Sorption Enhanced - Steam Methane Reformation in H_2 Production

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Abstract

Hydrogen gas has many important uses in the chemical engineering industry and will play an important role in renewable energies of the future. In this report, a sorption-enhanced membrane reactor (SEMR) is modeled to improve the current process of hydrogen production, known as steam methane reforming (SMR), in order to keep CO_2 emissions at a minimum while still being an affordable solution that meets market demands. The results show a product stream of hydrogen with lower emissions of carbon dioxide. Namely, the simulation for SEMR produces 8.86 kg H_2/kg CO_2 compared to the average SMR values of 0.24 kg H_2/kg CO_2. The economic evaluation of using an SEMR demonstrates that it is a largely profitable idea with the estimated yearly revenue being \$250,125,757.64 year^{-1}. Compared to other literature data, the results show both the H_2 and CO_2 output and cost efficiency is as expected or even improved compared to other SMR processes. However, there are several improvements that can be made to justify this conclusion. For example, investing more research into Pd membrane technology can improve cost efficiency, lifespan, and operating conditions. Also, to increase profits, $CaCO_3$ could be sold to offset operating costs. Overall, the proposal report aligns accurately with literature in order to create a more optimal model for hydrogen production.

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1 Introduction

Due to the growing concern for the climate, the need for environmentally sustainable and affordable methods of production have increased. One of these sectors is hydrogen production. Currently 48% of the world's total hydrogen production comes from SMR.[1] However, SMR is a high temperature process that produces large amounts of CO_2.[2] Therefore, Air Liquide is looking for more affordable and sustainable solutions to produce hydrogen while still being able to keep up with demand.

Currently, there are many alternatives to the SMR process, but one of the more promising alternatives is sorption-enhanced-steam methane reforming (SE-SMR). SE-SMR and SMR are similar processes, but where SE-SMR and SMR differ is the use of conventional reactors in the SMR process.[2] The use of conventional reactors leads to less pure H_2 being produced and thus more unit operations required to purify the H_2.[2] SE-SMR uses a SEMR which allows for separation and reaction to occur as one unit operation.[2] Thus, these membrane reactors require less energy and produce less CO_2 due to the less unit operations in the process. In addition, they are able to work at milder conditions compared to conventional reactors.[2]

Initially the proposal for this project sought to keep CO_2 emissions to 95.76 kmol h^{-1} and H_2 production to 29,936,673.36 kg year^{-1}. With a consumption of CH_4, H_2O, Pd, and CaO were determined to be 198,488,856.1 kg year^{-1}, 1,004,046,454 kg year^{-1}, 12,023 g year^{-1}, and 1000 kg year^{-1}, respectively. This resulted in a gross margin of $31,201,983.09. Therefore, this report attempted to accomplish these goals, which will be explored later on.

2 Basis of Design

The design for the project revolves around using a sorption enhanced membrane reactor (SEMR), which is comprised of a membrane, sorbent, and metal catalyst. First, a SEMR uses a membrane that is capable of separating H_2 and H_2O from the other byproducts.[3] The ability to efficiently separate H_2 during the reaction process allows less unit operations to be required for the purification of said H_2. Second, the SEMR uses a sorbent that is a mixture of both a catalyst and a carbon dioxide acceptor.[3] In the sorbent, the CO_2 acceptor would be the adsorbent while the catalyst is generally any metal catalyst. The use of sorbents is meant to improve the extent of chemical reactions in reactors down the line, such as the reactions in the methanation reactor (methantor), by having a high sorption capacity.[3] A high sorption capacity allows the adsorbent to pick up most of the sorbents that will be used for the reactors down the line. In addition, the adsorbent can be placed on either the retentate or permeate side of the membrane.[3] The retentate side would allow the CO_2 acceptor to be mixed with the catalysts, whereas the permeate side would separate both the acceptors and catalysts if needed.[3] Third, SEMR also uses metal catalysts to produce the essential byproducts for H_2 production.[3] Different metallic catalysts can be used, but a Pd catalyst was considered. This is due to SMR processes already using this catalyst for its high efficiency.[3] Therefore, despite the cost of Pd being expensive, the catalyst will be used as part of the project. All these components combine together to allow important to SEMR's function.

By using SEMR, this process reduces the CO_2 output and operating costs compared to SMR. For example, fewer unit operations results in less energy required and thus leads to a decrease in operating costs. For instance, the SEMR process uses $20 - 25\%$ less energy compared to other conventional processes with reactors.[3] In addition, having a high sorption capacity results in less output of CO and CO_2. Therefore, SE-SMR may be a plausible solution to produce sustainable and affordable H_2 that meets market demands.

3 Specifications

The incoming feed stream is at 298 K and 1 bar where it enters the jacketed SEMR where it reacts at 673 K and 3 bar. The retentate stream then leaves at 673 K and 8 bar. The permeate stream leaves the SEMR at 673 K and 0.15 bar. The permeate stream then goes to the flash drum operating at 216.81 K and 1 bar, where H_2 is separated to the top and H_2O is separated to the bottom at the same temperature and pressure as the flash drum. The retentate stream is then sent to the methanation reactor which is operating at 673 K and 1 bar. Part of the H_2 from the top of the flash drum is sent back to the methanation reactor at the same conditions as before. The rest of the H_2 is sent out the plant also at the same conditions as before. Finally, the tail gas from the methanation reactor leaves at 673 K and 1 bar.

4 Assumptions

One assumption that was made involved combining the calcinator/carbonator reactor with the chemical reactions that are included in the SEMR reactor. This was used to form an SEMR reactor. This assumption was made due to prior literature talking about how the model of both reactors are supposed to occur in one reactor, which is considered to be the SEMR reactor.[2] The SEMR reactor provides the same process as the calcinator/carbonator reactor, as well as applying the SMR reactions. Within the SEMR reactor, the process shows calcium oxide (CaO) reacting with CO_2 at the outer part of the membrane, which is where H_2 permeates through.

Another assumption that was made involved the efficiency of the SEMR reactor. The SEMR reactor modeled in Aspen calculates the flow rates in a best case scenario, to which calculates the thermodynamic equilibrium at a set temperature and pressure. As for the membrane, it is assumed to function at 90% because the membrane is unable to separate the reactants at 100%. This efficiency value has to be assumed since membranes are characterized by a permeance value instead of an efficiency value.

A third assumption that was made was the lifespan of the ceramic supported thin Pd-Ag membrane (c-tPd). Based on literature, the efficiency of the membrane will eventually drop due to the duration of the equipment being used over a long period of time.[4] Since the membrane is constantly being used with little to no breaks, the amount of hours the membrane is in use would build up, causing a drop in efficiency overtime.[4] This results in having to change the palladium membrane every month.

The last assumption that was made was the water utility source. One suggested area for the supply of water was listed to be in California. By comparing the cost of water utility in California, Burbank was both cheaper and more affordable options to have, resulting in being cheaper than cities like Santa Barbara.[5]

5 Methods

The flow rates were determined by initially using mass balances and taking the ratios between the different components in Lee et al.'s paper and were then scaled appropriately to fit the desired product flow rate of H_2.[2] First, a target flow rate of H_2 was selected, and 1×10^6 m^3/day was chosen since that's what's normally used in SMR plants currently.[6] Working backwards, ratios were determined for the various components in the process and were utilized to solve for the appropriate flow rates in the feed, permeate, retentate, and final product streams. To ensure that less CO_2 would be produced than typical SMR processes, the composition of CH_4 and CO_2 were varied until an appropriate amount of CO_2 exiting the plant was achieved. The various flow rates are given in Table 1, which originate from the simulations conducted in Aspen. For the heat duty values, as seen in Table 2, they also originate from Aspen. They were each pulled from the respective unit operation block that they belonged to.

Firstly, a Peng-Robinson equation of state was used since these reactions take place in the vapor phase.[2] Lee et al.'s setup used a 30:1 ratio of water to methane, so this same ratio was used this time with a different basis as seen in Figure 1 to adjust for the desired purity of H_2.[2] This comes in at a standard temperature of 298 K and a higher pressure of 1 bar since such conditions have been used before in previous literature.[7] The stoichiometric reactor was assumed to have a temperature of around 673 - 973 K since this was used in previous research.[7]

A membrane separator was then used to split the heated feed stream into two different product streams: retentate and permeate streams. The permeate stream is made up of hydrogen and water whereas the retentate stream is made up of carbon monoxide and carbon dioxide. The data for both hydrogen and water streams was collected via flash separator; the separator was then able to effectively separate the two components at a colder temperature with the same pressure as the other unit operations. As for the retentate stream, a methanation reactor was used to effectively convert CO and CO_2 to CH_4 and H_2O. In order to do so, H_2 was recycled from the flash drum to assist in the reduction of CO and CO_2 emissions.

6 Optimization and Comparison

To start, the initial design of the SE-SMR process consisted of a steam methane reforming membrane reactor where the SMR reaction occurred within the membrane reactor while the carbonation of the CO_2 produced from the reaction would be absorbed by the CaO embedded into the walls of the SMR membrane reactor. Inside the reactor, the membrane, a concentric tube within the SMR reactor, would allow the H_2 to permeate through itself where the H_2 would leave through the permeate stream and be separated from the sweep gas which in our case is the steam. The improvement made from this aspect of the base design starts with the kind of membrane used. In the base design, the membrane that was decided was a pure palladium membrane; however, with further research, the pure palladium membrane was found to not perform as well as the membrane chosen in this design. In this design, a c-tPd membrane is used because the permeance of the membrane is much higher than that of a purely palladium membrane.[8] With a higher permeance, the SMR membrane reactor can allow the H_2 to permeate more efficiently.

To continue, the base design had the carbonation occurring inside the SMR membrane reactor, and this design was based on Lee et al.[2] What changed with regards to the carbonation process was not having the CaO embedded in the walls of the SMR membrane reactor. Instead, in the optimized design, the CaO flows with the feed allowing for the CO_2 to react with the CaO. This change was done because the current membrane technology does not allow for feasible performance with CaO. The current research at this moment only has Pd membranes with other metal alloys. In a previous iteration of the design, the calcination/carbonation process occurred in two separate units; however, because the calcination/carbonation units were costly and not feasible, the calcination/carbonation reaction was, consequently, placed within the SMR membrane reactor.

Another aspect that was altered from the base design was the removal of the boiler and the heat exchanger. The purpose of the boiler in the base design was to use the methane that did not react in the SMR reactor, and burn the methane, to heat the retentate stream. The heated retentate stream is then fed to the heat exchanger that heats the methane and water feed. The improvement that was made in the optimized design was removing the boiler and heat exchanger. The boiler was not fed enough methane to heat the retentate and, consequently, heat the methane and water feed in the heat exchanger to the designed temperature. Another reason for the removal was to reduce the cost of heating the feed and, instead, heat the feed in each of the unit operations.

An important change to the base design of the process was the addition of the methanator. The purpose of the methanator was to reduce the CO emitted from the process. In the base design and during the optimization of the process, CO is being emitted at a relatively high quantity. To remedy the issue, the methanator was added to reduce CO; however, the downside of the methanator was the additional cost of a unit operation and the loss of the H_2 produced from the SMR membrane reactor.

7 Description Of Unit Operations

Overall, the process consists of several unit operations in order to produce H_2 with lower CO_2 emissions. Specifically, this process is based off of Lee et al.'s SE-SMR process.[2] The reactants enter the system at standard conditions i.e. at a temperature of 298 K and a pressure of 1 bar, since operating at low pressures such as 1 - 3 bar allows for more H_2 production.[2,3] Now, the temperature of the reactants are heated within the SEMR reactor block.[2] The reactants are heated to a higher temperature in order for the reaction to produce more H_2 and less CO_2.[2,3] Within this SEMR reactor block, the membrane used is c-tPd since that's been used in Nordio et al.[8] The one drawback to this is it's expensive price, which will be addressed later on in this proposal. Within the SEMR reactor block, a stoichiometric reactor is used for the SMR reactions and carbonation/calcination process, and it is then utilized in conjunction with the c-tPd membrane that will then be used to separate H_2.[2] The retentate of the membrane is sent to another stoichiometric reactor which is where the methanation reaction will take place. Leaving this stoichiometric reactor will be the tail gas. Finally, the permeate of the SEMR block is sent to a separator, specifically a flash drum, in order to further separate the H_2 from the H_2O as the final step of the process.[2] This flash drum is essential since it allows for H_2 to be fully separated from the water product. As seen here, a few modifications have been made in order to make the SMR process suit the needs of this project. The process flow diagram (PFD) diagram is provided below.

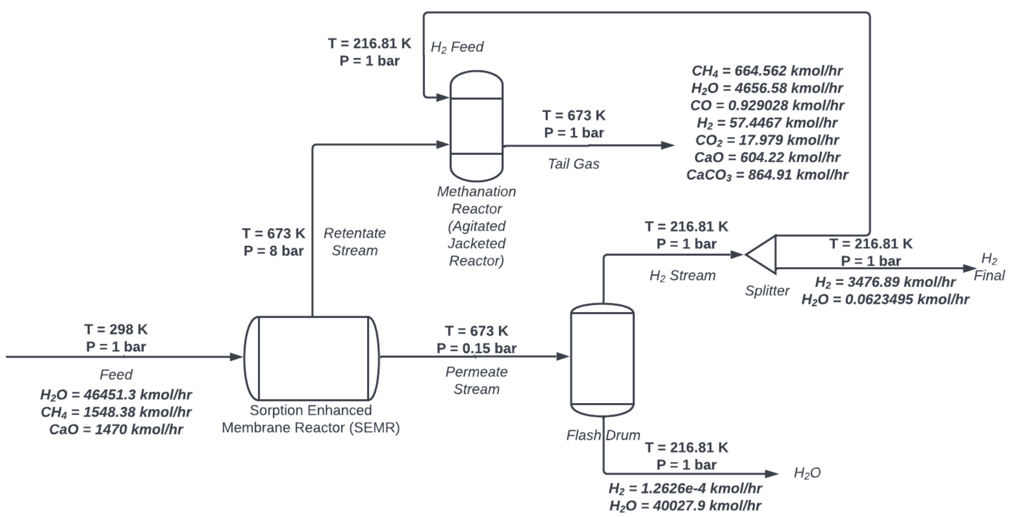

Figure 1: This figure shows the PFD of the SE-SMR process, which is based off of Lee et al.'s paper (*6*).

The process steps described earlier were modeled using the appropriate Aspen blocks. The reactants were heated up within the SEMR, which was modeled in Aspen as two separate blocks. The first block contained the actual SMR and carbonation/calcination reactions itself:

Table 1: Mole Balance of SE-SMR process.

Mole Balance ($kmol\,h^{-1}$)									
Species	Feed	H_2 Feed	H_2 Final	Hydrogen	Permeate	Retentate	SEMR	Tail Gas	Water
CH_4	1548.38	0	0	0	0	557.417	557.417	664.562	0
H_2O	46451.3	1.52424e-3	0.0623495	0.0638737	40028	4447.56	44475.6	4656.58	40027.9
CO	0	0	0	0	0	6.19352	6.19352	0.929028	0
H_2	0	84.9985	3476.89	3561.89	3561.89	395.766	3957.66	57.4467	1.2626e-4
CO_2	0	0	0	0	0	119.86	119.86	17.979	0
CaO	1469.13	0	0	0	0	604.22	604.22	604.22	0
$CaCO_3$	0	0	0	0	0	864.91	864.91	864.91	0

Table 2: Summary of Unit Operations

Summary Unit Table			
Unit	Heat Duty ($kJ\,s^{-1}$)	Temperature (K)	Pressure (bar)
SEMR (Retentate)	563028	673	8
SEMR (Permeate)	563028	673	0.15
Flash Separator	-744019	216.81	1
Methanator	-4953.32	673	1

$$CH_4 + H_2O \rightleftharpoons CO + 3\,H_2, \tag{1}$$

$$CO + H_2O \rightleftharpoons CO_2 + H_2, \tag{2}$$

$$CH_4 + 2\,H_2O \rightleftharpoons CO + 4\,H_2, \tag{3}$$

$$CO_2 + CaO \longrightarrow CaCO_3 \tag{4}$$

and they were modeled as a stoichiometric reactor since the stoichiometry of the reactions were known and the conversion was able to be specified.[2] Specifically, the stoichiometric reactor was just mapped out to the default option, which was a horizontal, jacketed, agitated reactor, in order for the temperature to be realistically controlled if built physically. Equation 1 and Equation 3 are used to produce hydrogen in two different stages. Equation 1 is first used to create CO and H_2, then Equation 3 is used to create additional H_2 afterwards. The reaction in between those two, Equation 2, is used to convert the CO to CO_2 and create more H_2. The final reaction, Equation 4, is used to capture CO_2 and convert it to $CaCO_3$ with the help of CaO in order to decrease the amount of CO_2 being emitted into the atmosphere.[7] All 4 of these reactions take place in this reactor since that's how it would be realistically modeled according to Lee et al.[2] The SMR reactions take place outside of the membrane where the retentate is located, while the calcination process occurs on the walls of the retentate space. The hydrogen then permeates through the membrane into the permeate stream. The reactor had a temperature of 673 K since that was the minimum temperature allowed for this project and a pressure of 8 bar; this temperature and pressure were chosen based off of previous literature values.[7,8]

After this block is a separator that acts as the membrane of the process. The separator was designed in such a way that allowed for the selection of the permeate and retentate streams leaving the membrane. Specifically, the permeate stream contains H_2 and H_2O while the retentate stream contains everything else. A membrane efficiency of 90% was chosen in order to realistically simulate a membrane in Aspen. However, membranes aren't typically given efficiencies; rather, they're given membrane permeance values to determine their effectiveness. Thus, this 90% value is the best and only way to simulate a membrane in Aspen. Consequently, the SEMR consists of just the stoichiometric reactor and the membrane. The products of the SEMR are sent to two different places. One of these is the flash drum that separates the hydrogen and water. The other place it'll be sent back to is the methanation reactor, which is also modeled as a stoichiometric reactor. The stoichiometric reactor contains the methanation reactions, which are shown below.[9]

$$CO + 3\,H_2 \longrightarrow CH_4 + H_2O \tag{5}$$

$$CO_2 + 4\,H_2 \longrightarrow CH_4 + 2\,H_2O \tag{6}$$

Leftover CO and CO_2 from the retentate stream reacts with some H_2 that leaves the vapor stream of the flash drum. The purpose of this reaction is to reduce the amount of CO being emitted into the atmosphere. However, the drawback of this is that CH_4 is produced; ultimately, CO and CO_2 are more dangerous, so it is a sacrifice that has to be made. The temperature of this reactor is at 673 K in order to minimize the amount of CO coming out of the tail gas. After trying different temperatures, 673 K was found to be the most ideal since it allowed for a small amount of CO leaving the reactor. Leaving this reactor is the tail gas, which consists of considerably smaller amounts of CO and CO_2 compared to normal SMR processes. In summary, a greater amount of H_2 is able to be produced while the amount of CO_2 entering the atmosphere is heavily reduced.

8 Discussion

As a result of the simulations done in Aspen, the SE-SMR process was found to be a more effective way of producing H_2 while reducing the amount of CO_2 emitted into the environment compared to the regular SMR process. This modification of the SMR process is what allows for the increased H_2 production and reduced CO_2 emissions. The emission of CO_2 in this model was calculated to be 17.979 $\mathrm{kmol\,h^{-1}}$ compared to the average CO_2 emissions of 750 $\mathrm{kmol\,h^{-1}}$ in the current SMR process.[6] This large difference in the CO_2 emissions demonstrates how environmentally friendly the SE-SMR process is compared to a standard SMR process. Modifications were done to previous literature on the SEMR process by separating the SEMR into two individual blocks: a stoichiometric reactor and a separator to act as the membrane. The separation of the SEMR block was done to model the process as closely as it can be in Aspen. Once simulations began in Aspen, the target flow rate of H_2 was actually surpassed; as seen in Table 1, 3476.89 $\mathrm{kmol\,h^{-1}}$ was produced. This is considerably more than the value that is produced by normal SMR plants, which is about 1858 kmol h^{-1}.[6] As a result, the process here is able to produce H_2 while emitting significantly less CO_2.

9 Sizing and Materials

Sizing of the major equipment and unit operations were done using hand calculations as well as Aspen Plus. The SEMR reactor was sized using a combination of hand calculations and Aspen. Within this reactor, 2 things needed to be sized: the actual membrane itself, and the reactor that will hold the membrane.

The membrane was sized using data acquired from Aspen simulation and equations from Nordio et al., Kim et al., and Lee et al. Firstly, the H_2 permeate flow rate equation from Lee et al. was utilized to calculate the value for DL, which was the diameter of the membrane multiplied by its length. The equation is listed below:

$$F_{\mathrm{H_2,\,perm}} = \mathrm{H}_{2,\mathrm{permeance}} \times \pi DL \times (P_{\mathrm{H_2,\,ret}} - P_{\mathrm{H_2,\,per}}) \tag{7}$$

where $F_{\mathrm{H_2,\,perm}}$ is the flow rate of H_2 in the permeate, $\mathrm{H}_{2,\mathrm{permeance}}$ is the permeance of H_2, $P_{\mathrm{H_2,\,ret}}$ is the partial pressure of H_2 in the retentate and $P_{\mathrm{H_2,\,per}}$ is the partial pressure of H_2 in the permeate.[2] Since Aspen isn't able to model a membrane accurately, data was taken from Nordio et al. in order to use for this equation. Specifically, the membrane permeance and retentate/permeate pressures were chosen from Nordio et al. depending on the type of membrane that was being worked on, which in this case was a c-tPd membrane.[8] Using this equation, a diameter/length lumped value, DL, was determined; a diameter of 3.4 m, the number of passes (n), and the number of stories (h) were then assumed in order to get the length of the membrane. Namely, n was set at 10 and h was set at 5. This resulted in a length L of approximately 10.5 m for each pass. In order to fit the membrane within the reactor, the plan was to have it snake within the reactor. Then, the ratio between the membrane and reactor lengths and diameters were calculated using Kim et al.'s data and were applied using the membrane length and diameter obtained earlier.[7] This resulted in a reactor length L_R of approximately 12.6 m, which had to be multiplied by 50 in order to get the total size since this was just accounting for one pass of the membrane; the reactor diameter D_R was then determined to be 6.8 m. The calculations for these are provided in the Appendix in Figure 3.

The flash drum was sized purely using hand calculations since Aspen returned an H/D ratio greater than 4, which is not an optimal ratio for flash drum sizes. The sizing process used originates from Seader's *Separation Process Principles*.[10] The flash drum height and diameter were determined to be approximately 13 m and 3.3 m respectively. It was then mapped to its default option, which is known as a "DVT cylinder", since this is just a regular flash drum. Finally, the methanator was sized using Aspen using the mapping option that was discussed previously. It was specifically mapped as a vertical, jacketed, agitated tank since the temperature had to be controlled so that the products won't exit at an exceedingly high temperature that would surpass the requirements set by the project statement. Its diameter and height were calculated to be approximately 2.9 m and 10.4 m respectively. Just like the calculations for the membrane and reactor, the flash drum and methanator calculations are provided in the Appendix in Figure 4.

In regards to the materials of construction for the unit operations, they were chosen based off what's commonly used as well as cost. Initially, the plan was to have all of the reactors made out of stainless steel, specifically SS 304. This is because stainless steel has been used in some SMR small scale processes before for its reactors.[11] However, after determining the economic costs of using stainless steel for the process, it was determined that it was too expensive. Although it's a good material because of its strength and corrosion resistance, it is a costly material. Thus, it was decided that stainless steel only be used for one of the pieces of equipment, the flash drum, while the others would be made out of carbon steel. The flash drum was chosen to be made out of stainless steel since it's operating at a colder temperature than the rest of the other pieces of equipment. It's operating at a lower temperature since a purity of 99.99% was desired, and having it at this temperature resulted in this purity when inputted into Aspen's design specification option. Since carbon steel is not good for lower temperatures, stainless steel was used for the flash drum. As for the SMR reactor and methanator, they were both chosen to be carbon steel since it's inexpensive and is operating within the temperature range of carbon steel. Additionally, although stainless steel has been used in some SMR processes before as mentioned earlier, carbon steel has also been used for SMR reactors as well.[12] Thus, it was decided that the two most commonly used materials would be used for this project's design. For the membrane, a Pd membrane was chosen due to the aforementioned benefits. Specifically, a c-tPd membrane was chosen since it was able to reduce the size of the SEMR reactor and had a better effective permeability compared to other membranes.[8] Other membranes were made out of different materials, such as alumina-carbon composite, but its performance was not as good as the c-tPd membrane and would've made the size of the SEMR reactor much bigger.[8] Thus, it was beneficial to select c-tPd as the membrane material.

10 Economic Analysis

To determine the economic viability of the SE-SMR process, an economic evaluation was done. To begin the economic evaluation, the price of the materials used in the process was determined. In Table 3 the price of each material is shown. The hourly use of each material is converted to the dollar amount needed each year for the production and is shown in Table 3. The sum of yearly cost of each material is summed to determine the total cost of yearly production. An important note to make is the addition of the c-tPd membrane in the list of raw materials. The c-tPd membrane was added due to the lifespan that these kinds of membranes have. Typically Pd membranes of this type only perform continuously for a month which would require its replacement every month.[4] The price of the c-tPd membrane was determined by the surface area the membrane required which was calculated in the Sizing & Materials section. The next aspect that was evaluated was the capital cost of each unit in the SE-SMR process. The units included in the SE-SMR and their prices are shown in Table 4. The cost of each of units were calculated by Aspen Plus Economic Analyzer. Aspen determined the cost of each unit by their dimensions and the material the units were made of. The sizing and materials of each unit were determined previously in the Sizing & Materials section. The sum of each of the units is the total capital cost.

Table 3: Annual Capital Cost of Production c-tPd: ceramic supported thin Pd-Ag membrane

Cost of Raw Materials & Monthly Membrane Replacement			
Raw Material	\$/Unit	Amount Required	Cost of Raw Material
CH_4	\$4.81 MM^{-1} Btu^{-1} [13]	198,489,433.5 kg $year^{-1}$	\$46,306,470.27 $year^{-1}$
H_2O	\$0.00337 kg^{-1} [5]	6,687,970,742 kg $year^{-1}$	\$5,971,947.39 $year^{-1}$
CaO	\$31.2 kg^{-1} [14]	658,420,845.4 kg $year^{-1}$	\$20,542,730.38 $year^{-1}$
c-tPd	\$7,517,040.66 /membrane	12 $year^{-1}$	\$90,204,487.87 $year^{-1}$
Total Cost of Raw Materials & Monthly Membrane Replacement			\$163,025,635.91 $year^{-1}$

Table 4: Fixed Capital Cost

Fixed Capital Cost	
Unit	Cost of Unit
Steam Methane Reactor	\$199,070,000.00
Flash Drum	\$857,500.00
Methanator Drum	\$1,375,100.00
Total Fixed Capital Cost of Investment	\$208,819,640.66 $year^{-1}$

Table 5: Annual Revenue

Hydrogen Produced & Selling Price	
Yearly Amount	56,019,206.64 kg $year^{-1}$
Price	\$4.47 kg^{-1} [15]
Total Revenue	\$250,125,757.64

To continue, the annual revenue is determined by taking the amount of hydrogen that is being produced annually and multiplying that by the price at which hydrogen is being sold at. In Table 5, the price of hydrogen is shown along with the amount that is expected to be produced. With the selling price of the hydrogen and the amount of hydrogen is being produced, the annual revenue can be determined.

In regards to the plant lifespan, the plant is estimated to operate for 20 years.[16] The estimated 20 year lifespan was attained from previous literature discussing lifespans of SMR plants. With the 20 years of operation the plant is expected to produce $916,509,041. This amount is only expected if the c-tPd membrane only requires monthly replacements and the price of all the materials mentioned stay constant.[4] The economic analysis of the SE-SMR process chart is displayed in the Appendix as Figure 5. Continuing with Figure 5, the NPV is calculated to be $115,991,829.87. With a positive net present value, this indicates that the SE-SMR process is a profitable invest with relatively large returns.

A few assumptions to mention with regard to the economic analysis is the performance of the c-tPd membrane. The c-tPd membrane is a relatively new technology and its performance can improve over the lifespan of the plant. The improvement of the c-tPd membrane may lead to increased cost which will affect the cash flow of the SE-SMR project or reduced cost; however, if the improvement is with regards to the short lifespan of palladium membranes. Another assumption to mention is the price of hydrogen. The price of hydrogen that is used based on how other green energy sources are performing and the cost of producing hydrogen energy. As a result, revenue will change throughout the plant's lifespan.

11 Safety and Environmental Considerations

Research has shown that the effects of the greenhouse gasses of CO and CO_2 produced from the system are very harmful for modern society. One of the main effects was the aspect on how the emitted CO and CO_2 pollutes the atmosphere to where it is affecting human health.[17] People, specifically children and those who have asthma, have trouble breathing when these greenhouse gasses are circulating around the Earth's atmosphere.[18] As more man-made machines are being manufactured, more CO and CO_2 are being pumped in from the energy sources, which allows more of the ambient concentrations to linger in the atmosphere.[18] The rise of these gasses could result in affecting the lives of both current and future generations.

Not only is human health affected, but also the effects of CO and CO_2 causes climate change to occur as well. Initially, the rise of pollutants from greenhouse gasses results in the rise in global temperature.[19] With the rise in temperatures, areas like the East and West parts of Amazonia are being in danger due to the rise of climate change and deforestation to occur.[20] The dry seasons alongside with the warm temperatures will increase deforestation, fire occurrence, rise in more carbon emission, and much more problematic issues for various ecosystems that are around the Amazon.[20] Amazonia is one of the many areas where the rise in CO and CO_2 emissions are affected by climate change.

There are many more issues that are taken into account with the rise of CO and CO_2 in the atmosphere, which includes consequences of other catastrophic events dealing with weather, infrastructures being weakened, rising ocean levels, and much more.[17] These issues are known to be problematic as population increases and demand for more efficient machinery are needed to sustain the demand for society. From the design created, the system tried to negate as much CO and CO_2 being produced while maintaining a high amount of H_2. This will help limit the amount of the toxic emissions being emitted into the atmosphere. Therefore, the most important take away from the HAZOP, as seen in the Appendix in Figure 6, is the minimal output of CO and CO_2. Sensors and alarms are required for the exit of the methanation reactor to measure the CO and CO_2 flow rates leaving the plant. Any higher flow rates than expected would signal an alarm and result in the closing of the valve to prevent excess CO and CO_2 exiting into the atmosphere. In addition, the main reactor, SEMR, is also an important consideration in the HAZOP in Figure 6. Without any feed flow, correct temperature, and pressure the entire process would fail to operate as intended. Therefore, multiple sensors and alarms have been equipped to the SEMR to ensure proper operating conditions, as seen on Figure 2.

The abbreviations in Figure 2 indicates the sensors in the system. TC and TI are the temperature controller and indicator, FC and FI are the flow controller and indicator, and PI is the pressure indicator.

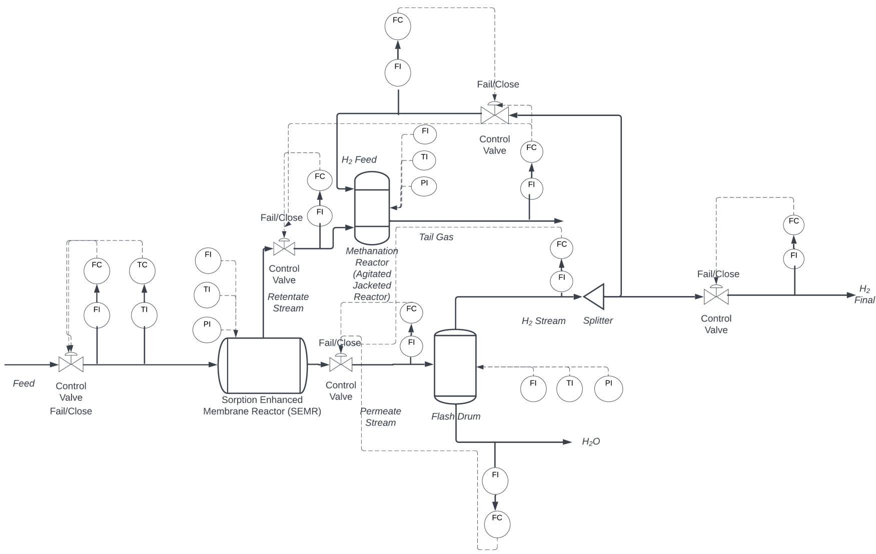

Figure 2: Shown above is the P&ID of the SE-SMR process, which implements various sensors and alarms based off the HAZOP.

12 Global and Societal Impacts

The amount of H_2 being produced from the system allows for an alternate energy source to be created. Instead of industries relying on burning fossil fuels for energy, H_2 can be an alternate energy source, and this gas can be implemented in many ways.[21] For example, companies in China are taking advantage of using hydrogen gas by creating these hydrogen-fuel-cell batteries that are used for modern vehicles.[22] The overall comparison to be made here is that H_2 is much more efficient to have rather than any kind of fossil fuels, as well as the gas itself is both renewable and eco-friendly.[21] By changing energy sources, it would result in the decrease in the world's carbon footprint due to less of CO and CO_2 being emitted into the Earth's atmosphere.

13 Conclusions & Recommendations

In conclusion, the SE-SMR process is a cost-effective process in reducing the amount of CO_2 and CO emitted to the environment. The use of the SMR c-tPd membrane reactor allows for the steam methane reforming process to occur efficiently while separating the H_2 from the CO_2 and CO. As mentioned previously, the SE-SMR process produces more H_2 and less CO_2 simultaneously compared to the regular SMR process.[6] Based off of Aspen simulations, the SE-SMR process produced 8.86 kg H_2/kg CO_2; when compared to the average SMR values of 0.24 kg H_2/kg CO_2, the SE-SMR process is shown to be substantially better. Over a 20 year plant lifespan, a little over $900,000,000 can be made assuming nothing goes wrong, which means that a sizable profit can be achieved. Since CO_2 emissions are able to decrease because of the SE-SMR method, less greenhouse gases will enter the atmosphere and thereby reduce the harmful effects of climate change and global warming. Furthermore, the H_2 produced is able to be used for a variety of different applications, such as the ones described earlier in the report. In the future, several recommendations can be implemented in order to further advance the use of the SE-SMR process in industry.

In regards to the recommendations of the SE-SMR process, more research can be invested in the Pd membrane technology. The issue that was present throughout the design of the SMR membrane reactor is the lifespan, the conditions the membrane needed to be to perform adequately, and the cost to manufacture the membrane. Improvements that can be made to the SMR membrane reactor are changing the operating conditions that the membrane must operate at, specifically the temperature and pressure. Reducing the pressure and the temperature would reduce energy costs, improving the viability of the SMR membrane reactor. Another improvement that can be made is increasing the lifespan, reducing the cost of using a SMR membrane reactor. Because the Pd membrane technology is relatively novel, the manufacturing methods at the moment are expensive, leading to a costly manufacturing process. Another recommendation that can be made to improve the viability of the SE-SMR process is doing more research on implementing the CaO into the SMR membrane reactor. The issue in the design discussed in this report was the inability to have the CaO be embedded into the outermost wall of the SMR membrane reactor. Current research on this design is not substantial enough to have a design with CaO. Consequently, the calcination/carbonation process could not be implemented into the SMR membrane reactor. Another item to note is selling or using the $CaCO_3$. The $CaCO_3$ can be sold to increase the revenue. Overall, the SE-SMR process is a potential green H_2 production method that may replace the traditional SMR process.

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Appendix

Relevant Equations

The equation used to calculate the heat duty in the flash separator is defined as,

$$Q = Lh_L + Vh_V - Fh_F \tag{8}$$

where h_L, h_V, and h_F are of the liquid, vapor, and the feed enthalpies, respectively. L, V, and F, are the molar flow rates of the liquid, vapor and feed streams, respectively. Q is the heat duty.

The equation used to calculate mass balance on the flash separator is defined as,

$$Fz_i = Vy_i + Lx_i \tag{9}$$

where F,V, and L are the feed, vapor and liquid flow rates, respectively. Z_i, y_i, and x_i are the mole fraction of the feed, vapor, and liquid components, respectively. The i subscript is used to denote the component.

The equation used to define the mass transfer in the stoichiometric reactors is defined below

$$\frac{dn_i}{dt} = \Sigma n_{i,out} - \Sigma n_{i,in} \pm R_i V \tag{10}$$

where n_i is the molar flow rate of the respective component, R_i is the rate of reaction for the respective component, and V is the volume of the reactor. The $\pm$ indicates whether the component is a product, a positive, or a reactant, a negative.

Finally, the energy balance for the stoichiometric reactors involves the two equations below:

$$\Delta G = \Delta H - T\Delta S \tag{11}$$

where T is the temperature, ΔG is the change in Gibbs free energy, ΔH is the change in enthalpy, and ΔS is the change in entropy. This equation can be rearranged to find ΔH. ΔH can be used in the following equation,

$$Q = n\Delta H \tag{12}$$

where n is the molar flow rate.

Sizing Calculations

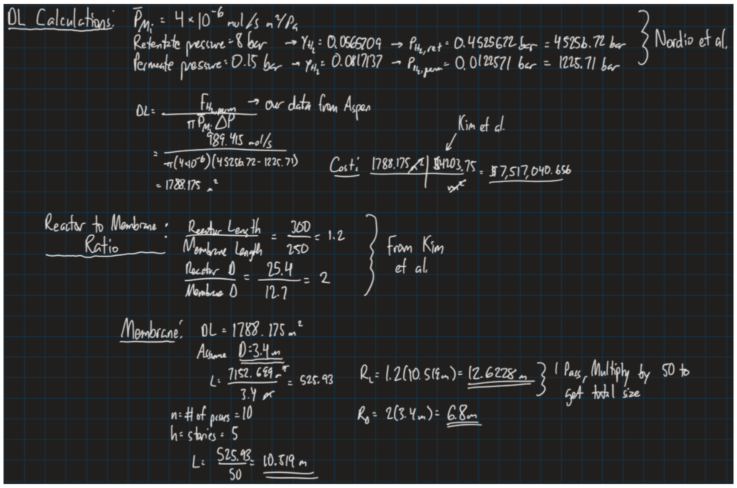

Figure 3: Shown above are the hand calculations done for the membrane reactor, which entailed the membrane and the reactor itself. The numbers that were used were from Aspen data, Kim et al., Lee et al., and Nordio et al.

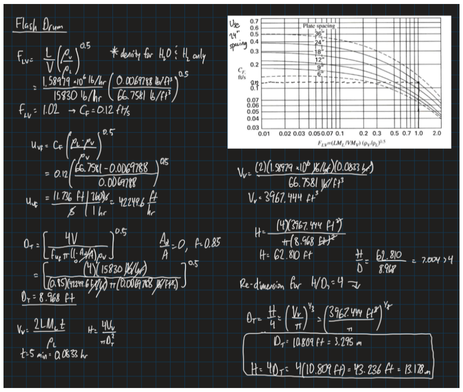

Figure 4: Shown above are the hand calculations done for the flash drum, which used the process from Seader et al.[10] Data was pulled from Aspen simulations.

Economic Analysis of SE-SMR Process

Fixed Capital Investment	Yearly Sales Revenue (After Startup)	Cash Cost of Production	Taxation Rate	Interest Rate %
$ 201,302,600.00	$ 250,125,757.64	$ 163,025,635.91	21%	0.12

Year	Gross Profit	Depreciation Charge	Taxable Income	Taxes Paid	Cash Flow After Tax	5-year recovery %	Discount Factor	n	Present Value
0	$ -	$ -	$ -	$ -	$ (100,651,300.00)	0.2	1	0	$ (100,651,300.00)
1	$ -	$ -	$ -	$ -	$ (100,651,300.00)	0.32	0.892857143	1	$ (89,867,232.14)
2	$ 87,100,121.73	$ 40,260,520.00	$ 46,839,601.73	$ -	$ -	0.192	0.797193878	2	$ -
3	$ 87,100,121.73	$ 64,416,832.00	$ 22,683,289.73	$ 9,836,316.36	$ (85,761,830.55)	0.1152	0.711780248	3	$ (61,043,577.00)
4	$ 87,100,121.73	$ 38,650,099.20	$ 48,450,022.53	$ 4,763,490.84	$ 82,336,630.88	0.1152	0.635518078	4	$ 52,326,417.44
5	$ 87,100,121.73	$ 23,190,059.52	$ 63,910,062.21	$ 10,174,504.73	$ 76,925,617.00	0.0576	0.567426856	5	$ 43,649,660.98
6	$ 87,100,121.73	$ 23,190,059.52	$ 63,910,062.21	$ 13,421,113.06	$ 73,679,008.66		0.506631121	6	$ 37,328,078.77
7	$ 87,100,121.73	$ 11,595,029.76	$ 75,505,091.97	$ 13,421,113.06	$ 73,679,008.66		0.452349215	7	$ 33,328,641.76
8	$ 87,100,121.73	$ -	$ 87,100,121.73	$ 15,856,069.31	$ 71,244,052.41		0.403883228	8	$ 28,774,277.86
9	$ 87,100,121.73	$ -	$ 87,100,121.73	$ 18,291,025.56	$ 68,809,096.16		0.360610025	9	$ 24,813,249.89
10	$ 87,100,121.73	$ -	$ 87,100,121.73	$ 18,291,025.56	$ 68,809,096.16		0.321973237	10	$ 22,154,687.40
11	$ 87,100,121.73	$ -	$ 87,100,121.73	$ 18,291,025.56	$ 68,809,096.16		0.287476104	11	$ 19,780,970.89
12	$ 87,100,121.73	$ -	$ 87,100,121.73	$ 18,291,025.56	$ 68,809,096.16		0.256675093	12	$ 17,661,581.15
13	$ 87,100,121.73	$ -	$ 87,100,121.73	$ 18,291,025.56	$ 68,809,096.16		0.22917419	13	$ 15,769,268.89
14	$ 87,100,121.73	$ -	$ 87,100,121.73	$ 18,291,025.56	$ 68,809,096.16		0.204619813	14	$ 14,079,704.36
15	$ 87,100,121.73	$ -	$ 87,100,121.73	$ 18,291,025.56	$ 68,809,096.16		0.182696261	15	$ 12,571,164.61
16	$ 87,100,121.73	$ -	$ 87,100,121.73	$ 18,291,025.56	$ 68,809,096.16		0.163121662	16	$ 11,224,254.12
17	$ 87,100,121.73	$ -	$ 87,100,121.73	$ 18,291,025.56	$ 68,809,096.16		0.145644341	17	$ 10,021,655.46
18	$ 87,100,121.73	$ -	$ 87,100,121.73	$ 18,291,025.56	$ 68,809,096.16		0.13003959	18	$ 8,947,906.66
19	$ 87,100,121.73	$ -	$ 87,100,121.73	$ 18,291,025.56	$ 68,809,096.16		0.116106777	19	$ 7,989,202.38
20	$ 87,100,121.73	$ -	$ 87,100,121.73	$ 18,291,025.56	$ 68,809,096.16		0.103666765	20	$ 7,133,216.41
				Total After Plant Life	$ 916,509,041.03			NPV	$ 115,991,829.87

Figure 5: Calculations of the SE-SMR process were done over a 20 year lifespan with a 5-year recovery period.

HAZOP

Guide Word	Deviation	Cause	Consequences and Action
*Vessel - **SEMR***			
Intention - to react CH_4 and H_2O to produce H_2			
NO	Flow	Blockage in SEMR	Unable to cause reactions; set an alarm and schedule maintenance
LESS	Temperature	Lack of heating	Reactions will not occur at desired specifications; add a temperature alarm and add heat
MORE	Pressure	Build up of products in reactor	Damage to reactor; add a pressure alarm and vent out products through product stream lines
Line Feed			
Intention - to carry CH_4 and H_2O to SEMR			
NO	Flow	Major leak in Line Feed	Hazardous materials exposed to workers; set a valve and alarm on Line Feed
NO	CH_4 Flow	Not enough CH_4 in storage	No reaction occurs in SEMR; routinely check CH_4 level
NO	H_2O Flow	Not enough H_2O in storage	No reaction occurs in SEMR; routinely check H_2O level
Line Permeate Stream			
Intention - to carry permeated H_2 and H_2O to flash drum for separation			
NO	Flow	Major leak in Line Permeate Stream	Hazardous materials exposed to workers and no materials being heated; set a valve and alarm on Line Permeate Stream
NO	H_2 Flow	Membrane is failing to separate H_2	H_2 can't be purified; add a flow sensor
NO	H_2O Flow	Membrane is failing to separate H_2O	Too much flow into Methanation Reactor and can damage reactor; add a flow sensor
Line Retentate Stream			
Intention - to carry by products in retentate to methanation reactor			
NO	Flow	Major leak in Line Retentate	Hazardous materials exposed to workers and no materials being heated; set a valve and alarm on Line Retentate Stream
MORE	H_2 Flow	Membrane is failing to separate H_2	H_2 can't be purified; add a flow sensor
MORE	H_2O Flow	Membrane is failing to separate H_2O	Too much flow into Methanation Reactor and can damage reactor; add a flow sensor
*Vessel - **METHANATION REACTOR***			
Intention - to convert residual CO and CO_2 into methane and water			
LESS	Temperature	Lack of heating	Reactions will not occur at desired specifications; add a temperature alarm and add heat
MORE	Pressure	Reaction not occuring	Damage to reactor; add a pressure alarm
NO	Flow	Blockage within reactor	Unable to cause reactions; set an alarm and schedule maintenance
Line Retentate Stream			
Intention - to carry by products in retentate to methanation reactor			
NO	Flow	Major leak in Line Retentate	Hazardous materials exposed to workers and no materials being heated; set a valve and alarm on Line Retentate
MORE	H_2 Flow	Membrane is failing to separate H_2	H_2 can't be purified; add a flow sensor
MORE	H_2O Flow	Membrane is failing to separate H_2O	Too much flow into Methanation Reactor and can damage reactor; add a flow sensor
Line Tail Gas			
Intention - to carry products of methanation reactor out of plant			
NO	Flow	Major leak in Line Tail Gas	Hazardous materials exposed to workers; set a valve and alarm on Line Tail Gas
MORE	CO_2 Flow	Reaction not occuring	Hazardous materials exposed; add a flow sensor
MORE	CO Flow	Reaction not occuring	Hazardous materials exposed; add a flow sensor
Line H_2 Feed			
Intention - to feed H_2 into methanation reactor			
NO	Flow	Major leak in Line H_2 Feed	Hazardous materials exposed to workers; set a valve and alarm on Line H_2 Feed
MORE	H_2 Flow	Splitter is not operating correctly	H_2 needlessly being wasted; add a flow sensor
LESS	H_2 Flow	Splitter is not operating correctly	Reactions will not occure at desired specifications; add a flow sensor

*Vessel - **FLASH DRUM***			
Intention - to separate H_2O and H_2			
HIGH	Temperature	Lack of cooling	Purity of H_2 decreases; add a temperature alarm
MORE	Pressure	Lack of cooling	Purity of H_2 decreases; add a pressure alarm
NO	Flow	Blockage within Flash Drum	Damage to Flash Drum; add level alarm
Line Permeate Stream			
Intention - to carry permeated H_2 and H_2O to flash drum for separation			
NO	Flow	Major leak in Line Permeate Stream	Hazardous materials exposed to workers; set a valve and alarm on Line Permeate Stream
NO	H_2 Flow	Membrane is failing to separate H_2	H_2 can't be purified; add a flow sensor
NO	H_2O Flow	Membrane is failing to separate H_2O	Too much flow into Methanation Reactor and can damage reactor; add a flow sensor
Line H_2O			
Intention - to carry H_2O out of plant			
NO	Flow	Major leak in Line H_2O	Flooding of plant; set a valve and alarm on Line H_2O
MORE	H_2 Flow	Flash Drum is failing to separate H_2	H_2 yield decreases; add a flow sensor
LESS	H_2O Flow	Flash Drum is failing to separate H_2O	Purity of H_2 decreases; add a flow sensor
Line H_2			
Intention - to carry H_2 into Splitter			
NO	Flow	Major leak in Line H_2	Hazardous materials exposed to workers; set a valve and alarm on Line H_2
MORE	H_2O Flow	Flash Drum is failing to separate H_2O	Purity of H_2 decreases; add a flow sensor
LESS	H_2 Flow	Flash Drum is failing to separate H_2	Purity of H_2 decreases; add a flow sensor
*Vessel - **SPLITTER***			
Intention - splits Line H_2 into H_2 Final and H_2 Feed			
LOW	H_2 Flow	Flash Drum is failing to separate H_2	Purity of H_2 decreases; add a flow sensor
MORE	H_2O Flow	Flash Drum is failing to separate H_2O	Purity of H_2 decreases; add a flow sensor
NO	Flow	Major leak in Splitter	Hazardous materials exposed to workers; set a valve and alarm on Splitter
Line H_2			
Intention - to carry H_2 into splitter			
NO	Flow	Major leak in Line H_2	Hazardous materials exposed to workers; set a valve and alarm on Line H_2
MORE	H_2O Flow	Flash Drum is failing to separate H_2O	Purity of H_2 decreases; add a flow sensor
LESS	H_2 Flow	Flash Drum is failing to separate H_2	Purity of H_2 decreases; add a flow sensor
Line H_2 FINAL			
Intention - carries pure H_2 out of plant			
NO	Flow	Major leak in Line H_2 Final	Hazardous materials exposed to workers; set a valve and alarm on Line H_2 Final
MORE	H_2O Flow	Flash Drum is failing to separate H_2O	Purity of H_2 decreases; add a flow sensor
LESS	H_2 Flow	Flash Drum is failing to separate H_2	Purity of H_2 decreases; add a flow sensor
Line H_2 Feed			
Intention - to feed H_2 into Methanation Reactor			
NO	Flow	Major leak in Line H_2 Feed	Hazardous materials exposed to workers; set a valve and alarm on Line H_2 Feed
MORE	H_2 Flow	Splitter is not operating correctly	H_2 needlessly being wasted; add a flow sensor
LESS	H_2 Flow	Splitter is not operating correctly	Reactions will not occure at desired specifications; add a flow sensor

Figure 6: HAZOP for all the unit operations and streams in the project.

Individual Contributions

1. Rodrick Alberto
    - Assumptions
    - Methods
    - Global and Societal Impacts
    - Safety and Environmental Considerations
    - P&ID
2. Jed Durante
    - Methods
    - Description of Unit Operations
    - PFD
    - Discussion
    - Aspen Simulation
    - Sizing
    - Materials of Construction
    - Conclusions and Recommendations
    - Poster
3. Phat Le
    - Abstract
    - Introduction
    - Basis of Design
    - Specifications

- HAZOP
- Aspen Simulation
- Poster

4. Rafael Vasquez

    - Mole Balance Table
    - Summary Unit Table
    - Aspen Simulation
    - Economic Analysis
    - Optimization & Comparison
    - Conclusion and Recommendations
    - Poster

Project Plan

Task	Start	Duration/Planned Finish Date	Responsible Person	Completed Status
Interim Report				
Basis of Design	4/5/22	5/1/22	Phat	Completed
Specifications	4/5/22	5/1/22	Phat	Completed
Assumptions	4/5/22	5/1/22	Rodrick	Completed
PFD	4/5/22	5/1/22	Jed	Completed
Description of Unit Operations	4/5/22	5/1/22	Jed	Completed
Stream Tables	4/5/22	5/1/22	Rafael	Completed
Summary Tables: Unit Operatio	4/5/22	5/1/22	Rafael	Completed
Methods	4/5/22	5/1/22	Rodrick	Completed
Discussion	4/5/22	5/1/22	Jed	Completed
Final Report				
Abstract	5/10/22	5/29/22	Phat	Completed
Intro	5/10/22	5/29/22	Phat	Completed
Optimization/Comparison	5/10/22	5/29/22	Rafael	Completed
Sizing	5/10/22	5/29/22	Jed	Completed
Materials of Construction	5/10/22	5/29/22	Jed	Completed
Fixed Cap Cost	5/10/22	5/29/22	Rafael	Completed
NPV	5/10/22	5/29/22	Rafael	Completed
Econ evaluation results	5/10/22	5/29/22	Rafael/Jed	Completed
HAZOP	5/10/22	5/29/22	Phat	Completed
P&ID	5/10/22	5/29/22	Rodrick	Completed
Environmental	5/14/22	5/29/22	Rodrick	Completed
Global and/or societal impacts	5/10/22	5/29/22	Rodrick	Completed
Conclusions and Recs	5/10/22	5/29/22	Rafael/Jed	Completed
Poster				
Designs	5/30/22	5/31/22	Phat/Rafael/Jed	Completed

Figure 7: Project schedule over the entire quarter.

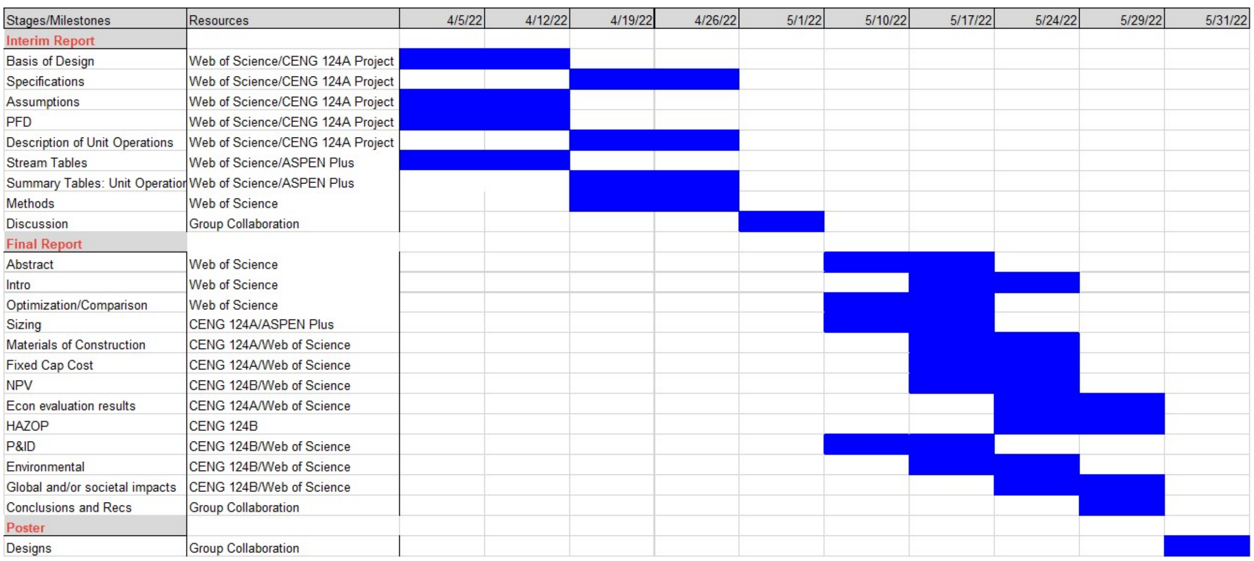

Figure 8: Gantt chart of project tasks.