

Shale Gas Decarbonization in the Permian Basin: Is It Possible?

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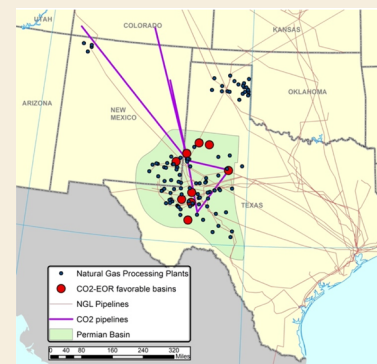
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ABSTRACT: The United States is unique in the energy reserves held in shale gas fields, which coproduce natural gas and natural gas liquids. Use of this resource, however, contributes to greenhouse gas emissions and, correspondingly, climate change. We explore how natural gas and natural gas liquids might build bridges toward low-carbon transportation fuels. For example, as petroleum refineries produce less gasoline in response to widespread electrification, natural gas liquids can be converted to fuel. We consider whether the greenhouse gas emissions from production and use of these fuels might be offset through three potential outcomes of converting coproduced natural gas to CO₂ through steam methane reforming. First, the CO₂ could be injected into conventional oil formations for enhanced oil recovery. Second, it could be sequestered into saline aquifers to avoid CO₂ emissions from the produced oil combustion. Third, it could be injected into unconventional gas formations in the form of CO₂-based fracturing fluids. Simultaneously, the coproduced hydrogen from steam methane reforming could be used to support the expansion of the hydrogen economy. The region of study is the Permian Basin. The results show sizeable emission benefits by decreasing net emissions of natural gas production and use to 28 from 88 g-CO₂e/MJ. For revenue generating pathways, a partial decarbonization of 3.4 TCF/year is possible. All of the natural gas can be partially decarbonized if the CO₂ is sequestered in saline aquifers. Overall, the results show that while greenhouse gas emissions can be reduced through decarbonization approaches relying on subsurface sequestration, full natural gas decarbonization is not achieved but must be pursued through other approaches.



KEYWORDS: shale gas, greenhouse gas emissions, enhanced oil recovery, CO₂ capture and storage, CO₂ fracturing fluid, Permian Basin

1. INTRODUCTION

The U.S. natural gas (NG) sector has expanded significantly, with dry natural gas production increasing from 18 TCF in 2005 to 33 TCF in 2020 (compounded annual growth rate: 4.1%).^{1–3} In this same time period, production of natural gas liquids (NGLs) grew from 1.7 to 5.1 million barrels per day.⁴ NGLs include ethane, propane, butane, isobutane, pentane, and other hydrocarbons. With the abundance of NG and NGLs in the U.S., it is interesting to consider how these resources might be used to meet near-term energy needs while building a bridge to a decarbonized energy future. One opportunity is to use NGLs to produce hydrocarbon transportation fuels^{1,2,5} that may fill gaps as refineries transition their product slate potentially away from liquid fuels (especially for the light-duty sector) as electrification increases. Doing so could ensure a stable and cost-effective fuel supply for the population of vehicles continuing to use an internal combustion engine for the next several decades.⁶

Yet, use of such fossil fuels will not contribute to decarbonization in the near term. Combustion of fossil fuels made from NGLs will emit CO₂. Furthermore, well drilling, liquids unloading, flaring, and equipment venting and leaking in the NG/NGL supply chain emit methane, a potent greenhouse gas (GHG). For instance, Alvarez et al.⁷ estimated the total U.S. methane emissions from the oil and gas sectors at

13 Tg-CH₄ annually, out of which 2.7 Tg-CH₄ are from the Permian Basin alone.⁸ Finally, the NG produced alongside NGLs is generally combusted, emitting ~61 g CO₂e/MJ. At first glance, it may seem undesirable to pursue the use of NGLs as a fuel feedstock, given the GHG emissions that accompany NG and NGL coproduction and use. However, at a systems level, considering an entire shale gas basin and its interconnections with other energy systems, it may be possible to achieve or approach decarbonization. Such considerations are important, given the importance of NG/NGL systems in the U.S. energy landscape.

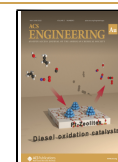
If NGLs are viewed as a liquid fuel feedstock, converting natural gas to hydrogen and CO₂ through steam methane reforming offers one route to potentially move toward net-zero emissions of shale basin products at a systems level, depending on the use of the CO₂ and H₂ that would be produced. The oil and gas industry^{9,10} and multiple organizations interested in decarbonization of downstream emissions^{11,12} see this route as

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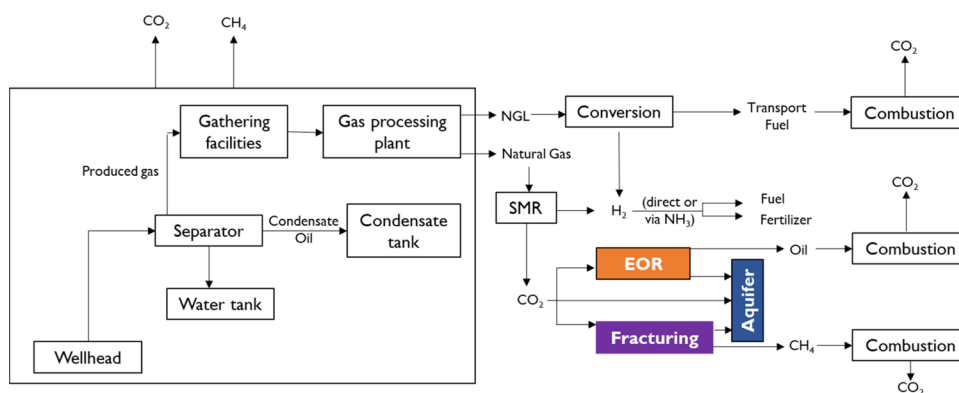


Figure 1. Illustrative pathways for conversion of shale gas products via hydrogen production and CO₂ separation. The end use of CO₂ in the three pathways is shown in colored boxes with white font. The box represents the shale gas production and processing stages, which have been evaluated in a previous study.¹⁵ The stages outside the box are representative of further conversion and decarbonization processes SMR: Steam methane reforming, EOR: Enhanced oil recovery.

promising. For example, hydrogen produced in this manner is known as “blue hydrogen,” and deploying it as a fuel could build the infrastructure that would be used by “green” hydrogen produced using renewable electricity-driven hydrolysis in the longer term. The best use for blue H₂ and the coproduced CO₂ depends upon multiple factors, notably what type of infrastructure and existing systems are available, which could be region-dependent.

In this paper, we select the Permian Basin as our region of focus. It is unique compared to other basins in the U.S. in its conventional oil formations and aquifers, which can store large quantities of CO₂. We quantify system-level GHG emissions from NG/NGL systems in this basin, which spans Texas and New Mexico. We consider a case in which NGLs are converted to liquid transportation fuels, and CO₂ and H₂ are produced from steam methane reforming of the coproduced NG. We consider three pathways for CO₂ use: enhanced oil recovery (EOR), fracturing, and disposal in saline aquifers. It should be noted that the first two pathways may be more attractive to shale gas producers because they produce saleable products. No revenue arises from CO₂ injection in an aquifer. We evaluate the amount of natural gas, the main product within the fracking systems of the Permian Basin, that could be decarbonized (e.g., produced and used with zero GHG emissions).^{13,14} In addition to evaluating system-level GHG emissions in these pathways, we consider both short-term and long-term utilization prospects for hydrogen in the Permian Basin region based in part on infrastructure (e.g., pipeline) availability.

2. METHODOLOGY

In this paper, we evaluate the decarbonization potential and broader environmental effects of three conceptual pathways (Figure 1) for the use of NG and NGLs as products from shale gas, based on the characteristics of the Permian Basin.

This analysis considers the production of NG and NGLs in wet and dry gas fields. Some regions may also produce substantial amounts of oil.¹⁵ Within life cycle assessments (LCA) of natural gas systems, a methodological choice arises in selecting a method to allocate greenhouse gas (GHG) emissions between NG and NGLs. Often, analyses view NG as the main product in a basin and assign all GHG emissions to it. NGLs, and, in some cases oil, are also coproducts, however, and it can be argued that they should bear some of the GHG

burden.¹⁶ Depending on allocation methods between NG and NGLs,¹⁶ upstream emissions for NGLs could range from 0 if all emissions are assigned to natural gas to 12 gCO₂e/MJ if emissions are divided among NG and NGLs based on their energy content. In our analysis, we assumed a functional unit of 1 MJ NG using energy allocation among NG, NGLs, and H₂. We considered the production of wet natural gas, where the energy content of produced methane and NGLs were in the ratio of 1.83:1. Numerous uses of the NGLs are possible.^{17,18} In this analysis, we consider their conversion to liquid transport fuels.^{1,2,5}

We assume that the produced methane undergoes steam methane reforming (SMR) to produce hydrogen. The coproduced CO₂ is captured. Accordingly, the hydrogen could be categorized as “blue hydrogen.” In Texas or New Mexico, hydrogen in the near term could be used to produce fertilizer. In the longer term, there are industrial and research outlooks,^{10,19} which project that hydrogen may be a prominent fuel and/or energy carrier for transportation. In our analysis, hydrogen is converted to and re-reformed from ammonia because direct hydrogen storage costs more and poses safety concerns. We treat the coproduced H₂ and CH₄ with energy allocation. Two pathways assume utilization of CO₂. The first entails CO₂ from the SMR process used in EOR operations. The second adopts it as a substitute for conventional aqueous fracturing fluids. CO₂ fracturing fluids are almost entirely CO₂ and are injected underground in the supercritical phase. Upon reaching the necessary depth, it expands to fracture the reservoir in the supercritical-CO₂ phase.²⁰ In the third pathway, CO₂ is injected into a saline aquifer.

2.1. Process Description

2.1.1. Production/Conversion of Shale Gas and Hydrogen. For each tonne of methane, the corresponding amount of hydrogen production under the state-of-the-art conditions is 0.37 t (0.27 t from the SMR process and 0.10 t from the NGL-to-fuels process).²¹ The SMR process also leads to CO₂ emissions of 2.6 t-CO₂ per tonne of methane—out of which 90% are captured.²² We assume that the rate of CO₂ emissions stays constant over the project lifetime based on a constant rate of natural gas conversion to hydrogen. The rate of upstream methane emissions from NG varies based on the approach to allocating these upstream emissions among coproducts (NG, NGLs, and oil).¹⁵

Compared to transportation of H₂, transportation of ammonia is more straightforward with today's infrastructure. Accordingly, as previously mentioned, this analysis assumes the conversion of H₂ to ammonia prior to use as a fuel. In the analysis, we account for a 7% loss in energy content²³ when hydrogen (lower heating value 142 MJ/kg) is converted to ammonia at 62 bar. Ammonia is then transported in liquid form for approximately 200 km and reformed to hydrogen at the point of use.^{24,25}

2.1.2. Pathway #1: CO₂ Utilization for Enhanced Oil Recovery. Currently, CO₂ used in enhanced Permian Basin oil recovery (EOR) operations originates from naturally occurring formations. In this analysis, CO₂ from the SMR process replaces some of this CO₂. Accordingly, we estimated the quantity of CO₂ these oil wells can accommodate over a 35-year well lifetime. To calculate this quantity, we relied on ARI projections²⁶ of CO₂ consumption in the Basin. Initially, the purchased CO₂ (from the SMR process) is directly injected underground to stimulate oil production. Within a few years (years 3–4), oil production starts, and the well produces CO₂ (produced CO₂ in Figure 2). This CO₂ is combined with the

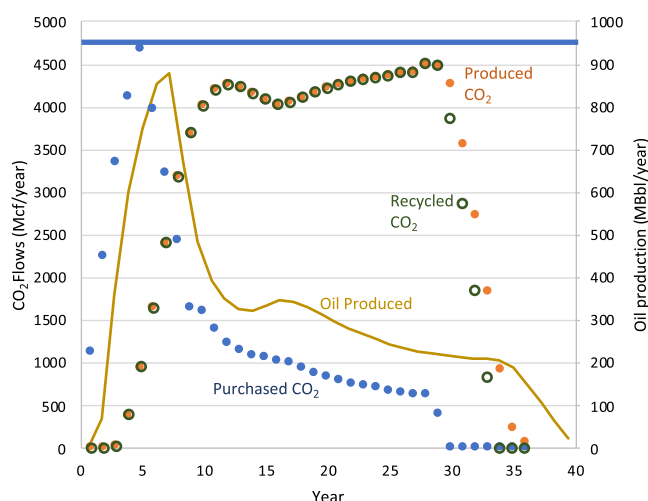


Figure 2. EOR parameters for the Permian Basin.²⁶ The horizontal line depicts constant CO₂ emissions from the SMR facility. “Purchased CO₂” values are injected for EOR, and the remaining CO₂ is injected into a saline aquifer. Year 0 here represents the starting point for injection of purchased CO₂ for incremental oil recovery.

purchased CO₂ and injected back into the well (recycled CO₂). Gradually, the amount of recycled CO₂ increases such that the amount of purchased CO₂ starts to drop off around years 6–7 and reaches zero in year 30. Concurrently, the amount of recycled CO₂ increases past that of purchased CO₂. Based on this CO₂ use profile, about 30% of the CO₂ captured from the SMR + water gas shift process can be injected permanently into the oil formation. The remaining CO₂ captured from the SMR process is assumed to be injected into a saline aquifer. The pumping energy consumption for injection into a saline aquifer is assumed to be 6.68 kWh/t-CO₂.²⁷

To calculate the net GHG emissions from these scenarios, we adopted several literature-based values. For example, we assumed that EOR's energy consumption is 1.78 kWh/bbl²⁸ (1 bbl = 5860 MJ). We adopt a CO₂ emission factor of 430 kg-CO₂e/bbl for crude oil refining and subsequent fuel product

combustion. Finally, we assign a displacement credit of 200 kg-CO₂/bbl to account for the use of SMR-derived CO₂ compared to extraction and compression of natural CO₂.²⁹ The CO₂ transport distance between the source of CO₂ (SMR facility) and the EOR site (conventional oilfields in the Permian Basin) is assumed to be 50 km because the EOR-suitable sites are very well spread throughout the Basin.³⁰ The pipeline construction emissions burdens were adapted from Melara et al.,³¹ in which the emission intensity of steel is 2.12 kg-CO₂e/kg steel. Pressurization requirements for CO₂ are 1.43 kWh/t-CO₂, while a leakage rate of 0.15% was assumed for pipeline systems.³² Use of CO₂ from SMR displaces extraction of CO₂ from natural formations for use in EOR. Use of natural CO₂ incurs upstream emissions of 0.22 t-CO₂/bbl.²⁹

2.1.3. Pathway #2: CO₂ Utilization for Production of Fracturing Fluids. In the analysis of applying the SMR-produced CO₂ as a fracturing fluid, the SMR and hydrogen conversion to ammonia parameters are the same as in the EOR case. The parameters for the use of CO₂ as a fracturing fluid are recorded in Table 1. Use of CO₂ from SMR in this case

Table 1. Key Parameters Associated with Use of CO₂ as a Fracturing Fluid per 1 GJ of Natural Gas Produced from the Fracturing Process²⁰

input	units	value
estimated ultimate recovery (EUR)	GJ	2.67×10^7
total fluid volume per well	m ³	6.04×10^4
flowback (% of injected fluid)	%	50
diesel fuel consumption	m ³	950
CO ₂ compression energy (source)	MJ/tCO ₂	241

displaces the production of fracturing fluids. The production of these fluids is 67% more energy-intensive than aqueous fracturing fluid (2.7 versus 2 MJ/GJ-natural gas extracted).²⁰

Typically, 50% of the CO₂ that is produced as flowback is assumed to be injected into a saline aquifer at an energy consumption of 6.68 kWh/t-CO₂.²⁷

2.1.4. Pathway #3: CO₂ Storage in Saline Aquifers. The EOR and fracturing fluid pathways described above result in additional CO₂ emissions due to the extraction of incremental oil and gas, respectively. We account for these emissions. Several sources have argued that EOR detracts from decarbonization because it promotes dependence on fossil fuels.^{33,34} While one past analysis reported that EOR has very high net carbon mitigation benefits (0.04 t-CO₂ emitted per 1 t-CO₂ gross injected),³⁵ other studies indicate that the combustion of the produced oil emits CO₂ and may undercut sequestration benefits.^{29,36} Differences in past analyses stem from variations in CO₂ sources, regional CO₂ production–injection patterns, system boundary considerations, and coproduct allocation strategies. Moreover, these pathways only retain 33–50% of the injected CO₂ in our analysis. The rest is assumed to be injected into a saline aquifer. Although it is unlikely from a process economics perspective compared to the pathways in which CO₂ is used to produce saleable products, we considered a third pathway that does not use CO₂ to produce fossil fuel products that will eventually be combusted. Furthermore, all of the CO₂ injected would remain sequestered. We adopt an energy consumption of 6.68 kWh per t-CO₂ injected into the aquifers. All other parameters remain the same as Pathways 1 and 2. When CO₂ is injected into saline aquifers, brine is produced. This requires treatment

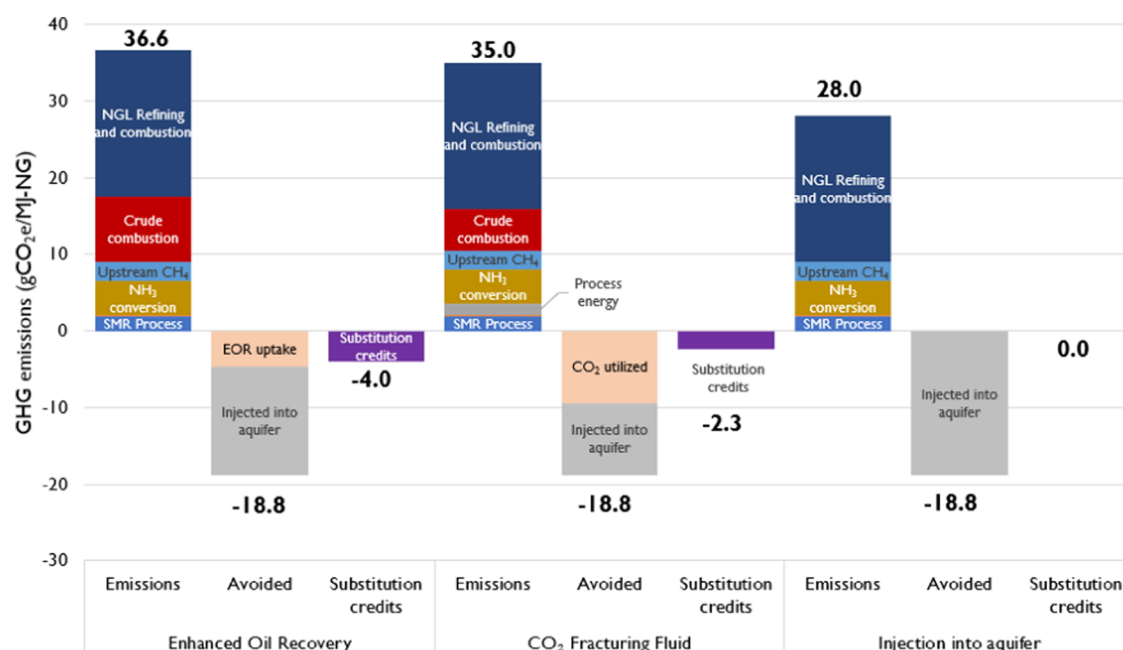


Figure 3. Emissions, avoided emissions, and substitution credits in SMR with CO₂ utilization for wet natural gas. Values above/below the bars indicate net emissions.

and/or other management due to environmental and regulatory reasons. Depending on the reservoir, saline aquifer injection may lead to a significant energy penalty for treating coproduced brine. However, the aquifers adjoining the Permian Basin are characterized by low salinity, and as such, the energy requirements for desalination may be anticipated to be lower than 5 kWh/t-CO₂.³⁷

2.2. Potential for CO₂ and H₂ Use in the Permian Basin

It is important to consider the need and capacity for CO₂-based EOR and fracturing in the Permian Basin. In the case of using CO₂ for EOR, the available pore space can accommodate 2.6 Bt-CO₂ over 35 years based on basin-specific projections on residual oil present.²⁶ This assumes residual oil extraction of 1 Mbbl/day via EOR. For the case of CO₂ fracturing fluids, 1800 new wells are drilled each year.³⁸ The estimated ultimate recovery (EUR) of oil from these wells is 2.67×10^7 GJ. Only 50% of the pore space is used because half of the CO₂ returns to the surface as flowback. In this case, sufficient pore space is available to accommodate all of the SMR-generated CO₂. CO₂ not retained underground during the fracturing process is assumed to be injected into saline aquifers. Texas and New Mexico have an abundant sink availability in the form of saline aquifers with a theoretical storage potential of 1700 Bt-CO₂ with likely viable potential > 200 Bt-CO₂.^{39,40} It is also important to consider the end use of the oil produced through EOR and the natural gas produced from fracturing. While liquid fuels' consumption in the light-duty sector will decline as electrification becomes more common, liquid fuels from petroleum will continue to be used in aviation and heavy-duty transport—and even light-duty transport—through 2050.^{6,41,42} Demand for natural gas is anticipated to rise in most scenarios in the EIA Annual Energy Outlook to 32–43 TCF/year, up from the current 30 TCF/year.

Along with CO₂, hydrogen is also produced from SMR. We considered that hydrogen would be used in fertilizer manufacturing in the short term. In the region of the Permian Basin, hydrogen use for fertilizer plants is around 60,000 t-H₂

annually,⁴³ which would correspond to only 20 BCF/year gas production. Such a small level of demand would not meaningfully influence system-level GHG emissions. Accordingly, we did not further evaluate this end use. On the other hand, demand for H₂ as a transportation fuel may reach a nominal demand level in 2050 of 22.6 Mt-H₂.¹⁹ We assume SMR-produced H₂ will be converted to NH₃ and correspondingly can be transported anywhere in the country for use. Conversion to NH₃ is assumed due to the absence of an existing pipeline network to transport hydrogen at high pressure. The scenario we have adopted assumes that SMR-based production of H₂ will increase by 16% between 2020 and 2050.¹⁹

3. RESULTS AND DISCUSSION

If it were possible to produce natural gas in a manner that resulted in net-zero GHG emissions from the system boundary diagram, as shown in Figure 1, we would consider shale gas to be fully decarbonized. Within this section, we report the extent of potential decarbonization in each pathway.

3.1. Upstream GHG Emissions

In our analysis, we accounted for three contributions to the net GHG emissions of one MJ of natural gas within the system boundary: GHG emissions into the atmosphere from the combustion of recovered oil, geologic CO₂ sequestration, and benefits resulting from systems expansion (Figure 3). Specifically, in the EOR and fracturing cases, respectively, the use of CO₂ from SMR displaces recovery and use of natural CO₂ for EOR and use of conventional fracturing fluids. The upstream emissions are associated with recovering shale gas, NGL conversion to fuel and subsequent combustion, the SMR process, and conversion of H₂ to ammonia. The sum of these emissions is 28 g-CO₂e/MJ. The dominant contributor to this total (7 g-CO₂e/MJ) is the production and combustion of fuels from NGLs, as these CO₂ emissions are not captured.

Converting hydrogen to ammonia by the GHG-intensive Haber–Bosch process, the only commercial process for this

purpose, emits 7 g-CO₂e/MJ. As noted earlier, conversion to ammonia occurs at 62 bar. Pressurization and later reforming back to hydrogen consumes 7% of the energy that the pressurized hydrogen contains. The final contributor to the upstream emissions is the SMR process, which contributes 2 g-CO₂e/MJ. In calculating these emissions, we assumed a CO₂ capture rate of 90%. It may be possible to achieve capture rates up to 96%,⁴⁴ but this increase comes at the expense of declining hydrogen yields. The only major scope for emissions reductions is to reduce methane leakage. We adopt a value of 6 g-CO₂e/MJ for methane leakage based on past work.^{15,16} Recent studies indicate that > 50% of the methane emissions from gas production may be reduced at a net economic benefit. That is, the market price of the recovered natural gas could offset the cost of changes to the infrastructure that limit emissions. We did not evaluate the potential influence of these advances on our system of study.

We note that we considered using a system expansion methodology and assigning credits for H₂ displacing gasoline as a transportation fuel on a miles per gasoline gallon equivalent basis. The resulting credit would be 139 g-CO₂e/MJ NG, which would greatly distort the results. Because the ratio of energy of H₂ and CH₄ produced is 3:4, energy allocation, the method we used, is more defensible than systems expansion.⁴⁵ Furthermore, as fuels used in the light-duty sector shift toward electricity, this substitution benefit would decrease.

3.1.1. Pathway 1 Results: CO₂ Use in EOR. In this pathway, on top of the upstage emissions described in the previous section, 9 g-CO₂e/MJ is emitted from the combustion of the produced oil. This comprises 25% of the overall emissions in this pathway. This result is similar to many other life cycle studies of EOR in which the emissions from combusting the recovered fuel undercut the avoided CO₂ in the CCS process.⁴⁶ These emissions, which are specific to the Permian Basin, may be lower in regions where the incremental oil productivity per tonne of CO₂ injected may be lower, such as the Gulf Coast.²⁸

In terms of the direct geologic sequestration of CO₂, it is useful to reiterate the key assumptions about CO₂ flows. In the case of EOR, most produced CO₂ in the latter years of the injection process is recycled, i.e., injected back into the oil reservoir, and the residual CO₂ is rerouted to a saline aquifer. We note that geologic sequestration (“Avoided” column in Figure 3) accounts for 83% of the overall mitigation benefits in the case of EOR. Prior position papers and analyses on the life cycle inventory of CCS^{47,48} have shown that the mitigation benefits are more defensible if they rely mostly on actual geologic sequestration (sum of the CO₂ injected in oilfield and saline aquifer in this analysis) instead of substitution benefits. Substitution benefits can vary with fluctuations in markets and as technology evolves. In our analysis, we diversified storage options for CO₂. When oilfields (Figure 2) were saturated with CO₂, the surplus CO₂ was geologically sequestered in aquifers. In our analysis, aquifers stored 14 g-CO₂/MJ in the EOR case. Displacement of existing products/processes reduces net GHG emissions. For instance, the current EOR practices in the Permian Basin use CO₂ from natural formations. Shifting to captured CO₂ from SMR process could lead to systems expansion benefits of 4 g-CO₂e/MJ.

3.1.2. Pathway 2 Results: CO₂ Use as a Fracturing Fluid. In pathway 2, emissions are lower than in pathway 1, but so are substitution credits. Accordingly, net emissions are

similar (~32 g-CO₂e/MJ). Fuel combustion emissions are somewhat less in pathway 2, i.e., 6 g-CO₂/MJ, because we assume that emissions from combustion of the produced natural gas will be captured with 90% CO₂ efficiency. If we consider that the system boundary in Figure 1 ended at the gate of the gas processing plant reflecting today’s conditions in the Permian Basin, the NG produced could also be combusted and the resulting emissions captured with 90% efficiency. In this case, system-level emissions would be around 45 g CO₂e/MJ, which exceeds pathway 2 emissions.

3.1.3. Pathway 3 Results: CO₂ Sequestration in Saline Aquifers. This pathway, for which we estimate total GHG emissions of 28 g-CO₂e/MJ, avoids combustion of produced oil and gas from the use of SMR-derived CO₂. Injection of CO₂ into saline aquifers contributes <1% of the total emissions. Essentially all of the emissions therefore stem from upstream processes. The total CO₂ stored in saline aquifers is 18.8 g-CO₂/MJ. Overall, TX has more than > 1500 Gt storage, and a large share of this is in the Permian Basin.⁴⁰

3.2. Summarizing Emission Reductions

Considering these avoided emissions and system expansion effects in Pathways 1–3, the life cycle GHG emission of 1 MJ of natural gas ranges from 28–32 g CO₂e/MJ. This value is approximately 52–56% lower than baseline life cycle GHG emissions associated with natural gas without any CO₂ sequestration or utilization (88 g-CO₂e/MJ). The main route for these reductions is avoided emissions through geologic sequestration (18 g-CO₂e/MJ). These reductions are analogous with avoided CO₂ emissions from fossil fuel systems when CCS is used.^{48,49} While a 52–56% reduction in life cycle GHG emissions is sizeable, these systems do not deliver fully decarbonized natural gas. The emission reductions are generally similar in all three pathways. Thus, the optimal pathway within the relevant geologic sequestration in the Permian Basin would likely be dependent on other factors (discussed in Section 3.3).

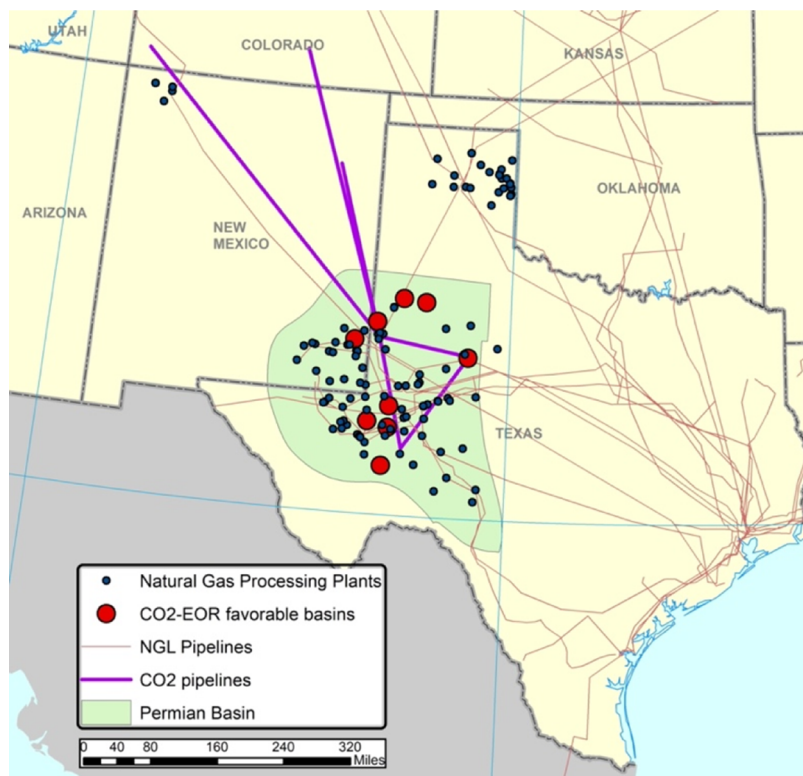
That said, it is important to put these results into context. Analysis of net-zero energy systems shows that the transport sector does not necessarily reach net zero even when global GHG emissions are net zero. Net-zero scenarios are generally characterized by the power sector becoming carbon neutral first, followed by a 1–2 decade lag for the transport sector. This is due to difficulty in decarbonizing liquid fuels. These results show that while geologic CO₂ sequestration does not lead to a fully decarbonized shale gas chain (i.e., with zero GHG emissions), it is much more effective than a counterfactual, where limited options for CO₂ injection underground and ammonia conversion of hydrogen exist.

3.3. Potential for the Use of Coproduced CO₂ and H₂

In the Permian Basin, there are factors that limit the extent of use of CO₂ and H₂, as we have conceptualized. For the EOR pathway, the available pore space in the Permian Basin will limit CO₂ demand. The ARI²⁶ analysis indicates that if 600,000 MJ-oil/day is incrementally recovered through CO₂ injection, the overall storage potential would be 2.6 Bt-CO₂ over a period of 35 years (Figure 2). This corresponds to ~75 Mt-CO₂/year. Based on the parameters we adopted in our analysis, this demand corresponds to a dry natural gas flow of 2 TCF/year. Similarly, the pore space availability for fractured shale reservoirs is 1.8 Bt-CO₂ based on 1800 new wells annually in the Permian Basin. It should be noted that this is a conservative estimate, as recent evidence indicates that the

Table 2. Emissions Reduction Potential of the Pathways Considered in This Study along with the Limiting Factors That Constrain Their Deployment

pathway	net GHG emissions (g-CO ₂ /MJ)	% reduction compared to baseline	amount of shale gas with reduction of emissions (BCF)	limiting factors	coproducts
EOR	32	52%	2000	incremental oil present in conventional hydrocarbon reservoirs	petroleum
fracturing fluid	32	52%	1400	shale gas demand; Limited financial incentives in the current policy (45Q) structure	shale gas
saline aquifer	28	56%	5800	economic incentives dictated largely by market incentives in the absence of beneficial coproduct	none

**Figure 4.** Map showing the proximity of relevant infrastructure (gas processing plants, EOR-favorable oilfields, NGL pipelines, CO₂ pipelines) within/near the Permian Basin. Data sources are 4, 26, and 57.

fracturing fluid requirement in the Permian Basin has increased 800% in the last decade.³⁸ This requirement is higher than for other basins, which have lower lateral depths. Based on this demand for CO₂ and the life cycle GHG emissions we report above, the amount of dry natural gas flow that be produced using reduced emissions using this pathway is 1.4 TCF/year. As noted before, saline aquifers in the Permian Basin have an extremely large pore space availability. Table 2 shows these options and the limiting factors that constrain the deployment of each pathway.

The National Renewable Energy Laboratory¹⁹ estimates that in 2050, the demand for hydrogen as a transportation fuel in the U.S. could reach 22.6 Mt-H₂. In the system we evaluated, this demand would correspond to 4.1 TCF-NG/year. Because we assume H₂ would be converted to ammonia for ease of transporting the energy carrier nationwide, this may be treated as the nominal natural gas demand for H₂ production at a national level. Because of the above-described limits on pore space, pore space availability in geologic sinks would limit the amount of natural gas that can “benefit” from the use of H₂ as a low-carbon transportation fuel to approximately 1.4 TCF/year.

The potential could be increased further if geologic sequestration is assumed without EOR (Pathway 3, 56% GHG emission reduction). For instance, saline aquifers have at least two orders of magnitude higher pore space availability than EOR in the Permian Basin as per the DOE Carbon Storage Atlas.⁴⁰ In the Permian Basin, however, storing CO₂ in oil formations is more technically mature than storing it in aquifers. Furthermore, in the absence of producing a useful product, economic incentives to store CO₂ in that manner would be less strong than using it for EOR. It is noteworthy, though, that the current federal 45Q tax credits do incentivize storage in saline aquifers at a higher rate (\$50/t-CO₂) than EOR (\$35/t-CO₂), which could bridge some of the difference in the available revenues.

An additional reduction of 28–32 g-CO₂e/MJ is needed for carbon neutrality in Pathways 1–3. Certainly, minor reductions may be possible by process improvements in CO₂ capture processes and the hydrogen-to-ammonia conversion process. Additionally, efforts are underway to reduce fugitive methane emissions and reduce energy consumption during gas refining. Additional options to reduce GHG emissions include

electrification of refining facilities and use of carbon capture and sequestration (CCS) to abate emissions from natural gas processing. Beyond technology-based approaches, policies are also being developed to drive down natural gas supply chain emissions. For example, the U.S. Senate recently ratified an act with a methane emissions reduction target of 50% over the next decade.⁵⁰ Another challenge is evaluating the full supply chain GHG emissions of any fuel or product that uses natural gas is how these upstream emissions should be allocated among multiple natural gas coproducts, including oil and natural gas liquids.¹⁶ Most of the emissions in these pathways, however, are due to combustion of NGL-derived fuels and oil, so improvements in CCS technologies and reductions in methane emissions in the NG supply chain are insufficient to achieve fully decarbonized natural gas systems.

It is also noteworthy that the potential for EOR here is estimated only for the Permian Basin. This potential is subjected to several adjustments. On one hand, CO₂-EOR potential is influenced by market conditions. Between 2019 and 2020, the oil produced through EOR in the Permian Basin decreased from 204 to 185 MB/day, with the corresponding CO₂ demand reducing from 1830 to 1010 MMcf/day.⁵¹ This was largely due to declining oil prices. There is decreasing demand for additional oil, especially as refineries are moving away from gasoline production.⁶ That said, there is also a possibility of higher EOR potential based on the exploration of shale reservoirs as CO₂ sinks.^{52,53} The Permian Basin has an estimated 16 Bt-CO₂ of storage potential in shale formations that could potentially recover an additional 47,000 MMB-oil.⁵⁴ New projects in the Permian Basin are making use of CO₂ captured from the ambient air (instead of fossil fuels) for CO₂-EOR, which could further the cause of carbon neutrality in the transport sector.⁵⁵

It is also noteworthy that the Permian Basin is uniquely poised to adopt systems such as the two we conceptualized. Multiple analyses suggest that the U.S.-wide EOR deployment could rise as high as 200–260 Mt annually, out of which a majority could be within the Permian Basin. There is a substantial experience of such projects in the region. For instance, the Petra Nova Project—located in Thomsons, Texas—had transported 3.3 Mt-CO₂ during 2017–2020, before oil prices declined due to the COVID-19 pandemic. The Port Arthur SMR project within the Valero refinery also captures more than a million tonnes of CO₂ each year for EOR as part of the DOE Industrial Carbon Capture Storage Initiative.⁵⁶ Such projects could be further scaled up as the Oil and Gas Climate Initiative eyes creation of market conditions for CCUS deployment in the region. Figure 4 shows the prominent EOR oilfields, where production is already taking place or is likely to take place. Also shown are existing NGL and CO₂ transport infrastructure and gas processing plants, where NG and NGL are separated.

Another appealing prospect for large-scale CCS in the region is the extent to which CO₂ and hydrogen transport infrastructure is already in place. Over 2600 miles of CO₂ pipelines already exist in the region, which represents the highest concentration of such infrastructure globally.⁵⁷ This includes six large-scale trunk pipelines (shown in Figure 4), with throughput > 200 MMcf/d, and several smaller-scale distribution systems, with a throughput of 50–200 MMcf/d.⁵⁸ Forward-looking analyses suggest that throughput capacity of an additional 20 Mt-CO₂ could be developed in the short term.⁵⁹ Moreover, an ammonia terminal already exists in

Galveston and could also be established near Houston to handle 1.3 Mt-NH₃ annually with an investment of \$1 billion.

This analysis illustrates that it is incredibly difficult to fully decarbonize shale gas systems that include fossil-fuel combustion relying mainly on subsurface sequestration of CO₂ from natural gas SMR. Nonetheless, it highlights opportunities to create lower-emitting shale gas systems that link to emerging low-carbon systems like the hydrogen economy as the energy landscape continues to evolve. As the U.S. considers how to phase down fossil fuels, it is worth continuing to evaluate how such links and bridges from shale gas systems to lower carbon transportation systems might be possible. Examples related to the conceptual scenarios we considered include developing technology to derisk biofuels or e-fuels by providing inexpensive H₂ for pyrolysis oil upgrading or carbon capture and utilization technologies that use CO₂ and H₂.

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Notes

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REFERENCES

- (1) Chen, Z.; Li, Y.; Oladipupo, W. P.; Rodriguez Gil, E. A.; Sawyer, G.; Agrawal, R. Alternative Ordering of Process Hierarchy for More Efficient and Cost-Effective Valorization of Shale Resources. *Cell Rep. Phys. Sci.* **2021**, 2, No. 100581.
- (2) Chen, Z.; Agrawal, R. Alternative Processing Sequence for Process Simplification, Cost Reduction, and Enhanced Light Olefin Recovery from Shale Gas. *ACS Sustainable Chem. Eng.* **2021**, 9, 13893–13901.
- (3) US EIA. Shale Gas Production. https://www.eia.gov/dnav/ng/ng_prod_shalegas_s1_a.htm (accessed Dec 8, 2021).
- (4) US EIA. US Field Production of Natural Gas Liquids. https://www.eia.gov/dnav/pet/hist/LeafHandler.ashx?n=PET&s=M_EPL2_FPF_NUS_MBBLD&f=A (accessed Dec 8, 2021).
- (5) Ridha, T.; Li, Y.; Gençer, E.; Sirola, J. J.; Miller, J. T.; Ribeiro, F. H.; Agrawal, R. Valorization of Shale Gas Condensate to Liquid Hydrocarbons through Catalytic Dehydrogenation and Oligomerization. *Processes* **2018**, 6, No. 139.

- (6) US EIA. 2021 Annual Energy Outlook. <https://www.eia.gov/outlooks/aeo/pdf/02/AEO2021/Petroleum.pdf> (accessed Jan 1, 2022).
- (7) Alvarez, R. A.; Zavala-Araiza, D.; Lyon, D. R.; Allen, D. T.; Barkley, Z. R.; Adam, R.; Davis, K. J.; Herndon, S. C.; Jacob, D. J.; Karion, A.; Kort, E. A.; Lamb, B. K.; Lauvaux, T.; Maasackers, J. D.; Marchese, A. J.; Omara, M.; Pacala, S. W.; Peischl, J.; Robinson, A. L.; Shepson, P. B.; Sweeney, C.; Townsend-Small, A.; Wofsy, S. C.; Hamburg, S. P. Assessment of Methane Emissions from the U.S. Oil and Gas Supply Chain. *Science* **2018**, *361*, 186–188.
- (8) Zhang, Y.; Gautam, R.; Pandey, S.; Omara, M.; Maasackers, J. D.; Sadavarte, P.; Lyon, D.; Nesser, H.; Sulprizio, M. P.; Varon, D. J.; Zhang, R.; Houweling, S.; Zavala-Araiza, D.; Alvarez, R. A.; Lorente, A.; Hamburg, S. P.; Aben, I.; Jacob, D. J. Quantifying Methane Emissions from the Largest Oil-Producing Basin in the United States from Space. *Sci. Adv.* **2020**, *6*, No. eaaz5120.
- (9) BP. Energy Outlook 2020 edition explores the forces shaping the global energy transition out to 2050 and the surrounding that <https://www.bp.com/content/dam/bp/business-sites/en/global/corporate/pdfs/energy-economics/energy-outlook/bp-energy-outlook-2020.pdf> (accessed Jan 1, 2022).
- (10) Royal Dutch Shell. Shell Scenarios: Sky - Meeting the goals of the Paris agreement. https://www.shell.com/promos/meeting-the-goals-of-the-paris-agreement/_jcr_content.stream/1521983847468/5f624b9260ef2625f319558cbb652f8b23d331933439435d7a0fc7003f346f94/shell-scenarios-sky.pdf (accessed Jan 1, 2022).
- (11) Chapman, A.; Itaoka, K.; Hirose, K.; Davidson, F. T.; Nagasawa, K.; Lloyd, A. C.; Webber, M. E.; Kurban, Z.; Managi, S.; Tamaki, T.; Lewis, M. C.; Hebner, R. E.; Fujii, Y. A Review of Four Case Studies Assessing the Potential for Hydrogen Penetration of the Future Energy System. *Int. J. Hydrogen Energy* **2019**, *44*, 6371–6382.
- (12) McPherson, M.; Johnson, N.; Strubegger, M. The Role of Electricity Storage and Hydrogen Technologies in Enabling Global Low-Carbon Energy Transitions. *Appl. Energy* **2018**, *216*, 649–661.
- (13) Deutch, J. Is Net Zero Carbon 2050 Possible? *Joule* **2020**, *4*, 2237–2240.
- (14) Jenkins, J. D.; Mayfield, E. N.; Larson, E. D.; Pacala, S. W.; Greig, C. Mission Net-Zero America: The Nation-Building Path to a Prosperous, Net-Zero Emissions Economy. *Joule* **2021**, *5*, 2755–2761.
- (15) Chen, Q.; Dunn, J. B.; Allen, D. T. Aggregation and Allocation of Greenhouse Gas Emissions in Oil and Gas Production: Implications for Life-Cycle Greenhouse Gas Burdens. *ACS Sustainable Chem. Eng.* **2019**, *7*, 17065–17073.
- (16) Allen, D. T.; Chen, Q.; Dunn, J. B. Consistent Metrics Needed for Quantifying Methane Emissions from Upstream Oil and Gas Operations. *Environ. Sci. Technol. Lett.* **2021**, *8*, 345–349.
- (17) Qyyum, M. A.; Naquash, A.; Haider, J.; Al-Sobhi, S. A.; Lee, M. State-of-the-Art Assessment of Natural Gas Liquids Recovery Processes: Techno-Economic Evaluation, Policy Implications, Open Issues, and the Way Forward. *Energy* **2022**, *238*, No. 121684.
- (18) Dutta, A.; Karimi, I. A.; Farooq, S. Technoeconomic Perspective on Natural Gas Liquids and Methanol as Potential Feedstocks for Producing Olefins. *Ind. Eng. Chem. Res.* **2019**, *58*, 963–972.
- (19) Ruth, M. F.; Jadun, P.; Gilroy, N.; Connelly, E.; Richard, B.; Simon, A. J.; Elgowainy, A.; Zuboy, J. *The Technical and Economic Potential of the H₂@Scale Hydrogen Concept within the United States*; National Renewable Energy Laboratory, 2020.
- (20) Wilkins, R.; Menefee, A. H.; Clarens, A. F. Environmental Life Cycle Analysis of Water and CO₂-Based Fracturing Fluids Used in Unconventional Gas Production. *Environ. Sci. Technol.* **2016**, *50*, 13134–13141.
- (21) Collodi, G.; Azzaro, G.; Ferrari, N.; Santos, S. Techno-Economic Evaluation of Deploying CCS in SMR Based Merchant H₂ Production with NG as Feedstock and Fuel. *Energy Procedia* **2017**, *114*, 2690–2712.
- (22) Bartlett, J.; Krupnick, A. *Decarbonized Hydrogen in the US Power and Industrial Sectors: Identifying and Incentivizing Opportunities to Lower Emissions*; Resources for the Future, 2020.
- (23) Bartels, J. R. *A Feasibility Study of Implementing an Ammonia Economy*; Iowa State University, 2008.
- (24) Ishimoto, Y.; Voldsund, M.; Nekså, P.; Roussanaly, S.; Berstad, D.; Gardarsdottir, S. O. Large-Scale Production and Transport of Hydrogen from Norway to Europe and Japan: Value Chain Analysis and Comparison of Liquid Hydrogen and Ammonia as Energy Carriers. *Int. J. Hydrogen Energy* **2020**, *45*, 32865–32883.
- (25) Fúnez Guerra, C.; Reyes-Bozo, L.; Vyhmeister, E.; Jaén Caparrós, M.; Salazar, J. L.; Clemente-Jul, C. Technical-Economic Analysis for a Green Ammonia Production Plant in Chile and Its Subsequent Transport to Japan. *Renewable Energy* **2020**, *157*, 404–414.
- (26) ARI. Basin Oriented Strategies for CO₂ Enhanced Oil Recovery - Permian Basin. <https://netl.doe.gov/sites/default/files/2021-03/permian.pdf> (accessed Jan 1, 2022).
- (27) Wildbolz, C. Life Cycle Assessment of Selected Technologies for CO₂ Transport and Sequestration. Diploma Thesis, Swiss Federal Institute of Technology Zurich, 2007. <https://www.doka.ch/CCSDiplomaWildbolz07.pdf> (accessed April 4, 2022).
- (28) Abotalib, M.; Zhao, F.; Clarens, A. Deployment of a Geographical Information System Life Cycle Assessment Integrated Framework for Exploring the Opportunities and Challenges of Enhanced Oil Recovery Using Industrial CO₂ Supply in the United States. *ACS Sustainable Chem. Eng.* **2016**, *4*, 4743–4751.
- (29) Hussain, D.; Dzombak, D. A.; Jaramillo, P.; Lowry, G. V. Comparative Lifecycle Inventory (LCI) of Greenhouse Gas (GHG) Emissions of Enhanced Oil Recovery (EOR) Methods Using Different CO₂ Sources. *Int. J. Greenhouse Gas Control* **2013**, *16*, 129–144.
- (30) Middleton, R. S.; Clarens, A. F.; Liu, X.; Bielicki, J. M.; Levine, J. S. CO₂ Deserts: Implications of Existing CO₂ Supply Limitations for Carbon Management. *Environ. Sci. Technol.* **2014**, *48*, 11713–11720.
- (31) Melara, A. J.; Singh, U.; Colosi, L. M. Is Aquatic Bioenergy with Carbon Capture and Storage a Sustainable Negative Emission Technology? Insights from a Spatially Explicit Environmental Life-Cycle Assessment. *Energy Convers. Manage.* **2020**, *224*, No. 113300.
- (32) McCoy, S. T.; Rubin, E. S. An Engineering-Economic Model of Pipeline Transport of CO₂ with Application to Carbon Capture and Storage. *Int. J. Greenhouse Gas Control* **2008**, *2*, 219–229.
- (33) Mabon, L.; Littlecott, C. Stakeholder and Public Perceptions of CO₂-EOR in the Context of CCS - Results from UK Focus Groups and Implications for Policy. *Int. J. Greenhouse Gas Control* **2016**, *49*, 128–137.
- (34) Gunderson, R.; Stuart, D.; Petersen, B. The Fossil Fuel Industry's Framing of Carbon Capture and Storage: Faith in Innovation, Value Instrumentalization, and Status Quo Maintenance. *J. Cleaner Prod.* **2020**, *252*, No. 119767.
- (35) Nagashima, S.; Miyagawa, T.; Matsumoto, M.; Suzuki, S.; Komaki, H.; Takagi, M.; Murai, S. Life Cycle Assessment Performed on a CCS Model Case in Japan and Evaluation of Improvement Facilitated by Heat Integration. *Energy Procedia* **2011**, *4*, 2457–2464.
- (36) Corsten, M.; Ramírez, A.; Shen, L.; Koornneef, J.; Faaij, A. Environmental Impact Assessment of CCS Chains - Lessons Learned and Limitations from LCA Literature. *Int. J. Greenhouse Gas Control* **2013**, *13*, 59–71.
- (37) Bartholomew, T. V.; Mauter, M. S. Energy and CO₂ Emissions Penalty Ranges for Geologic Carbon Storage Brine Management. *Environ. Sci. Technol.* **2021**, *55*, 4305–4313.
- (38) Kondash, A. J.; Lauer, N. E.; Vengosh, A. The Intensification of the Water Footprint of Hydraulic Fracturing. *Sci. Adv.* **2018**, *4*, No. eaar5982.
- (39) Martin-Roberts, E.; Scott, V.; Flude, S.; Johnson, G.; Haszeldine, R. S.; Gilfillan, S. Carbon Capture and Storage at the End of a Lost Decade. *One Earth* **2021**, *4*, 1569–1584.
- (40) NETL *Carbon Storage Atlas*, 5th ed.; National Energy Technology Laboratory, 2015.

- (41) Feijoo, F.; Iyer, G.; Binsted, M.; Edmonds, J. US Energy System Transitions under Cumulative Emissions Budgets. *Clim. Change* **2020**, *162*, 1947–1963.
- (42) McJeon, H.; Mignone, B. K.; O'Rourke, P.; Horowitz, R.; Khesghi, H. S.; Clarke, L.; Kyle, P.; Patel, P.; Edmonds, J. Fossil Energy Deployment through Midcentury Consistent with 2 °C Climate Stabilization. *Energy Clim. Change* **2021**, *2*, No. 100034.
- (43) US EPA. Commercial Fertilizer Purchased. <https://www.epa.gov/nutrient-policy-data/commercial-fertilizer-purchased> (accessed Dec 8, 2021).
- (44) Spallina, V.; Shams, A.; Battistella, A.; Gallucci, F.; Annaland, M. V. S. Chemical Looping Technologies for H₂ Production with CO₂ Capture: Thermodynamic Assessment and Economic Comparison. *Energy Procedia* **2017**, *114*, 419–428.
- (45) Wang, M.; Huo, H.; Arora, S. Methods of Dealing with Co-Products of Biofuels in Life-Cycle Analysis and Consequent Results within the U.S. Context. *Energy Policy* **2011**, *39*, 5726–5736.
- (46) Jaramillo, P.; Griffin, W. M.; McCoy, S. T. Life Cycle Inventory of CO₂ in an Enhanced Oil Recovery System. *Environ. Sci. Technol.* **2009**, *43*, 8027–8032.
- (47) Terlouw, T.; Bauer, C.; Rosa, L.; Mazzotti, M. Life Cycle Assessment of Carbon Dioxide Removal Technologies: A Critical Review. *Energy Environ. Sci.* **2021**, *14*, 1701–1721.
- (48) Singh, U.; Colosi, L. M. The Case for Estimating Carbon Return on Investment (CROI) for CCUS Platforms. *Appl. Energy* **2021**, *285*, No. 116394.
- (49) Müller, L. J.; Käthelhön, A.; Bachmann, M.; Zimmermann, A.; Sternberg, A.; Bardow, A. A Guideline for Life Cycle Assessment of Carbon Capture and Utilization. *Front. Energy Res.* **2020**, *8*, 1–20.
- (50) Vaughan, A. The Methane Mystery. *New Sci.* **2021**, *250*, 16–17.
- (51) ARI. *The U.S. CO₂ Enhanced Oil Recovery Survey An Interim Update of Enhanced Oil Production Totals and CO₂ Supplies for Active Survey at a Glance*; Advanced Resources International, 2021.
- (52) Singh, U.; Sharma, N.; Dunn, J. B. Revisiting Geologic Storage Potential in Unconventional Formations Is Key to Proactive Decision Making on CCS in India. *Front. Clim.* **2021**, *3*, No. 708320.
- (53) Menefee, A. H.; Ellis, B. R. Regional-Scale Greenhouse Gas Utilization Strategies for Enhanced Shale Oil Recovery and Carbon Management. *Energy Fuels* **2020**, *34*, 6136–6147.
- (54) ARI. Increasing Shale Oil Recovery and CO₂ Storage With Cyclic CO₂ Enhanced Oil Recovery. <https://adv-res.com/pdf/USEA-ARI-Shale-Recovery-Storage-CO2-EOR-SEP-22-2020.pdf> (accessed Jan 1, 2022).
- (55) Gupte, E.; VanLaningham, P. US' Occidental supplies first cargo of "carbon-neutral crude" to India's Reliance.
- (56) Medlock, K. B.; Miller, K. *To Reduce Industrial Emissions in Texas CARBON CAPTURE IN TEXAS: COMPARATIVE ADVANTAGE IN A LOW CARBON PORTFOLIO*; Baker Institute, 2020.
- (57) Edwards, R. W. J.; Celia, M. A. Infrastructure to Enable Deployment of Carbon Capture, Utilization, and Storage in the United States. *Proc. Natl. Acad. Sci. U.S.A.* **2018**, *115*, E8815–E8824.
- (58) Wallace, M.; Goudarzi, L.; Callahan, K.; Wallace, R. *A Review of the CO₂ Pipeline Infrastructure in the U.S.*; National Energy Technology Laboratory, 2015.
- (59) Friedmann, S. J.; Agrawal, M.; Bhardwaj, A. *Evaluating Net-Zero Industrial Hubs in the United States: A Case Study of Houston*; Columbia University, 2021.