Catalysis in Carbon Dioxide-Based Media: A Multiscale Approach toward Process Development

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CEBC: The mission of the Center for Environmentally Beneficial Catalysis (CEBC) is to generate technologies aimed at transforming the catalytic manufacture and use of chemicals into inherently safe and ecologically responsible processes, while retaining their economic viability. The Center is headquartered at the University of Kansas (KU), with core partners at the University of Iowa (UI), Washington University in Saint Louis (WUSt.L) and Prairie View A&M University (PVAMU). CEBC research projects cut across the four interdisciplinary thrust groups (TGs), that organize the expertise of CEBC: TG1, *Catalyst Design and Preparation*: TG2, *Media Design*; TG3, *Advanced Experimental Measurements Relating to Reactions and Reactors*; and TG4, *Molecular and Process Modeling/Optimization*. Chemists, biologists and engineers collaborate in the design, development and assembly of the "engineered system" consisting of the catalyst, media and reactors. Industry guidance and the deployment of state-of-the-art R&D tools (mechanistic studies, multiscale modeling, economic and environmental impact analyses) ensure that the engineered systems are not only environmentally beneficial but also economically viable for commercialization consideration.

Multiphase Catalysis in CO_2-based media: The near-critical region (roughly 0.9-1.2 T_c; and 0.9 – 2.0 P_c) provides access to unique fluid properties (liquid-like densities and gas-like transport properties) that are, in many instances, ideally suited for performing catalytic reactions. Potential advantages of near-critical reaction media include (a) significant alleviation of oxygen or hydrogen solubility limitations in multiphase reaction systems; (b) enhanced desorption and transport of heavy molecules (such as coke precursors) in mesoporous catalysts mitigating pore-diffusion limitations and improving catalyst effectiveness; (c) enhanced heat capacity ameliorating the problem of parametric sensitivity in exothermic fixed-bed reactors; and (d) facile separation of catalysts, reactants and products. Near-critical reaction systems are thus excellent examples of the 'multifunctional reactor' concept.

At the Center for Environmentally Beneficial Catalysis (CEBC), we employ a multiscale approach for developing bench scale catalytic process concepts that exploit CO_2-based media. Ways in which near-critical media, particularly dense phase carbon dioxide, lend themselves for exploitation in homogeneous and heterogeneous catalysis will be presented. In homogeneous catalysis, the emphasis is on recent developments in catalytic oxidations and hydroformylations. Specifically, the use of mixed solvents (carbon dioxide, water and organic media) as reaction media is highlighted. In heterogeneous catalysis, the emphasis is on solid-acid catalysis. It is shown how the multiscale approach involving concurrent catalyst, media and reactor design can be applied to systematically develop CO_2-based reaction systems.