235d CO2 Capture under Elevated Pressures and Temperatures Using Fluorinated Solvents

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The acid gas removal, including CO_2, from Integrated Gasification Combined Cycle (IGCC) power generation facilities has been conventionally carried out using: (1) a chemical process employing methyl-diethanolamine (MDEA) or (2) a physical process utilizing either chilled methanol (Rectisol) or a mixture of dimethylethers of polyetheleneglycol (Selexol). The MDEA process requires high thermal energy (heat) for solvent regeneration. The Rectisol process is complex, and refrigeration makes it the most expensive acid gas removal process. The Selexol process is more expensive than the MDEA process, and the chilling option could increase the process costs. In an IGCC application, these physical and chemical processes for acid gas removal, however, require cooling and subsequent reheating of the stream before the gas turbine which undeniably decreases the plant thermal efficiency and thusly increases the overall cost of the process. Thus, there is a need for the development of an alternative process which should be economical and absorb carbon dioxide without significant cooling of the gas streams.

Extensive literature review revealed that perfluorinated compounds (PFCs) have low reactivity and high chemical stability due to the high energy of their C-F bonds. They have high boiling points and low vapor pressures because of the strength of the C-F bond and the high molecular weight. They also have no dipole and very low molecular interactions due to the repulsive tendency of fluorine atoms. These unique properties lead to high gas solubility, minimum vapor losses, and low forces required for expelling the gas molecules upon decreasing pressure or increasing temperature. Thus, PFCs show an immense potential for CO_2 capture from post-shift fuel gas streams at elevated pressures and temperatures. The main objective of this study is to investigate the potential use of perfluorinated compounds as physical solvents for CO_2 capture from post water-gas-shift reactor streams under elevated pressures and temperatures. In order to achieve this objective an experimental program was devised to obtain the equilibrium gas solubility, and the hydrodynamic and mass transfer parameters (gas holdup, Sauter mean bubble diameter, and volumetric mass transfer coefficient) for CO_2 in three different PFCs, namely Perfluoro-perhydrofluorene ($C_{13}F_{22}$), Perfluoro-perhydrophenanthrene ($C_{14}F_{24}$), and Perfluoro-cyclohexylmethyldecalin ($C_{17}F_{30}$), known as PP10, PP11, and PP25, respectively.

The transient physical gas absorption technique was employed to measure the volumetric mass transfer coefficient; and the gas solubility was determined when the thermodynamic equilibrium was reached in the reactor. The expanded liquid height method and a photographic method were used to obtain the gas holdup and the Sauter mean bubble diameter, respectively. The data were statistically designed and obtained in a gas-inducing, one-gallon Zipper-Clave agitated reactor, equipped with sight-windows in wide ranges of operating conditions: pressures (6 - 30 bar), temperatures (300 - 500 K), mixing speeds (10 - 20 Hz), and liquid heights (0.14 - 0.22 m).

The equilibrium solubilities of CO_2 in PP10, PP11, and PP25 were found to increase with pressure at constant temperatures. The CO_2 solubilities in PP25 appeared to be greater than those in the other two

PFCs due to the fact that its molecular weight is greater than those of the other two PFCs. The results showed that CO_2 is more soluble in the Selexol solvent than in the PFCs only at low temperatures ($\leq$ 333 K). The Selexol process, however, is customarily operating at temperatures of about 312 K, indicating that the Selexol solvent would not be effective at high temperatures typifying those at the exit of the gasifier system. Thus, this study proved the thermal and chemical stability and the ability of the PFCs to absorb CO_2 at temperatures up to 500 K and pressures as high as 30 bar. The volumetric mass transfer coefficients (k_La) of CO_2 in PP10, PP11, and PP25, increased with increasing mixing speed, pressure, and temperature due to the increase of the gas-liquid interfacial area (a) and the liquid-side mass transfer coefficient (k_L). The increase of the gas-liquid interfacial area with these operating variables was attributed to the increase of the gas holdup (ε_G) and the decrease of the Sauter mean bubble diameters (d_S). The volumetric mass transfer coefficients of CO_2 in the three PFCs, however, decreased with increasing liquid height above the impeller due to the decrease of the gas holdup and increase of the Sauter mean bubble diameter, which led to the decrease of the gas-liquid interfacial area. The volumetric mass transfer coefficients for CO_2 in PP25 were smaller than those in PP11, and both were smaller than those in PP10, indicating that the volumetric mass transfer coefficients decrease with increasing the PFCs viscosity. Also, under the operating conditions investigated, the gas-liquid interfacial areas of CO_2 in the three PFCs appeared to control the behavior of the volumetric mass transfer coefficients in the gas-inducing reactor used.