Molecular Dynamics Modelling of Mechanical Activation and Catalysis in Ball-Milling Mechanochemistry

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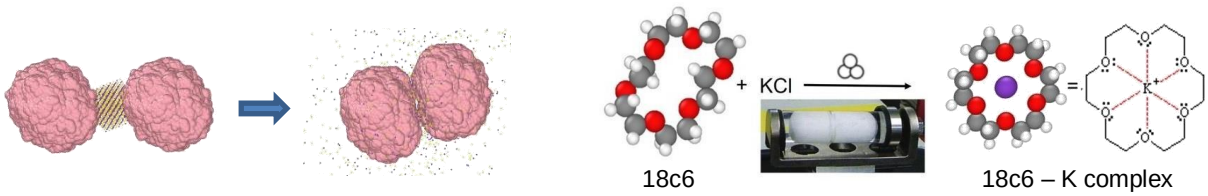
Mechanochemistry involves transformations driven or assisted by mechanical force [1]. Ball milling (BM) is the most widely used method in modern mechanochemistry, but its inherent nature makes it challenging to study individual impacts experimentally [2]. To overcome this limitation, molecular dynamics (MD) simulations offer a powerful tool to gain molecular-level insight into mechanical activation events.



We perform MD simulations using GROMACS package to describe mechanical activation by fragmentation of crystal particles down to individual atoms, ions and ion pairs and their subsequent recrystallization. To achieve this, we incorporate neutral, rigidly constrained amorphous balls to exert mechanical force on the crystal. Furthermore, we use the host-guest complexation reaction of 18-crown-6 ether (18c6) with KCl, as our model system and monitor subsequent product formation and rate of agglomeration under neat and liquid-assisted conditions. We demonstrate that a small amount of the liquid additive facilitated product formation, while too much of it destabilized the product by stabilizing the reactants. Our findings signify the importance of not only impacts for reactant activation, but also the immediate surroundings of the activated species for stability. This approach provides a fresh and detailed view of mechanochemistry that could allow for its theoretical optimization and guide the targeted use of catalysts in mechanochemical reaction.

[1] James, S.L. et al. Mechanochemistry: opportunities for new and cleaner synthesis. Chem. Soc. Rev. 41, 413–447 (2012).
[2] Carta, M., Vugrin, L., Miletić, G., Kulcsár, M. J., Ricci, P. C., Halasz, I., & Delogu, F. (2023). Mechanochemical reactions from individual impacts to global transformation kinetics.*Angewandte Chemie International Edition*, 62(33), e202308046.