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Synthesis of Conventional and Hybrid Cryogenic Distillation Sequence for Purification of Natural Gas

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Abstract: Synthesis of efficient cryogenic distillation sequence for purifications of natural gas having medium and high concentration of carbon dioxide content, has been explored in the present study. Calculations were performed for conventional and hybrid distillation column sequences by using the heuristic and evolutionary strategies. Different feed compositions were selected from literature and the possible sequence for different feed composition is computed. Three different sequences, namely direct, in-direct and mixed were chosen for each of the feed. Total minimum vapor flow and marginal vapor flow calculations were performed for each of the sequences. Capital and energy requirements were compared to show the advantage of hybrid network over conventional cryogenic network.

Key words: Process synthesis, cryogenic CO_2 separation, hybrid cryogenic network, distillation column network

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INTRODUCTION

Natural gas is one of the main sources for the energy production in the world. The increased use of natural gas will force the development of low quality reserves. The exploitation of these reserves has not yet made because of the economic, technical and environmental reasons. Over 13 tscf of hydrocarbon gas remains undeveloped in high CO_2 fields in Malaysia and CO_2 content in individual field ranges from 28-87% (Darman and Harum, 2006). Currently available conventional gas treatment technologies are unable to make use of these reserves economically. The heavy hydrocarbon contents may also vary in each gas field. Different configurations may be suitable for different amount of carbon dioxide, present in the feed and also the amount of heavy hydrocarbons could be an important factor for finding the optimum process sequence. Synthesis of process flowsheet for the different concentration of carbon dioxide present in the feed is a challenging work and needs attention.

Distillation is one of the oldest and developed separation process. Cryogenic distillation is one of the suitable process for the separation of carbon dioxide from methane. Substantial research focus was not given to cryogenic processing because of common perception about higher energy requirements. Though different processes have been reported in literature (Mullick *et al*., 2013) for the cryogenic separation of carbon dioxide from natural gas, research work needs to be carried out to make this process economical by minimizing the energy requirements. Abulhassan *et al*. (2013) reported the use of desublimation based countercurrent cryogenic switched beds for minimization of energy. A hybrid cryogenic network was proposed by Maqsood *et al*. (2013, 2014) and Mullick *et al*. (2013) for the purification of natural gas with high carbon dioxide content. In this study the conventional cryogenic distillation sequence along with the hybrid network have been studied for the optimal solution of carbon dioxide separation.

The synthesis of distillation sequence for multicomponent separation is often a basic step in the development of flowsheets. Alternative configurations of distillation columns may be used for separating multicomponent mixtures. Systematic synthesis of multicomponent separation sequence into different products of comparatively high purity is an important process design problem. Different methods have been proposed for synthesis of distillation sequences. It can be divided into three different categories: (1) Heuristic approach, (2) Evolutionary strategies and (3) Algorithmic techniques (Nishida *et al*., 1981).

Heuristics are normally used for the process synthesis steps (Malone *et al*., 1985). Earliest attempt for the optimum distillation sequence was made by Lockhart (1947). A distillation column network for the separation of three products of natural gasoline was synthesized.

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Harbert (1957) considered energy as the main economic parameter and a key requirement for optimum distillation network. Rod and Marek (1959) emphasized on the use of total vapor flow rate as main economic factor. A strategy of heuristics with different weightage was proposed by Powers (1972). Several researchers (Doukas and Luyben, 1978; Freshwater and Henry, 1975; Mahalec and Motard, 1977; Siirola *et al*., 1971; Thompson and King, 1972) suggested heuristics rule sets for the optimum sequences of distillation network. Based on reported literature (Biegler *et al*., 1997), the most useful heuristics are employed. The heuristics are listed in order of importance for the present study.

The separation sequence was designed to handle easy splits (i.e., those having the largest relative volatilities) first in the sequence. The next split targeted to the removal of the major component. Subsequently the most volatile component (i.e., choose the direct sequence) is removed. Finally the species leading to desired products appeared in a distillate product in the sequence. These heuristics did not require column design and costing. Sometimes these heuristics are conflicting with each other, such as "removal of the lightest component first," that depends on the volatility and also "removal of the most plentiful component first," depend only on feed composition. In this case, cost and other factors needed to be considered for the development of distillation network. Column design and cost estimation often leads to the evolution of most preferred sequence. Different methods are proposed by researchers (Gomez and Seader, 1976; Hendry and Hughes, 1972; Rodrigo *et al*., 1975; Seader and Westerberg, 1977). A marginal vapor rate (MY) method was proposed by Modi and Westerberg (1992). This method can be used without requiring complete column design and cost calculations and can provide satisfactory results (Seider *et al*., 2004).

CONVENTIONAL DISTILLATION NETWORK

For conventional distillation network three different feed compositions were selected: feed with medium content of carbon dioxide, feed with high contents of carbon dioxide and feed with higher contents of heavy hydrocarbons. The composition of feeds and flow rates are provided in Table 1. Three different configurations for the distillation column sequence have been selected as direct, in-direct and mixed sequence. Figure 1 shows the different configurations of the distillation column sequences. Firstly, heuristics were used to find the best combination of distillation columns. Secondly, calculation were performed for finding the minimum vapor flow and marginal vapor flow in the each column by the method provided by Biegler *et al*. (1997).

Table 1: Different feed compositions (mol fr.)

Component	Medium CO_2 (Berstad *et al*., 2012)	High CO_2 (Darman and Harum, 2006)	Higher H/C (Darman and Harum, 2006)
N_2	0.005	0.005	0.07
CH4	0.397	0.20	0.47
CO2	0.506	0.72	0.30
C_2H_6	0.035	0.025	0.04
C_3H_8	0.024	0.015	0.04
C_4H_{10}-02	0.009	0.01	0.03
C_4H_{10}-01	0.009	0.01	0.03
C_5H_{12}-02	0.006	0.005	0.03
C_5H_{12}-01	0.006	0.005	0.03
C_6H_{14}	0.002	0.005	0.02
Flow (kmol h^{-1})	24800	24800	24800

Marginal vapor flow rate in the columns are calculated using the underwood's method by using these three equations:

$$\sum_i \frac{\alpha_{ik}}{\alpha_{ik} - \phi} f_i = \sum_i \frac{\alpha_{ik}}{\alpha_{ik} - \phi} d_i + \sum_i \frac{\alpha_{ik}}{\alpha_{ik} - \phi} b_i = (1 - q)F \tag{1}$$

$$(R_{min} + 1)D = \sum_i \frac{\alpha_{ik}}{\alpha_{ik} - \phi} d_i = v_{min} \tag{2}$$

By using Eq. 1 and 2:

$$V_{min} = \sum_i \frac{\alpha_{ik}}{\alpha_{ik} - \phi} d_i = (1 - q)F - \sum_i \frac{\alpha_{ik}}{\alpha_{ik} - \phi} b_i \tag{3}$$

where, V_{min} is the sum of terms of species that exists in distillate and bottom.

Calculations were performed for each of the sequence and columns by following these equations and results are presented in Table 2 and 3 for conventional distillation sequence.

The best combination of distillation column was also found to be direct by other two approaches i.e., by minimum vapor flow and marginal vapor flow in the columns (Table 2 and 3). The minimum vapor flow is 50110, 113782 and 80848 kmol h^{-1} for 50% CO_2 in direct, in-direct and mixed combination, respectively. This trend is same for the 72% and higher hydro carbon feeds also. For the marginal vapor flows in the columns direct sequence has the minimum marginal vapor flow as 3738 kmol h^{-1}, whereas in-direct sequence has the maximum marginal vapor flow of 70840 kmol h^{-1} and mixed flow is always between these two. These results are because of the higher mole fractions of methane and carbon dioxide in the feed.

V_{min} in the direct sequence is minimum with higher composition of carbon dioxide. For the lower hydrocarbon feed V_{min} value for the direct and mixed sequence is very close. As the composition of the carbon dioxide decreases

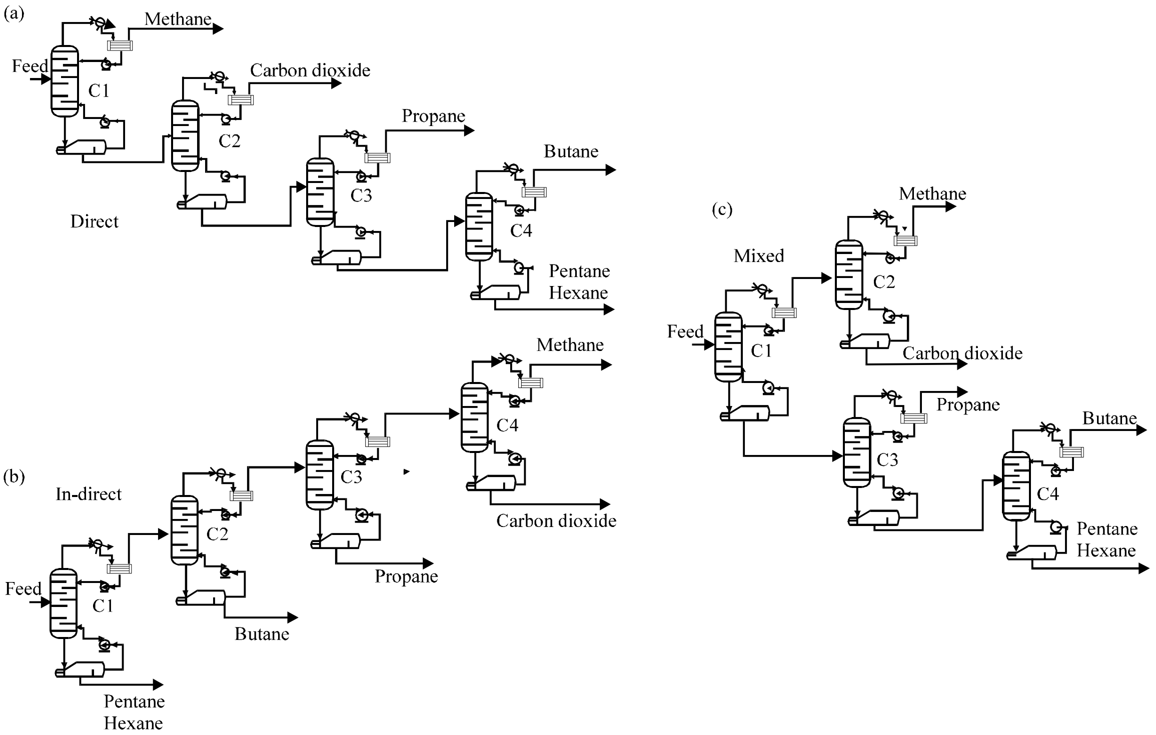

Fig. 1(a-c): Different cryogenic distillation sequences, (a) Direct, (b) In-direct and (c) Mixed

Table 2: Minimum vapor flow (kmol h^{-1}) in columns for conventional distillation sequence

	50% CO_2			72% CO_2			Heavy H/C		
Column	Direct	In-direct	Mixed	Direct	In-direct	Mixed	Direct	In-direct	Mixed
1	30325	26924	48058	11881	27277	53911	30973	26172	39604
2	17310	28678	30287	25667	28541	24480	13776	24781	16697
3	1349	30646	1329	635	28387	642	2332	24268	1684
4	1125	27533	1174	1183	25615	1283	3861	15060	3896
Total	50110	113782	80848	39367	109819	80316	50942	90280	61880

Table 3: Marginal vapor flow (kmol h^{-1}) in columns for conventional distillation sequence

	50% CO_2			72% CO_2			Heavy H/C		
Column	Direct	In-direct	Mixed	Direct	In-direct	Mixed	Direct	In-direct	Mixed
1	1102	27335	17304	620	27797	9884	1138	27275	23289
2	1264	28839	1155	1308	30125	888	3447	24627	3556
3	508	13581	528	545	7033	583	2319	16340	2176
4	864	1085	904	931	922	1027	3578	1198	3556
Total	3738	70840	19890	3403	65877	12381	10481	69440	32577

Table 4: Energy usage (MW) for conventional distillation sequences

	50% CO_2			72% CO_2			Heavy H/C		
Column	Direct	In-direct	Mixed	Direct	In-direct	Mixed	Direct	In-direct	Mixed
1	84	582	211	76	568	221	70	585	235
2	132	304	89	177	299	56	104	321	47
3	10	350	10	8	343	5	19	326	13
4	10	74	10	6	78	13	33	56	34
Total	235	1311	320	266	1288	294	227	1287	330

in the feed, mixed sequence may become the better choice. In case of the MV flow the value is minimum for the high content carbon dioxide feed. As the amount of carbon dioxide decreases in the feed, the MV flow increases in the direct sequence. The MV flow for the mixed sequence is also lowest for 72% carbon dioxide contents. Energy calculations has been conducted for the each case and is presented in Table 4. It is evident from the Table 4 that

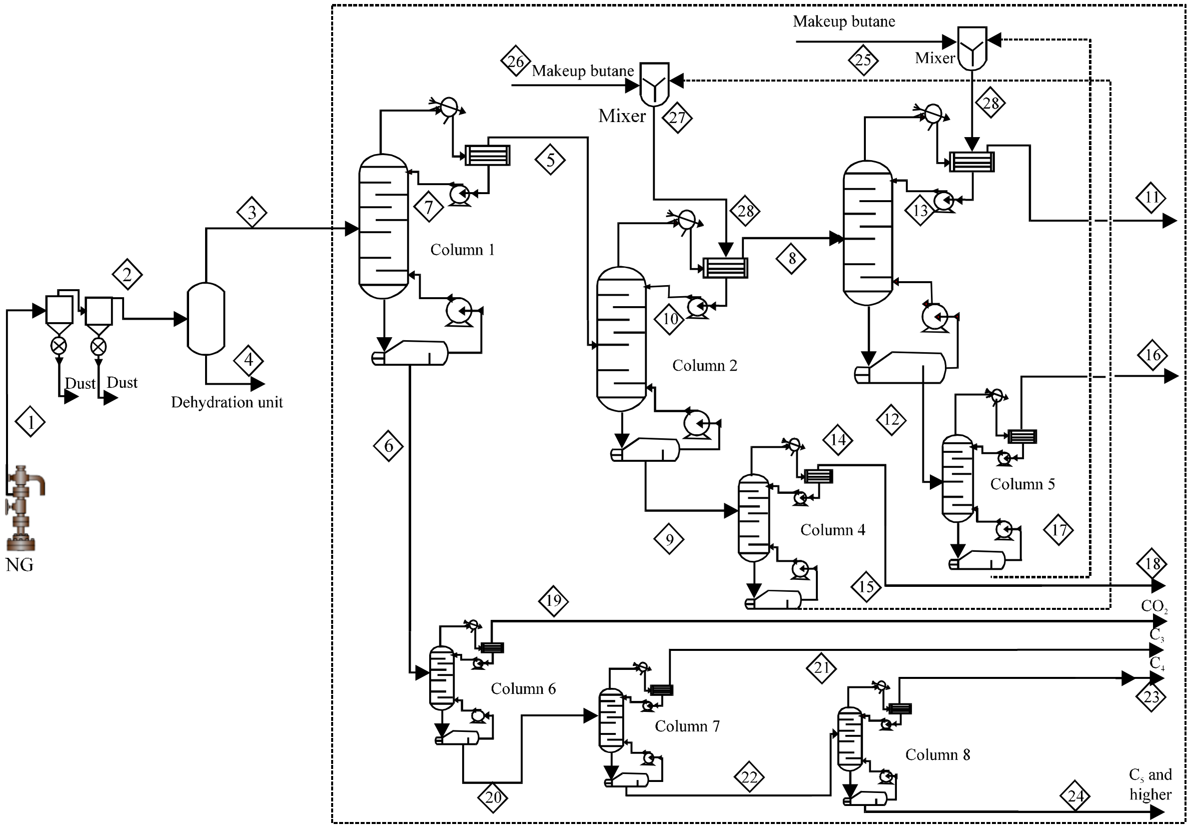

Fig. 2: Schematic diagram of conventional cryogenic distillation process for natural gas purification

energy requirement is minimum for the direct sequence and maximum for in-direct sequences, it is same in the case of minimum vapor flow and marginal vapor flows in the each distillation column sequence. Total energy requirement is minimum for direct sequence for all the feed compositions in conventional distillation column sequences. Indirect sequence is required almost the 5 times more energy than the direct sequence for each of the cases.

The energy requirements for the direct sequence for 72% CO_2 feed is the highest due to the energy requirements for the separation of CO_2 from hydrocarbons. After that is 50% CO_2 and high hydrocarbon feed, which contains 30% carbon dioxide.

This shows that with the increase of the hydrocarbons content in the feed the energy requirement as well as marginal flow rate decreases in the columns for the conventional distillation column sequences for direct sequencing.

Results show that direct sequence is better than other than two sequences in respect of vapor flows and energy requirements. For the purification of methane gas to the pipeline specifications, it was needed to add more distillation columns. The final schematic diagram of conventional distillation column is shown in Fig. 2.

HYBRID CRYOGENIC DISTILLATION NETWORK

A hybrid system can be defined as a process system that involves different unit operations interlinked and optimized as one network to achieve a specified task. A hybrid cryogenic network for the purification of natural gas is presented in the current study. This hybrid cryogenic network is a combination of conventional cryogenic distillation network and the multiple cryogenic packed bed separators proposed by Mohamad (2012) and Abulhassan *et al.* (2013).

Hybrid system is a combination of cryogenic packed beds and conventional distillation column as described by Abulhassan *et al.* (2013). Hybrid network maximizes the benefits of both desublimation or solid-vapor based separation as well as distillation or vapor-liquid equilibrium based separation during the separation of carbon dioxide from methane or natural gas. The amount of CO_2 solidification had been calculated by the experimental data provided by Pikaar (1961) and Donnelly

Table 5: Minimum vapor flow (kmol h^{-1}) in columns for hybrid sequence

	50% CO_2			72% CO_2			Heavy H/C		
Column	Direct	In-direct	Mixed	Direct	In-direct	Mixed	Direct	In-direct	Mixed
1	17935	14094	15772	6993	9431	9692	27709	18796	18587
2	1974	14214	12832	2102	9471	6488	1450	17587	18545
3	2251	13270	1608	1449	8580	731	4179	15125	2571
4	1176	13287	1196	1274	9504	1203	3968	15740	3410
Total	23337	54865	31408	11817	36986	18115	37305	67249	43114

Table 6: Marginal vapor flow (kmol h^{-1}) in columns for hybrid sequence

	50% CO_2			72% CO_2			Heavy H/C		
Column	Direct	In-direct	Mixed	Direct	In-direct	Mixed	Direct	In-direct	Mixed
1	941	14432	19226	612	9244	12811	2035	19607	22779
2	634	13714	845	609	9694	690	1758	16774	1060
3	1271	15676	519	924	10217	531	3208	17463	2219
4	957	815	938	1073	676	1016	3759	1041	3150
Total	3802	44638	21528	3218	29831	15048	10760	54886	29208

Table 7: Energy usage (MW) for hybrid sequence

	50% CO_2			72% CO_2			Heavy H/C		
Column	Direct	In-direct	Mixed	Direct	In-direct	Mixed	Direct	In-direct	Mixed
1	36	348	175	24	195	124	70	444	172
2	13	214	32	16	120	28	9	249	28
3	25	165	12	13	130	5	35	227	23
4	10	25	10	12	25	13	34	27	31
Total	84	752	230	64	470	170	148	947	254

and Katz (1954). This data by Pikaar (1961) is available in GPSA, Engineering Data Book (GPSA, 2012).

For the hybrid case, the calculation of carbon dioxide coming out after the packed bed has been calculated on the basis of thermodynamic data provided by Pikaar (1961).

Vapor flow and energy calculations for hybrid cryogenic network: The values of minimum vapor flow and marginal vapor flow were calculated along with the energy requirements in each case and presented in Table 5-7. Energy requirements for the packed bed provided in Table 8. As similar to the conventional case, the minimum vapor flow and marginal vapor flow along with the energy requirements were found to be lower in the direct sequence. The V_{min} and marginal vapor flow for 72% CO_2 feed found to be the lowest.

The total energy for the hybrid network can be calculated by adding the energy requirement provided in Table 8. Both minimum vapor flow and marginal vapor flow shows direct sequence as the best and this can be concluded from the energy requirements as well. The energy requirement for the direct sequence were found 84, 64 and 148 MW.

Total energy requirement for the 50% CO_2 feed in direct sequence was found to be lowest, 100 MW for the packed bed and 84 MW for the separation of hydrocarbons purification. The total energy requirement in all the cases for the hybrid network was significantly lower as compared to the conventional distillation column. Energy requirement for hybrid system is 184 MW as compared to conventional network that was found to be 235 MW. Similarly in case of 50% CO_2, the energy reduced to 206 MW as compared to the conventional network that was 217 MW.

Table 8: Energy usage (MW) in cryogenic packed bed

50% CO_2	72% CO_2	Heavy H/C
100	142	59

This energy requirement is significantly less as compared to the other sequences. The packed bed energy requirement was considered same in all the sequences and provided in Table 8.

The required purity of methane needs further processing and more distillation columns are needed to bring the CO_2 percentage to pipeline specifications i.e., <2%. This will increase the energy cost further in conventional cryogenic network. In the hybrid network the amount of CO_2 left after the first distillation column is very small and the energy required to separate this is significantly lower as compared to the conventional distillation column.

The schematic diagram of the hybrid cryogenic network is shown in Fig. 3.

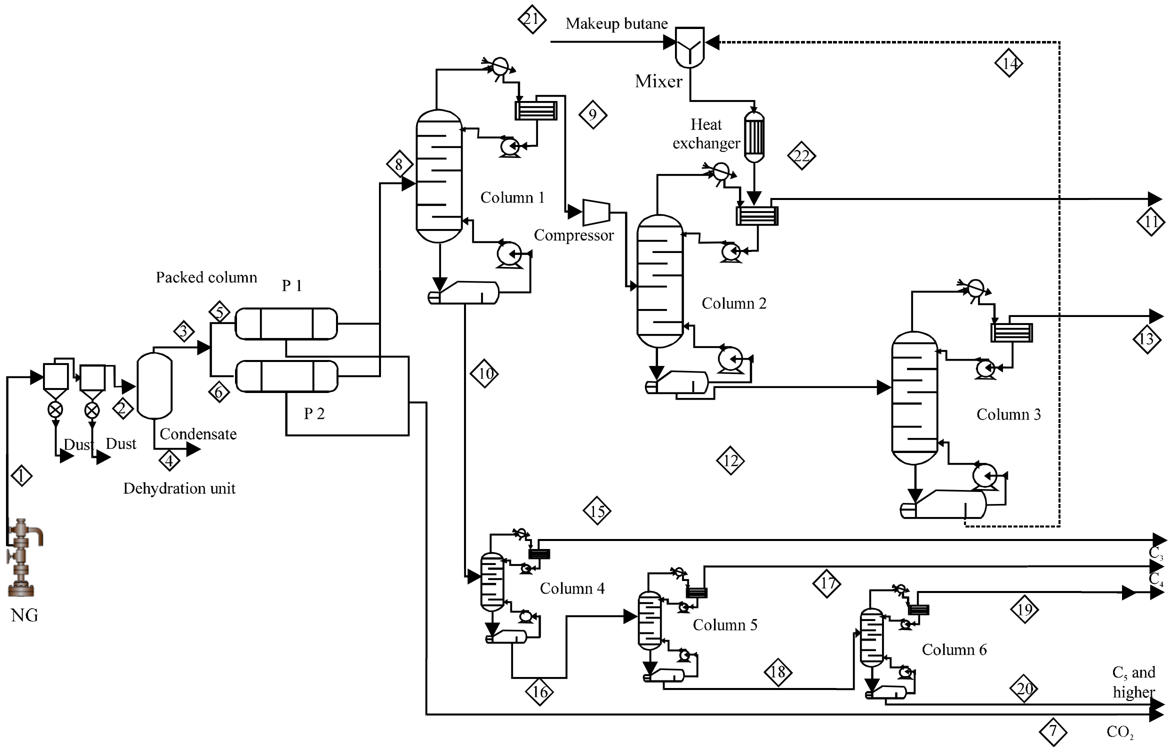

Fig. 3: Schematic diagram of hybrid cryogenic distillation network

Table 9: Column diameter (m) for conventional distillation sequence

	50% CO_2			72% CO_2			Heavy H/C		
Column	Direct	In-direct	Mixed	Direct	In-direct	Mixed	Direct	In-direct	Mixed
1	6.52	18.24	17.54	5.87	18.51	14.52	5.77	18.48	15.94
2	11.61	13.38	6.26	13.93	13.27	5.83	9.54	13.33	5.22
3	2.53	12.17	2.69	2.76	12.04	2.10	3.84	11.53	3.92
4	2.58	7.28	2.72	2.85	6.49	2.96	4.86	5.65	5.25

Table 10: Capital cost (M$) for conventional distillation sequence

	50% CO_2			72% CO_2			Heavy H/C		
Column	Direct	In-direct	Mixed	Direct	In-direct	Mixed	Direct	In-direct	Mixed
1	4.26	45.75	42.36	3.52	47.09	29.21	3.42	46.97	35.09
2	18.90	24.92	3.95	26.95	24.50	3.48	12.97	24.74	2.87
3	1.48	20.73	1.59	1.64	20.30	1.21	2.62	18.67	2.69
4	1.51	5.23	1.61	1.71	4.22	1.80	3.82	3.29	4.36
Total	26.14	96.63	49.52	33.82	96.12	35.70	22.82	93.68	45.01

CAPITAL COST ESTIMATION

The cost of columns and packed columns are calculated by the correlations published by National Energy Technology Center (Loh *et al.*, 2002) and subsequently updated with Chemical Engineering Plant Cost Index (CEPCI) for 2013. Material of construction has been assumed as stainless steel. The accuracy of capital cost for the network using conceptual cost estimation has an accuracy of 40% as reported in recent literature (Tuinier *et al.*, 2011). The diameters and capital costs of the major equipment in conventional distillation column sequences are provided in Table 9 and 10. In case of hybrid sequence, the diameters and costs of the major equipment is shown in Table 11 and 12. It is evident from Table 9 and 11 that there is a significant reduction of distillation column diameters with hybrid cryogenic network and the subsequent reduction in cost is possible for each of the feed composition. For a feed containing 50% CO_2 the capital cost reduces to

Table 11: Column diameter (m) for hybrid sequence

	50% CO_2			72% CO_2			Heavy H/C		
Column	Direct	In-direct	Mixed	Direct	In-direct	Mixed	Direct	In-direct	Mixed
Packed	0.72	0.72	0.72	0.71	0.71	0.71	0.74	0.74	0.74
1	5.64	14.44	9.78	3.58	10.70	9.18	6.55	16.80	12.65
2	2.98	11.13	5.09	3.21	8.34	3.67	3.80	12.03	5.34
3	4.00	9.87	2.65	2.87	8.05	2.07	5.03	10.41	4.21
4	2.13	5.11	2.70	2.79	3.67	2.49	4.76	5.72	4.69

Table 12: Capital cost (M$) for hybrid sequence

	50% CO_2			72% CO_2			Heavy H/C		
Column	Direct	In-direct	Mixed	Direct	In-direct	Mixed	Direct	In-direct	Mixed
Packed	4.01	4.01	4.01	4.01	4.01	4.01	4.01	4.01	4.01
1	3.28	28.92	13.60	1.56	16.15	12.04	4.29	38.92	22.33
2	1.81	17.42	2.75	2.00	10.06	1.62	2.57	20.25	3.18
3	2.78	13.84	1.56	1.73	9.41	1.19	4.04	15.33	3.01
4	1.22	2.76	1.60	1.66	1.62	1.45	3.69	3.36	3.60
Total	9.10	62.94	19.51	6.96	37.24	16.30	14.59	77.86	32.12

9.10 M$ from 26.14 M$. The decrease in cost is higher for higher CO_2 contents.

CONCLUSION

The impact of different compositions of carbon dioxide present in natural gas feed was observed for sequencing of cryogenic distillation network. The results indicate direct sequence is better than in-direct and mixed sequences for the separation of carbon dioxide from natural gas with different feed compositions in respect of minimum vapor flow, marginal vapor flow and energy requirements. The effect of hybrid cryogenic network was examined in energy calculations and it was found that hybrid cryogenic network required considerably lower energy as compared to the conventional cryogenic network. For low contents of carbon dioxide in the feed and in heavy hydrocarbon containing feed mixed network is closed to the direct network in minimum vapor flow and energy requirements. Finally the capital cost of the conventional and hybrid system was compared and a significant reduction in capital cost of columns had been observed by using hybrid cryogenic network.

NOMENCLATURE

$\propto$ = Relative volatility
F = Feed flow
f = Component flow in feed
q = Feed condition
D = Distillate flow
b = Bottom component flow
d = Distillate component flow
ϕ = Parameter, whose value lies between values of $\propto$ of the key components
R_{min} = Minimum reflux ratio
V_{min} = Minimum vapor flow
i = Index for component number
i_k = Species i to k

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