

Atmospheric Oxidation of Amines Relevant for Carbon Capture and Storage

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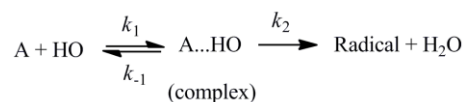
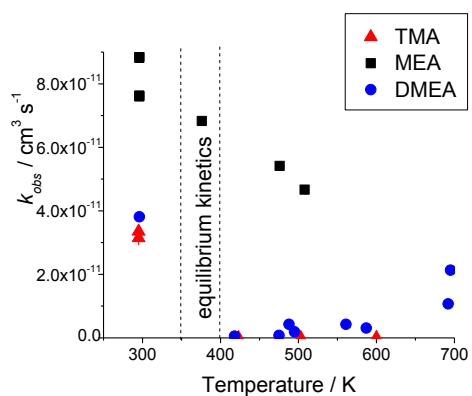
Amine-based compounds have been proposed for capturing CO₂ in Carbon Capture and Storage technology. However, in the case of large-scale CO₂ capture plants, there is potential for significant amine release to the atmosphere.[1] This study aims to fill in gaps in our knowledge about the atmospheric oxidation of such amines.

We have performed laser photolysis, laser induced fluorescence experiments on the reaction of OH radical with a range of amines: monoethanolamine (MEA, H₂NCH₂CH₂OH), dimethylethanolamine (DMEA, (CH₃)₂NCH₂CH₂OH) and trimethylamine (TMA, (CH₃)₃N). We have studied the decay of OH under pseudo-first-order conditions over the temperature range 296 – 700 K (Fig. 1). This is the first study on the temperature effect on these systems. We have found that all the amines initially form a weakly bonded complex with OH, k_1 . In the case of MEA the formation of this complex is followed by a fast OH abstraction to products, k_2 , at all temperatures (see Scheme 1). The value of the rate coefficient for the reaction of OH with MEA at 296 K is in a good agreement with that found very recently by Karl *et al.*[2] The reaction of DMEA with OH at room temperature is similarly characterized by a fast rate coefficient. However, at 380 K, equilibrium is observed, which is strong evidence for complex formation. Above 380 K, the reaction rate coefficient is much slower than at 296 K, but is increasing with temperature. In the case of TMA - OH system we observed similar behaviour except that, above the equilibrium temperature, no reaction is observed. The value of the rate coefficient for TMA - OH reaction at 296 K is in excellent agreement with that of Carl and Crowley.[3]

In order to find the site(s) for abstraction in these reactions between OH and amines we plan to perform further studies, using the same technique, by measuring OH kinetics with partially deuterated amines/methyl analogues and also to carry out product studies.

References

- (1) Thitakamol, B.; Veawab, A.; Aroonwilas, A. *Int.J.Greenh.Gas Control* **2007**, 1, 318-342.
- (2) Karl, M et al. *Atm.Chem.Phys.Disc.* **2011**, 11, 27763-27814.
- (3) Carl, S. A.; Crowley, J. N. *J.Phys.Chem A* **1998**, 102, 8131-8141.



A: MEA, DMEA, TMA

k_2 MEA - large, no overall barrier
 k_2 DMEA - small, small overall barrier
 k_2 TMA - zero, large barrier

Scheme 1

Figure 1. Rate coefficients vs. temperature for the reaction of OH with: (■) monoethanolamine (MEA), (●) dimethylethanolamine (DMEA) and (▲) trimethylamine (TMA).