

# Improvements in the Design of the Ammonia Synthesis Process Implementing Counter Current Gas Permeation Modules

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**S** Supporting Information

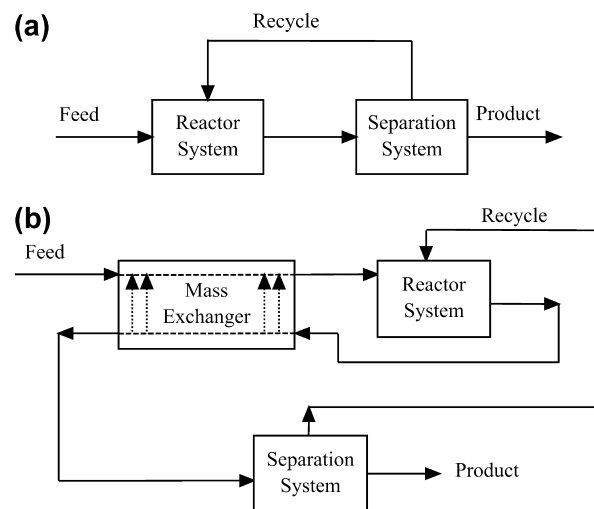
**ABSTRACT:** This paper explores mass exchanging the outlet and inlet streams of the reactor, following a design heuristic proposed by Fischer and Iribarren in *Industrial and Engineering Chemistry Research* 2011, 50 (11), 6849–6859 within the Hierarchical Process Design Procedure by J. M. Douglas in *Conceptual Design of Chemical Processes*, McGraw-Hill, 1988. When applied to the ammonia synthesis process, this design methodology generated a process alternative different from that previously proposed by other authors, resorting to ceramic membrane counter current gas permeation units to perform the mass exchange of hydrogen. This alternative design is shown to produce a reduction of the gas recycle stream (hydrogen and nitrogen) of up to an interesting 8.40%, reducing recompression associated costs. However, as the present cost of zeolite membranes is still high and their hydrogen–nitrogen selectivity moderate, in the optimal economical solution, the net annual income amounted to 4.56%, corresponding to U.S. \$817,793/year savings. The heuristic was used again at a later refinement stage, yielding an appreciable percent reduction in the cost of recovering hydrogen from the purge. As gas permeation technology (and hydrogen permeation, in particular) is a very active R&D area, we hope that the results of this paper bring some attention to this novel (concentration driven) counter current application for gas permeation modules, apart from the presently more widespread (pressure driven) cross-flow application.

## INTRODUCTION

This work explores the changes generated on an ammonia synthesis process when including the mass exchange networks (MENs) concept as a heuristic, in the design stage. When designing a new process following the hierarchical methodology of Douglas,<sup>1,2</sup> one moves toward designing more-detailed versions of the process with an increasing number of process blocks interconnected by new streams. The first level of decisions is the “input–output” structure of the flow sheet. Raw materials, end products, and processing routes are defined at this level, yielding the overall structure of the components that enter and exit the process. The second level of decisions adds detail to the selected process alternative, deciding about the recycle structure of the flow sheet and selecting the unit operations to perform the required separations. This is done following heuristics that recommend alternatives for recycling components and criteria for selecting the unit operations. The general structure arrived at is schematized in Figure 1a.

On the other hand, the technique for the synthesis of MENs<sup>3–5</sup> extrapolates pinch analysis of heat exchange networks (HENs) to mass exchange, designing mass exchangers in a counter-current arrangement. As in the case of heat integration, the technique for the synthesis of MENs requires as input information the list of streams to be integrated as well as their flow rates and concentrations (instead of temperatures, as in heat integration). Thus, both methodologies (for the integration of heat and mass) are usually applied in the last stage of the process design, once all the process streams have been generated.

In this work, we use the technique for synthesizing MENs at an earlier stage of the hierarchy, when deciding the recycle



**Figure 1.** (a) Douglas heuristic process design procedure standing alone. (b) Douglas process design procedure including the MENs heuristic.

structure of the process. This can be done after the reaction is defined, which usually requires operating conditions with reactants in excess, which must be removed after the reactor. The concept of MENs synthesis can be used as an additional

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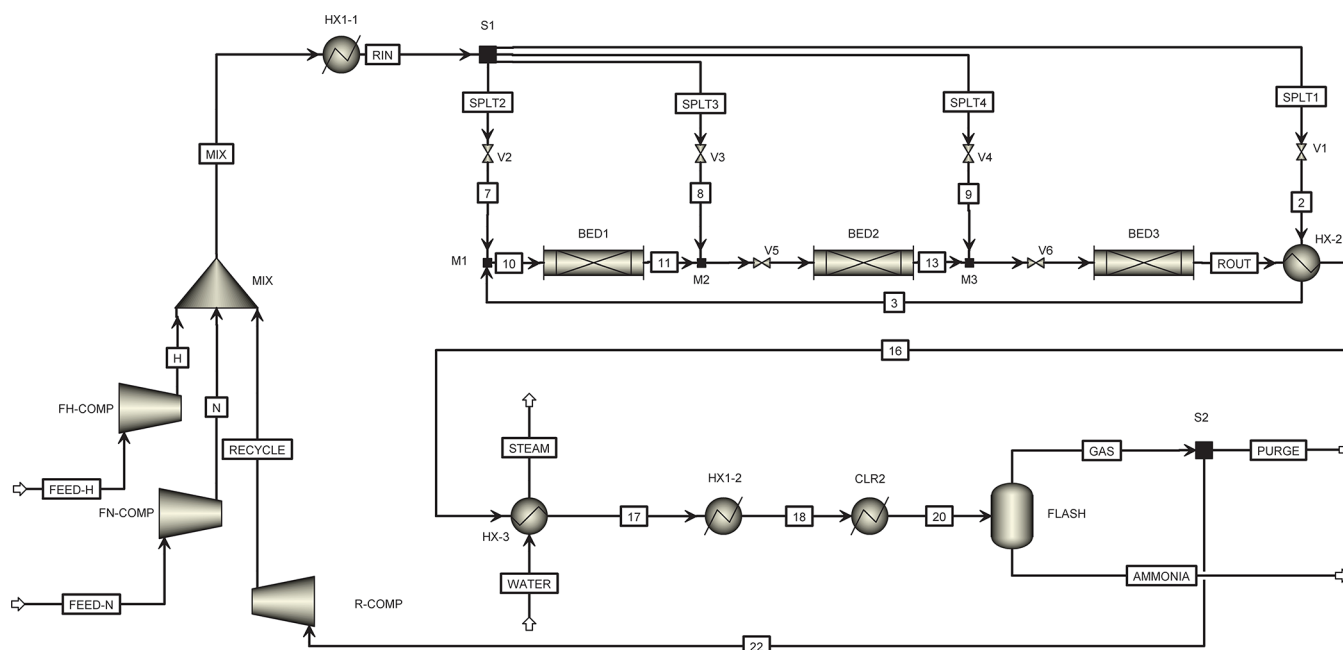


Figure 2. Ammonia synthesis loop.

Table 1. Main Streams in the Ammonia Synthesis Loop

	feed-H	feed-N	10	16	20	gas	purge	recycle	ammonia	
temperature (°C)	17.00	17.00	319.30	472.60	407.70	40.00	40.50	40.50	47.50	40.50
pressure (bar)	23.10	23.10	203.96	200.76	199.76	200.00	196.28	196.28	207.96	196.28
mole flow (kmol/h)	6410.29	2133.70	27 720.80	33 807.31	33 807.31	33 807.31	29 819.35	596.39	29 222.96	3987.97
mole flow (kmol/h)										
H <sub>2</sub>	6382.02	0.00	18 592.07	19 390.69	19 390.69	19 390.69	19 334.47	386.69	18 947.78	56.23
N <sub>2</sub>	0.00	2108.26	5455.32	5452.62	5452.62	5452.62	5432.72	108.65	5324.06	19.90
H <sub>3</sub> N	0.00	0.00	2855.54	7849.78	7849.78	7849.78	3969.86	79.40	3890.46	3879.92
CH <sub>4</sub>	28.27	0.00	459.97	626.63	626.63	626.63	610.67	12.21	598.46	15.96
AR	0.00	25.44	357.90	487.60	487.60	487.60	471.63	9.43	462.20	15.97
mole fraction										
H <sub>2</sub>	0.9960	0.0000	0.6710	0.5740	0.5740	0.5740	0.6480	0.6480	0.6480	0.0140
N <sub>2</sub>	0.0000	0.9880	0.1970	0.1610	0.1610	0.1610	0.1820	0.1820	0.1820	0.0050
H <sub>3</sub> N	0.0000	0.0000	0.1030	0.2320	0.2320	0.2320	0.1330	0.1330	0.1330	0.9730
CH <sub>4</sub>	0.0040	0.0000	0.0170	0.0190	0.0190	0.0190	0.0200	0.0200	0.0200	0.0040
AR	0.0000	0.0120	0.0130	0.0140	0.0140	0.0140	0.0160	0.0160	0.0160	0.0040

heuristic rule, in competition with other Douglas heuristics: “Explore the implementation of a mass exchanger between the streams exiting and entering the reactor”. This mass integration could eventually be performed with absorbers, strippers, or membrane systems arranged to operate in counter-current mode. If this material integration were possible, the subsequent separations and recycle are minimized (or even eliminated if a total integration between output and input could be achieved). The general structure, arrived at in this case, is schematized in Figure 1b.

In previous works, this concept was applied successfully, reducing the total annual cost of a biodiesel<sup>6</sup> process and the HDA<sup>7</sup> (hydrodealkylation of toluene to benzene) process originally used by Douglas<sup>1,2</sup> to present his methodology. Next, we get into the specific contribution of this paper: an in detail discussion of a different ammonia synthesis process, arrived at by using the concept of MENs, first, just after defining the reaction and, later, in a design refinement stage, improving the

process performance by recovering hydrogen from the purge stream. Finally, the last section draws the conclusions of this work.

## EXAMPLE: AMMONIA SYNTHESIS

In this paper, we apply the concept of MENs as a design heuristics after defining the reaction, to design a different alternative for an ammonia production process. The term “ammonia synthesis” is commonly used to refer to the entire industrial process for producing ammonia from different feed stocks. The entire process can be split into three sections to simplify the analysis. The first section is the hydrogen and nitrogen production; the second is the compression of the hydrogen and nitrogen produced up to the reaction pressure, and the third section is the synthesis of ammonia itself, normally termed “ammonia synthesis loop”. With regard to the section of hydrogen and nitrogen production, different technologies have been proposed, which cover a wide range

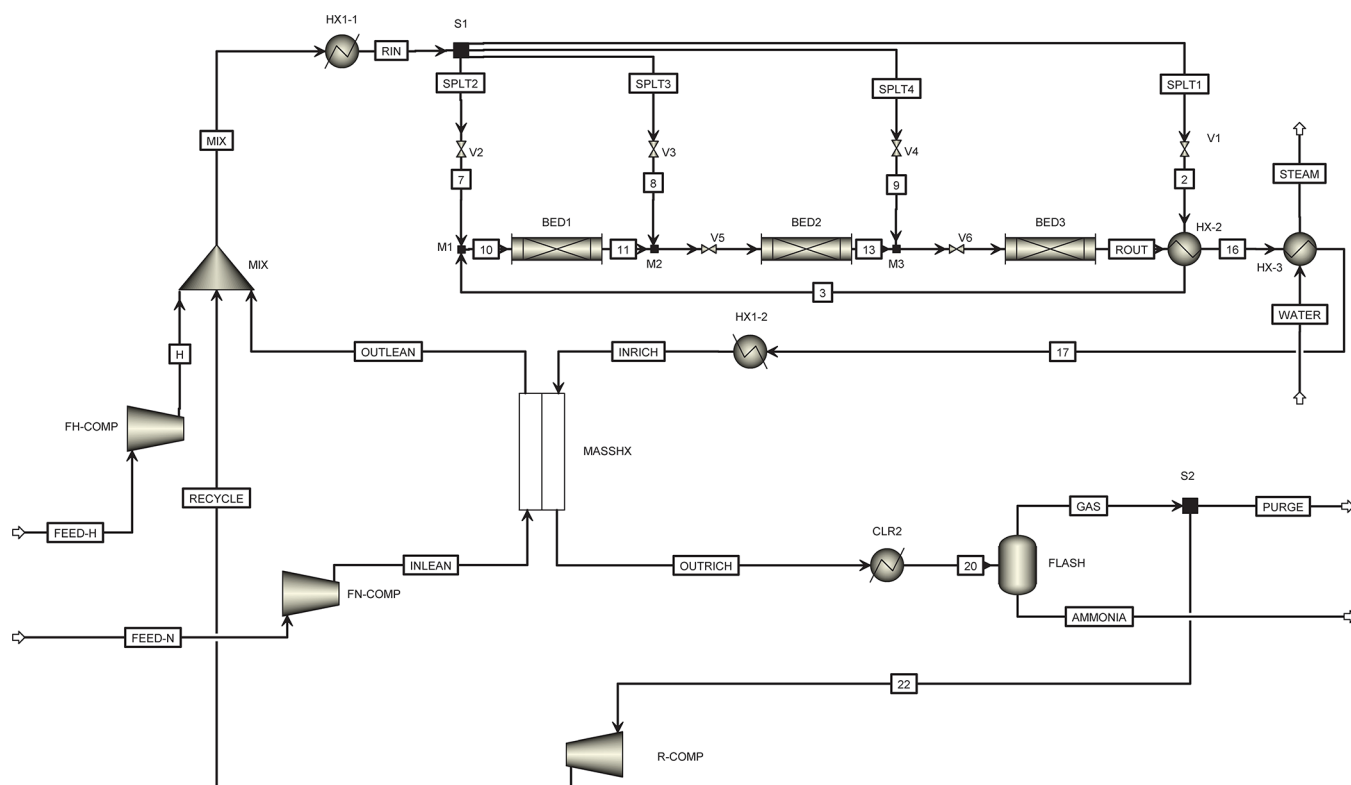


Figure 3. Ammonia synthesis loop with a mass exchanger.

of raw materials. This section is where there are more variations between the different processes for the production of ammonia. The compression section area is important because the process is performed at high pressure (even exceeding 200 bar), and there are many compressors designed specifically for this work. With respect to the section of the ammonia synthesis loop, there are different alternatives depending on the point where the synthesis gases are admitted and where the ammonia is separated. Detailed descriptions of each of the sections and different technologies can be found in the books of Max Appl<sup>8</sup> and Gary Maxwell.<sup>9</sup>

In this work, we will concentrate on the separation structure of the ammonia synthesis loop. We will consider a case study where the synthesis gas is generated such that the hydrogen is obtained with a small concentration of methane, and the nitrogen is obtained with some argon (when using an air separation process). Also, we assume that the gases are treated and already free of other compounds (CO, CO<sub>2</sub>, O<sub>2</sub>) that may contaminate the catalyst. Therefore, we will consider an ammonia synthesis loop where the synthesis gases are admitted and sent along with the recycle to the reactor.

Usually, the whole ammonia production process is strongly energy integrated, with numerous heat exchangers. Furthermore, the process is exothermic and this is exploited generating steam.<sup>8,9</sup> As we are redesigning the process at an early level of the hierarchical process design procedure, we will not consider the HENs. Instead, we use heaters and coolers in our flow sheets, which will be replaced by heat exchangers when synthesizing the HENs at the end of the design.

#### Brief Description of the Ammonia Synthesis Loop.

Figure 2 presents the flow sheet of a simplified process, and Table 1 shows the properties of the main streams. The ammonia synthesis loop consists of a few essential operations; to model them, we will use the parameter values and models

reported by Araujo and Skogestad.<sup>10</sup> This configuration uses a three stages quench converter whose first stage is operating at 319.30 °C and 203.96 bar, while its effluent from the third stage is at 472.60 °C and 200.76 bar. After the reaction, the gases are cooled to 40 °C and ammonia is separated through a flash. The separation is not complete, and the ammonia recovered has about 3% of dissolved gases (including synthesis and inert gases). The gases exiting from the flash are recompressed and sent to the reactor after taking a small purge to prevent accumulation of inert gases (argon and methane). The purge fraction is usually within 1% to 4.5%.<sup>8,9</sup> of the gaseous stream. In our case study, we use an average value of 2%.

There are several optimization variables in this process, mainly temperatures, pressures, and the purge fraction. We did not optimize these variables after modifying the process but stick to those reported in the literature so that, when we compare our proposed process with the one reported in the literature, the only relevant difference between them is the inclusion of the mass exchanger between the outlet and inlet of the reactor.

We select, as the hydrogen rich stream, the reactor exit stream after heat exchanger HX1-2 and, as the hydrogen lean stream, the nitrogen process feed stream after compressor FN-COMP. In particular, we will study how the main process variables behave when changing the mass exchange area of this counter current gas permeation module. We placed the custom model previously developed in ACM to contact these streams and simulated with Aspen Plus V7.2 to determine the effect on all the variables of interest.

There are different types of membranes that have a selective permeability between hydrogen and nitrogen at this temperature. The literature reports membranes of zeolite, carbon, silica, and metal alloys for such separation. In general, the membranes were developed to be tailored to each particular

application, and although there is not one specifically developed for our case, we note that a ceramic membrane of zeolite ZSM-5 is capable of operating in our process conditions.

The literature<sup>11</sup> has reported a ceramic membrane of zeolite ZSM-5 with a permeability for hydrogen of  $0.360 \text{ kmol}/(\text{m}^2 \text{ bar hr})$  and a hydrogen to nitrogen pure gases ideal selectivity of 61, that corresponds to a nitrogen pure gas permeability of  $0.0059 \text{ kmol}/(\text{m}^2 \text{ bar hr})$ . Although these figures may vary with pressure, temperature, and composition, using these pure gas permeability values give us an approximation to the performance that can be expected if used in the ammonia process.

Several sensitivity analyses were performed on the ammonia synthesis loop, from which it was decided to explore the effect of the additional mass exchange area in the range from 0 to  $1000 \text{ m}^2$ . Figure 3 shows the new process flow sheet with the mass exchanger, and Table 2 (in Supporting Information) presents the molar flows of hydrogen and nitrogen transferred in the mass exchanger.

A constant  $\text{H}_2/\text{N}_2$  ratio was kept at the inlet of the reactor, and the total amount of these two reagents was also kept constant, in both cases at the same value as in the process without the mass exchanger. This was achieved by controlling the amount of fresh input of these two components to the synthesis loop while varying the exchange area. This kept the initial conditions of the reaction equal to the original case but allowed the extent of the reaction be different.

Analyzing the flows in Figure 4, we note that, increasing the exchange area, the amount of hydrogen transferred grows

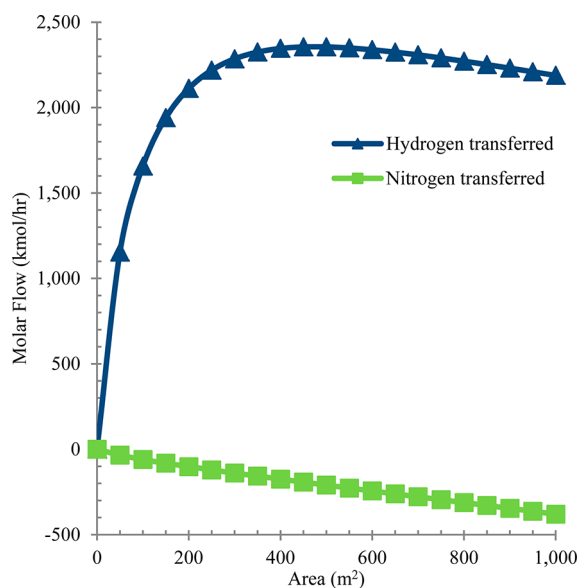


Figure 4. Hydrogen and nitrogen transferred.

rapidly to a maximum. The maximum is reached at  $500 \text{ m}^2$  of mass exchange area, and then, the amount of transferred hydrogen begins to decrease slightly.

Increasing the exchange area not only increases the amount of hydrogen transferred but also increases the amount of nitrogen transferred in the opposite direction. This leads to less nitrogen in the lean hydrogen stream and a greater molar fraction of hydrogen: the partial pressure gradient for the hydrogen (the driving force) diminishes and thus limits the exchange of hydrogen.

The explanation of this peak in the flow of hydrogen through the membrane is in the concentration profiles that arise in the mass exchanger when increasing the area. These are plotted in Figure 5.

In the case that there was no nitrogen flow from the lean stream to the rich stream, the peak would not exist. For simplicity, let us pretend that the total pressure on both sides of the membrane was the same, so that the differences in  $y_{\text{H}_2}$  was the driving force (in fact, it is the partial pressures).

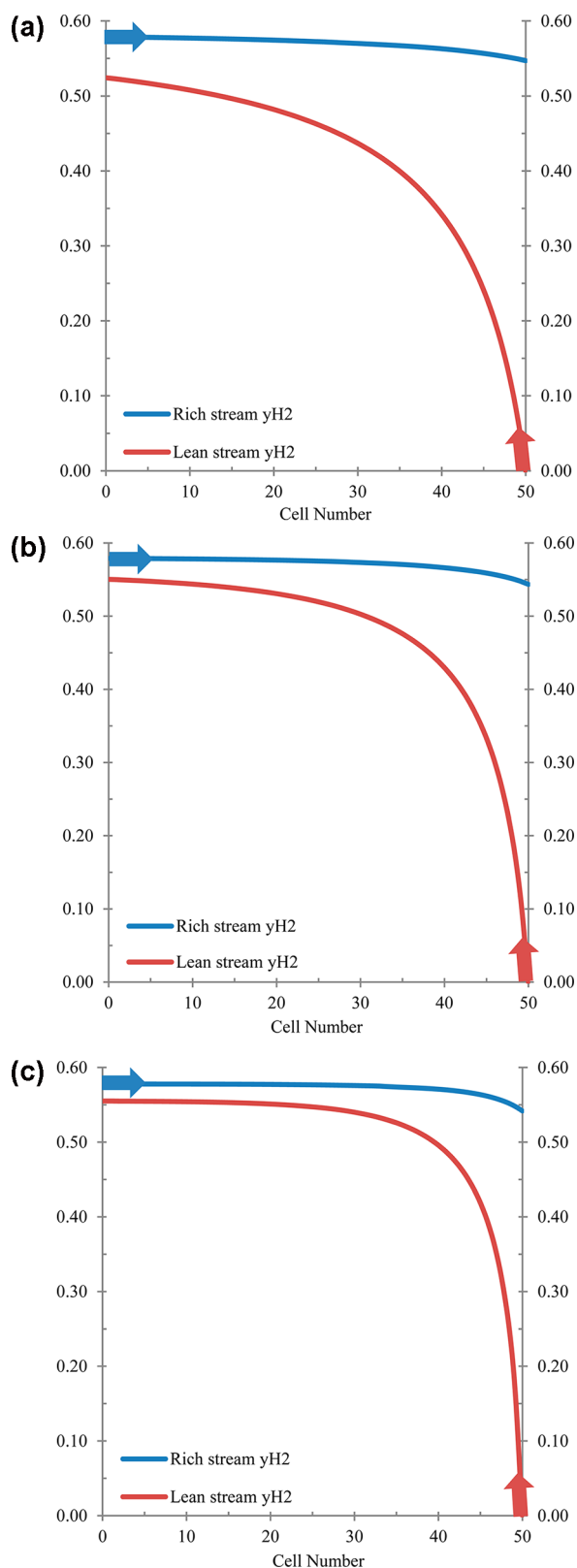
In this case, the maximum flow rate of hydrogen that could be transferred is  $2874.4 \text{ kmol/h}$  because this is the amount that added to the inlet of the lean stream would cause its outlet concentration to be  $y_{\text{H}_2} = 0.574$  (equal to the concentration of the inlet of the rich stream). At this end of the mass exchanger, the driving force for hydrogen transfer would vanish, and the required membrane area would be infinite. At the other end of the mass exchanger, this maximum amount of hydrogen transferable would cause an outlet concentration in the rich stream of  $y_{\text{H}_2} = 0.534$  with still an appreciable driving force to transfer hydrogen into the inlet of the lean stream with a concentration of  $y_{\text{H}_2} = 0.0$

Analyzing the results of the simulations, we see that the maximum flow of hydrogen  $2355 \text{ kmol/h}$  is achieved with a membrane area of  $500 \text{ m}^2$ . The simulations take into account the nitrogen flow from the lean stream into the rich stream which, as shown in Figure 4, increases almost linearly with the membrane area. This flow (although much smaller than that of hydrogen) contributes to reduce  $y_{\text{H}_2}$  in the rich stream and increase it in the lean stream. Increasing the exchange area, the driving forces are reduced all along the mass exchanger. The driving force is mainly reduced at the end of the mass exchanger where the rich stream enters, because the exiting lean stream has a larger  $y_{\text{H}_2}$  due to the nitrogen loss along the mass exchanger. Also, the driving force is reduced at the end of the mass exchanger where the rich stream exits, because its own  $y_{\text{H}_2}$  is lower due to the nitrogen received along the mass exchanger. The reduction of the driving force due to an increase in the membrane area, in the case that only hydrogen was transferred, would cause its flow rate to tend asymptotically to a maximum. By adding the effect of nitrogen transfer, it can be explained that, after passing through a maximum, the flow of hydrogen begins to decrease.

Now, we analyze the recycle stream, which is the rich stream leaving the mass exchanger. Table 3 (in Supporting Information) reports the molar flows of the main components in this stream, which are also plotted in Figure 6.

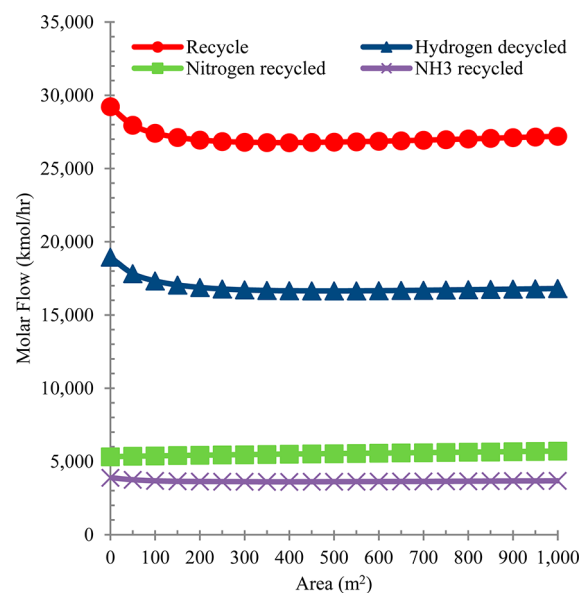
The first thing we notice in this figure is the reduction in the flow of recycle that must be handled by the compressor. As the exchange area is increased, the flow rate decreases, reaching a minimum with a reduction of 8.40%. After this minimum, the flow begins to increase slightly. This is associated with the transfer of hydrogen and nitrogen described above. As the membrane is permeable in both directions, the nitrogen of the lean stream (hydrogen-poor but rich in nitrogen) permeates into the rich stream.

This permeation of nitrogen is strongly influenced by the selectivity of the membrane and the concentrations of nitrogen in the streams selected to perform the exchange of hydrogen. The recycle stream reaches the minimum flow to an exchange area of  $400 \text{ m}^2$ , which is a different value as that for maximum transfer of hydrogen.



**Figure 5.** (a) Concentration profile in the mass exchanger with 250  $\text{m}^2$  of area. (b) Concentration profile in the mass exchanger with 500  $\text{m}^2$  of area. (c) Concentration profile in the mass exchanger with 1000  $\text{m}^2$  of area.

This exchange of hydrogen and nitrogen between the outlet and inlet of the reactor modifies the structure of the recycle and produces changes affecting the whole ammonia synthesis loop.



**Figure 6.** Main components molar flow rates in the recycle stream.

Table 4 (in Supporting Information) tabulates the values of the inputs and outputs of this loop. It also displays the powers of the compressors corresponding to the feed streams (FH-COMP and FN-COMP) and recycle stream (R-COMP).

The variations in the streams are minor, but since it has very large flow rates, these minimal changes represent large amounts of the components involved; so, we proceed to analyze them. In all cases, the flows of hydrogen and nitrogen entering the reactor are the same. If we analyze the hydrogen feed to the synthesis loop, we find a decrease in its consumption, reaching a minimum with a membrane area of 550  $\text{m}^2$  and a decrease of 0.8%. In contrast, the nitrogen feed always increased by increasing the membrane area. In the range analyzed, it increased 0.5%. This corresponds to the fact that, losing nitrogen and gaining hydrogen in the mass exchanger, the recycle (and therefore, also the purge) has a lower hydrogen and a higher nitrogen composition. These streams are plotted in Figures 7 and 8.

With regard to the amount of liquid ammonia obtained in the stream exiting the flash separator (AMMONIA), it can be seen in Figure 9 that it grows to a maximum of about 0.3% with 450  $\text{m}^2$  of exchange area. The amount of ammonia increases because, recycling hydrogen free of inert gas, the reaction proceeds a little more in the reactor. The reaction conversion always increases increasing the exchange area, although this is not reflected in the amount of liquid ammonia recovered in the flash separator. This occurs because, as the gaseous stream exiting the flash separator (GAS) increases, this increases the amount of ammonia which is removed with it. Therefore, the amount of ammonia recovered in the liquid stream starts decreasing slightly above an exchange area of 450  $\text{m}^2$ .

The ammonia in the gas stream (GAS) is recycled after the purge and increases the amount of ammonia fed to the reactor. This effect of an increased amount of ammonia in the reactor inlet would be greater in the case that some ammonia was transferred to the feed in the new mass exchanger. Given the similar values of kinetic diameters of ammonia and nitrogen, we expect that their permeability will be similar too. Besides, given that ammonia concentrations are about 10 times smaller than nitrogen, that this flux does not jeopardize the process but just

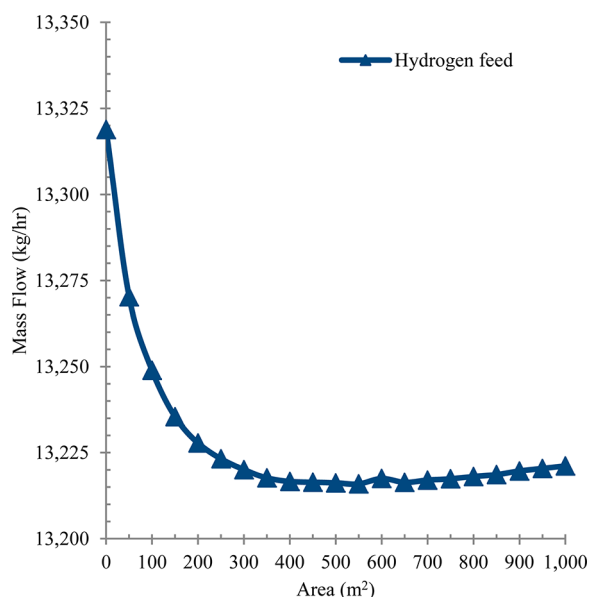


Figure 7. Hydrogen feed.

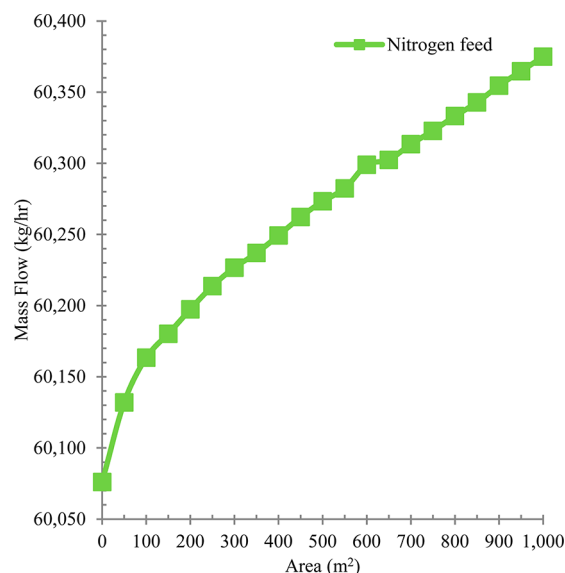


Figure 8. Nitrogen feed.

returns a small portion of ammonia to the reactors, and that we lack experimental permeability values for this compound, this effect was not modeled, nor did we model the small transfer of methane that can occur in the new mass exchanger, that does not really affect us.

We notice that the different parameters analyzed all vary with the membrane area but find their optimal values at different areas of exchange. Therefore, to find the optimum mass exchange area for the process, we must consider all the changes together and evaluate their impact on the cost of the process.

**Cost Analysis.** The main advantageous effect of the inclusion of a countercurrent mass exchange is that the downstream separation and recycle system will be smaller. However, this effect should offset the cost of installation of the mass exchanger.

In this paper, we consider the total annual cost (TAC) and the net annual income (NAI) for the process with the mass exchanger and compare them with the literature reported

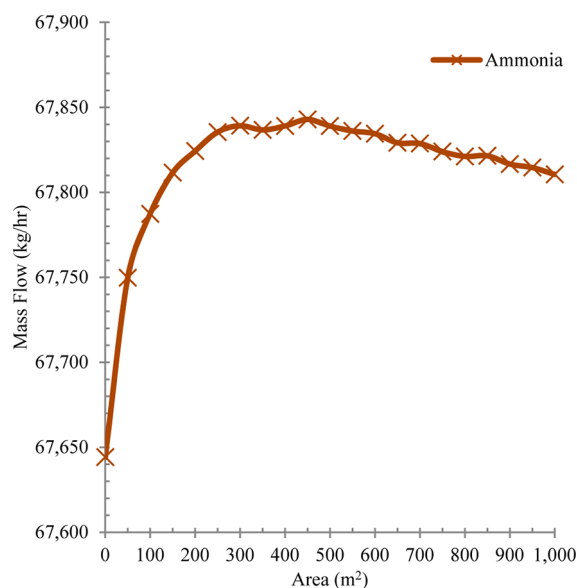


Figure 9. Ammonia in the liquid stream leaving the flash separator.

process taken as reference. Within the TAC, we consider the total costs of operation and installation of all compressors. We do not consider the costs of the reactor nor the flash separator, assuming their size and therefore their cost do not vary. We do consider the variations in the generation of steam and in the process input and output streams with their associated costs. We take the following values for these streams: hydrogen, 0.5058 U.S. \$/kg; nitrogen, 0.01 U.S. \$/kg; ammonia, 0.20 U.S. \$/kg; purge, 0.01 U.S. \$/kg; and steam, 0.017 U.S. \$/kg.<sup>10,12</sup>

Compressors for ammonia synthesis are usually specifically designed to take advantage of the steam generation process.<sup>8</sup> Here, for simplicity, we will consider centrifugal compressors driven by electric motors. To estimate the installation cost of the compressors, we use the correlations of Douglas<sup>2</sup> where  $M \& S$  is the Marshall and Swift inflation index  $M \& S = 1477.7$  for the year 2009,  $F_d$  is the design factor  $F_d = 1$  for centrifugal compressors, and  $bhp$  is the brake horsepower, calculated with Aspen Plus V7.2 considering an isotropic efficiency of 0.72. The expression for estimating the cost of installation of each compressor is:

$$\begin{aligned} \text{compressor installed cost (\$)} \\ = \left( \frac{M\&S}{280} \right) \cdot 517.5 \cdot (bhp)^{0.82} \cdot (2.11 + F_d) \end{aligned}$$

For the installation cost of the mass exchanger and the cost of zeolite membranes, we refer to Caro et al.,<sup>13</sup> who report a figure of about \$3000 per  $m^2$  of membrane area, but it is usual to use a value 10 times lower assuming that the costs of this type of membrane quickly lower as its use intensifies.<sup>14</sup> Therefore, to estimate the cost of installation of a zeolite membrane, we used U.S. \$300 per  $m^2$  for the module cost and an Installation Factor 3.7.<sup>15</sup> These values result in an installation cost around U.S. \$1,110 per  $m^2$ .

Installation costs are annualized using a capital charge factor of 0.351. To estimate the energy consumption of the compressors, we considered an efficiency of 0.9 and an electrical energy cost of 0.07 U.S. \$/(kW·h).

Computing the total annual income and subtracting the TAC, we get the NAI. Computing this value in the range of membrane area between 0 and 1000  $m^2$ , we determine the

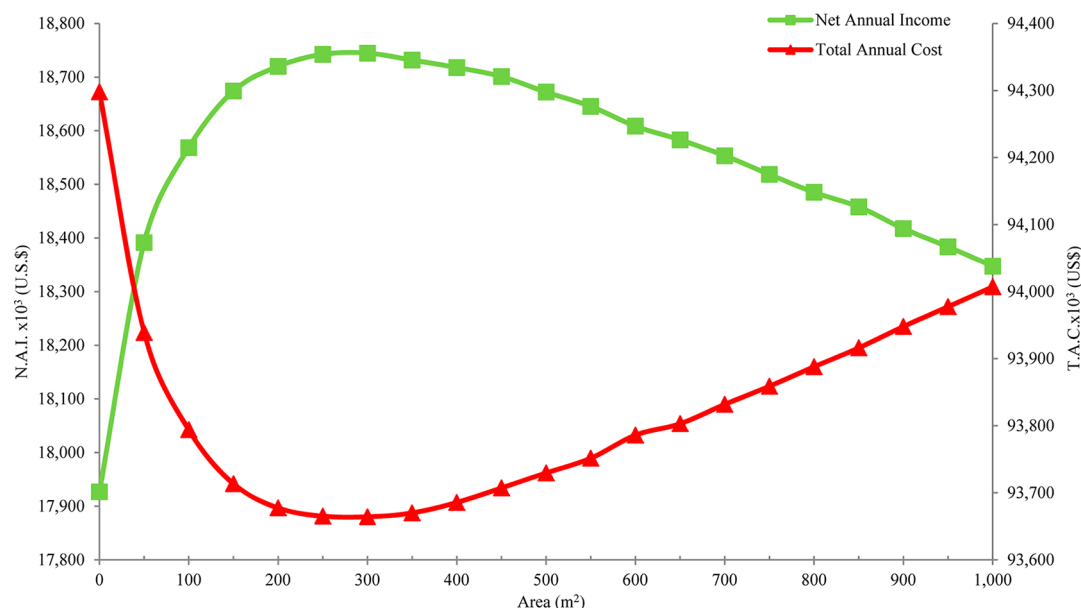


Figure 10. Net annual income and total annual cost.

optimal value of the mass exchange area. The costs described in this section are tabulated in Table 5 (in Supporting Information) and plotted in Figure 10.

The graph shows that the TAC has a minimum at an exchange area of 300 m<sup>2</sup> with a reduction of 0.67% from the cost of the process without the addition of the mass exchanger. This is a quite small percentage reduction of the TAC, due to the high cost of these membranes, with only the recycle compressor R-COMP significantly reduced. If we analyze the installed cost of this compressor, we notice that it drops a 7.12% from its original value as shown in Figure 11. Furthermore, the NAI reached a maximum at a membrane area of 300 m<sup>2</sup> with an increase of 4.56% compared to the original process. It's percent represents U.S. \$817,793/year savings.

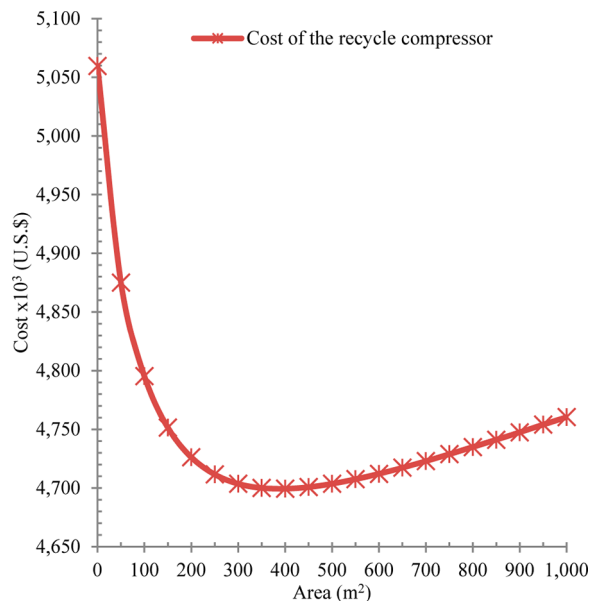


Figure 11. Cost of the recycle compressor.

**Design Refinement.** Present ammonia processes usually refine the design recovering most of the hydrogen contained in the purge stream (PURGE), which otherwise would be burned in the reformer. After reducing the content of ammonia to less than 200 ppm (to avoid damage to the polymeric membranes) scrubbing the vapors, this stream is fed to gas permeation units connected in cascade with a transmembrane pressure difference of about 60 bar.<sup>16</sup> The resulting permeate streams are sent to the admissions of the first and second stages of the feed compressor as shown in Figure 12. In this approach, the permeate streams must be recompressed up to the reactor pressure, with the cost of compression being an important component of the total cost of this hydrogen recovery.

Material integration can also be performed at the end of the synthesis process as a refinement of the design, which does not affect the operating conditions of the process. This has been formally proposed as a part of the hierarchical process design procedure in Fischer and Iribarren.<sup>6</sup> The standard methodologies for implementing MENs usually resort to absorbers and strippers. In the ammonia process, one can use the here proposed counter current gas permeation unit as a mass exchanger between the purge stream (PURGE) and the input nitrogen stream, already compressed (INLEAN-0) as shown in Figure 13. This approach could potentially result in significant cost reductions as it does not need any hydrogen compressor to recompress the permeate stream as in the conventional system.

The conventional pressure driven membrane separation displayed in Figure 12 would recover 298.9 kmol/h of the hydrogen available in the purge (87.9%) with 88 m<sup>2</sup> of polymeric membrane which has a cost of U.S. \$550 per m<sup>2</sup> and requires a compression power of 596.87 kW, accounting for the two stages of compression. Such an installation would have a TAC of U.S. \$1,104,116.

Recovering the same amount of hydrogen with the proposed mass exchanger displayed in Figure 13 would require only 12 m<sup>2</sup> of exchange area, with no compression costs (only the membrane ZSM-5) and a TAC of U.S. \$13,200 which amounts to a reduction of the TAC of 98.8%

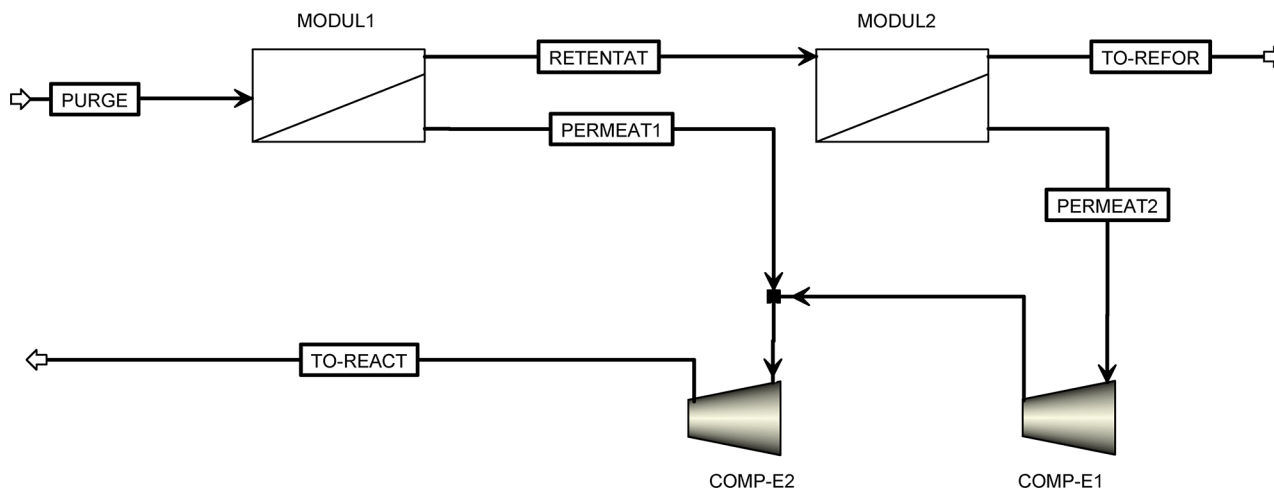


Figure 12. Present hydrogen recovery from the purge stream.

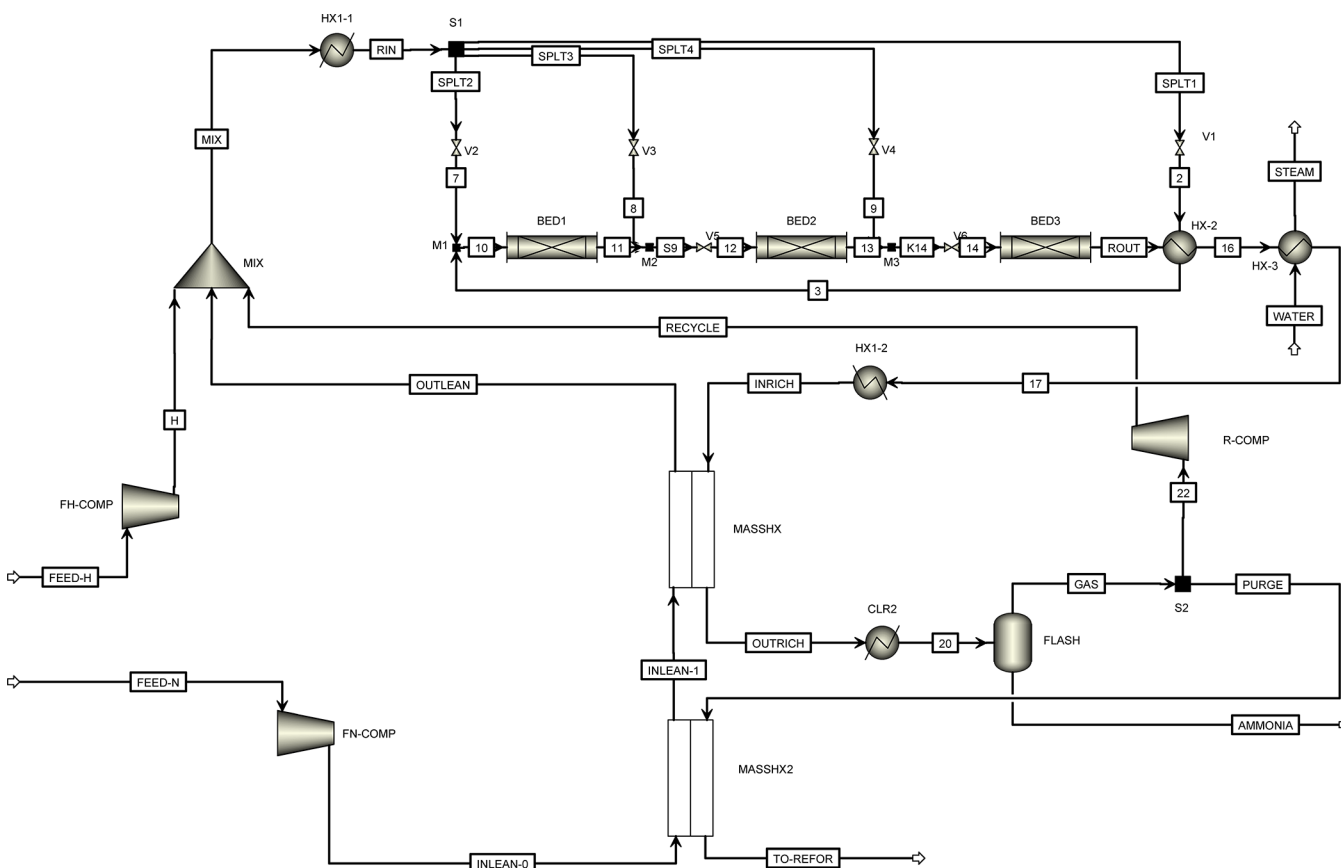


Figure 13. Proposed hydrogen recovery from the purge stream.

CONCLUSIONS

This paper discusses incorporating the concepts of mass exchange networks as heuristics in the hierarchical design procedure of Douglas.<sup>1,2</sup> These mass exchange decisions are made at two levels, first at the time of synthesizing the structure of the recycle and separation system and again at the end of the procedure, as a design refinement. The MENs synthesis methodology is a useful conceptual tool that when used at an early stage of the design procedure finds a new application, additional to the usual procedure of applying it to completely designed processes, with a list of defined streams. Each of these

newly generated alternatives should be evaluated to determine if it improves the design.

We applied the methodology to the design of an ammonia synthesis loop. First, we selected a stream exchange between the output from the reactor after being cooled and the nitrogen process feed after being compressed. We varied the mass exchange area and analyzed the behavior of different process variables, with special attention on the effect of the reverse permeation of nitrogen. We observed a reduction in the size of the separation and recycle system; with a membrane area of 400 m<sup>2</sup>, the recycle flow rate is decreased by 8.40% resulting in a reduction of the installed cost of the recycle compressor of



7.12%. Mainly, due to the high cost of zeolite membranes, the optimal design is achieved with 300 m<sup>2</sup> resulting in an increase of 4.56% of annual net income.

Then, we applied the concept of MENs again at the end of the procedure, as a process design refinement. In this case, we placed a mass exchanger between the purge stream and the nitrogen stream, already compressed and prior to its exchange with the reactor exit stream. This resulted in a very significant cost reduction (a 98.8% of the total annualized cost of the conventional purge hydrogen recovery system) as this approach does not need any recompression of the permeate stream.

As gas permeation technology (and hydrogen permeation in particular) is a very active R&D area, we hope that the results of this paper bring some attention to this novel (concentration driven) counter current application for gas permeation modules, apart from the presently more widespread (pressure driven) cross-flow application.

## ■ ASSOCIATED CONTENT

### 📄 Supporting Information

For more-interested readers, detailed tables. Tables 2, 3, 4, and 5: "Hydrogen and Nitrogen Transferred in the Mass Exchanger", "Main Components Molar Flow Rates in the Recycle Stream", "Inlets, Outlets, and Compressors Power" and "Costs", respectively. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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### Notes

The authors declare no competing financial interest.

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