Modeling the Phase Behavior of Fluid Systems Relevant for Carbon Capture Processes: The Importance of SOx and NOx

Andrew Haslam[1, S, C], Claire Adjiman[1], Amparo Galindo[1] and George Jackson[1]

[1]*Chemical Engineering, Imperial College London, London, United Kingdom*
a.haslam@imperial.ac.uk

Considerable research effort has been expended in recent years to enable accurate, quantitative modeling of the fluid phase equilibria of carbon dioxide in aqueous carbon capture solvents, such as monoethanolamine and other more sophisticated solvents. However, real flue-gas streams contain impurities, such as SO_x and NO_x, which can adversely affect carbon-capture plants, as well as influence the phase equilibria. The development of a modeling capability to assess the impact of these impurities on the thermodynamics of these fluid systems is a necessary precursor to the design and operation of successful carbon capture infrastructure. This work is part of a "world's-first" project to demonstrate a 10t/d CO_2 capture plant using a Rotating-Packed-Bed absorber and desorber with a bespoke designer solvent. We use the state-of-the-art group contribution SAFT-γ Mie equation of state and work within the framework of HiRECORD. We embark on the first step of this task via the development of models for sulphur dioxide (SO_2), nitrogen (II) oxide and nitrogen (IV) oxide, three of the more important SO_x and NO_x in this context. The chemistry of each of these fluids presents challenges to modeling; the behaviour of SO_2 is strongly influenced by the large dipole and quadrupole moments, while the free-radical nature of NO and NO_2 result in both "pure" nitrogen (II) oxide and nitrogen (IV) oxide comprising mixtures of two NO_x: NO + N_2O_2, and NO_2 + N_2O_4 (respectively). We use the developed models to examine the phase equilibria of mixtures relevant for carbon capture. The facility of SAFT-γ Mie to incorporate intermolecular association renders it an ideal tool to address these challenges.