Sabatier System Design Study for a Mars ISRU Propellant Production Plant

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As NASA looks towards human missions to Mars, an effort has started to advance the technology of a Mars in situ resource utilization (ISRU) Propellant Production Plant to a flight demonstration. This paper will present a design study of the Sabatier subsystem. The Sabatier subsystem receives carbon dioxide, CO_2, and hydrogen, H_2, and converts them to methane, CH_4, and water, H_2O. The subsystem includes the Sabatier reactor, condenser, thermal management, and a recycling system (if required). This design study will look at how the choice of reactor thermal management, number of reactors, and recycling system affect the performance of the overall Sabatier system. Different schemes from the literature involving single or cascading reactors will be investigated to see if any provide distinct advantages for a Mars propellant production plant.

Nomenclature

AES	=	Advanced Exploration Systems
CH_4	=	methane
CO	=	carbon monoxide
COCO	=	Cape Open to Cape Open Simulator
CO_2	=	carbon dioxide
ΔH	=	heat of reaction
H_2	=	hydrogen
H_2O	=	water
ISRU	=	in-situ resource utilization
LCH_4	=	liquid methane
LO_2	=	liquid oxygen
NASA	=	National Aeronautics and Space Administration

I. Introduction

THE ability of humans to be successful in the planned journey to Mars will be dependent on many things; however, one of the important components of this success will be the ability to utilize the resources that are available on-site. This concept of in-situ resource utilization (ISRU) for Mars has been around for decades[1] and has received increasing attention over the years as an effective method of reducing the required mass and overall cost of various space exploration activities. There are several ISRU technologies that are applicable for a human mission to Mars, such as the capture and conversion of solar energy to electrical power or the extraction of water ice from subsurface soil deposits.[2,3] The usable resources offered by the Martian environment include the atmosphere which

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consists of primarily carbon dioxide, CO_2, as well as both surface and subsurface water, H_2O, ice. Previous work has detailed the concept of using an ISRU propellant production plant as the means to provide on-site refueling capability for a Mars exploration mission.[1,3,4] Although there are several technologies capable of processing the available CO_2, making up 95.3% of the Mars atmosphere, into usable consumables, this paper will focus on the Sabatier reaction. The Sabatier reaction uses transition metal catalysts (such as ruthenium or nickel) to catalyze the methanation of CO_2 by reacting with hydrogen, H_2 (Eq. 1).

$$CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O \quad (1)$$

This is an exothermic reaction (ΔH = -165.4 kJ/mol) in which methane, CH_4, and H_2O are produced at an elevated temperature. This has traditionally been carried out using packed bed reactors[5–7], although recent advances have allowed for the use of new reactor designs[8,9]. All reactor designs have advantages and disadvantages. Regardless of the type of reactor, the challenges for efficient conversion are the same. The reaction is thermodynamically favored at low temperatures, but must be carried out at high temperatures so that the reaction kinetics[10,11] are fast enough. Brooks et al.[12] modeled thermodynamic and kinetic effects in their reactor. At temperatures above 375 °C, the kinetics were fast enough to allow the reaction to proceed to thermodynamic equilibrium but at higher temperatures, the thermodynamics dictated lower conversion. When the temperature was below 375 °C, the kinetics became too slow and conversion efficiency decreased even though thermodynamics predicted higher conversion. The reactor can be made bigger to overcome slow kinetics at low temperatures, but this introduces the complication that accurate reactor sizing equations are needed to trade between different reactor designs. Many reactors that employ different designs have been able to reach thermodynamic conversion efficiencies at their operating temperatures.

The choice and implementation of thermal management systems can be difficult. Many reactor styles have demonstrated thermal management with liquid[12–14] or air cooling[7,15]. Brooks et al. used a cooling fluid loop for temperature control of their microchannel reactor[12]. The reactor was kept at temperatures between 250 – 400 °C in their testing. Sometimes the reactor was kept very close to isothermal conditions and sometimes there was temperature gradient up to 50°C along the length of the channel. Two approaches developed for industrial applications use a thermal control fluid to create isothermal conditions. As the reaction proceeds, the thermal fluid reaches its boiling point and the vapors are condensed and returned to the system. This results in an isothermal reactor a temperature that is the boiling point of the thermal fluid. Blum et al.[13] built and evaluated a liquid phase methanation reactor where catalyst is placed in a liquid thermal fluid and the reactant gas passes through the fluid to reach the catalyst. This resulted in a fluidized bed reactor in a liquid medium. Another approach, submerging one or more traditional packed bed reactors into a thermal fluid was also demonstrated[14]. Air cooling has been successfully demonstrated on a Sabatier reactor employing a highly thermally conducting Microlith catalyst[15]. There is currently a Sabatier reactor on the International Space Station[7]. This is a packed bed reactor where the front third of the reactor operates around 593 °C and the back two thirds are air cooled to around 149 °C. The fast kinetics of the high temperature section allow for approximately 90% conversion and the cooler section allows another 5% conversion. In another effort, a carbon dioxide capture and reduction system was developed that used two Sabatier reactors[6] in series. The first reactor was adiabatic and the second was kept at lower temperatures by using the H_2 reactant gas as cooling fluid. A heat exchanger was in between the two reactors to cool the gas before it entered the second reactor. This setup takes advantage of the fast kinetics in the high temperature adiabatic reactor and better thermodynamic conversion efficiency in the cooled reactor.

Direct air cooling of a reactor will be challenging, and may not work, since Mars atmospheric pressure is so low. A liquid cooling system will increase the overall mass of the system and require a radiator, larger than would be required on Earth, since the heat must ultimately be transferred to the Mars environment. The thermal management system should be given consideration early in the Sabatier system design process. The overall ISRU propellant plant will give off heat[1], so thermal management needs to look not only at the subsystem level but at the system level. The heat flow from systems with either an adiabatic or isothermal reactor followed by a condenser are shown in Figure 1. The total amount of heat removed from the Sabatier subsystem is determined by the extent of the reaction. Heat must be put into the system to heat reactants to the reactor operating temperature. This requires less heat than is given off by the reaction and is normally done with waste heat from the reaction. The remaining thermal energy is removed from the condenser and/or the reactor. Adiabatic reactors, which have been developed for space applications[6,16], do not require a thermal management system. Systems that use adiabatic reactors typically use a second reactor to help conversion or recycle product gas back to the inlet of the reactor to keep the temperatures down. Without either of these schemes, the adiabatic reactor will get too hot to allow good conversion and may reach temperatures that damage the catalyst. Regardless if an isothermal or adiabatic reactor is chosen, a condenser

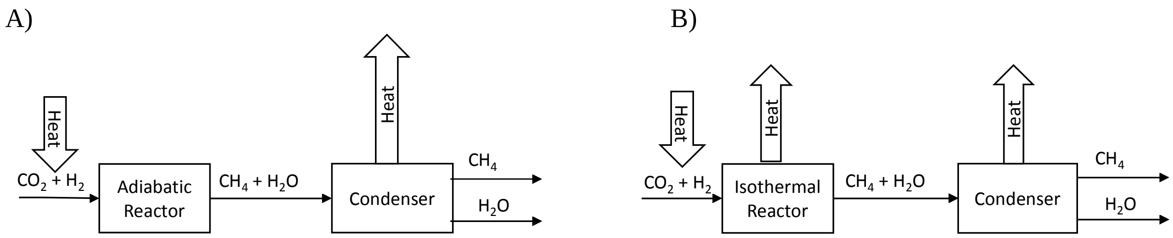

Figure 1. Mass and heat flow schematic for adiabatic, A, and isothermal, B, reactors followed by a condenser.

will always be needed to remove water from the product stream. The use of an adiabatic reactor means that the Sabatier system's thermal management only has to deal with one or more condensers. If an isothermal reactor is used, the thermal management system must cool two components, and those components will be at very different temperatures: the reactor around 400 °C and the condenser around 5°C. If the two cases shown in Figure 1 had the same CO_2 conversion efficiencies, the heat removed from the condenser in A) would be equal to the sum of the heat removed from the isothermal reactor and condenser in case B).

In this paper, a design study of the Sabatier subsystem proposed for use in a Mars ISRU Propellant Production Plant will be reviewed and discussed. Factors including reactor thermal management, single vs multiple reactor architectures, and possible recycling schemes were examined for possible performance effects on the overall conversion efficiency of the Sabatier process. The study looked to see if different configurations could produce the required CH_4, the purity of the CH_4 product, and the thermal loads of reactor and condenser.

II. Methodology

The Sabatier system has been modeled using the Cape Open to Cape Open (COCO) Simulator Environment. CO_2, CO, CH_4, H_2O, H_2, Ar, and N_2 were modeled in the system and the Peng Robinson equations of state were used. Ar and N_2 were not considered reactive species, but were included in the model to evaluate cases where the Mars atmosphere is fed directly into the reactor. The Mars atmosphere composition used in this study was 95.7% CO_2, 2.7% N_2, and 1.6% Ar. The extent of the chemical reaction was calculated using the equilibrium reactor unit operation in COCO. This approach was taken because much of the literature reports CO_2 conversion efficiencies greater than 90% of the thermodynamic limit at a given reactor temperature[12,17–19]. The reactor took into account the Sabatier reaction and the reverse water gas shift (RWGS) reaction using the equilibrium constants given in Swickrath and Anderson[20]. The reactor could be set to operate adiabatically or isothermally. Other unit operations used in the model were the flash separator (condenser) for cooling fluid streams and condensing water, a heat exchanger, mixers, and splitters for recycling. An example COCO flowsheet is shown in Figure 2. The lines are either material streams, which carry gases and liquids, or information streams, which carry values such as the amount of heat given off by a unit operation.

The CH_4 production requirement for this study was 0.34 kg/hr. This is based on a total methane requirement for a human Mars return mission[3] of 6978 kg produced in 428 days, and an assumption that the overall ISRU system will consist of three units, each with the capability to produce half the required product so that if one fails production can continue. The CH_4 production rate was one of the key outputs of the model showing configurations would meet the production requirement. When the production requirement was met, other parameters such as cooling needs and product purity were used to differentiate between different configurations.

This modeling study considered a number of parameters and operational scenarios. The CO_2 feed could either be pure, as would be the case if CO_2 was collected by freezing[21] or some other method that removed other gases, or the feed would have the same composition as the Mars atmosphere as would happen if mechanical compression were used to collect and pressurize CO_2. The operating parameters are given in Table 1. The CO_2 feed rate ranged from a lower limit, which was the stoichiometric amount to

Table 1. Operating parameters for design study.

Parameter	Values
CO_2 feed rate, kg/hr	0.935 – 0.984
H_2 feed rate, kg/hr	0.170 – 0.224
Operating Pressure, psia	8, 15, 75, 150

produce 0.34 kg/hr CH_4, to the high limit, the amount of CO_2 needed to meet the CH_4 requirement with 95% conversion efficiency. The lower bound of the H_2 feed rate range was the stoichiometric amount and the high bound was 5:1 H_2:CO_2 ratio for the highest CO_2 flow rate. The operating pressures were selected as 8, 15, and 75 psia based on project guidance, and 150 psia was evaluated in some cases to see if the enhanced conversion caused by higher pressures made a major difference to the system. When the input feed was Mars atmosphere, the CO_2 feed was the same but there was additional flow of Ar and N_2 in proportion to their concentrations in the atmosphere. Isothermal reactors were evaluated from 300 – 550 °C in 50 °C increments. Parametric studies were performed in COCO to evaluate the effects of CO_2 and H_2 flow rates, pressures, and reactor temperatures. This resulted in many different test cases for each configuration. Some configurations had no cases where requirements were met, some had a few, and sometimes many of the cases met the requirements.

There are many different systems configurations given in the literature and a number of them were evaluated here. Configuration 1 was one reactor followed by a condenser and was evaluated with adiabatic and isothermal reactors. This is the simplest configuration and was chosen to show the thermodynamic limits of conversion for these two types of reactors. Configuration 2 used two adiabatic reactors in series followed by a condenser. Two options for configuration 2 were evaluated: one with a heat exchanger in between the two reactors to remove heat, and the second with a condenser in between to remove both heat and water. Configuration 3 employed one adiabatic reactor followed by a condenser and was evaluated with a recycle stream.

Since reactor size was not considered in this study, some configurations were not included. When an adiabatic reactor is followed by a low temperature isothermal reactor[6], the motivation is normally to minimize the total size. Most of the reaction occurs in the high temperature adiabatic reactor which has fast kinetics, and the low temperature isothermal reactor is there to push the reaction towards completion. Because the low temperature reactor has slow kinetics, if it was the only reactor its size would be bigger than the combined size of the two reactors. However, when an isothermal equilibrium reactor is used in COCO as the final reactor in a series of reactors and size is not considered, it masks the conversion of the earlier reactors. For example, if a reactor operating at 550°C is followed by a reactor at 400°C, the product stream will be the same as if a single reactor at 400°C were modeled. In practice, these systems may have different sizes, but the CO_2 conversion will be the same. Similarly, reactors with temperature gradients[7,12] can be modeled in COCO as separate reactors with different temperatures, but the results will be the same as a model of a single reactor with the coolest temperature in the reactor.

III. Results and Discussions

A. Configuration 1: One reactor and condenser

Figure 2 shows Configuration 1 with an adiabatic reactor and pure CO_2 feed. This configuration was also evaluated with an isothermal reactor and with direct Mars atmosphere feed. When an adiabatic reactor was used, the 0.34 kg/hr CH_4 production rate was not met. The maximum CH_4 production rates and conversions achieved under those conditions are shown in Table 2. There is a slight increase in the CO_2 conversion when Mars atmosphere is fed as would be expected because the N_2 and Ar help to lower the temperature of the reactor. The reactor temperature was at least 664 °C, which exceeds the temperature limit of many catalysts and is not favorable for CH_4 production. A single adiabatic reactor will not meet the needs for propellant production on Mars.

When an isothermal reactor was used, many flow conditions and pressures met the CH_4 production requirement and theoretical CO_2 conversions reached close to 100% when the reactor was held at lower temperatures and higher pressures. The advantage of using an isothermal reactor is the ability to drive high conversion by keeping the reactor temperature low, so it is informative to compare different conditions by looking at the methane purity of the Gas Out stream. The flow conditions at each pressure with the highest methane purity are shown in Table 3 and Table 4. There was not much difference between feeding CO_2 and Mars atmosphere. Although the CO_2 conversion can be raised dramatically with an isothermal reactor, the

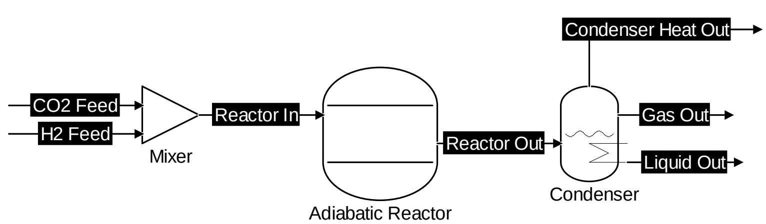

Figure 2. COCO Model input for Configuration 1 using an adiabatic reactor. CO2 Feed, H2 Feed, Reactor In, Reactor Out, Gas Out, and Liquid Out are material streams. Condenser Heat Out is an information stream giving the heat removal need from the condenser.

maximum CH_4 purity never exceeds 88%. There is currently no specification for the purity of CH_4 being produced by the Sabatier system. Even if the downstream liquefaction system and the rocket motor could handle a purity of 88%, this purity will result in lost reactants, putting a burden on commodity collection systems upstream of the Sabatier. A gas recycling system can return unreacted CO_2 and H_2 to the reactor limiting this effect.

Figure 3 plots CH_4 production rate and purity for an isothermal reactor at 350 °C and 15 psia in configuration 1. The production rate increases with both CO_2 feed rate and H_2:CO_2 ratio as expected. The CH_4 purity has the opposite trend. For this case, the best purity at an acceptable CH_4 production rate occurs at about 4.3 H_2:CO_2 ratio and the highest CO_2 feed rate. Figure 4 shows the CH_4 production rate and mole fraction at different isothermal reactor temperatures and operating pressures for a CO_2 feed of 0.935 kg/hr and 4.0 H_2:CO_2 ratio. Decreasing temperature and increasing pressure increase the production rate and purity of CH_4 in the Gas Out stream. These trends will be the same regardless of the CO_2 feed rate and H_2:CO_2 ratio.

The thermal management needs can be evaluated with this model. The adiabatic reactor does not require any thermal management, but the condenser had to remove 1.1 kW of thermal energy at the maximum CH_4 production rate. When an isothermal reactor is used, the reactor and condenser both need to remove heat. For the best conversions at 75 psia shown in Table 3 and Table 4, the reactor and condenser need to remove 780 and 750 W respectively. There was little difference whether pure CO_2 or Mars atmosphere was the feed. The total amount of heat removed is higher when the isothermal reactor is used since the reaction proceeds more towards completion and generates more heat.

Table 2. Maximum CH_4 production rate, kg/hr, and percent CO_2 conversion, in parenthesis, for Configuration 1 using an adiabatic reactor.

	System pressure			
Feed	8 psia	15 psia	75 psia	150 psia
CO_2	0.20 (55.7%)	0.21 (58.4%)	0.24 (65.6%)	0.25 (69.0%)
Mars Atm.	0.20 (56.3%)	0.21 (58.6%)	0.24 (65.8%)	0.25 (69.2%)

Table 3. The best CH_4 purities produced with Configuration 1 using pure CO_2 feed and an isothermal reactor.

	8 psia	15 psia	75 psia
CH_4 production rate, kg/hr	0.34	0.34	0.34
CH_4 purity, %	73.2	78.7%	88.0%
CO_2 feed rate, kg/hr	.984	0.984	0.955
H_2:CO_2 ratio	4.2	4.0	4.0
Reactor Temperature, °C	300	300	300

Table 4. The best CH_4 purities produced with Configuration 1 using Mars atmosphere feed and an isothermal reactor.

	8 psia	15 psia	75 psia
CH_4 production rate, kg/hr	0.34	0.34	0.34
CH_4 purity, %	73.1	78.6%	88.0%
CO_2 feed rate, kg/hr	.973	0.973	0.943
H_2:CO_2 ratio	4.2	4.1	4.1
Reactor Temperature, °C	300	300	300

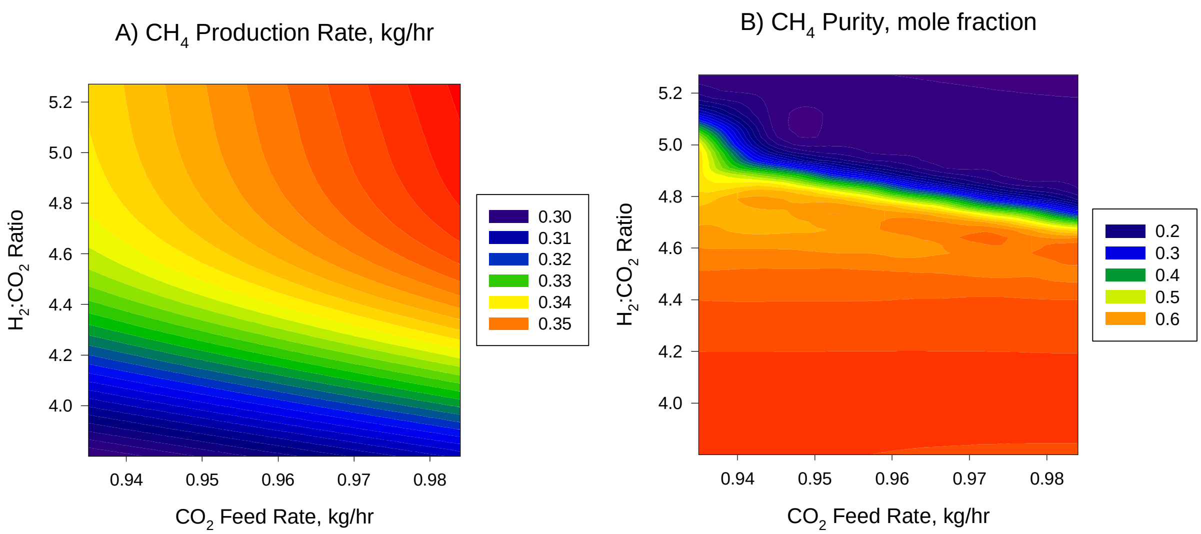

Figure 3. CH_4 production rate, A, and purity, B, as a function of CO_2 feed rate and H_2:CO_2 ratio for Configuration 1 with an isothermal reactor at 350 °C and 15 psia.

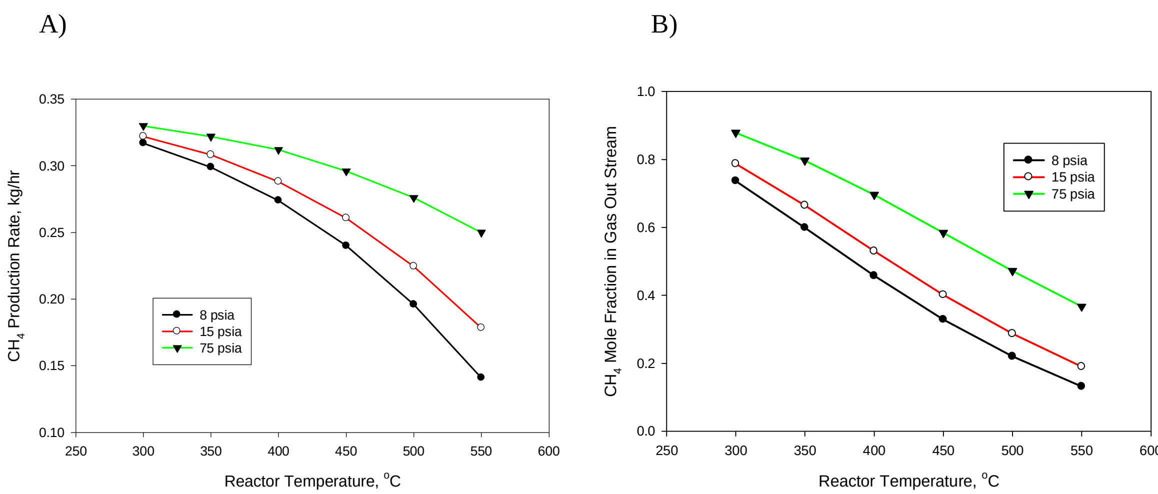

Figure 4. CH_4 production rate, A, and CH_4 mole fraction in Gas Out stream for different isothermal reactor temperatures in Configuration 1.

B. Configuration 2: Two adiabatic reactors

The flowsheet showing two adiabatic reactors with a heat exchanger in between and followed by a condenser is shown in Figure 5. The heat exchanger cooled the gas stream from the first reactor to either 250 or 300 °C. As the cooler temperature gas goes into the second reactor, the reaction proceeds further. This case was only modeled using Mars gas feed because that produced slightly better conversions than pure CO_2 feed when one adiabatic reactor was used.

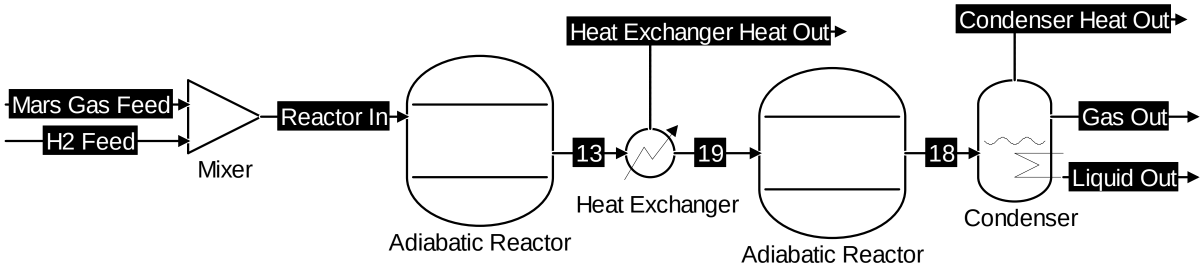

Figure 5. Two adiabatic reactors in series separated by a heat exchanger and followed by a condenser. Mars Gas Feed, H2 Feed, Reactor In, 13, 19, 18, Gas Out, and Liquid Out are material streams. Heat Exchanger Heat Out and Condenser Heat Out are information streams.

Table 5 gives the maximum CH_4 production rate and CO_2 conversion for Configuration 2 with a heat exchanger at each pressure. At 8, 15, and 75 psia operating pressures, the required CH_4 production rate is not met, however, it is met at 150 psia. This condition uses an H_2:CO_2 ratio of 5 and a high flow rate of Mars atmosphere feed, so the resulting methane purity is only 41.3%. In this case, the heat exchanger has to remove 460 W and the condenser 1050 W.

The second case for Configuration 2 uses a condenser in between two adiabatic reactors to both cool the gas and remove water before entering the second reactor. Cooling the gas before it enters the second reactor increases conversion in the second reactor just as in the first case, and removing water also pushes the reaction towards products. The flowsheet is shown in Figure 6. 8, 15 and 75 psia were evaluated under this configuration and only at 75 psia operating pressure was the CH_4 production requirement met. There were multiple flow conditions that met the requirement and the two that had the highest CH_4 purity are shown in Table 5. The purities are quite low, around 45%, due to the excess hydrogen needed to produce enough CH_4. Like the previous cases with adiabatic reactors, the temperature of the first reactor, about 640 °C, exceeds that which is normally considered safe for the catalyst. The second reactor had a temperature of 575°C. For the best results shown in Table 6, the first condenser removed 1030 W and the second 490 W.

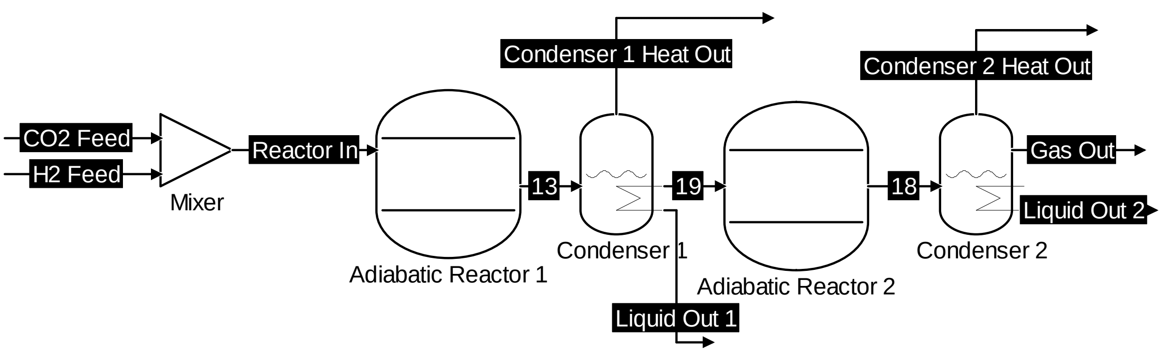

Figure 6. Configuration 2 with a condenser in between two adiabatic reactors. CO2 Feed, H2 Feed, Reactor In, 13, 19, 18, Gas Out, Liquid Out 1 and Liquid Out 2 are material streams. Condenser 1 Heat Out and Condenser 2 Heat Out are information streams.

Table 5. Maximum CH_4 production rate, kg/hr, and percent CO_2 conversion, in parenthesis, for Configuration 2 using a heat exchanger in between two adiabatic reactors.

	System pressure			
Feed	8 psia	15 psia	75 psia	150 psia
Mars Atm.	0.27 (75.8%)	0.28 (79.2%)	0.32 (89.4%)	0.34 (93.5%)

Table 6. Maximum CH_4 purity for Configuration 2 with a condenser between the reactors using pure CO2 and Mars atmosphere feeds.

	CO2 Feed	Mars Atm. Feed
CH_4 production rate, kg/hr	0.34	0.34
CH_4 purity	44.5%	45.4%
CO_2 feed rate	.9644	0.973
H_2:CO_2 ratio	5.1	4.9

C. Configuration 3: One adiabatic reactor with a recycle stream

When an adiabatic reactor is used in a single pass mode, the reactor reaches high temperatures which do not favor CH_4 production. A common method of lowering the temperature is using a recycle stream[22,23] which returns some of the product gas to the feed, keeping the reactor cooler. Condensing water from the recycle stream also promotes CH_4 formation. This is the basis for Configuration 3, which is shown in Figure 7. The splitter takes a fixed amount of the gas stream, the split factor, leaving the condenser (stream 18) and sends it back into the reactor. When the split factor is zero, no gas is recycled and all gas goes to the Gas Out stream. When the split factor is 0.5, half of stream 18 goes to Recycle and the other half goes to Gas Out. Configuration 3 was evaluated with only Mars atmosphere feed, and only at select recycle percentages. The flow conditions were 0.935 kg/hr CO_2 with a 4.0 H_2:CO_2 ratio. When the split factor is 0.9, this configuration was able to produce 0.336 kg/hr CH_4, very close to the production requirement. The reactor temperature was 318 °C, which is low enough that slow kinetics would be a concern. This configuration resulted in the highest CH_4 purity, 90.5%, of any of the configurations evaluated in this study. The composition of product gas is shown in Table 7. The condenser needed to remove 1.5 kW of thermal energy. A recycling stream that selectively recycles CO_2 and/or H_2, like a membrane or H_2 electrochemical pump, might improve this case. Split factors from 0.4 to 0.95 were also evaluated and results are shown in Figure 8. Increasing the split factor to 0.95 increases CH_4 production rate and purity, but the reactor temperature, 190 °C, is too low to consider this as a realistic case. Lower split factors result in lower production rates, purities, and higher reactor temperatures. Configuration 3 shows that adiabatic reactors with recycling can produce conversions comparable to isothermal reactors, although the low reactor temperature is a concern.

Table 7. Composition of the product stream for Configuration 3 with 90% recycling.

Component	Mol %
CO_2	2.4
H_2O	0.1
CH_4	90.5
H_2	2.7
Ar	1.6
N_2	2.6

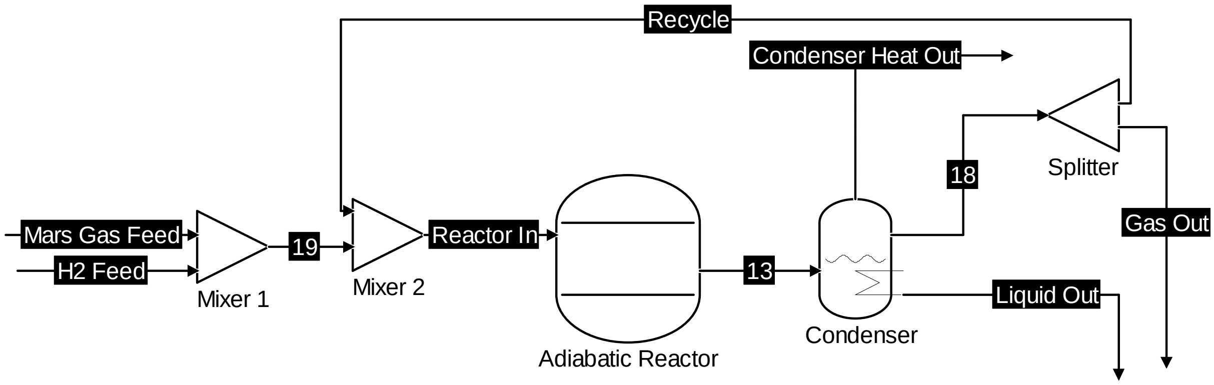

Figure 7. Flowsheet for Configuration 3. Mars Gas Feed, H2 Feed, Reactor In, 19, 13, 18, Recycle, Gas Out, and Liquid Out are material streams. Condenser Heat Out is an information stream.

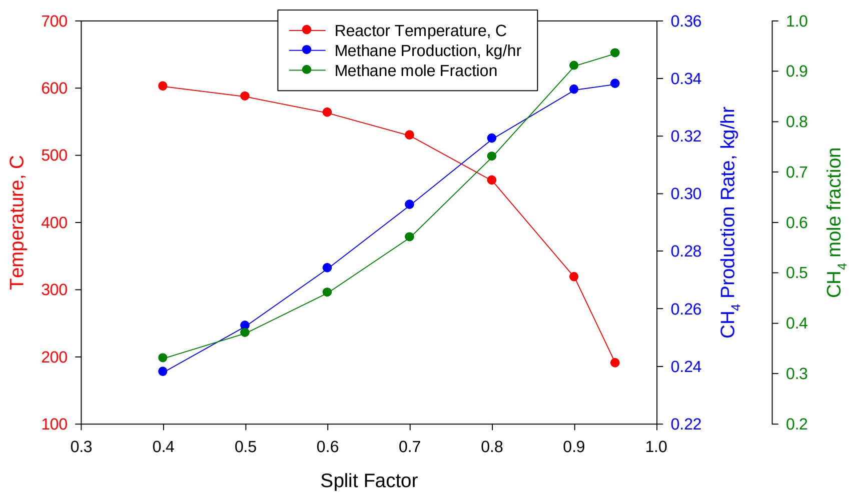

Figure 8. Reactor temperature, CH_4 production rate, and CH_4 mole fraction of product gas for configuration 3 with 0.935 kg/hr CO_2 feed and 4.0 H_2:CO_2 ratio at different split factors.

IV. Conclusion

This study has evaluated a few different Sabatier system configurations and shown that both adiabatic and isothermal reactors can be used successfully if put in the right configuration. Although the methane purity requirement is not yet known, it seems unlikely that even the best purity found in this study, 91.1%, would be acceptable. This means that either a different configuration, an alternative recycle system, or gas cleanup is needed. Future work with these models will implement different recycle systems, such as membranes which selectively recycle CO_2 and H_2 while allowing CH_4 to pass and H_2 electrochemical pumps. In addition, reactor sizing will be done using the material flow rates calculated in these models, when data for specific reactors is available. This will allow different configurations of reactors and recycling systems to be traded against each other on a mass and volume basis.

One area that still has open questions is thermal management and how to differentiate between adiabatic and isothermal reactors. For example, if both the adiabatic and isothermal reactors had the same mass and volume they would appear to be equivalent at the reactor level. However, the isothermal reactor requires a thermal management system while the adiabatic reactor does not. Both configurations require a condenser, but the condenser for the adiabatic system must remove more heat than the condenser with an isothermal reactor. It was not included in this study, but the adiabatic system with recycling requires a pump, so the system level trade in this case is thermal management of a reactor versus a mechanical pump. As new recycling components are added, it increases the trade space and these components will also need to be compared against reactor thermal management.

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