A Multi-scale Framework for CO₂ Capture, Utilization, and Sequestration: CCUS and CCU

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Abstract

We present a multi-scale framework for the optimal design of CO₂ capture, utilization, and sequestration (CCUS) supply chain network to minimize the cost while reducing stationary CO₂ emissions in the United States. We also design a novel CO₂ capture and utilization (CCU) network for economic benefit through utilizing CO₂ for enhanced oil recovery. Both the designs of CCUS and CCU supply chain networks are multi-scale problems which require decision making at material, process and supply chain levels. We present a hierarchical and multi-scale framework to design CCUS and CCU supply chain networks with minimum investment, operating and material costs. While doing so, we take into consideration the selection of source plants, capture processes, capture materials, CO₂ pipelines, locations of utilization and sequestration sites, and amounts of CO₂ storage. Each CO₂ capture process is optimized, and the best materials are screened from large pool of candidate materials. Our optimized CCUS supply chain network can reduce 50% of the total stationary CO₂ emission in the U.S. at a cost of \$ 35.63 per ton of CO₂ captured and managed. The optimum CCU supply chain network can capture and utilize CO₂ to make a total profit of more than 555 million dollars per year (\$9.23 per ton). We have also shown that more than 3% of the total stationary CO₂ emissions in the United States can be eliminated through CCU networks at zero net cost. These results highlight both the environmental and economic benefits which can be gained through CCUS and CCU networks. We have designed the CCUS and CCU networks through (i) selecting

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novel materials and optimized process configurations for CO₂ capture, (ii) simultaneous selection of materials and capture technologies, (iii) CO₂ capture from diverse emission sources, and (iv) CO₂ utilization for enhanced oil recovery. While we demonstrate the CCUS and CCU networks to reduce stationary CO₂ emissions and generate profits in the United States, the proposed framework can be applied to other countries and regions as well.

Keywords: Carbon capture and Storage, CO₂ utilization, CO₂ sequestration, CCUS, CCU, Optimization

1 Introduction

Global warming and recent climate change are often attributed to CO₂ emissions from burning and consumption of fossil fuels. Fossil fuels supply more than 85% of the current energy consumption worldwide, and contribute in similar proportions to the anthropogenic CO₂ emission. In the United States, large stationary sources such as power plants, cement production, iron and steel industries, refineries, petrochemicals, and gas processing plants contribute to more than 60% of the total stationary emissions. Stationary sources are point sources with large CO₂ emissions, and provide a realistic opportunity to reduce CO₂ emission. Recently, most of the stationary sources in the United States are geographically identified and their emissions are reliably estimated (National Energy Technology Laboratory, 2010a, 2012). CO₂ emissions from these sources can be reduced by capturing CO₂ in bulk and then utilizing and/or storing in large underground reservoirs for sequestration. The overall scheme for long-term CO₂ containment through capture, compression, transportation and utilization or storage is known as CO₂ capture, utilization and sequestration (CCUS) (Metz, Davidson, De Coninck, Loos, and Meyer, 2005; DOE, 1999; Chu, 2009; Haszeldine, 2009).

In a CCUS supply chain, CO₂ is targeted to be captured and compressed at the source plant, and then transported via pipeline to a site where it is utilized or injected for geological sequestration (Fig. 1). CCUS has the potential to reduce energy-related CO₂ emissions by 20% worldwide (IEA, International Energy Agency, Paris, 2008). As of mid-2012, at least 254 CCUS projects are in operation or under planning and development stages worldwide (National Energy Technology Laboratory, 2012). Furthermore, the utilization of CO₂ for tertiary oil recovery provides opportunities to supplement and eventually replace the naturally occurring CO₂ with CO₂ from anthropogenic sources for enhanced oil recovery (CO₂-EOR) (Godec, Kuuskraa, and Dipietro, 2013; Godec, 2012). CO₂-EOR can increase the oil recovery by as much as 15% of the original oil in place (OOIP) (National Energy Technology Laboratory, 2010a).

[Figure 1 about here.]

Capturing CO₂ from the right sources and storing in the right sites and the right amounts are crucial for a CCUS to be cost-effective. The selection of technologies and materials for CO₂ capture are equally important. Hasan *et al.* (Hasan, Baliban, Elia, and Floudas, 2012a,b) optimized the

capture and compression costs for leading capture technologies, namely absorption, membrane, and adsorption (PSA and VSA) for a range of stationary sources, and showed that no single technology is the best for all CO₂ compositions and flow rates. Even when the technology remains the same, the capture cost can significantly vary depending on the materials selected for CO₂ capture (Hasan, First, and Floudas, 2013).

Significant challenges remain to be addressed before CCUS can be deployed at the industrial scale. A major obstacle for the deployment of CCUS is the current high cost of CO₂ capture, compression and transportation. The overall CCUS cost depends on several factors. Each source is different than others in terms of the CO₂ flow rate, CO₂ composition in the exhaust flue gas, and moisture content of the flue gas. In our previous works (Hasan, Baliban, Elia, and Floudas, 2012a,b; Hasan, First, and Floudas, 2013; Hasan, Boukouvala, First, and Floudas, 2014), we quantitatively showed that these factors greatly affect the capture and compression costs. Furthermore, depending on the materials used, the capture costs can vary as much as 25%.

Recent results (Hasan, First, and Floudas, 2013; First, Hasan, and Floudas, 2014) also suggest that we must select the best materials and optimize processes in tandem for effective CO₂ separation. We need to design the CCUS network supply chain such that the sources are optimally connected with the utilization and sequestration sites (Hasan, Boukouvala, First, and Floudas, 2014). The cost of CO₂ transportation changes with the location of the sources, utilization sites, and sequestration wells. Most literature considers the transportation cost to be \$6-8 per ton of CO₂ transported. However, the transportation cost can vary from as low as \$0.20 to as high as \$40 for each ton of CO₂, depending on the distance between a source and a utilization/sequestration site (Hasan, Boukouvala, First, and Floudas, 2014). Connecting sources with appropriate sites is a combinatorial problem where the number of alternatives increases with the number of sources and utilization/sequestration sites.

While most studies considered CCS or CCUS activities, few considered the capture and utilization of CO₂ at the same time. In this work, we introduce the concept of a CO₂ capture and utilization (CCU) supply chain network to capture CO₂ from the source plants, and utilize the anthropogenic CO₂ for enhanced oil recovery purposes. While the goal of a CCUS supply chain network is to reduce the CO₂ emissions, the goal of a CCU supply chain network is to maximize the revenue or profit from CO₂ utilization. Therefore, the CCU supply chains are driven by eco-

conomic drivers, whereas the CCUS supply chains are primarily motivated by the environmental benefits through reducing emissions.

We present a novel, comprehensive multi-scale framework toward reducing the overall CCUS cost, and maximize the overall CCU profit. We consider the selection of source plants, capture processes, capture materials, CO₂ pipelines, locations of utilization and sequestration sites, and amounts of CO₂ storage. Each process is optimized based on overall cost, and the best materials are screened from large pool of candidate materials. The cost models include the capture, compression, transportation and injection costs, and revenues from CO₂ utilization. The capture and compression costs are derived using advanced modeling, simulation and optimization of CO₂ capture processes. The transportation and injection costs are computed using models from literature (Ogden, 2004). Our explicit input-output models for the capture and compression costs consider the variation in source flue gas compositions and flow rates. We consider real geographic locations and storage estimates from the available database, and incorporate rigorous cost estimations based on material selection and process optimization.

The novel features of this work are:

- (i) systematic formulation and optimization of a novel CO₂ capture and utilization (CCU) supply chain network,
- (ii) a multi-scale framework for the design of optimal CCUS and CCU supply chain networks,
- (iii) a zero-cost CCU network to reduce CO₂ emissions in the United States,
- (iv) screening and selection of novel materials and optimized process configurations for CO₂ capture for CCUS and CCU,
- (v) simultaneous selection of materials and capture technologies for CCUS and CCU,
- (vi) CCUS and CCU supply chain networks for minimizing emissions and maximizing profits by implementing CO₂ capture from diverse emission sources in the United States, and
- (vii) a systematic framework for CO₂ utilization for enhanced oil recovery through CCU supply chain networks.

We design a novel CO₂ capture and utilization (CCU) network based on a large-scale mixed integer linear optimization (MILP) to maximize the profit from CCU supply chain through the utilization of anthropogenic CO₂ for enhanced oil recovery. While we demonstrate the CCUS and CCU networks to reduce stationary CO₂ emissions in the United States, the proposed framework

can be applied to other countries and regions.

In the sequel, we provide a brief overview of CCUS and CCU, and their challenges. We then describe the overall multi-scale framework for the design and synthesis of optimal CCUS and CCU supply chain networks while considering materials screening, process optimization, technology selection, and supply chain network optimization toward reducing the CCUS cost. We describe a hierarchical *in silico* screening method to select candidate materials and optimize process conditions for CO₂ capture. We also discuss the explicit expressions that we have developed for the investment and operating costs to determine the most cost-effective materials and processes when addressing diverse emission scenarios. We present a framework for the optimization of nationwide CCUS and CCU supply chain networks to reduce the stationary CO₂ emissions at reasonable cost, and maximize the profits from CO₂-enhanced oil recovery in the United States, respectively. Lastly, we present and discuss a CCU supply chain network that reduces CO₂ emissions without additional cost (a zero-cost network where the costs are balanced out by the revenues).

2 CCUS and CCU Overview

The main components of a CCUS supply chain network are (i) CO₂ source, (ii) capture plant, (iii) pipeline for transportation, (iv) utilization site, and (v) sequestration site. The capture plant uses a process to separate or capture CO₂ from the sources. Depending on the separation technology used for CO₂ capture, each process uses a material (e.g., solvent, adsorbent, membrane, etc.) that is critical for the selective separation of CO₂ from the flue gas. Once the CO₂ is captured, it is compressed and transported to the utilization or sequestration site. CO₂ emitting industrial sources are the main suppliers of CO₂, and the demands of CO₂ come from various utilization and sequestration sites. Transport-ready CO₂ product is produced in capture plants attached to the sources, and pipelines serve as the main workhorse in CO₂ logistics.

A CCUS supply chain network can have multiple sources, capture plants with specific materials used for CO₂ separation, and utilization and sequestration sites. To this end, a CCUS supply chain network can be represented as a network as shown in Fig. 2.

[Figure 2 about here.]

Several terminologies used in this work are defined in Table 1. The descriptions provided herein are relevant to this work. For instance, while CO₂ utilization may include CO₂ conversion to other chemicals, using as solvents, and so on, we only consider CO₂-EOR as the only utilization of CO₂ in this work. In this paper, we consider two different CO₂ networks, namely (i) CO₂ capture, utilization and sequestration (CCUS) network, and (ii) CO₂ capture and utilization (CCU) network. CCUS network includes both utilization and sequestration of CO₂, while CCU network only considers utilization.

[Table 1 about here.]

3 CCUS and CCU Challenges

CCUS and CCU are complex and large-scale activities which pose several challenges. A major challenge is the current high investment and operating costs of CO₂ capture and compression. The costs of CO₂ capture and sequestration (without utilization) assessed for power plants range from \$72 to \$114/ton CO₂ avoided (Report DOE/NETL-2007/1281, August, 2007; Report DOE/NETL-2010/1397, November, 2010; Interagency Task Force on Carbon Capture and Storage, August, 2010). CO₂ avoidance from high-purity CO₂ sources (\$30–70), biomass conversion (\$35–80), refineries (\$45–120), cement (\$55–150), and iron and steel (\$60–80) are considered on an ad hoc basis. Such high costs are primarily due to costly capture of CO₂ from sources which are diluted in CO₂. In fact, CO₂ capture is the most cost-intensive entity in a CCUS supply chain (Yang, Xu, Fan, Gupta, Slimane, Bland, and Wright, 2008; MacDowell, Florin, Buchard, Hallett, Galindo, Jackson, Adjiman, Williams, Shah, and Fennell, 2010).

CO₂ sources also pose significant challenges for CCUS and CCU. All sources have differences in plant types, flue gas CO₂ compositions and flow rates. Most stationary sources in the United States (Fig. 3) are scattered in different regions and have varying emission amounts, which diminish the likelihood of a central CO₂ collection grid. All the 3317 sources shown in Fig. 3 have different CO₂ flow rates (from several thousand tons to more than 25 Mt per year). The largest sources are the power plants (630 coal-fired, 831 natural gas-fired, and 414 using other fuels). The costs of CO₂ capture for these large sources would be different than those of the smaller sources.

[Figure 3 about here.]

It is important to note that not all stationary sources have the same CO₂ compositions in their CO₂ containing streams or flue gases. Coal and natural gas-fired power plants, which are the major sources of CO₂ with large flow rates, have only 4–14% of CO₂ in the flue gases. On the other hand, iron and steel production, agricultural processing and sugar production often produce streams containing more than 40% CO₂. Depending on the CO₂ composition and flow rate of the flue gas, the cost of CO₂ capture would vary (Hasan, Baliban, Elia, and Floudas, 2012a,b). The stationary sources in the United States have a wide range (4 to 47%) of flue gas CO₂ compositions. Based on their flue gas CO₂ compositions and plant types, we can classify these sources into twenty different categories as follows (typical CO₂ compositions are shown in brackets: agricultural processing (47%), ammonia (8%), asphalt production (7%), cement (47%), refineries/chemical plants (7%), compressor stations (petroleum/natural gas, 4%), coal-fired power plants (14%), natural gas-fired power plants (4%), oil and other fuels based power plants (11%), ethanol plants (13%), petrochemicals (7%), industrial, manufacturing, aluminum, glass, mining (15%), gas processing (25%), iron & steel (corex, 24%), iron & steel (blast furnace, 20%), iron & steel (CCF, 44%), paper and wood products (13%), sugar (47%), utility (11%), and others (11%). A complete list of these source categories with compositions and number of sources in the United States can be found in the Supplementary Material (Supplementary Table 1).

Although many separation-based technologies exist for CO₂ capture, not all are economically attractive. Each technology has many alternatives for processes and materials for CO₂ capture. The suitability of a capture technology depends on several factors, which include CO₂ composition and moisture content of the feed flue gas, feed flow rate, type of capture, capture performance rating, and cost.

Fig. 4 lists the major CO₂ capture technologies, processes and classes of materials for CO₂ capture. Absorption is the most mature technology for CO₂ capture (Wang, Lawal, Stephenson, Sidders, and Ramshaw, 2011; Hasan, Baliban, Elia, and Floudas, 2012a; Rao and Rubin, 2002; Mores, Scenna, and Mussati, 2011; Oyeneke and Rochelle, 2006; Pellegrini, Moioli, and Gamba, 2011; Oyeneke and Rochelle, 2009; Rodríguez, Mussati, and Scenna, 2011; Oexmann, Hensel, and Kather, 2008; Harkin, Hoadley, and Hooper, 2010). However, the cost of post-combustion CO₂ capture from power plants using absorption technology, based on monoethanol amine (MEA)

solvent, is currently over \$70 per metric ton of CO₂ avoided (Raksajati, Ho, and Wiley, 2013). Furthermore, high energy requirement for regeneration, solvent degradation and loss, corrosion, etc., often make this technology prohibitive to use. Absorption can be physical or chemical, and different solvents can be used to capture CO₂. Not all solvents perform equally, and it is a challenge to get the optimal process configuration even for a fixed solvent.

[Figure 4 about here.]

Adsorption-based CO₂ capture (Hasan, Baliban, Elia, and Floudas, 2012b; Reynolds, Ebner, and Ritter, 2006; Reynolds, Mehrotra, Ebner, and Ritter, 2008; Ho, Allinson, and Wiley, 2008b; Zhang, Webley, and Xiao, 2008; Radosz, Hu, Krutkramelis, and Shen, 2008; Ko, Siriwardane, and Biegler, 2003; Agarwal, Biegler, and Zitney, 2010; Hasan, First, and Floudas, 2013; Haghpanah, Majumder, Nilam, Rajendran, Farooq, Karimi, and Amanullah, 2013) is a promising technology for CO₂ capture. Major adsorbents for CO₂ separation include microporous/mesoporous silica or zeolites, activated carbonaceous materials and metal organic frameworks (MOFs) (Samanta, Zhao, Shimizu, Sarkar, and Gupta, 2012; D'Alessandro, Smit, and Long, 2010; Li, Ma, McCarthy, Sculley, Yu, Jeong, Balbuena, and Zhou, 2011; Lin, Berger, Martin, Kim, Swisher, Jariwala, Rycroft, Bhowm, Deem, Haranczyk, et al., 2012; Krishna and van Baten, 2012; Hasan, First, and Floudas, 2013). A key challenge in adsorption-based CO₂ capture is that not all adsorbents exhibit similar performance. Adsorbent selection is a challenging problem given the vast pool of candidate zeolites and MOFs which can be used for CO₂ capture.

Membranes are not yet commercially popular for post-combustion CO₂ capture, mainly because of the high capital costs (Ho, Allinson, and Wiley, 2006; Ho, Allinson, and Dianne, 2008a) associated with feed gas compression to increase the driving force for separating CO₂ with low partial pressure. However, recent works on multi-stage separation and vacuum permeate pumping have shown promise toward cost-effective CO₂ capture using membranes (Hasan, Baliban, Elia, and Floudas, 2012a; Zhang, He, and Gundersen, 2013; Ho, Allinson, and Wiley, 2006; Kaldis, Skodras, and Sakellariopoulos, 2004; Hägg and Lindbråthen, 2005). Similar to adsorption-based processes, membrane-based processes also require the selection of an appropriate membrane materials from different polymeric, facilitated transport, inorganic and mixed matrix membranes which show potential for CO₂ capture (Powell and Qiao, 2006; Ramasubramanian, Zhao, and Winston Ho,

2013; Shah, McCarthy, Sachdeva, Lee, and Jeong, 2012; Bernardo, Drioli, and Golemme, 2009).

Presence of water in the feed flue gas is another challenge for CO₂ capture technologies such as adsorption and membrane. Depending on the source, flue gas can contain 5–15% H₂O. While alternatives exist for flue gas dehydration (Hasan, Baliban, Elia, and Floudas, 2012a), dehydration incurs an additional cost of \$10.22 per ton of CO₂ to the overall CCUS and CCU chains Hasan et al. (2012a,b, 2013, 2014).

Technologies for CCUS schemes are historically developed independently of one another. Such standalone development complicates the integration. For large-scale CCUS implementation, it is critical to optimally integrate capture, compression, transportation, utilization and sequestration activities. Although CO₂ management using CO₂ capture and sequestration infrastructure has been considered, these studies did not include utilization opportunities (Tan, Aviso, Bandyopadhyay, and Ng, 2013; Kazmierczak, Brandsma, Neele, and Hendriks, 2009; Johnson and Ogden, 2011; Zheng, Larson, Li, Liu, and Williams, 2010; van den Broek, Brederode, Ramírez, Kramers, van der Kuip, Wildenborg, Faaij, and Turkenburg, 2009; Brunsvold, Jakobsen, Husebye, and Kalinin, 2011; Fimbres Weihs, Wiley, and Ho, 2011; Svensson, Odenberger, Johnsson, and Strömberg, 2004; Van den Broek, Ramírez, Groenenberg, Neele, Viebahn, Turkenburg, and Faaij, 2010; Strachan, Hoefnagels, Ramírez, Van den Broek, Fidje, Espegren, Seljom, Blesl, Kober, and Grohnheit, 2011; Bakken and von Streng Velken, 2008; Middleton, Kuby, and Bielicki, 2012; Alhajaj, MacDowell, and Shah, 2013).

A CCUS supply chain network also requires to screen utilization and sequestration sites from a large number of potential locations. For instance, analyzing the NATCARB data (National Energy Technology Laboratory, 2010a), we have screened 161 CO₂-EOR sites as potential utilization locations (Fig. 5), and 491 saline formations (Fig. 6) and 1837 unmineable coal areas (Fig. 7) for potential sequestration locations for the United States.

[Figure 5 about here.]

[Figure 6 about here.]

[Figure 7 about here.]

Finally, while studies (Odenberger, Kjärstad, and Johnsson, 2008; Kjärstad, Ramdani, Gomes, Rootzén, and Johnsson, 2011; Mikunda, van Deurzen, Seebregts, Kerssemakers, Tetteroo, and

Buit, 2011; Klok, Schreiner, Pagès-Bernaus, and Tomasgard, 2010; Strachan, Hoefnagels, Ramírez, Van den Broek, Fidje, Espegren, Seljom, Blesl, Kober, and Grohnheit, 2011; Bakken and von Streng Velken, 2008; Middleton and Bielicki, 2009b,a; Middleton, Kuby, and Bielicki, 2012; Kuby, Bielicki, and Middleton, 2011; Han and Lee, 2011; Han, Ryu, and Lee, 2012; Tan, Aviso, Bandyopadhyay, and Ng, 2012; van den Broek, Faaij, and Turkenburg, 2008; Zheng, Larson, Li, Liu, and Williams, 2010; Johnson and Ogden, 2011; Figueroa, Fout, Plasynski, McIlvried, and Srivastava, 2008) addressed regional CO₂ capture and sequestration and CO₂ pipeline networks (Morbee, Serpa, and Tzimas, 2012; McCoy and Rubin, 2008; Benson and Ogden, 2003), it remains an open question whether a nationwide CCUS deployment would be economically feasible or not. Diverse emission scenarios, difference in CO₂ emissions, different geographic locations, technological complexities, etc., preclude a simplistic study toward the design of a cost-effective CCUS. This implies that the design of CCUS supply chain network should consider decisions at multiple scales, starting from materials screening to process synthesis to technology selection to supply chain topology determination.

4 Multi-scale Framework for CO₂ Capture, Utilization, and Sequestration (CCUS and CCU)

In this work, introduce a multi-scale framework to answer the following questions:

- Q1. What is the best way to capture CO₂ from diverse emission sources?
- Q2. Can we capture CO₂ at reasonable costs?
- Q3. Can CCUS reduce the stationary CO₂ emissions by 50% in the United States?
- Q4. Can CCUS reduce the CO₂ emissions at reasonable costs?
- Q5. What are the current best technologies, processes and materials for CCUS in the United States?
- Q6. How to design an optimal CCU supply chain network?
- Q7. What is the maximum profit that we can gain from CO₂ utilization for EOR?
- Q8. How much is the reduction of stationary CO₂ emissions that can be achieved as net zero-cost in the United States?

[Figure 8 about here.]

The overall framework is illustrated in Fig. 8. The framework applies advanced modeling and optimization at material, process, technology and supply chain levels. Our hierarchical approach considers the following steps: materials screening, process optimization, process technology selection, materials screening and process optimization, materials screening and process optimization and technology selection, and supply chain optimization. Each step starts with the updated information from the previous steps. To this end, we acquire and utilize information starting from micro-scales (atomistic calculation and geometric characterization of materials) to meter-scale (process optimization and technology selection) to kilometer-scale (supply chain optimization). As we proceed, we apply a hierarchical *in silico* screening method to select candidate materials and optimize process conditions for CO₂ capture. We also develop explicit expressions for the investment and operating costs to determine the most cost-effective materials and processes when addressing diverse emission scenarios, based on detailed cost-based modeling and optimization studies. All the information related to CO₂ estimates from sources, cost estimates for technologies, and demand estimates for utilization and sequestration are used for the optimization of nationwide CCUS and CCU supply chain networks.

While several alternatives exist, we only consider the four leading technologies for CO₂ capture, namely absorption, membrane, pressure swing adsorption (PSA), and vacuum swing adsorption (VSA). We consider amine-based chemical absorption, zeolite-based physical adsorption of CO₂ for PSA and VSA, and polymeric membranes. All capture processes are considered to be modular in design so that they can be easily integrated with the sources. However, the complete integration of the capture processes with thousands of different sources is considered beyond the scope of this work. The CCUS costs are calculated without considering additional SO_x removal from flue gases.

The multi-scale and hierarchical framework for CCUS and CCU is discussed below in more detail.

4.1 *in silico* Materials Screening

In this step, we consider the screening of candidate materials for a single technology and single process. This first-cut screening is applied to reduce the overall search from hundreds of materials

to a list of selected candidate materials for more detailed analysis.

To illustrate, we have applied materials screening to screen zeolites based on different selectivity criteria for adsorption-based CO₂ capture. Our screening begins with the three-dimensional pore characterization method ZEOMICS (First, Gounaris, Wei, and Floudas, 2011). This method applies graph, geometry, and optimization algorithms to identify portals, channels, cages, and their connectivity from the crystallographic data for a material. This comprises a complete description of all void spaces within the microporous material that guest molecules can access. From a database of known silica zeolite structures (First, Gounaris, Wei, and Floudas, 2011) with pore characterizations, we calculate accessible volume, accessible surface area, pore limiting diameter (PLD), and largest cavity diameter (LCD) First et al. (2011). The potential sorbents are then screened for shape selectivity using the approach described in First et al. (First, Gounaris, and Floudas, 2013). This method predicts three-dimensional shape selectivity of zeolites by considering the energetic cost of transport through possible pathways. Depending on the energetic cost, E , of transport, a guest molecule may have free passage ($E = 0$), hindered diffusion (low E), or no passage (high E) (Gounaris, Floudas, and Wei, 2006a; Gounaris, Wei, and Floudas, 2006b; Gounaris, Wei, Floudas, Ranjan, and Tsapatsis, 2009). Considering a flue gas as a binary mixture of CO₂ and N₂, we compute the shape selectivity between CO₂ and N₂, $\gamma_{CO_2-N_2} \in [0, 1]$, as a measure of the degree to which the zeolite can separate one molecule from the other based on hindrance to molecular transport through the most dominant pathway. A value of $\gamma_{CO_2-N_2} \rightarrow 1$ indicates that CO₂ diffusion is severely hindered, while N₂ enjoys free passage, and a value of $\gamma_{CO_2-N_2} \rightarrow 0$ indicates that both molecules have similar energetic penalties to travel through the zeolite.

We also compute the size selectivity of CO₂ over N₂ inside zeolites, which takes into account the entire distribution of pore sizes. Size selectivity, S_{size} , is a measure of relative difference in accessible pore volume between CO₂ and N₂ (First, Gounaris, Wei, and Floudas, 2011). We select zeolites with $\gamma_{CO_2-N_2} > 0$ or $S_{size} > 0.15$ for further consideration, and generate CO₂ and N₂ adsorption isotherms for these structures at 25 °C to calculate adsorption selectivity, S_{ads} , defined as the ratio of the Henry’s constants for CO₂ and N₂. We filter out zeolites with $S_{ads} < 10$, and for the remaining structures we construct CO₂ and N₂ adsorption isotherms at four additional temperatures (50 °C, 75 °C, 100 °C, 125 °C), which are fit to a dual-site Langmuir model. These equilibrium isotherm data are generated using grand canonical Monte Carlo (GCMC) simulations,

as implemented in MCCCS Towhee, a Monte Carlo molecular simulation code.

In summary, we rank zeolites based on shape selectivity and size selectivity, and select the top structures to generate CO₂ and N₂ adsorption isotherms. We subsequently calculate the Henry constants and perform a new filtering step based on adsorption selectivity. The list of candidate zeolites that are screened based on shape, size and adsorption selectivities can be found in the Supplementary Material.

It should be noted that the shape, size and adsorption selectivity-based screening of zeolites and other materials do not provide any information on cost. While material-centric metrics are typically used for screening materials, we combine materials screening with process optimization as we proceed through the framework.

4.2 Process Optimization

The purpose of this step is to optimize the process configuration and find the optimal operating conditions of a capture process based on a specific technology and material. For example, we have designed and optimized a 4-step PSA process to find the optimal column dimensions, operating pressures and step durations to develop an adsorption-based technology. The overall optimization framework can be applied for any given zeolite. We have also optimized processes for absorption and membrane-based technologies for CO₂ capture and compression. Each process consists of the following sections: flue gas dehydration (if required), CO₂ capture, and CO₂ compression. While the feed flue gas is allowed to have variable CO₂ composition and flow rate, the captured CO₂ is at least 90% pure, and has a pressure of 150 bar. These requirements are set to capture injection-ready CO₂ for utilization and sequestration purposes. We also restrict the processes to capture at least 90% of the total CO₂ present in the flue gas to reduce CO₂ emissions. Our objective is to minimize the total cost of CO₂ capture and compression while satisfying the minimum purity and recovery specifications. Each process is optimized and energy-integrated to minimize the total cost which includes the annualized investment, operating and material costs.

The flue gas dehydration section is placed before the capture and compression sections. The modeling and optimization of these sections for different process technologies are discussed below:

4.2.1 Flue gas dehydration Processes

The adsorption (e.g., PSA, VSA), and membrane-based processes often fail to capture CO₂ effectively, if the flue gas is wet and is saturated with water, which is typical for most flue gases from power plants. We address this challenge by designing novel processes for flue gas dehydration. Water can be removed from a wet flue gas in several ways, namely refrigeration, compression and cooling, membrane separation, sorbent-based guard beds, and TEG (triethylene glycol) absorption (Hasan, Baliban, Elia, and Floudas, 2012a). While a detailed description of various alternatives for the flue gas dehydration is presented in the Supplementary Material, we discuss the most cost-effective dehydration process that we have developed for power plant flue gases. This process is based on tri-ethylene glycol (TEG)-based absorption of water from a mixture of gases containing CO₂, N₂, and O₂. The dehydration process is shown in Fig. 9. The flue gas is first cooled using a direct-contact cooler. Cooling reduces the water content to 5.5% in the flue gas, which is the composition of a saturated flue gas at 35 °C. This composition is reached irrespective of the initial water content which can be up to 15% at the flue gas temperature of about 55 °C. Once the flue gas is cooled, it is fed to the absorber where H₂O contacts the TEG solvent in counter-current flow. The dry gas leaves the absorption column from the top. The H₂O rich solvent from the bottom of the column is heated in a heater at 73 °C. It is then flashed and regenerated under vacuum at 0.04 bar. The regenerated solvent is recycled back to the absorption column after being cooled to 65 °C. The TEG-based flue gas dehydration reduces the water content of the flue gas to less than 0.1% at a cost of \$10.20 per ton of CO₂.

[Figure 9 about here.]

4.2.2 Absorption-based Processes

We have modeled, simulated and optimized amine-based chemical absorption processes for CO₂ capture (the detailed methodology can be found elsewhere (Hasan, Baliban, Elia, and Floudas, 2012a)). The absorber and stripper are modeled and optimized using detailed reaction scheme and kinetics, and vapor-liquid equilibrium (VLE) model. We use the rate-based modeling approach and select the RADFRAC model in Aspen Plus process simulator for both the absorber and stripper. Both columns have the same packing specifications. IMTP#40 type metal packing is used for

all stages. We use the VLE models as used by Aboudheir et al. (Aboudheir, Tontiwachwuthikul, Chakma, and Idem, 2003) for monoethanol amine (MEA), and by Bishnoi and Rochelle Bishnoi and Rochelle (2000) for Piperazine, respectively. The transport property models used in this work were selected based on the suggestion of Zhang et al. (Zhang, Chen, Chen, Plaza, Dugas, and Rochelle, 2009). These include the Clarke density model for electrolytes solutions with mixed solvents to calculate density, the Jones-Dole model for liquid solutions with electrolytes for viscosity, the Onsager-Samaras surface tension model for the liquid mixture surface tension, and the Riedel thermal conductivity model for the liquid mixture thermal conductivity. We use the Nernst-Hartley electrolyte model which is the default model in Aspen to calculate diffusivity, instead of the Wilke-Chang model.

Fig. 10 shows the completely heat integrated and optimized absorption-based process for CO₂ capture and compression. The feed flue gas is first compressed, and then cooled using a direct contact cooler before feeding to the absorber where CO₂ is reacted with the lean solvent. The solvent exiting the absorber column is loaded with CO₂ and pumped to the stripper pressure. The pumped solvent then exchanges heat with a number of process streams. The heat is supplied by the intercoolers for CO₂ compression and additional utility heaters. The heated solvent is then sent to the stripper where CO₂ is transferred to the gas phase. While the solvent is cooled and recycled back to the absorber, the gaseous CO₂ released from the stripper is cooled, partially condensed and flashed in a flash drum. The pure CO₂ stream is finally sent to the 6-stage compressor train which compresses CO₂ to 150 bar.

[Figure 10 about here.]

The entire process is simulated in Aspen Plus, while the heat integration is performed using a heat exchanger network synthesis (HENS) model (Hasan, Baliban, Elia, and Floudas, 2012a) solved in GAMS. The optimal matching between hot and cold streams is obtained through a three-stage heat exchanger network synthesis method. While the reboiler and the heater use low-pressure steam as hot utility, the cooler uses cooling water as cold utility. The aftercondenser of the stripper is completely heat integrated with the rich solvent coming from the absorber. The economizers placed between the absorber and stripper also do not require any utilities.

4.2.3 Membrane-based Processes

We model and optimize multi-stage membrane-based processes for CO₂ capture. We consider a multistage configuration of the process, since using only a single-stage cannot always achieve the desired CO₂ purity of 90% across the membrane unit. The process flow diagram (Hasan, Baliban, Elia, and Floudas, 2012a) of the membrane process is shown in Fig. 11. Each stage in the multistage membrane system has the same equipment layout and configuration. The feed gas is first dried using a triethylene glycol (TEG)-based absorption process. This reduces the water content of the feed flue gas to less than 0.1%. In each stage, the gas is first split into two substreams. While the first substream undergoes the separation process, the second substream is merely a bypass stream which allows the feed gas to bypass the entire stage. The first substream is sent to a compressor and then cooled to achieve the desired driving force (pressure) and temperature for separation. The compressed and cooled gas then enters the membrane module as the feed. CO₂-free clean gas is collected as retentate and vented to the atmosphere, whereas CO₂ is separated and extracted on the permeate side using a vacuum pump. The vacuum pump produces vacuum at the inlet while discharging CO₂ at the atmospheric pressure. The mixture gas after the pump outlet is then sent as the feed gas to the next stage. Once the product CO₂ comes out of the vacuum pump of the final stage at 1 bar pressure, it is compressed along with the stage bypass to 150 bar.

[Figure 11 about here.]

We use a nonlinear optimization (NLP) model for the optimization of the above described membrane process for CO₂ capture (the detailed NLP model can be found in Hasan et al. (Hasan, Baliban, Elia, and Floudas, 2012a)). Features of this model include multi-stage and multi-component separation of CO₂ from industrial flue gas and gas mixtures. For given feed gas characteristics (e.g., flow rate, composition, pressure, temperature, etc.), and membrane material properties (e.g., permeability, thickness), this model would provide the optimal feed pressure and temperature of each stage, total membrane area, stage permeate pressure, utility consumption level for the minimum total annualized cost (investment plus operating) of the overall process.

4.2.4 Adsorption-based Processes

To evaluate and optimize the ultimate performance of each selected zeolite during the *in silico* materials screening step, we have developed (i) a process configuration, (ii) an adsorption/desorption cycle, and (iii) a detailed adsorption model for the process. Adsorption is a cyclic process in which each column undergoes a number of operational steps. Even the simplest cycle requires at least two steps, namely adsorption and desorption, but for improved performance, additional steps are used. For each zeolite on the short list of remaining candidates, we optimize a process that supports both pressure-swing adsorption (PSA) and vacuum-swing adsorption (VSA) modes. PSA and VSA are cyclic processes in which each column undergoes a number of operational steps.

We consider the process configuration as illustrated in Fig. 12a for both PSA and VSA-based CO₂ capture and compression. We use a 4-step adsorption cycle (Hasan, First, and Floudas, 2013; Hasan, Baliban, Elia, and Floudas, 2012b) and can operate in either PSA or VSA mode. A compressor compresses the feed gas in PSA mode, while the feed gas enters the column at atmospheric pressure in VSA mode. Multiple columns packed with solid adsorbent are used. Each column undergoes a cycle consisting of four steps, which in order are (i) pressurization, (ii) adsorption, (iii) forward blowdown, and (iv) reverse evacuation or desorption. Each step is operated with a pressure profile and duration indicated in Figure 12(b). During pressurization, feed gas enters each column from one end (green lines) while keeping the other end closed. During adsorption, both ends are open (red lines) while CO₂ is adsorbed at the adsorption pressure (P_{ads}) and N₂ is rejected as clean gas. During blowdown, additional N₂ is purged (blue lines) at the blowdown pressure (P_{bd}) using a vacuum pump. During evacuation, CO₂ is extracted (yellow lines) at the evacuation pressure (P_{evac}) using a second vacuum pump. Extracted CO₂ is compressed to 150 bar using a 6-stage compressor train with intercooling. Starting from an initial bed condition, the process undergoes a transient state for a number of cycles before reaching cyclic steady state.

[Figure 12 about here.]

The minimum cost of capture and compression is obtained by a detailed nonlinear algebraic and partial differential equation (NAPDE) model which is described in detail in the Supplementary Material. The PSA/VSA process includes 7 major independent variables, namely column length (L), adsorption pressure (P_{ads}), blowdown pressure (P_{bd}), evacuation pressure (P_{evac}), and the step

durations for adsorption (t_{ads}), blowdown (t_{bd}), and evacuation (t_{evac}). The pressurization time (t_{pr}) is fixed and the total number of columns is calculated using a procedure outlined in Hasan et al. (Hasan, Baliban, Elia, and Floudas, 2012b).

Coupled and nonlinear partial differential equations and constraints, which describe the mass and energy balances and fluid flow through porous media, make the NAPDE model complex and difficult to solve, even for fixed conditions. We use a robust and automated approach (Hasan, First, and Floudas, 2013; First, Hasan, and Floudas, 2014) that includes (i) solving the original NAPDE model at several fixed conditions to generate input-output data (samples), (ii) developing a Kriging-based surrogate model using the samples, (iii) optimizing the surrogate model for subsequent sampling, and (iv) repeating steps (ii) and (iii) until convergence to an optimal solution. The details of the Kriging-based grey-box optimization approach is also presented in the Supplementary Material.

4.3 Process Technology Selection

The purpose of this step is to provide a quantitative measure to select the most cost-effective CO₂ capture technology based on commercially available materials. The process optimization studies described in the previous sections provides a foundation for the cost-based selection of processes and technologies for CO₂ capture and compression. To enable this for a range of flue gas CO₂ compositions and flow rates from all types of stationary sources, we perform extensive process optimization studies considering different feed flue gas CO₂ compositions and flow rates. We obtain the optimum investment and operating costs for absorption, membrane, and adsorption technologies. In terms of processes, we select amine-based chemical process for absorption, multi-stage process for membrane, and both PSA and VSA processes for adsorption-based technologies. For each process, we use the most popular or commercially available material. We select MEA (monoethanol amine) as the solvent material for the chemical absorption process, FSC PVAm as the membrane material at each stage for the multi-stage membrane process, and zeolite 13X for both the PSA and VSA processes to capture CO₂. Each process includes capture of at least 90% of CO₂ from the dehydrated feed and compression to at least 90% pure CO₂ for utilization or sequestration at 150 bar. We optimize each process and obtain the costs for feed CO₂ compositions

ranging 1–70% CO₂, and at four different flue gas flow rates (0.1, 1, 5 and 10 kmol/s). These ranges cover the flue gas characteristics of most of the stationary CO₂ sources in the United States.

[Figure 13 about here.]

The results are shown in Fig. 13. The total cost (\$/ton of CO₂ captured and compressed) decreases with CO₂ composition in the feed flue gas. MEA-based absorption is the technology of choice when the flue gas contains very dilute amount of CO₂ (e.g., flue gas from natural gas fired power plants). However, at higher compositions, the costs for the absorption-based process are high compared to those for the other three processes. For chemical industries for which the emission scenarios are different and the CO₂ compositions are higher than those of the power plants, adsorption-based processes are more economical. Our results suggest that adsorption-based PSA and VSA processes are economically feasible options for CO₂ capture at moderate and high CO₂ compositions.

For a fixed CO₂ composition, the total cost decreases with increasing feed flue gas flow rate. Similarly, costs are higher at low feed flow rates. Interestingly, cost increases more sharply with decreasing feed flow rate at low CO₂ compositions. This highlights the benefits of economies of scale.

4.4 Materials Screening and Process Optimization

The cost-based comparison between the absorption-, membrane-, PSA-, and VSA-based CO₂ capture technologies presented in the previous section suggests that the flue gas CO₂ composition and flow rate significantly effect the CO₂ capture and compression costs. However, the costs would change if we change the material for each technology. Therefore, it is important that we simultaneously identify the top materials and optimize processes to reduce the overall cost.

To this end, we describe the methodology that we have developed to perform material screening and process optimization in tandem for adsorption-based CO₂ capture using PSA and VSA processes with zeolites as materials. We use a novel and hierarchical computational framework (shown in Fig. 14) that combines the steps involving *in silico* materials screening and process optimization to efficiently screen a large database of zeolites, optimize the PSA and VSA processes, and generate a rank-ordered of list of the top zeolites based on cost. From the zeolite database

ZEOMICS, we first identify potential zeolites based on their pore sizes using a three dimensional pore characterization method. Starting from ZEOMICS, the three materials-centric metrics (shape, size, and adsorption selectivities) provide 30 zeolites for which we apply detailed process optimization. After optimizing the PSA/VSA process for these 30 selected zeolites, we find that all but 4 zeolites satisfy both purity and recovery requirements, showing the effectiveness of our *in silico* screening approach to select zeolites that can satisfy the systems-level requirements of CCUS. The 26 zeolites (Fig. 15) that satisfy the purity and recovery constraints, have a combined cost of feed dehydration, CO₂ capture and compression that ranges from \$33.83 to \$51.65 for each ton of CO₂ captured. The costs include annualized investment, operating and material costs, and are inclusive of the cost of feed dehydration, which uniformly adds \$10.20/ton to the total cost. The top three zeolites for CO₂ capture include AHT, MVY and WEI (Hasan, First, and Floudas, 2013).

[Figure 14 about here.]

[Figure 15 about here.]

4.5 Materials Screening, Process Optimization, and Technology Selection

With the capability to optimize different processes for different materials, we now consider multiple materials, processes and technologies for comparison. We obtain optimum investment and operating costs for absorption, membrane, PSA and VSA processes. Aqueous MEA (30 wt%) and piperazine (40 wt% PZ) are selected as the solvents for the absorption technology. MEA is selected since it is the most commercially used solvent for CO₂ capture. Piperazine (PZ) is selected due to its energy efficient CO₂ capture and stable thermal performance (Rochelle, Chen, Freeman, Van Wagener, Xu, and Voice, 2011). POE1 and POE2 are screened based on CO₂/N₂ selectivities from more than 380 polymeric membranes (Powell and Qiao, 2006). Poly-vinylamine (FSC PVAm) is a fixed-site-carrier membrane that is selected because of its high CO₂ permeance (Hägg and Lindbråthen, 2005). We consider four zeolites (13X, AHT, MVY, and WEI) for PSA and VSA. 13X is the most widely used zeolites for CO₂ capture with experimental validation (Xiao, Zhang, Webley, Li, Singh, and Todd, 2008). AHT, MVY and WEI are identified in the previous step (Hasan, First, and Floudas, 2013) as the top zeolites for CO₂ capture based on cost. For each material, the capture processes are modeled and optimized.

Using the cost data from the rigorous optimization, we develop input-output models to express the investment and operating costs (Hasan, Baliban, Elia, and Floudas, 2012a,b; Hasan, Boukouvala, First, and Floudas, 2014). Specifically, we develop the following expressions for the investment (IC) and operating costs (OC) as functions of feed CO_2 composition (x_{CO_2}) and feed flow rate (F):

$$IC = \alpha + (\beta x_{CO_2}^n + \gamma) F^m \quad (1)$$

$$OC = \alpha' + (\beta' x_{CO_2}^{n'} + \gamma') F^{m'} \quad (2)$$

where, α , β , γ , n and m are model parameters. α , β , γ , n , and m for the absorption, membrane, PSA and VSA-based technologies and for different materials are estimated based on optimized investment and operating costs, using the maximum likelihood parameter estimation for the best fit. Their values are provided in the Supplementary Material. These cost expressions can be combined and used for the cost-based comparison of technologies and materials for CCUS. To illustrate, we present the combined investment and operating costs (converted to \$/ton) in Fig. 16 for the top technologies (absorption, membrane, PSA and VSA) and materials (MEA and PZ for absorption, PVAm, POE1 and POE2 for membrane, and 13X, AHT, MVY and WEI for PSA and VSA).

[Figure 16 about here.]

Results show that there is no single material or process that is always the most cost-effective for CO_2 capture and compression. For instance, while MEA-based absorption has the lowest cost at low CO_2 compositions, MVY-based PSA and WEI-based PSA are among the most cost-effective materials at moderate and high CO_2 compositions. Also, not all technologies are feasible (i.e., capable of capturing with at least 90% purity and recovery) for all CO_2 compositions. Only MEA and PZ based absorption technology can capture CO_2 at CO_2 compositions lower than 5%. AHT, MVY and WEI are feasible if the feed CO_2 compositions are 5% or more for PSA, and 10% or more for VSA. 13X is feasible only at 10% or higher CO_2 compositions.

Therefore, it is critical to consider the variabilities in feed CO_2 compositions, flue gas flow rates, technologies used, process configurations considered, and materials employed for capturing

CO₂ from different sources. Since a CCUS supply chain network involves different CO₂ sources, we must consider all plausible alternatives and select appropriate technology for each source, if we want to design a CCUS network to manage CO₂ at reasonable cost. The explicit cost expressions that we have developed are efficient and, provide an accurate representation of the costs involved for various technologies. These expressions provide a critical information derived from molecular-scale and process-scale modeling and optimization, and hence provide a link for the multi-scale analysis of CCUS supply chain networks.

4.6 CCUS and CCU Supply Chain Network Optimization

The objective is to minimize the overall cost of the CCUS supply chain network. The overall CCUS cost includes the total CCUS costs minus the overall CCUS revenue:

$$\begin{aligned}
 \text{Net CCUS network Expenditure} &= [\text{total CCUS costs}] - [\text{total CCUS revenue}] \\
 &= [\text{total dehydration cost}] \\
 &\quad + [\text{total capture \& compression cost}] \\
 &\quad + [\text{total transportation cost}] \\
 &\quad + [\text{total injection cost}] \\
 &\quad - [\text{total revenue from utilization (CO}_2\text{-EOR)}]
 \end{aligned}$$

The above can be calculated in dollars per year, or dollars per ton of CO₂. The dehydration cost for adsorption and membrane technologies is considered to be \$10.2 per ton of CO₂. The total capture and compression costs depend on the selection of material, process and technology. These costs are calculated based on the explicit expressions (Eqs. 1–2) for the investment and operating costs that we described in the previous section. The transportation cost includes the levelized investment and operating costs of the CO₂ pipelines, and are calculated using a piecewise linearization of the pipeline equations by Ogden (Ogden, 2004), as functions of the CO₂ flow rates and transportation

distances. The levelized piping cost is given by:

$$LC_{piping,j,k}(\$/\text{yr}) = (CCR + OM_{piping}) \left\{ C_{base,piping} \left(\frac{M_{transport,j,k}}{M_{base}} \right)^\eta \right\} \left\{ L_{pipe,j,k} \times 10^3 \left(\frac{L_{pipe,j,k}}{L_{base}} \right)^\nu \right\} \quad (3)$$

The above calculates the total levelized cost for a given CO₂ flow rate $M_{transport,j,k}$, and length of the pipeline $L_{pipe,j,k}$ from a location j to a location k . The description and the values of the parameters are given in the Supplementary Material. The length of a pipeline, $L_{pipe,j,k}$, depends on the geographic distance between a source and a utilization or sequestration site, and is calculated as follows:

$$L_{pipe,j,k} = \cos(Lat_j) \cdot \cos(Lat_k) \cdot \cos(Long_k - Long_j) + \sin(Lat_j) \cdot \sin(Lat_k) \quad (4)$$

where, Lat_j and Lat_k are latitudes, and $Long_j$ and $Long_k$ are longitudes of j and k , respectively.

The levelized cost of CO₂ injection for utilization/sequestration is given by (Ogden, 2004):

$$LC_{injection,j,k}(\$/\text{yr}) = (CCR + OM_{injection}) (m_1 d_{well,k} + m_2) n_{well,j,k} \quad (5)$$

where, $d_{well,k}$ is the depth of well k , CCR is the capital cost recovery and OM is the maintenance cost of an injection well. The parameters m_1 and m_2 are given in the Supplementary Material. The number of CO₂ injection wells ($n_{well,k}$) are calculated as follows:

$$n_{well,k} = \left\lceil \frac{\sum_j M_{transport,j,k}}{M_{well}^{max}} \right\rceil \quad (6)$$

where, M_{well}^{max} is the maximum injection capacity of a well.

The revenue from CO₂ utilization is earned by selling CO₂ to CO₂-EOR sites. We consider the CO₂ purchase cost for CO₂-EOR to be \$2/Mcf or \$38.5 per ton of CO₂, considering an oil price of \$70 per barrel (National Energy Technology Laboratory, 2010b).

With the aforementioned estimation of CCUS costs and revenues, the CCUS supply chain network problem can be represented using the following mixed inter programming (MIP) model:

$$MIN \quad \sum_{(i,k) \in OS^{CCUS}} (TC_{i,k} - TR_{i,k}) Y_{i,k} \quad (7)$$

$$s.t. \quad \sum_{k \in K} Y_{i,k} \leq 1 \quad \forall i \in I \quad (8)$$

$$\sum_{(i) \in I} (F_i R_i) Y_{i,k} \leq M_k^{\max} \quad \forall k \in K \quad (9)$$

$$\sum_{(i,k) \in OS^{CCUS}} (F_i R_i) Y_{i,k} \geq \epsilon^{\min} \sum_{i \in I} F_i \quad (10)$$

$$Y_{i,k} = \begin{cases} 1 & \text{if source } i \text{ supplies CO}_2 \text{ to sink } k \\ 0 & \text{otherwise} \end{cases}$$

$$\forall (i,k) \in OS^{CCUS}$$

where, $TC_{i,k}$ is the total cost that includes the costs of flue gas dehydration (\$10.2 per ton for membrane and adsorption technologies), CO₂ capture and compression (using Eqs. 1–2) from source i , CO₂ transportation from source i to utilization or sequestration site k (using Eq. 3), and CO₂ injection in site k (using Eq. 5). $TR_{i,k}$ is the total revenue from selling CO₂ to utilization site k after capturing from source i , and is calculated based on the total CO₂ utilization. F_i is the total CO₂ emission from source i . We only capture $F_i R_i$ (where $0 \leq R_i \leq 1$) amount of CO₂ from source i , if it is selected. R_i is the CO₂ recovery in the capture plant. In the most general case, R_i can vary between 0 and 1. However, in such a case, the supply chain model becomes a mixed-integer nonlinear optimization (MINLP) problem. In this paper, we consider $R_i = 0.90$. This means that if we select a source, 90% of the total available CO₂ is captured. This also reduces the model to a MIP model, and reflects the most realistic scenario. $\epsilon^{\min}$ is the minimum reduction of CO₂ emissions from stationary sources, and $M_k^{\max}$ is the maximum storage capacity of site k . The sum of all F_i is equal to the total CO₂ emissions from all sources. $Y_{i,k}$ is the 0-1 variable to select a specific source i with a capture plant equipped with the most cost-effective capture technology and material (these are known *a priori* based on the explicit cost expressions), and injection site k . The set OS^{CCUS} is created by listing the distances between sources and utilization/sequestration sites

which are smaller than 200 miles. This restriction on the maximum pipeline distance between a source and a sink reduces the problem combinatorics and makes the CCUS supply chain network model computationally tractable.

For a CCU network, where we consider CO₂ utilization only for CO₂-EOR purposes in oil and gas reservoirs and no sequestration, the overall MIP model remains similar to that of the CCUS model, except that (i) the objective is changed from minimizing cost to maximizing profit (revenue–cost) from utilizing CO₂, and (ii) there is no restriction on the minimum CO₂ reduction level. Therefore, the CCU supply chain network problem is represented by the following MIP model:

$$\text{MAX} \quad \sum_{(i,k) \in OS^{CCU}} (TR_{i,k} - TC_{i,k}) Y_{i,k} \quad (11)$$

$$\text{s.t.} \quad \sum_{k \in K} Y_{i,k} \leq 1 \quad \forall i \in I \quad (12)$$

$$\sum_{(i) \in I} (F_i R_i) Y_{i,k} \leq M_k^{\max} \quad \forall k \in K \quad (13)$$

$$Y_{i,k} = \begin{cases} 1 & \text{if source } i \text{ supplies CO}_2 \text{ to sink } k \\ 0 & \text{otherwise} \end{cases}$$

$$\forall (i,k) \in OS^{CCU}$$

5 Nationwide CCUS Supply Chain Networks

In this section, we present the results for proposed nationwide CCUS activities for the United States. We select the optimum topology using a mixed-integer (MIP) optimization model. The design decisions are: (i) how many sources and which sources to select, (ii) which technology-material pair to select for capture from each selected source, (iii) how many and which utilization sites to select, (iv) which sequestration sites to select, (v) how much to utilize/sequester in a selected site, and (vi) which source supplies CO₂ to which site. We also meet CO₂ supply and demand constraints, restrictions on transportation distances and achieve a prespecified CO₂ reduc-

tion level. We obtain the minimum overall CCUS cost which is defined as the sum of all capture and compression costs, transportation costs and injection costs, subtracted from the revenues from CO₂ utilization.

As mentioned earlier, we have identified 3317 stationary sources, 161 utilization sites, 491 saline formations, and 1837 unmineable coal areas, which can be considered for nationwide CCUS activities. If our primary goal is to achieve an environmental benefit through reducing the overall CO₂ emissions in the United States to a certain level (e.g., reduce nationwide stationary CO₂ emission by 50%), then we opt for designing a CCUS supply chain network with the goal to design it at minimum cost while achieving the desired level of CO₂ reduction. If our goal is, on the other hand, to utilize CO₂ for economic benefit, then we develop a CCU supply chain network to maximize the overall profit. For both CCUS and CCU networks, we consider 13 technology-material pairs (MEA and PZ-based absorption; FSC PVAm, POE1 and POE2-based membrane; 13X, AHT, MVY and WEI-based PSA; 13X, AHT, MVY and WEI-based VSA). Therefore, there exist $3317 \times (161 + 491 + 1837) \times 13 = 107,328,169$ distinct CCUS scenarios with different supply chain network topologies. We reduce the complexity by restricting the maximum distance of a single pipeline to be 200 miles. This reduces the number of binary variables for CCUS network to 2,379,249. The CCUS supply chain network model has 5,414 linear constraints. Both the CCUS and CCU models are solved to optimality using GAMS/CPLEX 12.4.0.1 on a single computer containing 8 Intel Xeon 2.83 GHz processors.

The optimal nationwide CCUS supply chain network (Fig. 17) is designed to manage 50% of the *stationary* emissions, which is about 1.534 Gt of CO₂ per year. The total cost (TC) is 58.093 billion dollars per year. This includes the annual flue gas dehydration costs (15.677 billion dollars), CO₂ capture and compression costs (38.238 billion dollars), CO₂ transportation costs (2.551 billion dollars), and CO₂ injection costs (1.627 billion dollars). While dehydration, capture and compression, transportation and injection adds to the overall cost, CO₂ utilization generates a total revenue of 3.430 billion dollars per year. Therefore, the overall annual CCUS cost is 54.663 billion dollars. When calculated per ton basis, this corresponds to \$35.63 for each ton of CO₂ *captured* (Fig. 18A-D shows the breakdown of costs for the selected sources). This is a significant reduction in CCUS cost. The selected sources are tabulated in the Supplementary Material.

[Figure 17 about here.]

Among the 444 selected sources, 331 sources use MVY-based PSA process, 103 sources use WEI-based PSA process, only 10 sources use MEA-based absorption process, and no membrane-based capture processes are selected (Fig. 18(A)). Most selected sources have moderate to high CO₂ compositions, for which MVY and WEI-based PSA processes are among the most cost-effective (Hasan, First, and Floudas, 2013). While MEA-based absorption is the most popular technology for CO₂ capture, it is not always cost-effective. The selected MEA-based absorption processes incur the highest costs in the CCUS network. To compare, when we consider a CCUS network using only MEA-based absorption (results provided in the Supplementary Material), the overall CCUS cost is \$46.15/ton, which is 29.5% higher than the most general case of \$35.63/ton. The overall results suggest that the selection of the most cost-effective materials is critical to reduce CCUS cost. The selected sources in the optimal CCUS network also have wide ranges of CO₂ flow rates and costs. The range of CO₂ capture and compression (Fig. 18(A)) is \$14.5–58.1/ton of CO₂, which highlights the diversity of selected sources. Among the selected sources, 150 sources supply CO₂ for CO₂ utilization for CO₂-EOR. CO₂ from 286 sources are sequestered in saline formations, and only 8 sources supply CO₂ for sequestration in unmineable coal areas. In the optimal CCUS network, coal-based power plants are by far the largest suppliers of CO₂, supplying 86.48% of the total CO₂ captured from 230 selected plants. These correspond to an annual management of more than 1.326 Gt of CO₂.

[Figure 18 about here.]

Similar observations are made for 60% (1.841 Gt/yr) and 80% (2.454 Gt/yr) reduction of CO₂ from the stationary sources (while the results are summarized in Table 2, detailed results for 60% and 80% reduction are presented in the Supplementary Material. The overall CCUS network expenditure is \$71.509 billion dollars and \$106.616 billion dollars per year for 60% and 80% CO₂ reduction levels, respectively. The networks also generate annual revenue of 3.52 billion and 3.60 billion dollars, respectively, through CO₂ utilization.

[Figure 19 about here.]

The breakdown of costs and revenues for different levels of CO₂ reduction (50%, 60% and 80%) are shown in Fig. 19. It is evident that the overall CCUS cost is dominated by the costs of

dehydration, capture and compression for all cases. The overall CCUS costs are \$35.63, \$36.93 and \$43.44 per ton of CO₂ managed (captured, transported and utilized or sequestered) for 50%, 60% and 80% CO₂ reduction, respectively. Clearly, the overall CCUS cost increases with minimum CO₂ reduction. This is an interesting trade-off between the CCUS cost and CO₂ reduction, which highlights the fact that CCUS costs should be assessed based on the targeted reduction in CO₂ emissions. Therefore, a major decision even before the CCUS implementation is the level of desired CO₂ reduction that one wants to achieve.

[Table 2 about here.]

[Table 3 about here.]

Our results suggest that it is possible to reduce more than 50% of the total stationary CO₂ emissions in the United States using a nationwide CCUS network which would cost \$35.63–43.44 for each ton of CO₂ captured, utilized and sequestered. A pulverized coal power plant emits 0.736–0.811 ton of CO₂ for each 1000 kWh of electricity generated (Rubin, Chen, and Rao, 2007). To this end, an overall CCUS cost of \$35.63/ton of CO₂ would correspond to increase in electricity price of \$0.026–0.029 per kWh. Considering a base electricity price of \$0.07/kWh, the electricity price inclusive of CCUS cost does not exceed \$0.10 per kWh. Furthermore, all our results correspond to the capture and isolation through utilization and sequestration of at least 90% CO₂ from the source. Therefore, if implemented, each source would mitigate 90% of their current CO₂ emissions. This would enable the continued consumption of fossil fuels without emitting excess CO₂ from energy related activities.

6 Nationwide CCU Supply Chain Networks

We design a nationwide CO₂ capture and utilization (CCU) supply chain network by considering the following:

Problem: Design a CCU network that will supply CO₂ through capture, compression and transportation of anthropogenic CO₂ from major stationary sources in the United States.

Objective: Maximize the overall profit from selling the anthropogenic CO₂ for enhanced oil recovery purposes. Profit is defined as the difference between the revenue obtained from CO₂ sales and the overall cost of CO₂ capture, compression and transportation.

Given: All stationary source locations and the maximum CO₂ flow rates available from each source, locations and maximum CO₂ demands of all potential utilization sites, potential capture technologies, processes, and materials, CO₂ selling price (\$38.5 per ton) and CO₂ capture, compression and transportation costs obtained as described in the previous sections.

We have considered 3317 sources, 13 technological (technology, process, and material) alternatives, and 161 utilization sites. Each of the 3317 sources can select any of the 13 technological alternatives for CO₂ capture, and can supply to any of the 161 utilization sites. Therefore, a total of 6,942,481 alternative topologies exist for the CCU supply chain network. If we do not allow any pipeline of more than 200 miles between a source and a utilization site, then the total possible topologies reduce to 47,128.

We apply the MIP optimization (Eqs.11–13) to obtain the most profitable topology of the CCU supply chain network. The MIP model includes the objective function, 2890 constraints, 47,128 binary variables, and 176,941 non-zero elements. The model is solved to optimality using GAMS/CPLEX 12.4.0.1.

[Figure 20 about here.]

The optimal CCU supply chain network is shown in Fig. 20, and the major results are tabulated in Table 4. The network includes 63 sources selected from 3,317 sources, and 45 selected sites from a pool of 161 EOR sites. The network is designed to capture and transport 60,162,292 tons of CO₂ every year. While this amount is only about 2% of the total stationary CO₂ emissions in the United States, the utilization of this CO₂ generates an overall annual profit of \$555,419,217. This corresponds to an overall profit of \$9.23 for each ton of CO₂ captured, compressed and transported. Therefore, our CCU network is economically attractive. Considering a typical oil recovery of 2.5 barrels for each ton of CO₂ injected and stored (Godec, Kuuskraa, and Dipietro, 2013), a nationwide CO₂-EOR of 60.16 Mt/yr would additionally produce more than 150 million barrels of oil annually. Assuming an oil price of \$70/barrel, it would mean that it would be possible to produce annually about 10.5 billion dollars worth of oil using a nationwide CCU supply chain. While CO₂

capture is alone currently not economically viable, optimal incorporation of CO₂ utilization with capture can potentially increase the possibility of industrial scale deployment of CCU and CCUS technologies.

[Table 4 about here.]

It is important to note that the optimal CCU network maximized the total profit, and at maximum total profit we do not require to utilize all 161 utilization sites. While maximizing the utilization opportunities increases the revenue, we find that the overall profit does not increase if we consider more than the selected 45 utilization sites. An increase in CO₂ utilization or CO₂ flow rate also increases the network costs. More CO₂ needs to be captured at higher costs and transported to longer distances for any additional utilization. Therefore, the CCU network is designed by optimally considering the trade-offs between the utilization revenues and costs.

It is also important to note that when we do not have any restriction on the minimum CO₂ capture or reduction level, we use the total profit (\$/yr) as the objective function. If we use average profit (\$/ton), then the resultant CCU network will only consider very few sources for which the costs are the lowest, since the revenue per ton of CO₂ is fixed at \$38.5.

All 63 selected sources in the CCU supply chain network use adsorption-based PSA process for CO₂ capture. Only two materials, namely MVY and WEI (both are zeolites), are selected. 34 sources use MVY, 29 sources use WEI as the material for CO₂ capture. The total CCU costs of these sources are shown in Fig. 21 where blue and red colors represent the sources with costs associated with PSA processes using MVY and WEI, respectively.

CCU costs include the costs of CO₂ capture and compression, CO₂ transportation, and CO₂ injection. The CO₂ capture and compression costs vary from source to source (\$15.13–\$34.02 per ton). This highlights that sources with different CO₂ compositions and flow rates have been selected in the optimal network. In fact, 27 cement plants (47% CO₂), 16 gas processing plants (25% CO₂), 11 coal-fired power plants (14% CO₂), 6 iron & steel plants (corex, 24% CO₂), 2 sugar plants (47% CO₂), and 1 ethanol plant (13% CO₂) are selected for CO₂ capture and supply to the utilization sites. Hence, sources with high CO₂ compositions in the feed flue gases are preferred, since these correspond to reduced capture and compression costs. However, not all sources with high CO₂ compositions are selected. The selection also depends on the distance

between the sources and utilization sites. If sources with low capture costs are excluded, then it is because either the sources are not feasible in terms of CO₂ supply or the overall costs are high when capture and transport are considered together. The levelized transportation costs vary from \$0.17–\$20.04 per ton. This variation is mainly due to the distances (4.95 – 194.3 miles) between the selected sources and utilization sites. The CO₂ injections in the EOR sites also vary significantly (\$0.93–7.53 per ton). Different CO₂ flow rates and different number of CO₂ injection wells are the two primary reasons behind this variability.

The overall CCU costs vary from \$21.9–\$38.2 per ton of captured CO₂. Since the selling price of CO₂ is set to be \$38.5 per ton, all 63 sources make profit. While these profits vary from \$0.3–\$17.6 per ton of CO₂ for individual sources, the average profit is equal to the overall CCU profit, which is \$9.23 per ton of CO₂.

[Figure 21 about here.]

7 Zero-cost CCU Network

When we consider a minimum CO₂ reduction level, the CO₂ supply chain network is dominated by the costs. For instance, our optimal CCUS supply chain network for a 50% reduction in the nationwide stationary CO₂ emissions requires \$35.63 for every ton of CO₂ captured. Although utilization of CO₂ is allowed and an annual revenue of 3.43 billion dollars is generated, even such large-scale CO₂ utilization is not sufficient to balance the costs. We have also observed that the overall cost reduces with the CO₂ reduction targets. For example, the nationwide CCUS costs reduce from \$43.44 to \$36.93 to \$35.63, when the overall CO₂ reduction targets are reduced from 80% to 60% to 50%, respectively. However, we have also shown that the CCUS economics can be altered from loss to profit by considering CO₂ utilization without specifying any CO₂ reduction targets. For the optimal CCU supply chain network which is free from any restrictions on emission reduction, the CCU activities in fact generate \$9.3 dollars of profit per ton of CO₂, by capturing and transporting CO₂ at \$29.2 per ton and selling it at \$38.5 per ton). Moreover, this network reduces the overall stationary emissions by about 2%. While the goal of a CCUS with specified CO₂ reduction targets is primarily for environmental benefits, a CCU with profit maximization is primarily driven by economic benefits.

We are also interested in addressing this trade-off between the environmental and economic drivers for CO₂ capture. Specifically, we want to know the maximum level of CO₂ emissions reduction that can be targeted without any net cost. One way to obtain this is to perform a sensitivity analysis by optimizing costs at different reduction levels, until we get the supply chain network for which the cost is zero. To this end, we present a sensitivity study for CCU supply chain networks to finally obtain the CO₂ reduction level for which there is not net profit or cost. The results are obtained by using the same MIP model for CCU supply chain network optimization, except for the fact that we incorporate Eq. 14 in addition to Eqs.11–13.

$$\sum_{(i,k) \in OS^{CCUS}} (F_i R_i) Y_{i,k} = \epsilon \sum_{i \in I} F_i \quad (14)$$

where, ϵ is a parameter that denotes the CO₂ reduction level. In other words, ϵ is the fraction of the total stationary CO₂ emissions which is captured and utilized through the CCU supply chain network.

We specify the reduction level (ϵ) for each run and obtain the CCU network profit. We start with a CO₂ reduction level of 0.02 (i.e., 2% reduction for which the overall profit is maximized at \$9.3 dollars per ton). We increase the CO₂ reduction level at each run until the profit becomes zero. If we increase the minimum reduction level even higher, then the CCU network does not make any overall profits and starts incurring costs, as shown in Fig. 22. As we can see from Fig. 22, the overall CCU cost or profit is zero when the CO₂ reduction level is set to be 3.08%. This means that using the CCU supply chain network, it is possible to reduce the total stationary CO₂ emissions by 3.08% in the United States without any overall cost or profit. While we will need investment and operating costs to implement and operate the CCU supply chain network, the revenues earned from selling the captured CO₂ will exactly compensate those costs.

[Figure 22 about here.]

8 Conclusions

We presented a novel multi-scale framework to screen materials, processes and technologies for CO₂ capture for the design and optimization of cost-effective (i) CO₂ capture, utilization and stor-

age (CCUS), and (ii) CO₂ capture and utilization (CCU) supply chain networks. Through process synthesis and optimization, we explicitly showed how the investment and operating costs of absorption, membrane and adsorption-based post-combustion CO₂ capture technologies would change from plant to plant, and how the carbon mitigation effort could benefit by adopting industries which have large flow rates and diverse feed CO₂ compositions. We also demonstrated the effectiveness of combining atomistic, geometric and process understanding to screen materials and optimize processes in tandem for CO₂ separation. Using a hierarchical framework that combines material screening with process optimization, we uncovered top materials, optimized processes, and identified best technologies for CCUS and CCU activities for diverse emission scenarios. We designed nationwide CCUS and CCU supply chain networks by considering simultaneous selection of materials and processes for CO₂ capture, CO₂ sources, and utilization and sequestration sites. We also showed that at least 3% of the total stationary CO₂ emissions in the United States can be eliminated through CCU networks at zero net cost. It is found to be crucial to consider both the selection of materials and processes as integral parts of a large-scale CCUS and CCU networks. Although the CCUS and CCU methods are currently applied to design CO₂ networks to reduce stationary CO₂ in the United States, the method is general and, with the available database for CO₂ emission and storage estimates, can be applied to any country worldwide.

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Figure 1: A conceptual CCUS supply chain.

Figure 2: A network representation of a CCUS supply chain showing the nodes for sources, capture materials and processes, and utilization and sequestration sites.

Figure 3: Stationary sources of CO₂ in the United States, (a) U.S. map locating the stationary sources, where each circle represents a source. Sources are shown based on their locations and relative CO₂ emissions. Different colors are used to show the twenty source types, (b) number of sources belonging to twenty source types are shown using a pie-chart, (c) the histograms showing the distribution of sources based on the amount of CO₂ emitted annually. All CO₂ source data are collected and processed from the National Carbon Sequestration Database and Geographic Information System (NATCARB) and *Carbon Sequestration Atlas of the United States and Canada (Atlas III)* (National Energy Technology Laboratory, 2010a), as released by the DOE's NETL in November, 2010.

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Table 1: Some terminologies related to CCUS.

Term	Description
Source	Stationary emitter of CO ₂ (e.g., flue gases from power plant, cement, iron and steel, refinery, etc.)
Flue gas	CO ₂ containing gaseous mixture with other components such as N ₂ , H ₂ O, O ₂ , NO _x , SO _x , etc. Flue gases are produced from burning or consumption of fossil fuels.
CO ₂ capture	separation of CO ₂ from flue gas
CO ₂ utilization	Use of captured CO ₂ for economic benefit
CO ₂ -EOR	High pressure CO ₂ injection for enhanced oil recovery (EOR)
CO ₂ sequestration	Long-term geological storage of CO ₂ in underground saline formations or underground reservoirs
CCUS	Activity that includes CO ₂ capture, compression, transportation, utilization and sequestration of CO ₂ for both environmental and economic benefit
CCU	Activity that includes CO ₂ capture, compression, transportation, and utilization of CO ₂ for economic benefit
CCUS supply chain network	Network of multiple CO ₂ sources, capture plants, utilization sites and sequestration sites which are connected through CO ₂ pipelines
CCU supply chain network	Network of multiple CO ₂ sources, capture plants, and utilization sites which are connected through CO ₂ pipelines. No sequestration sites are present

Table 2: Nationwide CCUS costs.

	CO ₂ reduction level		
	50%	60%	80%
Total stationary CO ₂ emission (Gt/year)	3.068	3.068	3.068
Total CO ₂ captured (Gt/year)	1.534	1.841	2.454
Total flue gas dehydration cost (billion dollars/year)	15.677	18.815	25.080
Total capture & compression cost (billion dollars/year)	38.238	46.823	70.906
Total transportation cost (billion dollars/year)	2.551	3.870	7.636
Total injection cost (billion dollars/year)	1.627	2.001	2.994
Total CCUS cost (billion dollars/year)	58.093	71.509	106.616
Total CCUS revenue (billion dollars/year)	3.43	3.52	3.60
Overall CCUS cost (billion dollars/year)	54.663	67.989	103.016
Overall CCUS cost (dollars/ton CO ₂ captured)	35.634	36.930	43.445

Table 3: CO₂ capture materials and processes for nationwide CCUS supply chain.

Process	Material	CO ₂ reduction level		
		50%	60%	80%
Absorption	MEA	10	13	255
Absorption	PZ	0	2	33
PSA	MVY	331	499	1029
PSA	WEI	103	112	126
	total	444	626	1443

Table 4: Nationwide CCU supply chain network results.

description	value
Total CO ₂ sources	63
Total CO ₂ -EOR utilization sites	45
Total CO ₂ captured (Mt/year)	60.16
Total CCU revenue (billion dollars/year)	2.32
Total CCU cost (billion dollars/year)	1.76
Overall CCU profit (billion dollars/year)	0.555
Overall CCU profit (dollars/ton)	9.23