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Research Note

Toward Carbon Neutrality for Natural Gas Liquids Valorization from Shale Gas

Zewei Chen, Edwin Rodriguez, and Rakesh Agrawal*

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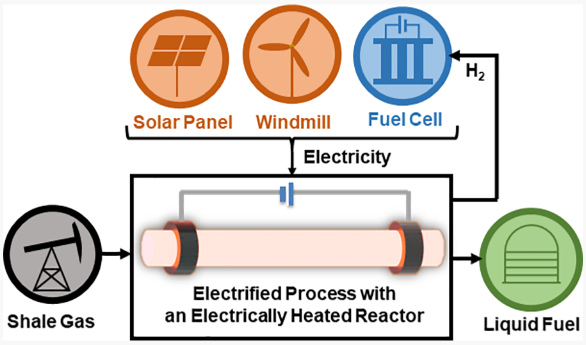
ABSTRACT: Electrification and decarbonization of the chemical industry are the keys to achieve carbon neutrality for human society, which necessitates the transition from fossil resource-based chemical production to renewable energy-based chemical production. To facilitate this transition, intermediate solutions with fossil resources as carbon sources and renewable electricity as energy sources are desirable steps. Shale resources, although often regarded as a bridge fuel, are still processed in an energy-intensive manner, and even worse, large amounts of associated gas are directly flared on site. In this research note, we introduce several decarbonization flowsheets for the process to first convert natural gas liquids (NGLs) to mainly ethylene in an energy-intensive dehydrogenation reactor and subsequent conversion of ethylene into value-added and easy-to-transport liquid fuels. Decarbonization of such an energy-intensive process through direct electrical heating of the thermal dehydrogenation reactor in conjunction with electricity from the byproduct hydrogen and a fuel cell has potential to reduce storage of the intermittently available renewable electricity to a manageable scale. Use of such processes could enable valorization of shale gas NGLs at a distributed scale without any carbon dioxide emissions during the chemical conversion step.



■ INTRODUCTION

Electrification and decarbonization of the chemical industry are necessary to achieve carbon neutrality for human society.[1−5] While chemical engineering evolved against the backdrop of an abundant supply of fossil resources for chemical production and energy, the economic use of renewable energy resources such as solar and wind will be the main theme in a sustainable future.[4] To facilitate this transition, intermediate solutions for chemical production with fossil resources as carbon sources and renewable electricity as energy sources to mitigate carbon dioxide release during transformation and recovery of chemicals are crucial steps. Shale resources, often regarded as a bridge carbon source, is therefore of special interest during this transition. However, processing shale gas at remote locations is challenging due to the lack of transportation infrastructure. Some remote shale basins remain unexploited, and even worse, large amounts of associated gas is directly flared on site, contributing toward wastage of shale resources and increased greenhouse gas emissions. According to the satellite data from the National Oceanic and Atmospheric Administration (NOAA), 10.0 billion cubic meters of shale gas was flared every year from 2012 to 2017 in the United States,[6] resulting in approximately 7 million metric tons of CO_2 each year.

Converting natural gas liquids (NGLs) in shale gas into value-added and easy-to-transport liquid fuels and chemicals has been proposed and evaluated as a promising route for shale resource valorization.[7−9] However, the proposed processes generally contain energy-intensive activation steps to activate NGLs into reactive molecules such as olefins. Among them, steam cracking is the single most energy-consuming process in the chemical industry and globally uses approximately 8% of the sector's total primary energy use.[10] Decarbonization of such an energy-intensive process would reduce significant amounts of CO_2 emissions per year. In this work, we report process flowsheets with zero carbon emissions, by integrating the shale gas valorization process with renewable electricity. We mention that any emissions during shale gas handling and transportation are not included in our discussion, and such emissions should be avoided. The synthesis procedure in this work has the potential to be generalized to decarbonization of other energy-intensive chemical processes.

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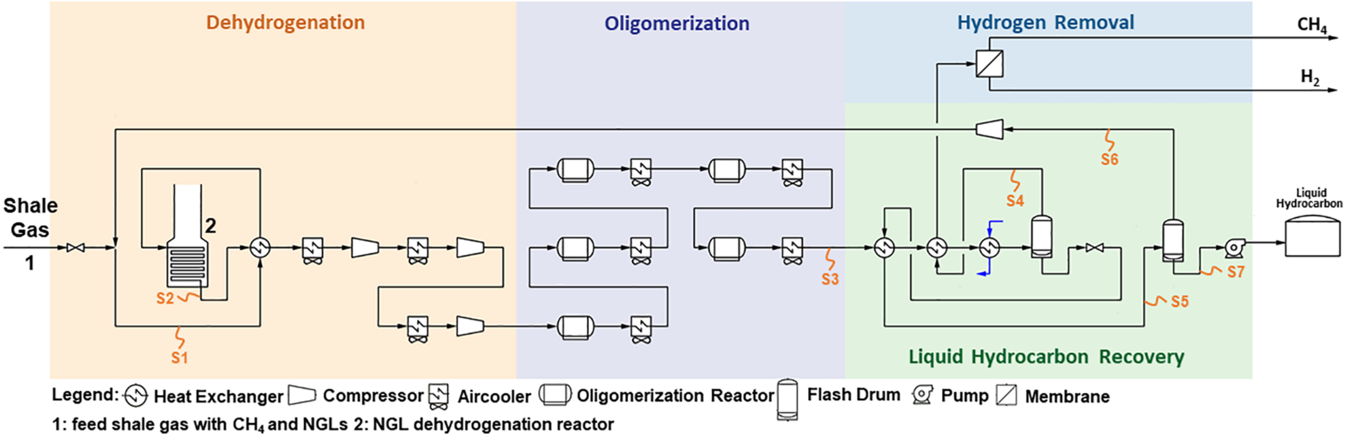

Figure 1. Proposed process for shale gas processing at remote areas (process I).

Table 1. Representative Shale Gas Composition of Bakken Field[14]

Component	CH_4	C_2H_6	C_3H_8	C_4H_{10}	C_5H_{12}	CO_2	N_2
Composition (mol %)	57.81	19.98	11.35	3.79	1.26	0.57	5.22

Table 2. Key Stream Information of Process I

Stream	Units	S1	S2	S3	S4	S5	S6	S7
H_2	%	0.0	13.8	15.8	17.0	0.4	1.3	0.0
N_2	%	5.1	4.0	4.8	5.2	0.3	0.8	0.0
CH_4	%	57.7	55.1	65.7	69.9	0.4	40.5	0.3
C_2H_6	%	20.0	3.7	4.4	4.3	6.5	18.2	0.8
C_2H_4	%	0.2	20.6	2.5	2.5	2.4	6.8	0.2
C_3H_8	%	11.2	0.2	0.2	0.1	0.9	2.0	0.3
C_3H_6	%	0.2	1.9	0.8	0.6	3.5	8.3	1.2
C_4H_{10}	%	2.8	0.0	0.0	0.0	0.3	0.4	0.2
C_4H_8	%	0.3	0.0	0.9	0.3	7.7	11.8	5.7
C_5H_{12}	%	0.5	0.0	0.0	0.0	0.0	0.0	0.0
C_5H_{10}	%	0.2	0.1	0.7	0.1	9.5	0.0	10.8
C_{6-11}	%	0.4	1.1	2.3	0.0	31.2	3.1	44.8
C_{12-15}	%	0.0	0.0	1.3	0.0	13.6	0.0	20.2
C_{16-20}	%	0.0	0.0	0.5	0.0	10.3	0.0	10.5
Flow rate	MMSCFD	10.2	13.9	10.8	10.2	0.9	0.3	4.0
Pressure	bar	2.2	1.6	25.8	25	1.6	1.4	1.2
Temperature	°C	50	850	65	−20	26	25	25

■ BASE CASE

We start with a base case process adapted from the processes synthesized in our earlier work,[11,12] which were demonstrated through detailed process simulation and economic analysis to be economically attractive for small scale installations. In one process,[11] shale gas was proposed to convert NGLs to liquid fuel (Figure 1). While the conventional shale gas process follows a "front end separation−NGL activation−NGL upgrading" processing sequence, for this process, we introduced a new processing sequence of "NGL activation−NGL upgrading−back end separation". In the process of Figure 1, the entire shale gas including methane is sent to a thermal dehydrogenation reactor (thermal cracker) to mainly produce ethylene from the constituent NGLs. While the ethylene could be separated from the reactor effluent and used for the synthesis of any number of chemicals, in Figure 1, it is fed to a series of oligomerization reactors, and then, an absorption unit is used to recover heavier liquid hydrocarbons. Comparing to the processes in our earlier publication, this process uses a two-flash back end separation system to further simplify the process and reduce energy consumption. In another process,[12] we suggest an alternative process sequence for process simplification, cost reduction, and enhanced recovery of butylene, propylene, and ethylene from shale gas. Again, methane separation is performed at the end of all the dehydrogenation reactors. A common feature of both the processes is that the delaying of methane separation from the feed shale gas and choice of novel process sequencing dramatically intensified and simplified the proposed processes by eliminating repeated or unnecessary unit operations and associated equipment. In both the processes, dehydrogenation reactors are major energy consumers and responsible for large quantities of CO_2 release from methane combustion. In this work, we focus on the decarbonization of the thermal dehydrogenation unit in the process of Figure 1, where all the NGLs in the shale gas are primarily cracked into ethylene.

The base case flowsheet, along with all other flowsheets in this research note, are simulated with a feed of 10 million standard cubic feet per day (MMSCFD) of shale gas from the Bakken field at 30 bar and ambient temperature with its composition listed in Table 1. The Bakken field is of special interest since its shale gas has a high NGL concentration, with wells in remote locations, and there is substantial flaring.[13] Key stream information on this process is shown in Table 2. General simulation assumptions are described in Table 3.

Table 3. General Assumptions in Simulations

Item	Values
Physical property	Peng−Robinson
Pressure drops across each unit operation	0.21 bar
Ambient temperature	25 °C
Isentropic compressor efficiency	70%
Minimum temperature approach	5 °C
Dehydrogenation model	Kinetics from ref 16
Oligomerization	RGibbs
% Furnace heat to conventional dehydrogenation reactor	42%
Fuel cell efficiency	60%
Battery storage efficiency	80%
H_2 storage efficiency	94%[17]

Although dramatically simplified and intensified, this base case process still has power consumption of 10.9 MW, which includes 8.6 MW needed as heat by the dehydrogenation reactor and the remaining 2.3 MW of power for pumps and compressors. If the dehydrogenation reactor is fueled by CH_4 and byproduct H_2, all the H_2 and 7.7% of CH_4 in the shale gas feed need to be burned at the rate of 20.5 MW heat, and 9.8×10^3 metric ton of CO_2 would be emitted per year. If the power for pumps and compressors is from a natural gas power plant with 50% efficiency, then another 5.6% of CH_4 in the feed has to be burned and an additional 7.1×10^3 metric ton of CO_2 would be emitted per year. The higher energy consumption is due to the energy efficiency limitation in the conventional dehydrogenation reactor furnace. The residence time of the flowing stream within the dehydrogenation reactor is small, which requires a high heat transfer rate from the furnace to the reactor. Due to the limitation of the thickness of the coil material, the temperature difference between the inside and the outside of the reactor tube is usually above 200 °C to achieve this high heat transfer rate. As a result, about 42% of the energy obtained from the fuel combustion process is generally transferred to the reactor coils in a commercial dehydrogenation furnace.[15] Most of the remaining conbustion heat is recovered as steam in an ellaborate heat exchanger network. This adds not only cost and process complexity but also the need to gainfully utilize resulting steam energy.

■ INTEGRATION WITH RENEWABLE ELECTRICITY

Step 1: Electrification of Heat Driven Unit Operations. Renewable energy is usually available in the form of electricity.[4,18] Redesign of heat-driven unit operations so they become amenable for integration with renewable electricity is necessary for decarbonization. In process I, the only heat-driven unit is the thermal dehydrogenation reactor. Here, we propose the use of a direct electrically heated reactor tube, which provides a compact, simple, and easy-to-operate alternative compared to current methods of using furnaces, as illustrated in our previous patent application (Figure 2a).[19] An electrically heated reactor has been proposed and studied for multiple industrial applications, especially for processes that generate H_2 as a byproduct.[20,21] For example, Wismann et al.[22] described a direct electrically heated catalyst-coated metal tube reactor for steam methane reforming (SMR), which if implemented on all SMR reactors globally was estimated to result in a reduction of nearly 1% of global CO_2 emissions. The design of an electrically heated steam cracker has also been initiated by companies for demonstration at an industrial scale.[23] The direct electrically heated reactor tube is especially advantageous in thermal dehydrogenation, as limiting heat transfer steps are eliminated, resulting in increased heat transfer efficiency.

It is worth noting that besides endothermic reactions, heat-driven separation units are also subject to electrification if necessary. Although electrification of separation units is not applicable to the proposed process, and hence not discussed in this work, readers may refer to Chavez Velasco et al.'s work for a systematic analysis on heat- and electricity-driven separations.[24]

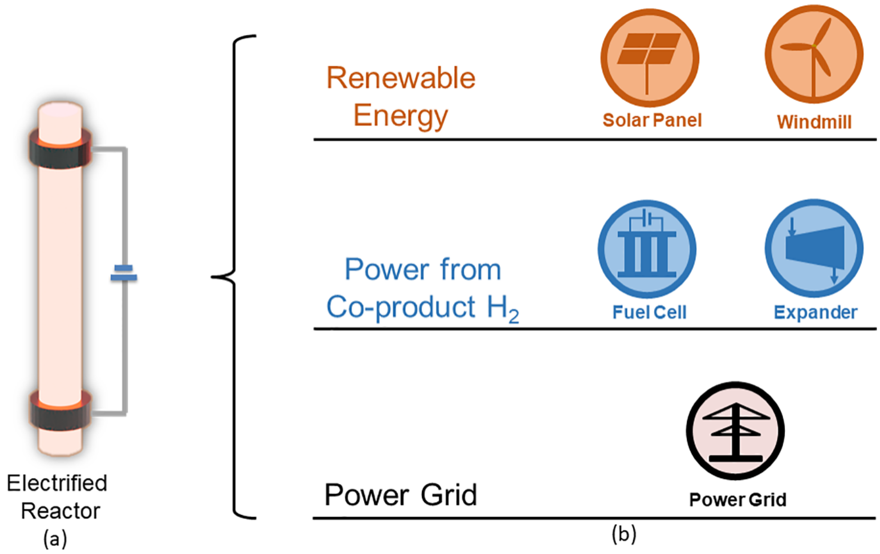

Figure 2. (a) Direct electrically heated reactor tube. (b) Possible electricity sources to supply needed electricity.

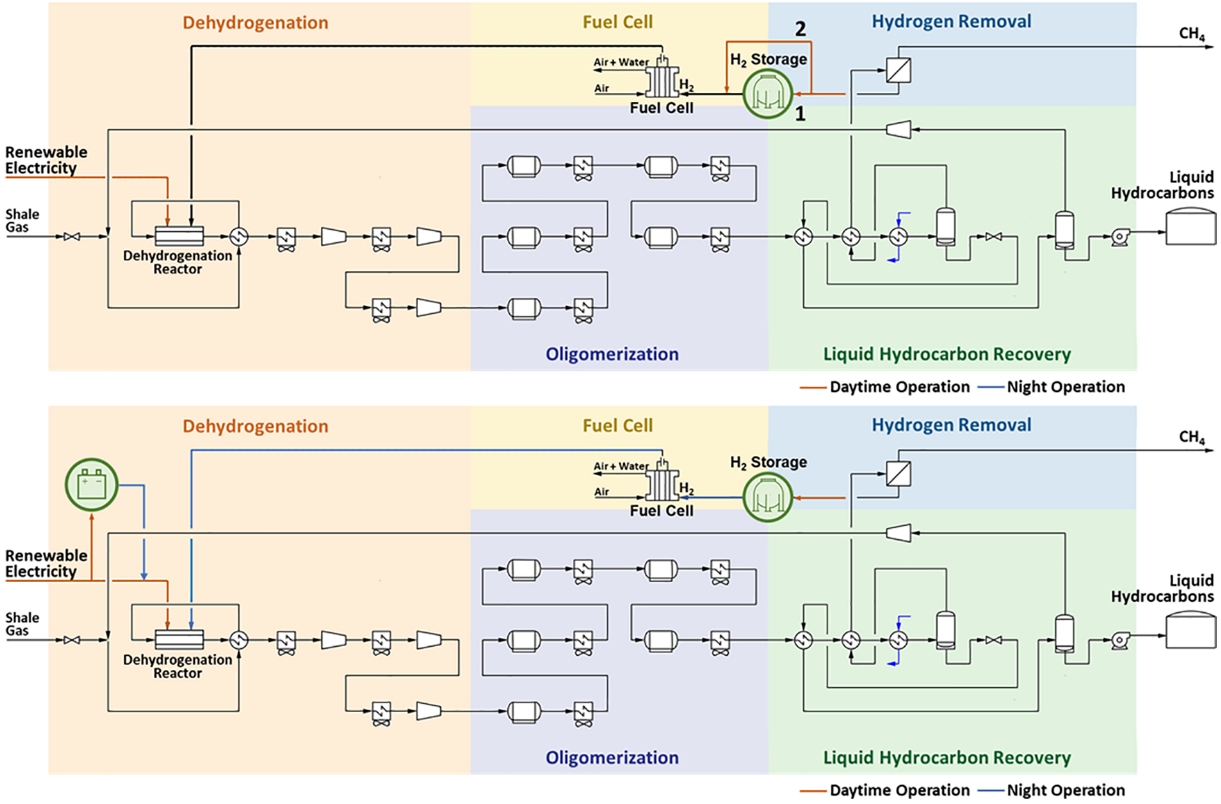

Figure 3. Proposed processes for electrified NGL dehydrogenation reactor with liquid fuel production: (a) long sunlight hours (process II) and (b) short sunlight hours (process III). Dehydrogenation reactor is electrically heated. Legend for other equipment is given in Figure 1.

Step 2: Identification of Electricity Sources. We classify electricity sources into three categories. The first category is the electricity supply directly from the local renewable facilities, such as solar panels and/or wind turbines. This category of electricity is essentially free of carbon emissions but necessitates the accessibility to renewable facilities.[25] The second category of electricity emanates as a byproduct from the process itself. This may include running a fuel cell with byproduct H_2, as well as utilizing a turbo expander to extract electricity from any available high pressure stream. Our base process is an example of this category. The H_2 from the dehydrogenation reactor effluent, in conjunction with a fuel, can be used to generate electricity. Similarly, any pressurized steam from the exothermic oligomerization reactors is a source for in situ electricity generation. The third category of electricity source is a power grid. Carbon emission of this type of electricity source depends on the percentage of renewable electricity in the power grid. In a 100% renewable future, this electricity source could be carbon free, but we expect a hybrid power grid with a significant amount of fossil fuel-based electricity in the near future. In our work, to achieve zero carbon emissions, we only consider the first and the second types of electricity sources. To be specific, we consider solar electricity and H_2 fuel cells as our electricity sources. Of the total 10.9 MW electricity requirement of the plant, 76% can be supplied by the H_2 fuel cell, and only 24% of the electricity needs to be supplied by renewable electricity. The electricity from H_2 fuel cells, if used solely for a thermal dehydrogenation reactor, can supply 95% of the power demand of this reactor.

Step 3: Design of Storage Cycles for Intermittent Electricity Sources. The biggest challenge of implementing renewable electricity from wind and solar in the chemical industry is its intermittency. This challenge can be overcome by designing energy storage cycles, in which energy is stored in the daytime and used at night.[26−28] Depending on the average sunlight availability hours within a 24-h day, we propose two process alternatives as shown in Figure 3a and 3b.

Long Sunlight Hours. In this scenario, due to availability of sunlight for relatively long hours, the energy stored in H_2 could be sufficient to supply electricity at night, and any extra H_2 may be used during the daytime to reduce demand for electricity from the renewable source. In the daytime, a portion of H_2 in stream 1 is stored in the H_2 storage unit. Another portion in stream 2 may be sent to the H_2 fuel cell to generate electricity. The remaining electricity is supplied by renewable electricity. At night, the stored H_2 is utilized to run the fuel cell and generate the needed electricity. Supplementary electricity due to storage of renewable electricity is not necessary in the night. For the 10 MMSCFD shale gas plant under consideration, the minimum sunlight time under this scenario is 5.76 h per 24-hour cycle, assuming a 60% efficiency of the H_2 fuel cell. The minimum sunlight time is calculated using the following equation:

$$\text{Minimum sunlight time} = 1 - \frac{\text{Energy generated from the fuel cell}}{\text{Energy needed for the process}} \times 24 \qquad (1)$$

Short Sunlight Hours. In this scenario, the electricity stored in byproduct H_2 is not sufficient to supply electricity demand at night. When the average sunlight time is less than 5.76 h per day, all the byproduct H_2 is preferably stored in the daytime, and the required electricity is totally supplied by renewable

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electricity. Also, a portion of the renewable electricity will need to be stored in a battery and used at night.

In North Dakota where the Bakken shale gas basin is located, the average peak sunlight hours[29] is 5.00 h per day, which belongs to the second scenario. The calculated required daily average power for a solar panel is 13.58 MW with battery storage to provide 8.3 MWh of electricity per day, and H_2 storage of 13.8 tons. These solar panel and average energy storage requirements are techinically achievable. Ten MW solar farms are already built around the world.[30−32] Here, 8.3 MWh battery capacity is about equal to the batteries on 83 Telsa cars, and 13.8 ton of H_2 can be stored in pressurized vessels.[33] It should be noted that the battery storage capacity of 8.3 MWh supplies 0.76 h worth of the plant's needs. On the days when there is a greater shortfall, much greater battery storage capacity will be needed. Alternatively, long-term hydrogen storage capacity may be built and used on such days to somewhat mitigate the need for extensive battery storage.

It is worth noting that for this short sunlight hours case, use of wind energy, when available in conjunction with photovoltaic panels, is likely to eliminate any need for battery storage. For example, in the Bakken shale gas basin of the United States, wind energy is quite prevalent.[34] Since wind energy is typically available for more than 30% of a 24-h day including night times,[35] its usage will likely ensure availability of renewable electricity in a 24-h cycle.

CONCLUSION

In this study, we proposed a synthesis procedure to decarbonize an energy-intensive process which includes conversion of natural gas liquids in shale gas to olefins in an endothermic dehydrogenation reactor. In the process, the primary product ethylene, from the dehydrogenation reactor, is converted to liquid fuel for ease of transportation from remote locations. Successful implementation of the decarbonized processes not only reduces flaring of shale resources at remote areas but also produces chemicals and liquid fuel without carbon emissions. Table 4 shows the power consumption, CO_2 emission, H_2 storage, and battery and solar panel requirements for processes I and III. Comparing to the base case process I, on average, process III requires daily storage of 13.8 ton H_2, an 8.3 MWh battery, and installation of a 13.08 MW solar panel. As a future work, the battery and H_2 storage capacities vis-a-vis daily variation in solar/wind energy are being studied. In addition, economic analyses can be conducted to determine the economic feasibility of the process or provide cost targets for these additional equipments to make the entire process economically attractive.

Table 4. Summary of Processes I and III for a 10 MMSCFD Shale Gas from Bakken field

	Process I	Process III
Power demand	10.9 MW	10.9 MW
CO_2 emission	1.70×10^4 ton/year	0
Average daily H_2 storage	0	13.8 tons
Average daily battery storage	0	8.3 MWh
Solar panel	0	13.58 MW

AUTHOR INFORMATION

Corresponding Author

Rakesh Agrawal − *Davidson School of Chemical Engineering, Purdue University, West Lafayette, Indiana 47906, United States;* orcid.org/0000-0002-6746-9829; Phone: +1 765 586 7259; Email: agrawalr@purdue.edu

Authors

Zewei Chen − *Davidson School of Chemical Engineering, Purdue University, West Lafayette, Indiana 47906, United States*

Edwin Rodriguez − *Davidson School of Chemical Engineering, Purdue University, West Lafayette, Indiana 47906, United States*

Complete contact information is available at:
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Notes

The authors declare no competing financial interest.

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