

An extractive distillation technique for producing CO₂ enriched injection gas in enhanced oil recovery (EOR) fields

B. ZareNezhad^{a,*}, N. Hosseinpour^b

^aSchool of Chemical, Petroleum and Gas Engineering, Semnan University, P.O. Box 35195-363, Semnan, Iran

^bSchool of Chemical Engineering, University of Tehran, P.O. Box 11155/4563, Tehran, Iran

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ABSTRACT

Sharp Separation of CO₂ (carbon dioxide) from CO₂-rich gases issuing from enhanced oil recovery (EOR) fields is investigated by different processing alternatives. A new extractive distillation technique without using an external solvent is introduced for debottlenecking of CO₂ separation problem encountered in cryogenic distillation units. The technique involves the addition of a portion of produced C₄⁺ (NGL) stream (natural gas liquid stream comprising iso-butane and heavier components) to the top section of a CO₂ stripper followed by C₂/C₃ (ethane and propane) stripping of recovered C₂⁺ (ethane and heavier components) stream. Detail description of the proposed technique for producing greater than 96% CO₂-rich injection gas without external solvent is presented. According to the proposed approach, high quality C₂/C₃ and C₄⁺ products with the corresponding purities of 89.6% and 99.6% suitable as feedstocks for gas and petrochemical processing units are also obtained. Sharp separation of CO₂ from recovered hydrocarbon mixtures by the proposed technique and recycling it to petroleum reservoirs is an important issue regarding the optimization of EOR processes and greenhouse gas emission control.

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1. Introduction

In order to increase oil production in many reservoirs, the injection of gas for enhanced oil recovery (EOR) has been carried out in numerous projects [1]. The gas injected may be CO₂ with little or no hydrocarbons. Thus large volumes of CO₂ are produced with the oil and gas which must be handled in the gas processing facilities. The CO₂ that is injected into the reservoir is typically purchased from third party suppliers and is the single greatest operating cost in the EOR project [2]. Therefore, the CO₂ produced with the associated gas is valuable and must be recovered and recycled to the reservoir.

The CO₂ produced in an EOR project can be separated from the hydrocarbon components using solvent or membrane processes. However, solvent processes such as amines, potassium carbonate, and physical solvents, as well as membrane systems, were not designed to handle the large volumes of CO₂ which are present in the EOR gas. The capital and operating costs of these systems increase in proportion to the acid gas content [3]. Additionally, the produced CO₂ through the solvent processes is at low pressure and typically saturated with water. The EOR project needs high pressure, dry CO₂ for reinjection [4]. Fractionation based on cryogenic distillation process is a useful alternative for treating high CO₂

gas stream associated with CO₂ miscible flood. In a typical cryogenic system, methane is produced as the overhead product of a demethanizer distillation column [5]. Carbon dioxide plus ethane and heavier hydrocarbons are also produced as the demethanizer bottom product. CO₂ is then separated from the remaining hydrocarbons in the second distillation column (CO₂ stripper) [6]. Unfortunately, ethane and CO₂ form an azeotrope such that a sharp separation can not be obtained [7].

In the initial part of this work, the separation of CO₂ from a CO₂ rich feed (e.g. demethanizer bottom stream) by employing cryogenic distillation process is investigated. The process is then modified for sharp separation of CO₂, C₂/C₃ and C₄⁺ from the incoming feed by using a new extractive distillation process without using an external solvent.

2. Modeling

In this study, the cryogenic (Fig. 1) and extractive (Fig. 2) distillation processes are simulated using a rigorous tray-by-tray method. Given the column configuration, a steady state solution may be computed that satisfies basic equations describing the column behavior. Different set of equations, including overall material balances [8], component material balances, phase equilibrium relations, enthalpy balances [9] and performance specifications are simultaneously solved by employing the Newton–Raphson method [10].

* Corresponding author.

E-mail address: zarenezhad@yahoo.com (B. ZareNezhad).

Nomenclature

CO ₂	carbon dioxide
C ₂	ethane
C ₂ ⁺	ethane and heavier components
C ₃	propane
C ₃ ⁺	propane and heavier components
C ₂ /C ₃	mixture of ethane and propane
C ₄ ⁺	iso-butane and heavier components
EOR	enhanced oil recovery
NGL	natural gas liquid
J	square jacobian matrix of order n
ΔX	conformable column vector of corrections
X	conformable column vector of independent variables
f	conformable column vector of material and energy balances functions
A	n by n matrix defined in Eq. (2)
R , R_1 and R_2	n by m matrices defined in Eq. (2)
I	identity matrix
k	iteration number

$\ f_k\ $	Euclidean norm of vector f at iteration k
N	number of stages in the CO ₂ stripper

Superscript

-1	inverse of a matrix
T	transpose of a matrix

Subscript

k	iteration number
m	order of an identity matrix

Greek symbol

\sum	summation
λ	a parameter between 0 and 1 defined in Eq. (5)
α	relative volatility of CO ₂ with respect to ethane at each stage
$\alpha_{average}$	average relative volatility of the CO ₂ stripper ($\sum \alpha/N$)

The complete set of Newton–Raphson equations may be stated in the following matrix form [11]:

$$J_k \Delta X_k = -f_k \quad (1)$$

where J_k is the square jacobian matrix of order n and ΔX_k and f_k are conformable column vectors. The stage temperature and interstage component molar flow rates are the tear variables. In this work, an efficient algorithm suitable for systems which contain recycle streams is used for solving Eq. (1). This can be derived by writing the matrix J as follows:

$$J = A + R = A + R_1 I_m R_2^T \quad (2)$$

where A is an n by n matrix, R_1 and R_2 are of order $n \times m$ and I_m is an identity matrix of order $m \times m$. Thus we have:

$$\Delta X = [A + R_1 I_m R_2^T]^{-1} (-f) \quad (3)$$

Application of Householder's identity [12] gives:

$$\Delta X = [A^{-1} - A^{-1} R_1 (I_m^{-1} + R_2^T A^{-1} R_1)^{-1} R_2^T A^{-1}] (-f) \quad (4)$$

To improve the convergence properties, the calculated correction ΔX_k can be modified as:

$$X_{k+1} = X_k + \lambda \Delta X_k \quad (5)$$

The value of λ (between 0 and 1) is chosen to produce a decrease in the Euclidean norm, e.g. in the sum of squares:

$$\|f_{k+1}\| < \|f_k\| \quad (6)$$

For the case of extractive distillation which involves a recycle stream between two or more columns, the above approach leads to a rapid convergence.

The Peng–Robinson equation of state with optimum values of binary interaction parameters is used for phase equilibrium calculations [13]. The logic diagram of the simulation algorithm for the proposed extractive distillation technique is displayed in Fig. 3. A solution of the problem is obtained when the CO₂ concentration of the CO₂ stripper overhead is at least 95 mol% and the C₃ concentration of the C₂/C₃ stripper bottom is less than 0.5 mol%.

3. Results and discussion

Process flow diagram of a conventional cryogenic distillation for CO₂ separation is shown in Fig. 1. The first column (demethanizer) is used to facilitate the separation of methane from the CO₂ and less-volatile constituents, rejecting CO₂ and C₂⁺ to the next column. The objective of the second column (CO₂ stripper) is to produce a CO₂-rich stream overhead at cryogenic condition and reject ethane

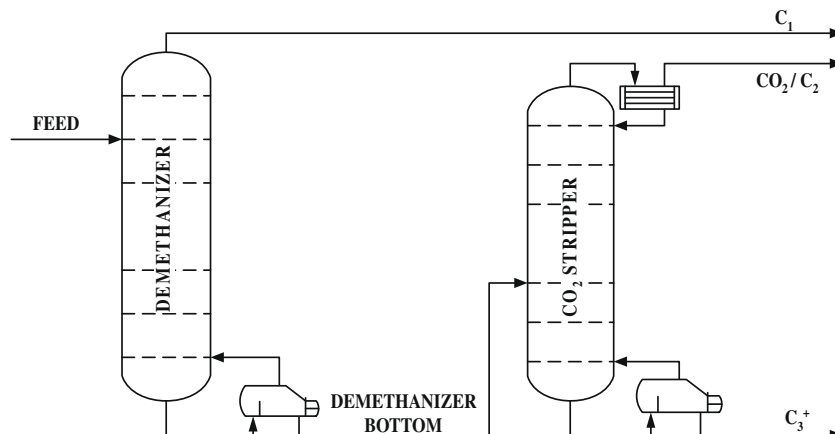


Fig. 1. Process flow diagram of a conventional cryogenic distillation unit for CO₂ separation.

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