Revealing the elementary processes controlling the conversion of CO2 to added-value chemicals & materials

Abstract:

The transformation of the abundant small molecules such as carbon dioxide (CO2) has the potential to contribute toward establishing an environmentally sensible circulation of energy and materials. In this talk, I will discuss recent results conducted in my group at Queen Mary University on the use of computational chemistry as a tool to reveal the early stages of these conversion processes and support the design of catalysts and operation conditions promoting CO2 conversion.
In the first part, I will focus on CO2 mineralization into magnesite (MgCO3). Magnesite can be formed via aqueous carbonation of Mg2+ ions and represents a promising route to carbon capture and reuse, albeit limited by the slow precipitation of MgCO3. The principal difficulty arises from the very strong Mg2+···H2O interaction, raising barriers to Mg-dehydration. We have used atomistic simulations, complemented by spectroscopic experiments, to investigate the influence of solution additives on the various stages of aqueous MgCO3 formation: Mg2+ dehydration; pre-nucleation Mg2+···CO32– pairing; surface growth. Results show which solution conditions lower the barrier to Mg2+ dehydration and subsequent incorporation into the lattice of Mg-carbonates. I will report the design of a carbonation rig to perform real-time, in-situ neutron measurements of CO2 mineralization at the Rutherford Appleton Laboratory in the UK. In the second part, I will present ab initio random structure searching methods to discover Cu-based nanoclusters for the electrocatalytic CO2 reduction reaction. I will show that the most stable and active catalysts are amorphous copper clusters and report a detailed investigation of CO2 activation and conversion on these nanoclusters.καλησπέρα