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Molecular-Level Insights into CO_2 Capture and Conversion by Biocompatible Materials Using Multiscale Theory and Modeling

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Abstract

The escalating urgency to reduce anthropogenic CO_2 emissions from fossil fuel usage has underscored the critical role of carbon capture, utilization, and storage (CCUS) technologies in addressing global climate and energy challenges. While conventional aqueous amine-based solvents dominate current CO_2 capture infrastructure, they are hindered by high regeneration energy demands, degradation, and environmental risks. In pursuit of safer, more efficient alternatives, our research focuses on the tailoring, design, and discovery of biocompatible solvent materials, particularly deep eutectic solvents (DES) and amino acid ionic liquids (AAILs), as next-generation media for CO_2 capture and conversion.

Despite their promise, the performance of these solvents is intricately governed by molecular-level interactions and rare, thermally activated events that remain poorly understood. To bridge this knowledge gap, we employ a comprehensive multiscale simulation framework that integrates quantum mechanical calculations, classical molecular dynamics (MD), and advanced enhanced sampling techniques. This hierarchy allows us to investigate both the electronic structure-level mechanisms of CO_2 activation and proton transfer, and the condensed-phase solvent dynamics that modulate reactivity and transport under realistic thermodynamic conditions.

Our studies[1,2] focus on two key phenomena: (1) proton transfer processes within hydrogen-bond-rich environments, which are essential to many CO_2 conversion pathways, and (2) the influence of solvent microstructure, including cation-anion interactions and hydrogen bond donor–acceptor motifs, on the thermodynamics and kinetics of CO_2 activation. In DES, we investigate how the presence of multiple hydrogen bond networks affects CO_2 solubility, reactivity, and speciation (e.g., formation of carbamate, bicarbonate, or carbonate species). In AAILs, we study how amino acid side chains and ionic coordination environments modulate proton mobility and facilitate or hinder reactive pathways. These effects are captured using both reactive force fields and electronic structure-based dynamics (e.g., DFTB and GFN2-xTB), enabling us to bridge the atomistic accuracy of quantum chemistry with the larger system sizes and timescales accessible via classical approaches.

Our results[1,2] reveal that rare events such as proton hopping, zwitterion formation, and ring-opening/closure reactions play pivotal roles in determining the overall efficacy of CO_2 conversion in these media. Importantly, we show that the rate-limiting steps are often not simple diffusion or solvation but rather involve complex, collective solvent rearrangements that require an intimate understanding of the local hydrogen-bonding network and dynamic solvent-solute coupling. These findings emphasize the necessity of capturing slow, activated events to design materials with optimized performance under realistic operational conditions.

From a broader perspective, this work contributes directly to the advancement of CCUS technologies by enabling the rational

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design of sustainable solvent systems guided by predictive molecular insights. Our computational framework provides transferable knowledge that can inform the synthesis of new DES and AAIL formulations with tailored properties such as enhanced CO_2 uptake, lower regeneration energy, and selective product formation. Furthermore, our methodology is generalizable to other energy-relevant systems, including electrochemical CO_2 reduction[3], biomass valorization, and gas separation membranes.

Ultimately, this research aims not only to explain how CO_2 interacts with biocompatible solvents at the molecular level but also to elucidate why certain structures outperform others—and how to design better materials. By aligning computational chemistry with sustainable materials discovery, our work contributes directly to the development of scalable, low-energy, and environmentally friendly CCUS systems. These findings offer valuable predictive insights for both academic and industrial communities seeking to advance carbon management technologies in the context of long-term climate mitigation.

References:

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