CO_2 Utilization Via Carbonate-Promoted C–H Carboxylation and CO_2 Hydrogenation

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Chemistry Seminar on CO_2 utilization

4:00 p.m.
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Via Zoom

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Abstract: With the CO_2 level in the atmosphere at 400 ppm and rising, finding new and efficient ways to recycle this greenhouse gas is of paramount importance. Most previous efforts have focused on converting CO_2 to low-value C1 compounds such as CO, formic acid, methanol and methane. The use of CO_2 as a feedstock for the synthesis of multi-carbon compounds is advantageous because these targets generally have higher value and greater energy density. The key chemical challenge is to form C–C bonds to CO_2 without using energy-intensive reagents. In this talk, I will describe a novel carboxylation reaction in which a C–H bond and CO_2 are transformed into a carboxylate ($C–CO_2–$) using alkali carbonates as a promoter, in absence of any catalyst or solvent. Alkali carbonate salts are capable of deprotonating C–H bonds that are ordinarily very weak acids (pKa>40 in organic solvent) to generate carbanions (C–) at intermediate temperatures (200-360 °C). In the presence of CO_2, the carbanions react rapidly to form carboxylates. The chemistry has been applied in the synthesis of 2,5-furandicarboxylic acid (FDCA) from 2-furoic acid, which can readily be made from lignocellulose. FDCA is an attractive green replacement for fossil fuel-derived terephthalic acid, used in polyethylene terephthalate (PET) polymer synthesis. Based on the above-described approach of C–C bond formation, I will also speak about how a mixture of alkali carbonate, CO_2 and H_2 can be readily converted to formate, oxalate and other C2+ carboxylates like acetate, propionate and succinate. The reaction occurs at intermediate temperature and pressure and requires H_2O vapor for hydrating the carbonate salt. Isotopic labelling studies are consistent with a mechanism that proceeds by a series of H_2 and C–H deprotonations to form H– and C-centred nucleophiles that undergo H–C and C–C bond formations.

About the speaker: Aanindeeta Banerjee hails from India, where she completed her bachelor's and master's in chemistry from Presidency College (Kolkata) and IIT (Kanpur), respectively. She came to the U.S. for Ph.D. studies at Stanford University and joined the lab of Prof. Matt Kanan. In her graduate work, she invented a new process for CO_2 utilization with applications in polymer and commodity chemical production. Upon completing her Ph.D. in 2018, she continued developing this technology with support from ARPA-E, as a postdoctoral fellow. In 2019, based on the same core technology, Banerjee and Professor Kanan founded ReSource Chemical with the vision to recycle CO_2 to sustainable chemicals and fuels at low cost.

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