

The Prediction of Mechanochemically Productive Reactions

Ryan W. Kwok^{1,2}, Leah Tamayev², Tzu-Hsuan Chao^{3,4}, Mark Boyer^{3,4}, Nick Hopper^{3,4}, Yeu-Shiuan Ho^{3,4}, Ivy S. Agyemang^{1,4,5}, Senad Skejovic^{1,4,5}, Adam B. Braunschweig^{1,2,4,5}, Daniel P. Tabor³, Mateusz Marianski^{1,2,4}

¹ Ph.D. Program in Chemistry, The Graduate Center of the City University of New York, New York, NY 10016

² Department of Chemistry, Hunter College, 695 Park Avenue, New York, NY 10065

³ Department of Chemistry, Texas A&M University, College Station, TX 77843

⁴ NSF Center for the Mechanical Control of Chemistry, Texas A&M University, College Station, TX 77843

⁵ Nanoscience Initiative, Advanced Science Research Center, New York, NY 10031

While traditional solvothermal chemistry uses heat and solvent to drive its chemical reactions, mechanochemistry harnesses mechanical force. In addition to more efficient energy usage and a significant reduction in solvent waste, mechanochemical conditions have also been shown to allow for faster reaction times and access to otherwise inaccessible reaction products. The prediction of these mechanochemically productive reactions however remains difficult due to the lack of fundamental design principles with which to guide experimental design. Previously, experiments using polymer-pen lithography have demonstrated that the Diels-Alder reaction can be accelerated via the application of a relatively small uniaxial force. The current understanding is that force alters the reaction trajectory from its solvothermal pathway onto an entirely new, mechanochemical pathway. This is due to force-induced molecular distortions in the reactant and transition state geometries. In my poster, I will present recent efforts towards developing a theoretical framework for the prediction of mechano-susceptible reactions. In particular, we first screened the chemical space of Diels-Alder reactions to identify potential reactants to use that will be accelerated under distortion. We then further analyze the most promising candidates to determine the origins of mechanochemical productivity and propose a means of experimentally verifying our model. A deeper understanding of the fundamental principles that govern mechanochemical reactions will help contribute to the widespread adoption of sustainable mechanochemical processes.