

Exploring Competitive Adsorption Behavior of Polymer Chains in Silica Nanopores by Small Angle Neutron Scattering (SANS)

Xuchun Wang and Byeongdu Lee*

X-ray Science Division, Argonne National Laboratory, 9700 S. Cass Ave., Lemont, IL 60439, USA

Nowadays, the billions of tons of plastic production and mismanagement of end-of-life waste plastic, incinerated or landfilled, cause severe environmental pollution and safety issues for humans and wildlife. Chemical upcycling is one of the strategies to convert waste plastics into value-added products. Among numerous catalysts, processive hydrogenolysis catalysts, featuring metal nanoparticles (e.g., Pt) embedded at the base of cylindrical silica nanopores, exhibit the ability to repeatedly cleave polymer chains, yielding value-added hydrocarbon products with carbon number ranging from 10 to 30.[1-3] To enable the processive mechanism, the mesoporous shell should exhibit a preference for adsorbing longer polymers. This ensures that virgin chains (long polymers) remain at active sites for extended periods, increasing the likelihood of cleavage, while cleaved chains exit swiftly to avoid secondary cleavage. Thus, we hypothesize that competitive adsorption occurs among polymer chains of varying lengths within spatially constrained nanopores.

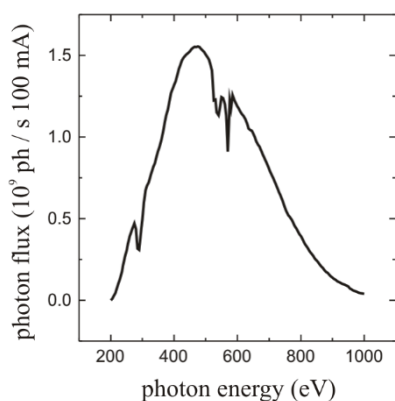


Figure 1: Example of a figure.

This research [3-4] presents a model study aimed at elucidating the competitive adsorption behavior of binary blends with distinct molecular weight in melting state, mimicking conditions akin to the intermediate stage of the catalytic reaction. Taking advantage of the contrast difference between hydrogenous and deuterated polymers in the neutron beam, we utilized small angle neutron scattering (SANS) to measure the relative composition of the polymers within the silica nanopores. Our findings demonstrate preferential adsorption of longer chains within the silica nanopores, which is consistent with the theoretical

prediction in the literature for the case of the enthalpic attraction between polymers and pore walls.

References

- [1] A. Tennakoon *et al.*, *Nat. Catal.* **3**, 893-901 (2020).
- [2] X. Wu *et al.*, *J. Am. Chem. Soc.* **144**, 5323-5334 (2022).
- [3] X. Lyu *et al.*, *ACS Appl Mater Interfaces* **15**, 27369–27379 (2023).
- [4] X.Wang *et al.*, *Macromolecules* ASAP (2024)