

Creating a zoo of crystal structures *via* tunable interactions

Julia Dshemuchadse¹, Michael Engel², Sharon C. Glotzer^{1,3,4}

¹*Department of Chemical Engineering, University of Michigan, Ann Arbor, MI, USA*

²*Department of Chemical & Biological Engineering, Friedrich-Alexander-University Erlangen-Nuremberg, Erlangen, Germany*

³*Department of Materials Science & Engineering, University of Michigan, Ann Arbor, USA*

⁴*Biointerfaces Institute, University of Michigan, Ann Arbor, MI, USA*

djulia@umich.edu

Structural complexity is often justified with building-block complexity: by their sheer size or odd shape, molecules of all kinds tend to form crystal structures with large unit cells. In intermetallic systems, an increasing number of complex structures with up to thousands of atoms per unit cell or exhibiting aperiodic order has been discovered over the last decades [1–3]. This is perceived as particularly perplexing, as metals typically exhibit the simplest structures imaginable—sphere packings—and due to the lack of directionality of the metallic bond. A similar scenario presents itself in a completely different class of materials on the mesoscale: systems of soft condensed matter—*i.e.*, polymer micelles, colloids, and nanoparticles—have been found to exhibit complex structures despite the apparent simplicity of their building blocks [4–8].

To approximate these simple systems, we studied attractive particles interacting *via* isotropic pair potentials. These short-ranged interactions had previously been found to form highly complex structures [9]. We examined two large parameter spaces of these one component systems and found a variety of crystal structures—some novel, some known; some simple, some complex.

We find structures that are equivalent to atomic compounds—of a chemical variety that ranges from tetrahedral environments that are characteristic for covalent bonding, to highly-coordinated, close-packings that is typical for the metallic bond. On top of that, we also discovered geometries that fall in the lesser-known area in between those extremes, exhibiting almost all coordination numbers between 4 and 14 (see Fig. 1), which is indicative of our ability to tune the inter-particle potentials in this agnostic model beyond limitations that the periodic table of elements poses on chemical interactions on the atomic scale.

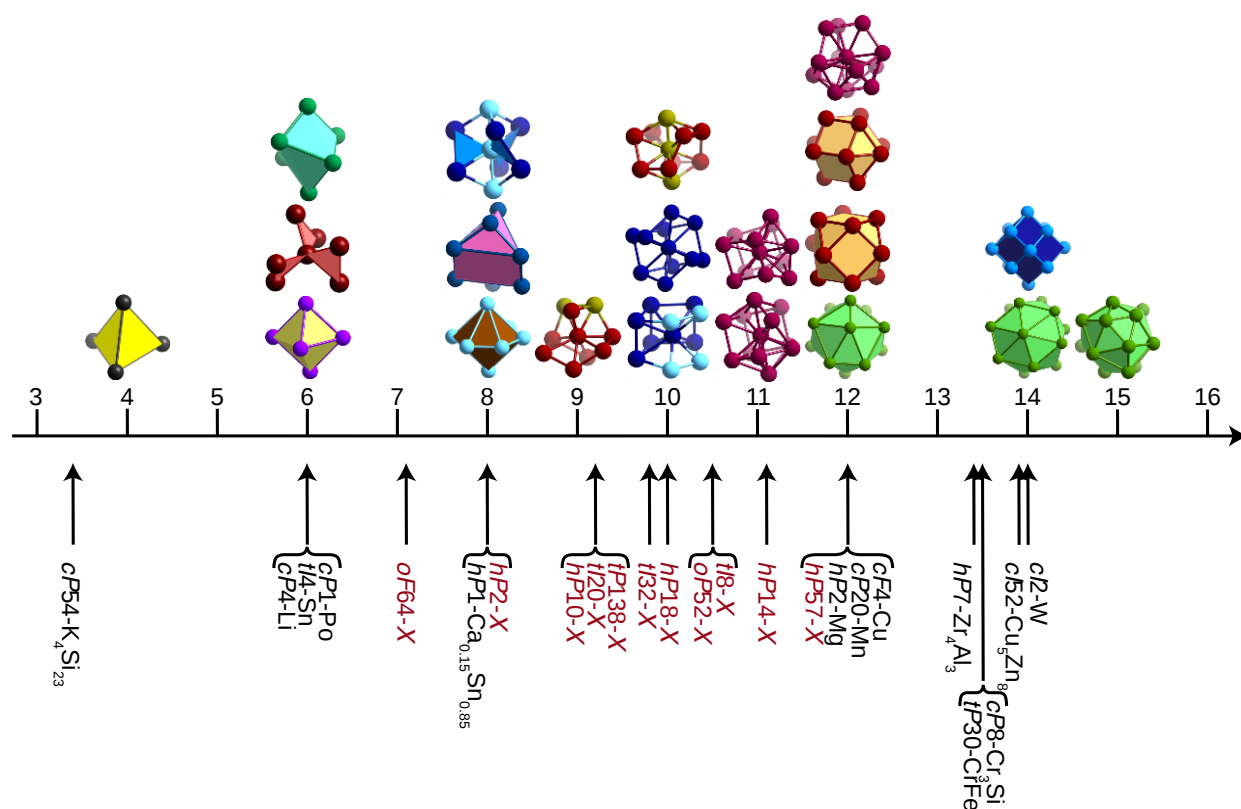


Figure 1. Coordination polyhedra in the range $CN = 4-15$, as they appear in the structures observed in this study. The average coordination number of the reported structures is indicated below the CN -axis ($CN = 4-14$). Novel structure types are displayed in red color.

1. S. Samson, *Nature* **195**, (1962), 259–262.
2. D. Shechtman, I. Blech, D. Gratias, J. W. Cahn, *Phys. Rev. Lett.* **53**, (1984), 1951–1953.
3. J. Dshemuchadse, W. Steurer, *MRS Proceedings* **1517**, (2012), KK1.01.
4. G. Ungar, X. Zeng, *Soft Matter* **1**, (2005), 95–106.
5. S. Lee, C. Leighton, F. S. Bates, *PNAS* **111**, (2014), 17723–17731.
6. M. B. Zanjani, J. C. Crocker, T. Sinno, *Soft Matter* **13**, (2017), 7098–7105.
7. S. Hajiw, B. Pansu, J.-F. Sadoc, *ACS Nano* **9**, (2015), 8116–8121.
8. H. Lin, S. Lee, L. Sun, M. Spellings, M. Engel, S. C. Glotzer, C. A. Mirkin, *Science* **355**, (2017), 931–935.
9. M. Engel, P. F. Damasceno, C. L. Phillips, S. C. Glotzer, *Nature Materials* **14**, (2015), 109–116.