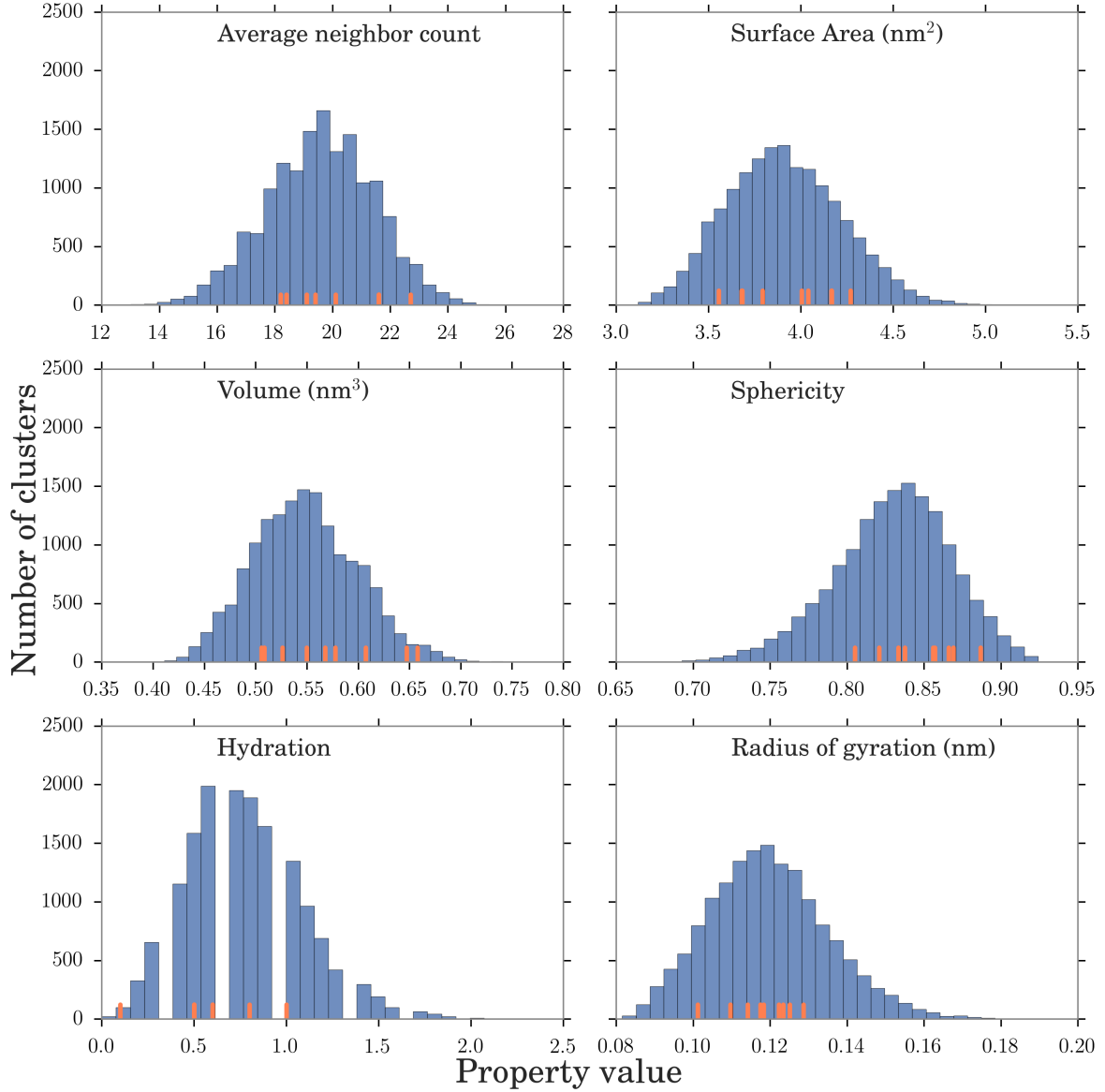


FIG. S1. Distributions of various properties for failed clusters (blue histogram) of size 10. Clusters that achieved nucleation are indicated by orange lines (for some properties, e.g. hydration, a single orange line might represent more than one cluster). Note that the property values of the clusters that achieve nucleation tend to lie around the mean of the distribution.

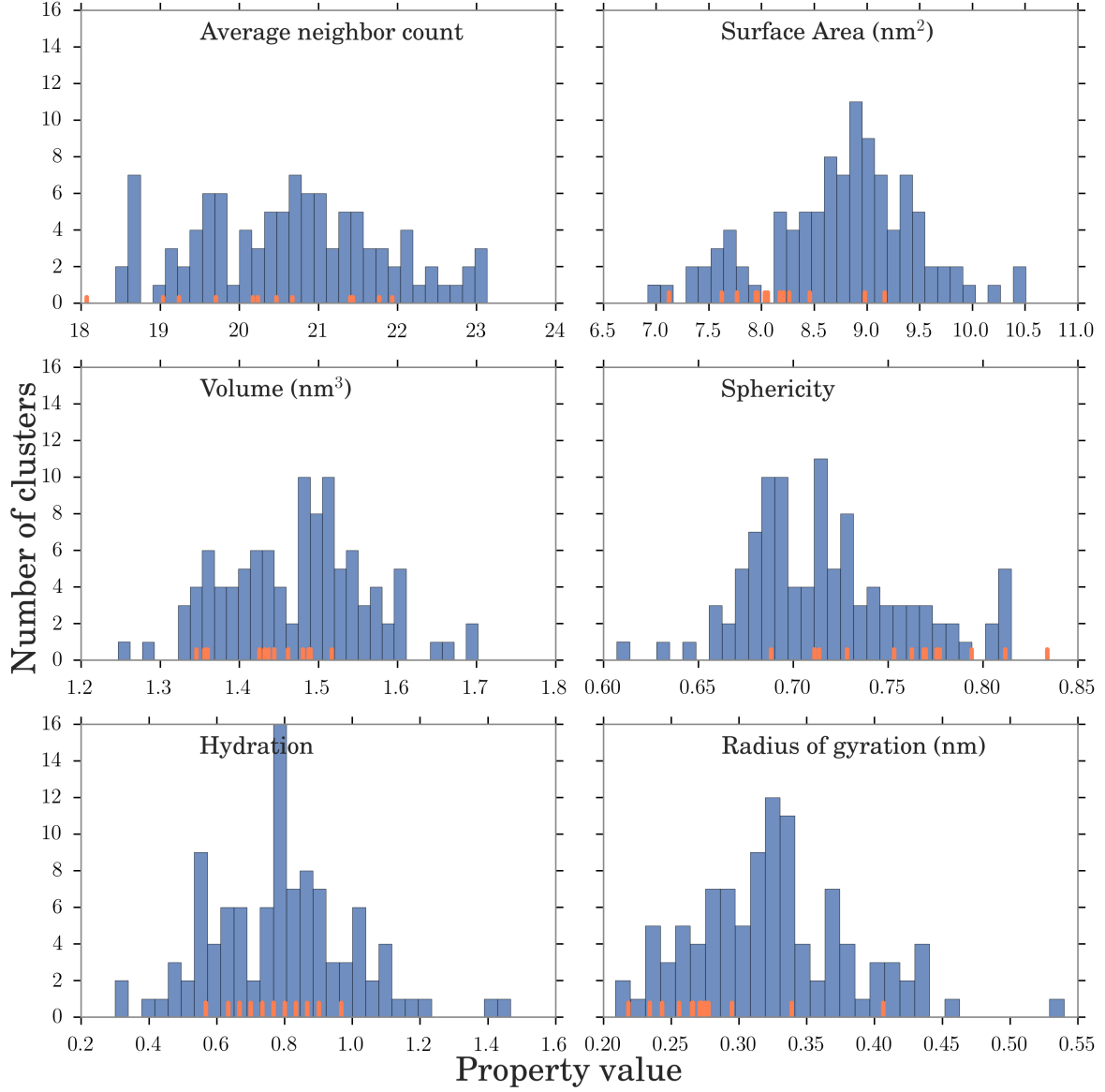


FIG. S2. Distributions of various properties for failed clusters (blue histogram) of size 30. Clusters that achieved nucleation are indicated by orange lines (for some properties, e.g. hydration, a single orange line might represent more than one cluster). At this size, some preference for a low radius of gyration, low surface area, and higher sphericity can be observed.

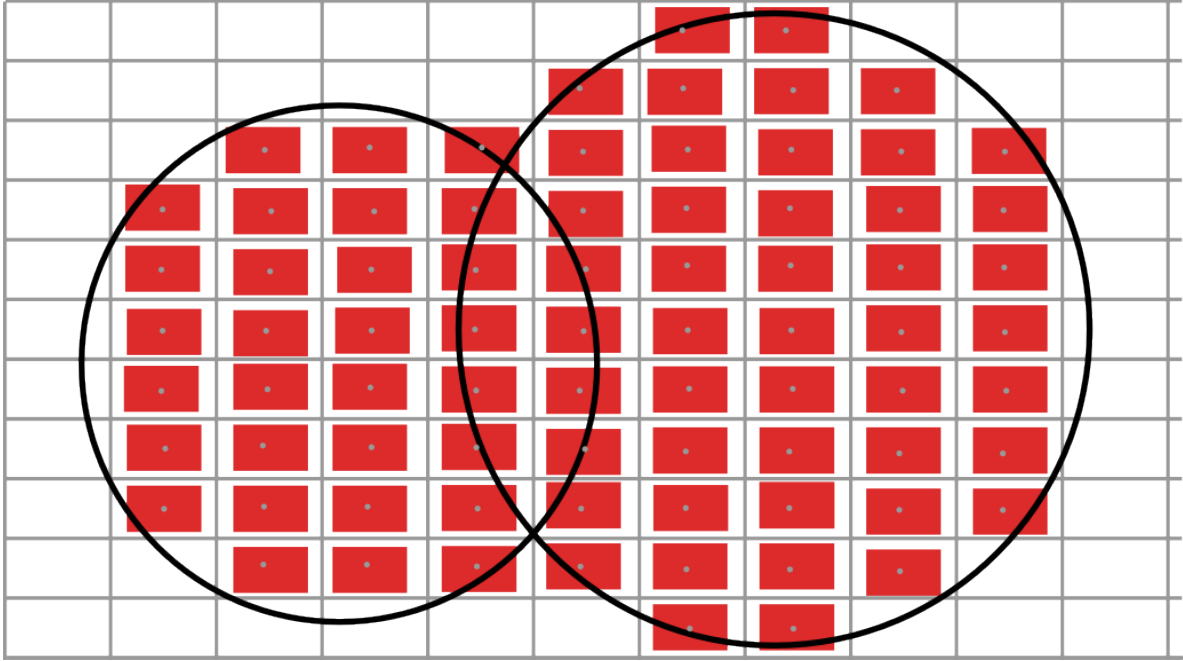


FIG. S3. A two dimensional representation of the algorithm used to estimate the volume. The cluster is encased in a grid, and the grid points that lie within the cluster are indicated in red.

REFERENCES

- ¹ Richmond, T. J. Solvent Accessible Surface Area and Excluded Volume in Proteins: Analytical Equations for Overlapping Spheres and Implications for the Hydrophobic Effect. *J. Mol. Biol.* **1984.** *178*, 63–89.
- ² Lorensen, W. E.; Cline, H. E. Marching Cubes: A High Resolution 3D Surface Construction Algorithm. In *ACM siggraph computer graphics*, volume 21. ACM, **1987** 163–169.
- ³ Xu, D.; Zhang, Y. Generating Triangulated Macromolecular Surfaces by Euclidean Distance Transform. *PloS one* **2009.** *4*, e8140.