

1.0 Research Question and Justification of Modeling Tools

Research Question:

How does polymer concentration influence the aggregation behavior of “MXene” flakes through depletion interactions? I am investigating at what concentrations of polymers to flakes I can begin to observe entropy-driven aggregation at. What is responsible for attractions between the flakes are depletion forces, as the closer the flakes get, the more thermodynamically favorable it is for the system, due to less free energy in the system. To test this, I am running two flake simulations in various polymer concentrations, isolating purely entropic depletion using purely repulsive interactions.

2.0.

The molecular dynamics simulations were implemented in Python using flowerMD, a simulation engine built on top of Hoomd-blue. It is a custom workflow to handle simulation setup, execution, and data analysis. The environment provided in the [flowerMD](#) documentation provides all the necessary software that is required to run this repository.

2.1

Each simulation began by constructing a system of graphene flakes embedded in a polymer solution. Graphene() was used to generate the flake particles, with custom geometry reflective of C-C bond lengths. Each flake consists of carbons arranged in a 2D lattice to mimic the geometry of a MXene sheet.

2.2

For interactions, the goal is to simulate entropy driven aggregation. To model purely entropic (depletion-driven) interactions, all nonbonded particle pairs—whether between monomers, between monomers and flakes, or between flakes themselves—interact via a Lennard-Jones potential truncated and shifted at a $2^{1/2}\sigma$, eliminating the attractive tail of the potential, producing a WCA potential that is purely repulsive. This is to ensure that no attraction exists between anything. Therefore, any aggregation observed in the simulations must arise from entropic origins.

2.3.

First, the system was relaxed at a lower $kT = 0.001$, then was ramped up to $kT = 2.0$, in order to allow any potential overlaps to resolve without blowing up the system. Every run had identical decorrelation times, as well as the same amount of independent samples.

2.4.

Smaller system sizes were used as a result of computing constraints, such as the VPN not allowing me to stay connected to it for very long. The lower the amount of polymers, the more aggregation over time occurs. I only used a two flake setup as I wanted to investigate how depletion is a pairwise, localized effect between colloids, which in my case, are flakes.

Answer to my research question from my work:

My simulations show that polymer concentration does influence the aggregation behavior of the flakes, consistent with depletion interaction theory. By simulating systems with purely repulsive interactions (no energetic attractions), we isolate entropic effects and observe how flake–flake spacing evolves under varying polymer concentrations.

As shown in the plot generated, the system with the fewest polymer chains, 50 chains, exhibits a pronounced decrease in average flake–flake distance over time, signaling clear aggregation. Larger systems exhibit less aggregation behavior, due to the gain in entropy from reducing excluded volume is less effective in more dense systems, reducing effective attraction between flakes. At higher polymer densities, the crowding causes the depletion forces to become weaker.

Literature:

<https://iopscience.iop.org/article/10.1088/0953-8984/21/3/035101>

<https://pubs.rsc.org/en/content/articlehtml/2014/sm/c3sm53058b>

<https://pubs.aip.org/aip/jcp/article-abstract/127/11/114903/295925/Effect-of-polymer-size-and-chain-length-on?redirectedFrom=fulltext>