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Improving the Capture of CO₂ by Substituted Monoethanolamines: Electronic Effects of Fluorine and Methyl Substituents

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Abstract

The influence of electronic and steric effects on the reaction between CO₂ and monoethanolamine (MEA) absorbents is investigated using computational methods. The pK_a of the alkanolamine, the reaction enthalpy for carbamate formation, and the hydrolytic carbamate stability are important factors for the efficiency of CO₂ capture. The steric and electronic effects of CH₃, CH₂F, CHF₂, CF₃, F, dimethyl, difluoro, and bis(2-trifluoromethyl) substituents at the alpha carbon of MEA on this reaction are investigated. Density functional theory (DFT) (B3LYP, M06-2X, M08-HX and M11-L) and ab initio methods [spin component-scaled second-order Moller-Plesset theory (SCS-MP2), G3], each coupled with solvent models [conductor-like polarizable continuum model (CPCM) and universal solvation models (SM8 and SMD)], are shown to yield accurately calculated pK_a values of the substituted MEAs. Specifically, G3, SCS-MP2, and M11-L methods coupled with the SMD and SM8 solvation models perform well with a mean unsigned error (MUE) of only 0.15, 0.24 and 0.25 pK_a units, respectively. SCS-MP2 is used to calculate the reaction enthalpy for carbamate formation and the carbamate stability towards hydrolysis. With the introduction of beta-fluoro substituents (especially the CH₂F moiety) the reaction enthalpy for the formation of carbamates can be fine-tuned to be less exothermic than that using the unsubstituted MEA. This implies a reduced energy requirement for the solvent-regeneration step in the post-combustion carbon-capture method, which is currently the energy-limiting step in efficient CO₂ capture. beta-Fluoro-substituted MEAs are also shown to form less stable carbamates than MEA. Thus, beta-fluoro-substituted MEAs display a great potential for the use in the post-combustion carbon-capture process. Finally, a clear correlation is observed between the gas-phase basicity and the tendency to form carbamates. This allows for the rapid prediction of which species will be formed experimentally, and thus the CO₂-absorbing capacities of alkanolamines can be estimated.

Keywords

Author Keywords: alkanolamines; basicity; carbamates; carbon dioxide fixation; computational chemistry

KeyWords Plus: MAIN-GROUP THERMOCHEMISTRY; FREE-ENERGY PERTURBATIONS; GAS-PHASE BASICITIES; AB-INITIO; NONCOVALENT INTERACTIONS; CARBON-DIOXIDE; AMINES; MOLECULES; ABSORPTION; ACCURACY

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