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# TECHNO-ECONOMIC MODELING OF $CO_2$ HYDRATE SLURRY FORMATION FOR CARBON SEQUESTRATION

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## ABSTRACT

*Significant carbon sequestration capacity (up to 10 Gigatons/yr) will be needed by 2050 to limit the Earth's temperature rise to < 1.5 ºC. The current worldwide capacity is ~40MT/yr, which highlights the need for the development of new and scalable sequestration approaches. One novel technology for long-term sequestration of $CO_2$ is the deposition of $CO_2$ hydrates (ice-like solids made with water and $CO_2$) on the seabed (under marine sediments or with artificial sealing). This involves rapid formation of $CO_2$ hydrate slurries in a bubble column reactor (BCR) by bubbling $CO_2$ gas at high flow rates in a BCR with the unreacted $CO_2$ being recirculated; this approach is being pioneered by the present research group. This study utilizes recent experimental results on ultra-fast hydrate formation to conduct a techno-economic analysis of the hydrate slurry-making process. All analysis is conducted for a 1 Megaton/yr sequestration project, which is expected to run for 30 years. Our analysis shows that the total cost of hydrate slurry production is $16.2/ton. Such projects would require an initial investment of $74M, and the energy requirement will be 641 MWh/day. Contributions of each part of the process to the total cost are identified. Our results show that gas recirculation in a BCR contributes minimally (0.04%) to the overall energy requirement. Furthermore, the cost of BCR is only 0.3% of the total investment cost. This suggests that a low conversion of gas into hydrates in each pass of the BCR is not detrimental from a techno-economic standpoint. The findings of this study set the stage for more detailed analysis of hydrates-based sequestration, which is essential to add this technology to the existing bank of established carbon sequestration solutions.*

Keywords: Carbon capture and sequestration, $CO_2$ hydrates, bubble column reactor, techno-economic modeling, hydrate slurry.

## 1. INTRODUCTION

Carbon dioxide ($CO_2$) levels have been steadily increasing in the atmosphere and oceans since the industrial revolution. The resulting increase in global temperatures is attributable in large part to anthropogenic emissions. The United Nations established the Paris Agreement in 2015 which targets limiting global temperature rise to <1.5 $^0$C above pre-industrial levels [1]. To achieve this, net-zero $CO_2$ emission targets need to be met by 2050. Carbon capture and sequestration (CCS) technologies are increasingly seen as essential to meet this target [2] in view of the slower pace of decarbonization. According to the Intergovernmental Panel on Climate Change (IPCC), up to 10 gigatons of $CO_2$/yr will need to be captured and sequestered by 2050 [3]. However, the existing global CCS capacity as of 2020 was only ~40MT/yr [2]. This highlights the urgency for the development of new and scalable CCS approaches.

Conventional carbon capture technology involves the use of chemical solvents like monoethanolamine (MEA), and there exists a wide range of deployable carbon capture solutions. On the other hand, there is only one carbon sequestration technology that is currently being deployed on a large scale [4]. This state-of-the-art sequestration technique is $CO_2$ injection in reservoirs or saline aquifers. However, suitable geology for this technology is scarce in many parts of world. Even in the US, despite decades of research, there exist only 2 Class VI reservoirs which allow injection for long-term sequestration [5]. Notably, despite more than 70 pending applications to the Environmental Protection Agency (EPA), no permits have been issued since 2020, highlighting the challenges and risk assessments needed for permitting [4], [6]. Key challenges with this technology include high monitoring costs associated with large areas and decades-long duration, high risks associated with leakage, risks of seismic activity and lack of appropriate geology in many large carbon emitting nations like China, India, Japan etc. [7], [8]. Alternative technologies include $CO_2$ mineralization in geological sites of

rocks like basalt [9], [10], microbial $CO_2$ sequestration [11], embedding $CO_2$ in concrete, chemicals, etc [12]. While these technologies seem promising, they all have significant challenges. Importantly no such technology by itself is adequate to address gigascale sequestration requirements. Overall, additional options for sequestration need to be urgently added to the basket of available solutions. This study looks into $CO_2$ hydrates-based carbon sequestration; this topic has gained significant interest in the scientific community over the last decade [7].

Clathrate hydrates were discovered in 1810 by Sir Humphrey Davy and are water-based crystalline solids consisting of a guest molecule (such as methane, $CO_2$, ethane, propane, etc.) trapped in a lattice of hydrogen-bonded water molecules [13]. $CO_2$ hydrates are synthesized at medium-pressures (>400psig) and low temperatures (<5 $^0$C), with 6 water molecules trapping 1 $CO_2$ molecule (on-average) [7], [13]. The potential geological sites where $CO_2$ hydrates would be thermodynamically stable (and where long-term sequestration would be technically viable) include subsea porous media under marine sediments, on the seabed with appropriate sealing, and deep under the permafrost regions [7], [14], [15].

Among these, sequestration on the seabed poses the least engineering challenges [4]. Seabed sequestration would involve compact hydrate formation with subsequent sealing to prevent dissociation of hydrates in seawater, as was seen in previous field tests [16], [17]. Additional key advantages include its high storage capacity wherein 1 Megaton $CO_2$ can be sequestered in a 1 $km^2$ area as a 3-meter thick layer of solid hydrate on the seabed [7]. This area requirement is at least 10 times smaller than the area footprint of geological injection projects. The higher density of hydrates (1040-1160 $kg/m^3$) compared to seawater further helps long-term sequestration from a technical standpoint. A key value proposition of hydrates-based sequestration is that hydrates can be made from relatively impure $CO_2$ streams (50-60% purity), thereby reducing overall CCS costs.

Nucleation and growth of $CO_2$ hydrates has been extensively studied in the last few decades due to the potential use of $CO_2$ hydrates for water desalination, gas storage, gas separation, gas transport, etc. [13], [18]–[21]. The economic feasibility of hydrate-based desalination has also been investigated and obtained to be only \$1.11/$m^3$ of water [22]. While hydrates-based sequestration is the focus of this work, hydrates-based carbon capture (HBCC) is also considered a promising technology and its techno-economic viability has been studied [23], [24]. The $CO_2$ intake capacity of HBCC is found to be much higher than conventional MEA based $CO_2$ absorption for the same amount of water [25]. HBCC was found to produce a hydrogen stream as pure as 92 vol% from syngas (60 vol% $H_2$, 40vol% $CO_2$) [26]. Aspen HYSYS was used to simulate a HBCC process with and without chemical promoters [24]. It was found that combination of membrane separation with HBCC with TBAB as a promoter had the least total energy consumption. The corresponding cost was obtained to be \$24.9/ton $CO_2$. The energy consumption for hydrates-based $CO_2$ separation from $CO_2/H_2$, along with subsequent transport and sequestration of slurries has also been studied [27]. It was found that gas separation accounted for 70% of total energy consumption.

While several studies on $CO_2$ hydrate formation exist, with some analyzing the techno-economics of hydrates-based carbon capture, there is no study on the techno-economics of hydrates-based carbon sequestration using captured $CO_2$. This work focuses techno-economic assessment of the hydrate slurry formation process. This slurry will then be transported and compacted for eventual sequestration; however, the techno-economics of those processes are not considered presently. Hydrate slurry formation in a bubble column reactor for a 1Megaton/yr project is considered as this is typical of industrial sequestration applications. Hydrate formation rates used for this model are based on recent experimental results from our group [28]. Capital expenditure (CAPEX) and operating expenditure (OPEX) are estimated for a 30-yr long project. The fraction of gas converted to hydrates in each pass (conversion fraction) is a key process parameter, which determines the extent of recirculation. The influence of conversion fraction on the techno-economics is analyzed. Contributions of various processes and components to the total cost of slurry production are quantified.

## 2. METHODOLOGY

### 2.1 Analysis domain

Figure 1 schematically shows the process for producing $CO_2$ hydrate slurries by bubbling $CO_2$ gas in stagnant water in a bubble column reactor. The dotted region indicates that domain that is analyzed in the present techno-economic model. A feed gas stream of pure $CO_2$ is supplied at $P_{Cs}$, $T_{Cs}$, equal to STP. The cost of $CO_2$ is not considered as the focus of this study is to evaluate sequestration costs only. The feed gas is first compressed to operating pressure ($P_o$) using the gas compressor and then cooled to operating temperature ($T_o$). The feed gas is mixed with the recirculated gas (exiting reactor at $P_o$, $T_o$) using a gas mixer, the cost of which is included as supplementary costs. The combined gas stream undergoes further compression and intercooling (represented by RGC in figure 1) so that it can be pumped into the bubble column reactor for hydrate formation. The corresponding inlet pressure is $P_i$, as represented by equation 1 shown below. The inlet gas emerging from the sparger with initial bubble diameter ($\eta$) equal to 0.25 mm is assumed to be distributed uniformly across the reactor.

$$P_i = P_o + \rho_g g H_w + 2\gamma/\eta \quad (1)$$

It was observed from experiments [28], [29] that hydrate shells that turn into hydrate slurry accumulate either on the gas-water interface or sink to the bottom of the reactor. Two slurry extraction pumps are therefore located at these two sites on the bubble column reactor. As mentioned previously, techno-economic analysis of these pumps is not included in the present study. The heat generated from hydrate formation, heat gain from walls of reactor and other heat gain processes need to be compensated for by the cooling jacket placed around the reactor

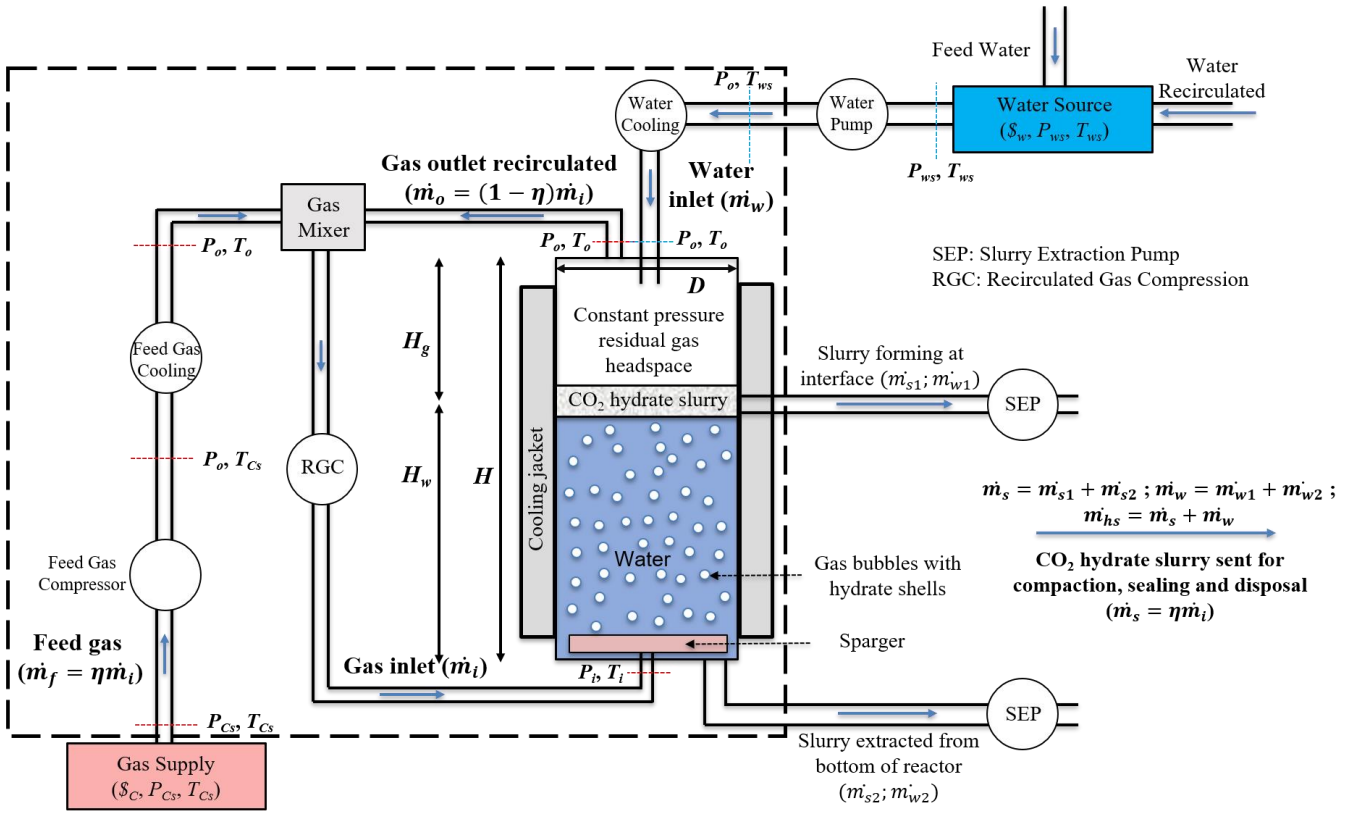


**FIGURE 1:** SCHEMATIC DEPICTION OF PRODUCTION OF $CO_2$ HYDRATE SLURRY (DOTTED LINES SHOW THE DOMAIN CONSIDERED IN THE PRESENT TECHNO-ECONOMIC ANALYSIS)

to maintain its operating temperature. To maintain continuous hydrate formation, a constant feedwater stream is provided to the reactor from the gas headspace. The feed water stream is seawater, our recent study has shown that we can get comparable hydrate formation rates with seawater as compared to freshwater. The use of seawater increases the techno-economic proposition of this concept as it eliminates the cost of procurement and transportation of freshwater (also $P_{ws}$, $T_{ws}$ is at STP). The feedwater is cooled to operating conditions before injection into the reactor. It is noted that the techno-economics associated with feed water pumping have are not included in this study.

The operating conditions and performance of bubble column reactor were based on our inhouse experimental results and assumed to linearly scale with size of the reactor. Our experiments [28] are conducted in a 650 ml reactor and yield very high hydrate formation rates of ~2kg/hr/lit of reactor volume, which would translate to 12.5 tons of $CO_2$ sequestered per year. Presently, we assume that a 1 Megaton/yr reactor can be linearly scaled from our current reactor. This corresponds to an inlet gas flow rate of 1.54*1e5 kg/hr and water flow rate of 7.83*1e5 kg/hr for a 12 m tall ($H_g$ = 2m, $H_w$ = 10m) and 3m diameter reactor operating at $P_o$ = 3.54 MPa and $T_o$ = 1$^0$C. The corresponding hydrate formation rates were based on 50-70% conversion fraction of the inlet gas. This study considers a wide range of conversion fraction (25-75%) for the same inlet gas flow rate. A 1MT/yr sequestration plant would correspond to a 75% conversion.

## 2.2 Analysis of costs associated with hydrate slurry production

The capital expenditure (CAPEX) in this study accounts for compressors, refrigeration, bubble column reactor, and supplementary costs (assumed 20% of total CAPEX). The operational expenditure (OPEX) is comprised of compression, intercooling, cooling cost (to cool feed gas, feed water and reactor), labor cost, maintenance costs, and other costs. Maintenance and other costs were assumed to be 2% and 1% of the total CAPEX respectively [30]. To evaluate the labor costs, it was assumed that 5 operator-equivalents would be employed at any given time to supervise the operation and an hourly wage of 30$/hr was used. In practice, the entire process will be automated. All of the energy requirements required for slurry production are assumed to be met by electricity. An electricity price of $30/MWh was used to estimate the OPEX. The sensitivity of OPEX to the price of electricity was considered for a range of $10-60/MWh.

The compression process to the operating pressure is carried out in three stages based on the guidelines for designing compressors [31], and an intercooler pressure loss of 10 psi is considered. The outlet temperature after each compression stage is estimated using an isentropic efficiency of $\eta_s$ = 0.83. After each compression stage, the temperature is brought back to inlet temperature. Further details on energetics of gas compression are reported in [31], [32]. The investment cost of compressors is estimated using the Douglas correlation [33] as shown in

equation 2 where M&S is the Marshall and Swift equipment cost index (2171.6 for 2020), *bhp* is the brake horsepower evaluated as described in [31], and $F_{c,c}$ is the correction factor equal to 1.15 for a centrifugal compressor [34].

$$Compressor\ Cost = \left(\frac{M\&S}{280}\right) 517.5(bhp)^{0.82}(2.11 + F_{c,c}) \quad (2)$$

The thickness of the bubble column reactor is estimated based on a working pressure of 10MPa (such that a factor of safety of 3 is established). This thickness is sufficient to withstand the hoop and longitudinal stresses, and was evaluated using the guidelines in ASME section VIII, paragraph UG-27. The investment cost of the reactor is estimated using equation 3 provided below [32], where M&S is the same as that of compressors, *D* and *H* are diameter and height of the reactor vessel, and $F_{c,v}$ is the correction factor for the reactor vessel and equals 9.175.

$$Reactor\ Cost = \left(\frac{M\&S}{280}\right) 101.9D^{1.066}H^{0.802}(2.18 + F_{c,v}) \quad (3)$$

To estimate the overall refrigeration costs, the refrigeration load or total cooling requirement is evaluated which includes cooling of the reactor, gas and water, cooling to compensate for exothermic hydrate formation (ΔH = 60kJ/mol), and other minor sources of heat gain by the reactor (which would slow down hydrate formation). The electrical energy requirement is then evaluated using a COP of 4. The capital cost of refrigeration is evaluated using a single-stage ammonia-based refrigeration system with a scaling factor of 0.6 with the refrigeration load as described in the work of Luyben [35].

To compare CAPEX and OPEX, the overall investment cost (total CAPEX) is converted into an annual CAPEX by using the capital recovery factor (CRF) as shown in equation 4. A discount rate of 10%, and a project period of 30 years is used to obtain a CRF of 0.106. The values for the life and discount rate are consistent with existing sequestration projects [4] and other established practices on valuations of energy and water-related projects [36].

$$Annual\ CAPEX\ (\$/ton) = \left(\frac{CRF * Total\ CAPEX}{Sequestration\ scale}\right) \quad (4)$$

Some key assumptions/limitations associated with the current modeling approach include:

- Costs associated with water pumping are not included.
- Challenges associated with materials compatibility with seawater have not been analyzed. The reactor is considered to be made of stainless steel. If seawater degradation is indeed an issue, water contacting surfaces will need to be coated with a seawater-resistant material or the reactor will need to be built of specialty alloy steels.
- Pressure head losses in heat exchangers for gas and water cooling are unaccounted for.
- Experimental results for hydrate formation in a 650 ml reactor are assumed to scale linearly with volume for the ~85m$^3$ reactor vessel used in the present model.

## 3. RESULTS AND DISCUSSIONS

### 3.1 Initial investment cost and energy requirement

Figure 2 shows the breakdown of initial investment costs for a 1Megaton/yr sequestration project (corresponding to 75% conversion fraction). The total investment cost was estimated to be $74M, with compressors account for the largest share (50.5%). This is due to the moderately-high pressure requirement and the multi-stage compression process. This contribution can further increase once water pumping costs are also included in the model. The cost of bubble column reactor (BCR) was negligible and only accounted for 0.3% of total initial investment. This suggests that if the conversion fraction from a single BCR is too low (say 25%), the pressurized gas can be passed to another BCR without significantly increasing the total costs. This will increase the overall conversion of the pressurized gas, and therefore the scale of sequestration being achieved.

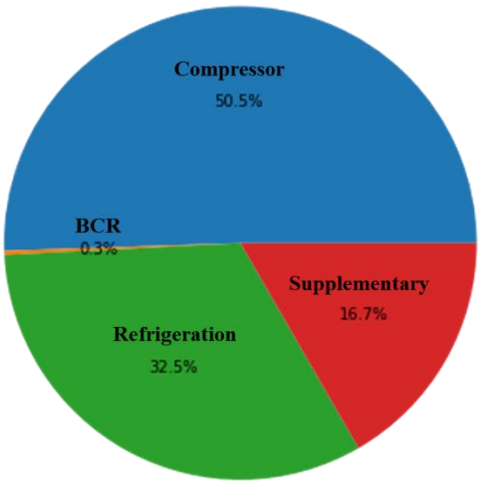


**FIGURE 2:** RELATIVE CONTRIBUTION OF INITIAL INVESTMENT COSTS (TOTAL INVESTMENT OF 74M$) FOR 1 MEGATON/YR $CO_2$ HYDRATE SLURRY PRODUCTION PROCESS.

The corresponding total electrical energy requirement for a 1Megaton/yr project is estimated to be 641MWh/day. It was observed that the compression associated with recirculation was only 0.04% of the total energy consumption. This clearly suggests that a low conversion factor does not hamper economics of the overall approach significantly. Low conversion will result in a lower scale of sequestration, or will need more water for the same extent of sequestration. The corresponding cost of water compression and cooling will be significant but they can also be avoided by removing excess water from hydrate slurry after

hydrate compaction and recirculating it back to the bubble column reactor for further slurry formation.

Feed gas cooling also has a low contribution to the overall energy requirement (0.91%), suggesting that atmospheric temperature conditions for feed $CO_2$ streams align well with a hydrates-based sequestration approach. Furthermore, even if the feed stream temperature was closer to 50$^0$C, the gas cooling requirement will only increase twofold and still remain insignificant compared to gas compression requirements. This suggests that if reducing the temperature increases hydrate formation more drastically than increasing the operating pressure, the latter would be more economically beneficial. This motivates further experimentation to study the change in hydrate formation rates versus lower temperatures and higher pressures.

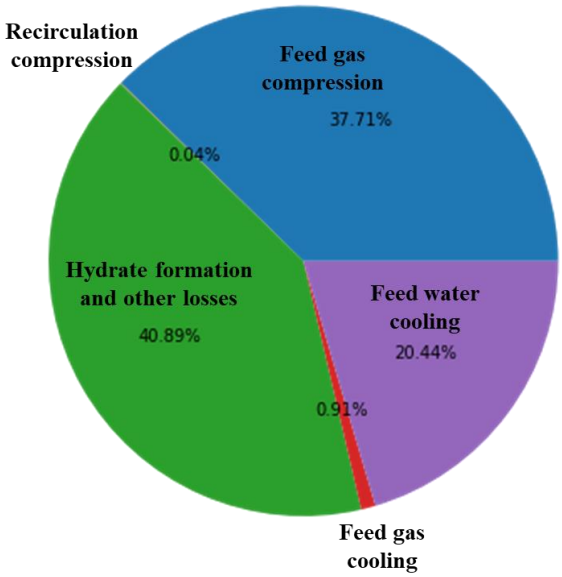


**FIGURE 3:** RELATIVE CONTRIBUTION OF ELECTRICAL ENERGY REQUIREMENT FOR EACH PROCESS. TOTAL ENERGY REQUIREMENT IS 641MWH/DAY FOR A 1 MT/YR HYDRATE SLURRY PRODUCTION PROCESS.

### 3.2 Influence of conversion factor and price of electricity on total cost

The influence of the conversion fraction on the scale and cost of slurry production is depicted in figure 4. The linear increase in sequestration rate with increasing conversion fraction was expected as per the modeling framework used in this work. The total cost was observed to decrease with increasing conversion factor. This is attributed to the fact that even though the feed gas flow rate is higher for the higher sequestration rate, the compressor cost increases with flow rate by a power of 0.82 [31], however the sequestration rate increases linearly. This results in total cost reduction from ~$26/ton to ~$16/ton for a conversion fraction change from 25% to 75%. The relative contribution of CAPEX and OPEX is observed to stay the same across the domain.

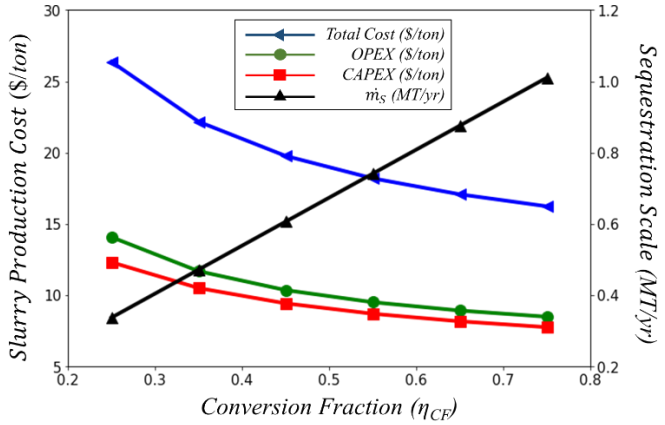


**FIGURE 4:** INFLUENCE OF $CO_2$ CONVERSION FRACTION (PER PASS) ON SCALE OF SEQUESTRATION AND COST OF SLURRY PRODUCTION.

Electricity prices vary significantly and depend on the location, time and the source of electricity (renewable, non-renewables). The influence of the price of electricity on slurry production costs was estimated for a 1Megaton/yr project and the results are shown in figure 5. The CAPEX remains constant since it is independent of electricity price. The OPEX increased linearly from ~$4/ton to ~$15/ton for an increase in electricity prices from $10/MWh to $60/MWh.

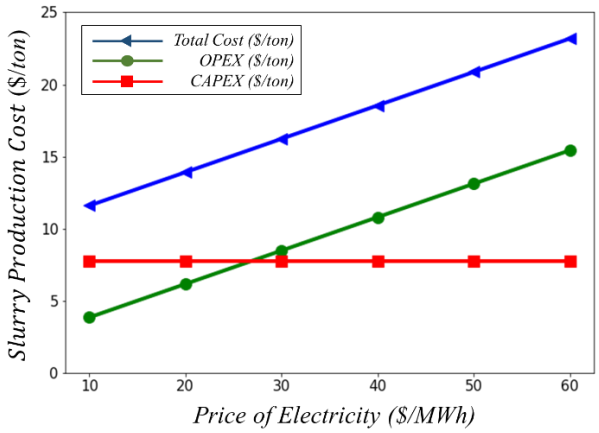


**FIGURE 5:** INFLUENCE OF PRICE OF ELECTRICITY ON THE COST OF PRODUCING HYDRATE SLURRIES.

### 3.3 Contribution of various components/processes to the total annualized cost (includes CAPEX and OPEX)

The relative contribution of each cost category on the total cost for a 1Megaton/yr project is depicted in figure 6. The corresponding slurry production cost for eventual sequestration is $16.2/ton. As reported by U.S. Department of Energy, the sequestration cost for a reservoir injection project lies in the range of $8-20/ton based on the plant location and basin [37]. The cost of slurry compaction, sealing and disposal on the seabed would need to be added to the slurry production cost to get the total cost for hydrates-based sequestration on the seabed, and it

is expected to be close to the range reported for reservoir injection projects. The OPEX:CAPEX ratio was 52.3%:47.7%. The largest component of CAPEX was from compressors (24.1% of total), while the largest component of OPEX was cooling (26.7% of total) which included gas cooling, water cooling, reactor cooling, and cooling to compensate exothermic hydrate formation and other heat gain mechanisms. The contribution of compression to OPEX is expected to increase when water pumping is included, despite which it contributed to 15% of the total cost.

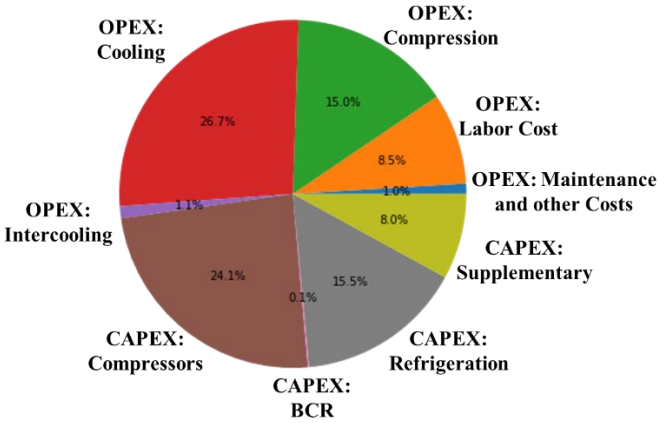


**FIGURE 6:** CONTRIBUTION OF VARIOUS COMPONENTS/PROCESSES TO THE TOTAL COST ($16.2/TON) OF A 1MEGATON/YR HYDRATE SLURRY PRODUCTION PROJECT.

## 4. CONCLUSIONS

A techno-economic model (based on inhouse experimental results) to assess $CO_2$ hydrate slurry formation in bubble column reactors is developed; this targets hydrates-based carbon sequestration applications. For a 30-yr long 1Megaton/yr project, the total cost of slurry production is $16.2/ton, with an initial investment cost of $74M, and a daily energy requirement of 641 MWh. The energy requirement associated with recirculation of $CO_2$ is minimal (0.04%), suggesting that operation at low conversion fractions is a viable option. Dominant contributions to the total cost come from compression (39.1%) and refrigeration (43.3%), with the costs actually associated with the reactor being low (~0.1%). Overall, this study sets the stage for further detailed studies on hydrates-based sequestration of carbon on the seabed.

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