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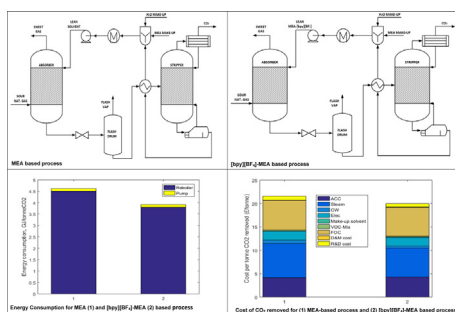
Study of CO₂ removal in natural gas process using mixture of ionic liquid and MEA through process simulation

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GRAPHICAL ABSTRACT



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ABSTRACT

There has been a shift to less carbon intensive fuels such as natural gas to meet energy demand due to increasing pressure to cut CO₂ emissions. This has prompted a need to assess unconventional and contaminated natural gas reserves (which contains CO₂ concentration of 20 mol% or more). The CO₂ capture process with MEA as the solvent is mostly adopted to treat contaminated natural gas. In this study, the option of using a blend of ionic liquids (IL) and MEA as a promising solvent in the process was investigated through modelling and simulation. A detailed rate-based model was developed for both MEA (30 wt%) solvent and IL (30 wt%)-MEA (30 wt%) blend using Aspen Plus® to assess both process and economic performances. The 1-Butylpyridinium ([bpy][BF₄]) ionic liquid was selected in this study. The physiochemical properties of [bpy][BF₄], predicted using Aspen Plus®, showed good accuracy compared with experimental data. The results from this study showed about 15% and 7.44% lower energy consumption in the reboiler duty and CO₂ removal cost respectively with aqueous [bpy][BF₄]-MEA solvent compared to 30 wt% MEA solvent. It is concluded that the aqueous [bpy][BF₄]-MEA solvent is therefore a promising solvent that could replace 30 wt% MEA solvent in this process.

Abbreviations: ACC, Annual Capital Cost; AOC, Annual Operating Cost; [bpy][BF₄], 1-butylpyridinium tetrafluoroborate; [bheaa], Bis(2-hydroxyethyl) ammonium acetate; [bmim][BF₄], 1-butyl-3-methylimidazolium tetrafluoroborate; [bmim][DCA], 1-butyl-3-methylimidazolium dicyanamide; CW, Cooling Water; DEA, Diethanolamine; DHVLB, Heat of Vaporization at T_b; D&M, Distribution and Marketing; Elec, Electricity; ENRTL, Electrolyte Non-Random Two Liquid; FC-CS, The Fragment contribution – corresponding states; FOC, Fixed Operating Cost; IL, Ionic Liquid; IEA, International Energy Agency; LNG, Liquefied Natural Gas; MDEA, Methyl-diethanolamine; MEA, Monoethanolamine; Mis, Miscellaneous; R&D, Research and Development; RK, Redlich Kwong; RKTZRA, Rackett/Campbell-Thodos Mixture Liquid Volume; RTILs, Room temperature ionic liquids; TSILs, Task specific ionic liquids; V_B, Liquid Molar Volume at T_b; VOC, Variable operating cost; VLSTD, Standard liquid Volume

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Nomenclature

$C_{1i}-C_{3i}$	Equation coefficients for (7)
$C'_{1i}-C'_{3i}$	Equation coefficients for (8)
Z_i^{RA} and d_i	Equation coefficients for (9)
A_i-C_i	Equation coefficients for (10)
$C_{1i}-C_{5i}$	Equation coefficients for (11)
D_i-D_{iii}	Equation coefficients for (12)
H_{ij}	Henry Constant
P_i^v	Vapour pressure of component i, Pa
P_c	Critical pressure, bar
Q_{reb}	Reboiler duty, kJ/kg _{co2}
Q_{cond}	Condenser duty, kJ/kg _{co2}
Q_{cooler}	Cooler duty, kJ/kg _{co2}
T	Temperature, K

T_b	Normal Boiling temperature, K
T_c	Critical temperature, K
T_r	Reduced temperature, K
V_c	Critical Volume, cc/mol
W_{pump}	Pump power kJ/kg _{co2}
Z_c	Critical Compressibility factor
σ_i	Surface tension, mN/m
η_i	Liquid Viscosity, cP
ρ_i	Liquid molar density, mol/cc
Ω	Omega
ΔH_f^0	Standard heat of formation, kJ/mol
ΔH_c^0	Standard heat of combustion, kJ/mol
λ_i	Thermal conductivity, kcal-m/hr-m ² -K

1. Introduction

1.1. Background

The need to reduce emissions has favoured a shift towards low carbon fuels such as natural gas for energy generation [1]. It is predicted that a switch to low carbon fuels will contribute about 15% in expected CO₂ emission cuts by 2050 [2]. Globally, mineable natural gas reserves are far smaller in comparison to that of carbon intensive fuels (*i.e.* coal) and as such, natural gas supply is less secured and expensive. This has prompted the need to re-assess the development of unconventional, stranded, contaminated and sour natural gas reserves [3]. However, raw natural gas is known to contain acid gas such as CO₂ with concentration of about 20 mol% and more, which makes these reserves economically unviable. These natural gas reserves are predominantly in SE Asia, NW Australia, Central USA, North Africa and the Middle East [4]. These locations are far from the established gas markets in Western Europe, Japan and South Korea. Thus, a large amount of natural gas must be conveyed either via long distance pipeline or as Liquefied Natural Gas (LNG) [3]. The presence of CO₂ in natural gas limits its quality (heating value) and the liquefaction process performance.

Natural gas sweetening technologies are adopted to remove CO₂, so natural gas can meet acceptable standards for pipeline transport to end-users and/or liquefaction process for LNG [5]. Natural gas sweetening process involves CO₂ separation from the gas mixture using techniques such as physical absorption, chemical absorption, adsorption, cryogenic separation and membrane separation among others [5]. Chemical absorption is the most commonly and widely used separation method in natural gas sweetening processes [6,7]. However, it is expensive especially due to the high energy penalty of the process [8]. Thus, there is a need to explore different options for reducing the high-energy penalty of the process.

1.2. Motivation

Amine-based CO₂ absorption/desorption process has been in use for decades in the industry for CO₂ removal from gas mixtures such as natural gas among others [3,6,9,10]. This is primarily due to its relatively rapid kinetics of the amine solvents [3,6]. However, amine solvents generally require high energy for regeneration. They also tend to stimulate equipment corrosion and degrade rapidly during operation. This makes the operating cost for amine-based process generally high

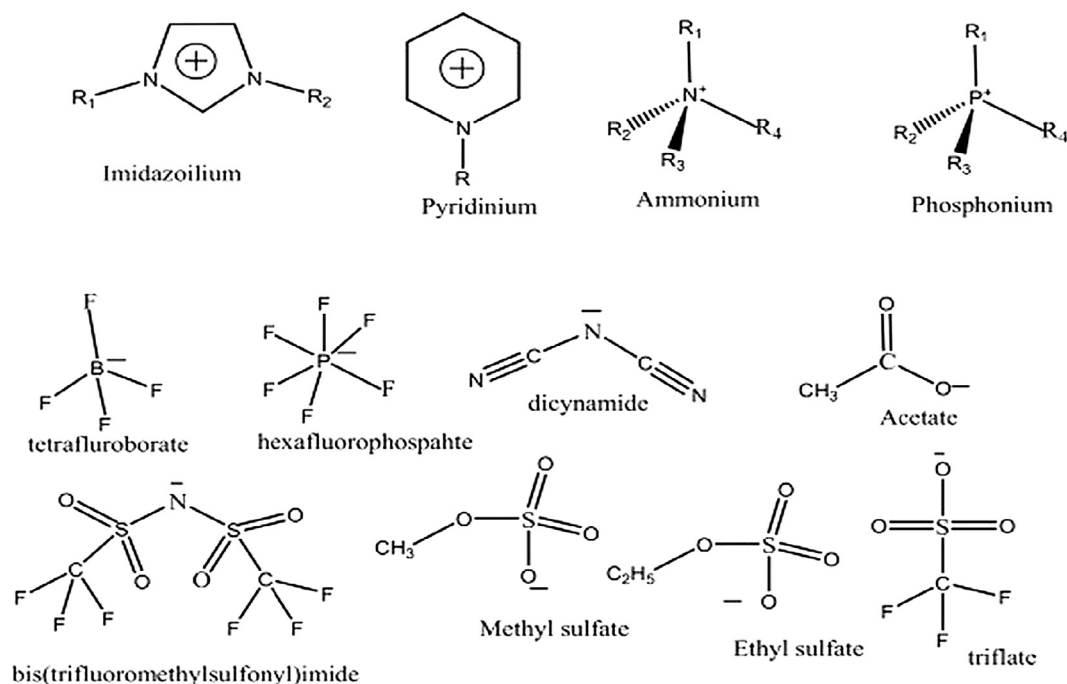


Fig. 1. General Structure of various cations and anions used for Ionic liquid formulation [5].

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