

2P04

Theoretical modeling of the removal of carbon dioxide by absorption in aqueous alkanolamines solutions

○Mohamed Ismael¹、源谷 夏奈¹、岩淵 翼¹、坪井 秀行¹、古山通久¹、畠山 望¹、遠藤 明¹、高 羽洋充¹、久保 百司¹、Carlos Del Carpio¹、清水信吉²、宮本 明^{1,3}

¹東北大学大学院工学研究科応用化学専攻(〒980-8579 仙台市青葉区荒巻字青葉 6-6-11-1302)

²財団法人地球環境産業技術研究機構(〒619-0292 京都府相楽郡木津町木津川台 9-2)

³東北大未来未来科学技術共同研究センター(〒980-8579 仙台市青葉区荒巻字青葉 6-6-10)

【Introduction】

In the gas absorption industry, carbon dioxide removal from gaseous mixtures with alkanolamine solutions is classified as adiabatic gas absorption. Recently, aminomethylpropanol (AMP) solution has been introduced as commercially attractive over conventional amines, because it has a loading capacity of up to 1 mol of CO₂ per mole of AMP, and it has excellent absorption characteristics, superior stripping qualities, higher degradation resistance, and a lower corrosion rate [1]. In this study, theoretical calculations were done to find out the probable mechanism of the reaction of CO₂ and AMP.

【Method】

The CO₂/alkanolamine bulk interaction was studied using our in house molecular dynamics (MD) program NEW-RYUDO. DMol³ code based on the density functional theory was used for studying the reaction of CO₂ and AMP in gas and aqueous phases by applying the COSMO method. Moreover one water molecule was introduced to the reaction model to investigate the role of base in the reaction.

【Results and Discussion】

Optimization of (AMP-CO₂) complex in a gas phase shows no interaction as the distance between the CO₂ carbon and the amine nitrogen increases to 2.47Å. By addition of one water molecule to the model it can be observed that this water molecule influences the interaction between the amine and CO₂ because it decreases the amine nitrogen and CO₂ carbon distance to 1.62Å as shown in Fig. 1. These results strongly suggest that, for the CO₂ to react with an amine center, basic solvent molecules must be present. According to our results the energies of the reactants, products, and intermediates were found to be more stable in the COSMO solvation models than the gas phase. When the formation energies for the carbamic acids of different alkanolamines are compared, it can be seen that the order of stability is (primary amine > secondary amine) as shown in Table 1. The aim of the MD simulation was to investigate the kinetic interaction through the absorption process (e.g. the proton transferring while the carbamate product formed).

Table 1. Formation energies for the formation of carbamic acids.

Compound	ΔH_f (kcal/mol)	
	Gas phase	Aqueous phase
Monoethanolamine	-4.38	-7.70
Aminomethylpropanol	6.84	-3.62
Methylaminoethanol	-4.53	-6.97
Propylaminoethanol	-4.79	-7.01
Ethylaminoethanol	-4.46	-0.50

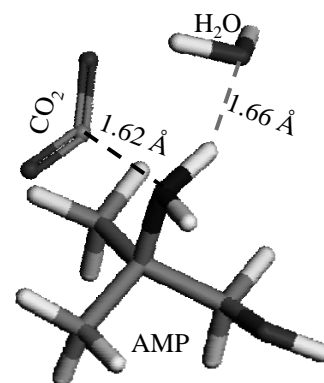


Fig. 1 (AMP-CO₂-H₂O) complex.

Reference

[1] I. Baek *et al.*, *Int. J. Thermophys.*, **21**, 1175 (2000).