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Low-Temperature Carbon Capture Using Aqueous Ammonia and Organic Solvents

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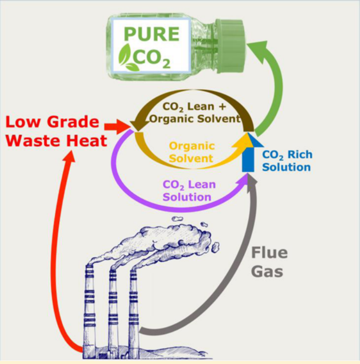
Supporting Information

ABSTRACT: Current postcombustion CO_2 capture technologies are energy intensive, require high-temperature heat sources, and dramatically increase the cost of power generation. In this work, we introduce a new carbon capture process requiring significantly lower temperatures and less energy, creating further impetus to reduce CO_2 emissions from power generation. In this process, high-purity CO_2 is generated through the addition of an organic solvent (acetone, dimethoxymethane, or acetaldehyde) to a CO_2 rich, aqueous ammonia/carbon dioxide solution under room-temperature and -pressure conditions. The organic solvent and CO_2-absorbing solution are then regenerated using low-temperature heat. When acetone, dimethoxymethane, or acetaldehyde was added at a concentration of 16.7% (v/v) to 2 M aqueous ammonium bicarbonate, 39.8, 48.6, or 86.5%, respectively, of the aqueous CO_2 species transformed into high-purity CO_2 gas over 3 h. Thermal energy and temperature requirements for recovering acetaldehyde, the best-performing organic solvent investigated, and the CO_2-absorbing solution were 1.39 MJ/kg of CO_2 generated and 68 °C, respectively, 75% less energy than the amount used in a pilot chilled ammonia process and a temperature 53 °C lower. Our findings exhibit the promise of economically viable carbon capture powered entirely by abundant low-temperature waste heat.



■ INTRODUCTION

Among human activities, CO_2 emissions from electricity generation and industry make up 65% of global greenhouse gas emissions.[1] Considering the world's growing energy demand and continued dependence on fossil fuels,[2] there is an unprecedented need to develop technologies to significantly reduce CO_2 emissions.

One promising means of reducing CO_2 emissions is postcombustion CO_2 capture and utilization (CCU),[3] which transforms low concentrations of CO_2 in emissions into high-purity CO_2 for utilization.[4] However, implementation of these technologies, such as the chilled ammonia and monoethanolamine (MEA) carbon capture processes,[5] has been limited to pilot plants because of enormous operating costs.[6] The most effective current processes require high-temperature heat, generally supplied by steam diverted from power generation,[7,8] increasing electricity costs by >70%.[9] High-temperature heat constitutes >80% of the energy consumption in current carbon capture processes and is the costliest component of CO_2 capture.[10] A lower-operating cost postcombustion CO_2 capture system is necessary to make CCU an effective means of reducing CO_2 emissions from power generation.[11]

Pure CO_2 is a valuable product with a commercial market of 80 Mt/year.[12−14] Because of the cost prohibitive nature of current CO_2 capture systems, >80% of the demand for pure CO_2 is supplied by the unsustainable drilling of CO_2 source fields,[12,15] which contain CO_2 that has been sequestered for millions of years.[16] An effective system that captures CO_2 from flue gas below market prices would displace the production of pure CO_2 from these unsustainable and counterproductive sources.

A compelling solution is a CO_2 capture process powered entirely by abundant low-temperature waste heat, allowing for the conversion of flue gas to pure CO_2, while negligibly impacting power plant efficiency.[17] Waste heat accounts for >68% of the energy generated by the U.S. thermal power industry,[18] with a weighted average temperature of 88.6 °C.[19] Low-grade power plant waste heat is generally untapped because of thermodynamic limitations on the conversion of low-temperature heat into electricity.[18] A low-temperature CO_2 capture system could be a more effective use of low-temperature waste heat.

In this study, we present a novel, regenerable carbon capture process powered entirely with low-temperature heat. The system generates high-purity CO_2 via the addition of a water-soluble organic solvent to a CO_2 rich aqueous ammonia/carbon dioxide solution, such as would be generated from the absorption of flue gas CO_2 in aqueous ammonia. The organic solvent is subsequently distilled using low-temperature heat, resulting in recovery of the solvent and remaining aqueous ammonia/carbon dioxide solution. The system generates pure CO_2 under room-temperature and -pressure (RTP) conditions and employs only low-cost, abundant reagents. Notably, our

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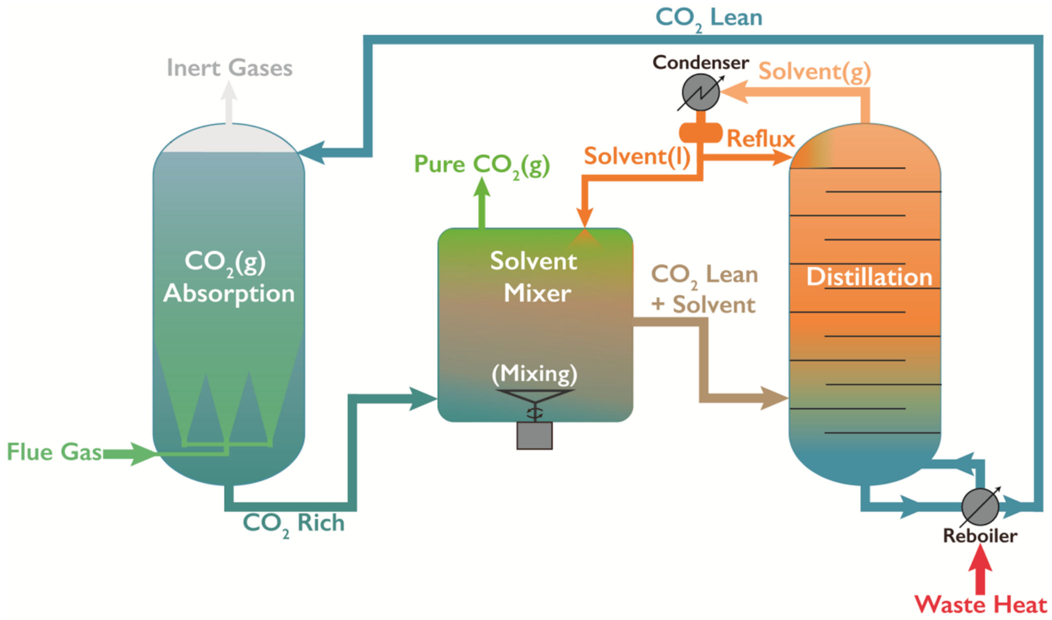

Figure 1. Schematic of the solvent addition ammonia/carbon dioxide carbon capture process. The process includes three stages: (1) CO$_2$ absorption column, (2) organic solvent addition and pure CO$_2$ generation (in a mixer), and (3) organic solvent distillation and solution recovery. An organic solvent is added under room-temperature and -pressure (RTP) conditions and distilled using low-temperature waste heat. The CO$_2$ lean and rich solutions contain NH$_3$:CO$_2$ molar ratios of >1.5 and <1.5, respectively. We note that a full-scale process would employ water washes to remove organic solvent vapor that may escape with the "inert gases" from the absorption column, and organic solvent vapor in the generated CO$_2$ from the solvent mixer/desorber.

study is the first to describe this novel solvent addition mechanism in CO$_2$ capture. Our results demonstrate that the carbon capture process is capable of converting flue gas into high-purity CO$_2$ with temperature and energy requirements significantly lower than those of current technologies.

■ MATERIALS AND METHODS

Experimental Setup and CO$_2$ Generation Measurements. Measurements were taken using an in-house constructed gas flow setup with online mass spectrometry (Figure S1). An Omega mass flow controller was used to control the flow rate of the carrier gas (ultra-high-purity helium, 50 mL/min). The outlet line was heated to prevent solvent condensation. For each experiment, an appropriate amount of ammonium bicarbonate (≥99.5%, Sigma-Aldrich) was dissolved in deionized (DI) water to form 100 mL of total solution at a desired molarity (1.0, 1.5, or 2.0 M). A 250 mL glass medium bottle containing the solution was attached to a three-port cap containing a helium carrier gas inflow port, a gas mixture outlet port, and an organic solvent injection port. Helium gas flowed into the headspace at a rate of 50 mL/min until no traces of air gases (N$_2$, O$_2$, and Ar) were present and the flow stabilized. Stirring was performed at a consistent angular velocity for all trials. An appropriate amount of organic solvent [acetaldehyde (≥99.5%, Sigma-Aldrich), acetone (≥99.5%, Fisher Scientific), or dimethoxymethane (99%, Sigma-Aldrich)] was injected. These solvents were selected on the basis of their high solubility in water, low molar mass, low toxicity, high volatility relative to water, and lack of irreversible reactivity with NH$_3$ or CO$_2$. A needle valve connected to a vacuum chamber with an SRS 100 residual gas analyzer was used to sample the outlet gas and obtain the CO$_2$ partial pressure. CO$_2$ partial pressures were converted to molar flow rates using a calibration curve derived from previous measurements of mass flow-controlled ultra-high-purity CO$_2$ and by normalizing the signal intensity to the helium carrier gas. Integration of CO$_2$ flow rates over 1 h yielded the values for total pure CO$_2$ generation.

Total CO$_2$ generation was determined by extrapolating results from 1 h experiments with an ExpConvExp fitting function using the Multipeak Fit package in Igor Pro (WaveMetrics Inc.). During long time frame experiments with 20 mL of organic solvent added, CO$_2$ generation tapered off after a 3 h period. Correspondingly, CO$_2$ generation from 1 h experiments was extrapolated to 3 h. Three-hour extrapolations deviated <12.5% from experimental results.

Modeling of Organic Solvent Distillation. The heat duty and temperature requirements for the recovery of acetaldehyde, acetone, or dimethoxymethane from their respective aqueous solutions were determined using an industrial process modeling software (Aspen HYSYS) with the UNIQUAC fluid package. The simulation used a 1.5 m diameter distillation column with 10 sieve trays and a 0.1 m^3 reboiler and condenser. The feed stream flow rate was 1 m^3 of solution/h and contained the optimal organic solvent mole fraction (x_f) to generate pure CO$_2$ (0.0467 for acetone, 0.0393 for dimethoxymethane, and 0.0599 for acetaldehyde) in water. According to vapor−liquid equilibrium studies, only slight traces of NH$_3$ and CO$_2$ vaporize at the low temperatures employed.[20,21] The simulated distillation column was at a scale sufficient for 0.43−1.83 t of CO$_2$ captured/day, or a scale similar to that of the referenced current process pilot plants, 1.7 and 4.0 t of CO$_2$/day for chilled ammonia and MEA, respectively.[22,23] The optimal organic solvent mole fraction was experimentally determined by adding the organic solvent under RTP conditions to a 100 mL 2 M aqueous ammonium bicarbonate solution until the injection of additional organic solvent had no discernible influence on CO$_2$ generation (Figure S2). The feed solution was distilled to the operational organic solvent mole fraction (x_b) in the regenerated solution (0.0216 for acetone, 0.0181 for dimethoxymethane, and 0.0279 for acetaldehyde). The operational organic solvent mole fraction was experimentally determined by adding small amounts of organic solvent under RTP conditions to a 100 mL 2 M aqueous ammonium bicarbonate solution until the injection of additional organic solvent resulted in CO_2 generation (Figure S2).

■ RESULTS AND DISCUSSION

System Overview. Figure 1 shows a schematic of the proposed carbon capture process. The process is composed of three steps: (1) flue gas CO_2 absorption in a CO_2 lean aqueous ammonia solution, (2) pure CO_2 generation through mixing in an organic solvent, and (3) recovery of organic solvent via low-temperature distillation.

In the first stage, the CO_2 absorption column, CO_2 in flue gas is absorbed by a CO_2 lean aqueous ammonia/carbon dioxide solution (NH_3:CO_2 molar ratio of >1.5), forming a CO_2 rich solution (NH_3:CO_2 molar ratio of ∼1). The remaining gases after the CO_2 is absorbed are released from the absorption column ("Inert Gases" in Figure 1). Similar CO_2 absorption columns are currently employed in the chilled ammonia process.[24]

In the second stage, the solvent mixer, the CO_2 rich ammonia/carbon dioxide solution from the CO_2 absorption column is mixed with an organic solvent (acetone, acetaldehyde, or dimethoxymethane) under mild temperatures and pressures, such as RTP conditions, generating high-purity CO_2. The solution becomes CO_2 lean as pure CO_2 is generated.

In the last stage, the solvent distillation column, the solution formed in the solvent mixer enters a distillation column, where the organic solvent is distilled from the CO_2 lean aqueous solution. The aqueous solution is recirculated to the CO_2 absorption column, and the organic solvent is recirculated to the solvent mixer.

Pure CO_2 Production Mechanism. Pure CO_2 was generated by adding acetone, dimethoxymethane (DMM), or acetaldehyde to aqueous ammonium bicarbonate solutions under RTP conditions. Figure 2 shows the amount of pure CO_2 generated over 1 h (experimentally observed) and 3 h (extrapolated) periods when 20 mL of acetone and DMM were added to 100 mL of 1, 1.5, or 2 M aqueous ammonium bicarbonate solutions under RTP conditions. The level of CO_2 generation per 20 mL of organic solvent increased with ammonium bicarbonate concentration. The level of generation of pure CO_2 from 2 M ammonium bicarbonate solutions was 51% greater with acetone and 36% greater with DMM than the corresponding results with 1 M ammonium bicarbonate solutions. Furthermore, DMM generated amounts of pure CO_2 larger than those generated with acetone from 10 to 30 mL of organic solvent added (shown in Figure S2), despite possessing a lower solvent mole fraction (x_f = 0.0393 for 20 mL of DMM; x_f = 0.0467 for 20 mL of acetone).

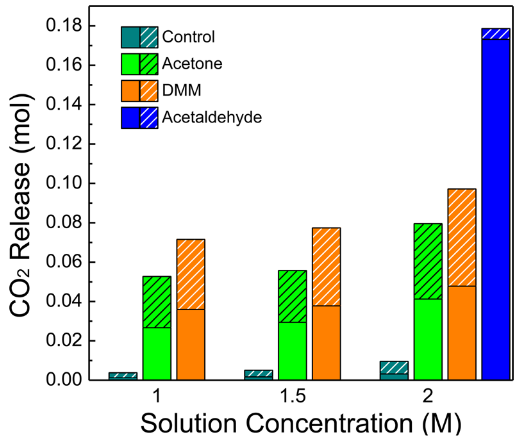

Figure 2. CO_2 generated at different ammonium bicarbonate solution concentrations with different organic solvents injected. Experiments were conducted using the online mass spectrometry setup (Figure S1) and 20 mL of solvent added to a 100 mL aqueous ammonium bicarbonate solution. The control was the CO_2 generated from solution with no organic solvent injected under room-temperature and -pressure (RTP) conditions. Solid bars represent pure CO_2 generated over 1 h, determined experimentally, and hatched bars represent the additional CO_2 generation during 3 h of operation, from extrapolation. We note that the CO_2 capacity for dimethoxymethane and acetaldehyde added to a 2 M solution is similar to those of current MEA[42] and chilled ammonia[43] processes.

Reports on desalination processes suggest organic solvents precipitate dissolved salts by reducing the dielectric constant (ε_r) of an aqueous solution.[25−29] Specifically, a reduction in dielectric constant from organic solvent addition weakens the hydration shells surrounding the solvated ions[25,26,28,29] and increases the extent of ion association due to the Coulombic attraction between oppositely charged ions,[25−28] thereby triggering salt precipitation.[25−29] In our study, an organic solvent was added to generate $CO_2(g)$ rather than a solid precipitate. According to studies of the CO_2 absorbing mechanism in aqueous ammonia,[30,31] aqueous ammonia performs multiple roles as a reactant, catalyst, base, and product controller,[31] thus allowing aqueous phase CO_2 to dissolve at a concentration significantly greater than that at which it would dissolve in the absence of ammonia.[30,31] We posit the addition of an organic solvent weakens the hydration shells surrounding the dissolved CO_2 due to reduction of the solution dielectric constant, thus prompting the generation of $CO_2(g)$ because of the significantly lower solubility of aqueous phase CO_2 when its interaction with ammonia is inhibited.[31] We attribute DMM's greater level of CO_2 generation to its significantly lower dielectric constant ($\varepsilon_r = 2.6$ for DMM;[32] $\varepsilon_r = 20.7$ for acetone[32]), as DMM requires a solvent mole fraction lower than that of acetone to decrease the solution dielectric constant by the same magnitude.[25,26,28]

Acetaldehyde generated more pure CO_2 than both DMM and acetone, despite having a greater dielectric constant ($\varepsilon_r = 21.7$ for acetaldehyde[32]) because it possessed a greater solvent mole fraction and a reversible reaction with ammonia species.[26,33,34] Acetaldehyde reacts with ammonia under anhydrous conditions to form a trimer.[26,33,35] Under aqueous conditions, the acetaldehyde−ammonia trimer is stable above pH 10, forms the acetaldehyde−ammonia adduct ion between pH 7 and 10, and reversibly dissociates into acetaldehyde and free ammonia below pH 7.[34] The aqueous acetaldehyde−ammonia adduct ion, which forms at the pH of aqueous ammonia/carbon dioxide solutions (CO_2 rich, pH ∼8; CO_2 lean, pH ∼9),[36] decomposes into acetaldehyde vapor and aqueous ammonia upon the volatilization of acetaldehyde.[34] Correspondingly, in our process, acetaldehyde generates more CO_2 than DMM and acetone and is effectively recovered from the aqueous ammonia/carbon dioxide solution during low-temperature distillation.

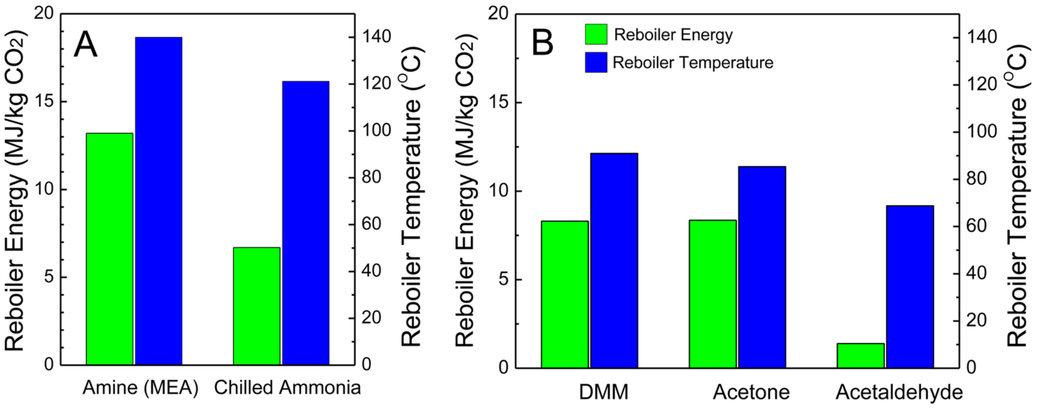

Figure 3. (a) Literature values for heat energy (green) per kilogram of CO_2 captured and reboiler temperature (blue) for the chilled ammonia[22] and monoethanolamine (MEA)[23] carbon capture processes. (b) Heat energy (green) per kilogram of CO_2 captured and reboiler temperature (blue) as a function of solvent type. The shown values are for 2 M ammonium bicarbonate with 16.7% (v/v) organic solvent added and use Aspen HYSYS simulation data (Figure S3).

Negligible CO_2 generation occurs at low solvent concentrations.[37] At 5 mL of organic solvent added to 100 mL of 2 M ammonium bicarbonate, the amount of CO_2 generated was 8% less than the amount in the no solvent case. This is consistent with a previous study that investigated the use of low concentrations of water-soluble organic solvents to prevent the release of ammonia from solution.[37] Specifically, it was found that low concentrations of organic solvents did not influence the rate of CO_2 absorption and desorption.[37]

At high ammonium bicarbonate and solvent concentrations, a plateau in CO_2 generation occurred, as shown in Figure S2. The amount of CO_2 generated from 2 M ammonium bicarbonate solutions under RTP conditions with 30 mL of added solvent was 2 and 8% smaller than the amounts with 20 mL of acetone and DMM added, respectively, and was accompanied by the immediate formation of a solid precipitate. At lower ammonium bicarbonate concentrations (1 M), the level of CO_2 generation increased with larger solvent volumes (20−30 mL) and no precipitate formed. We attribute these phenomena to an equilibrium between the formation of CO_2 gas and solid precipitate, which shifted toward solid precipitate at higher ammonium bicarbonate and solvent concentrations.[38,39]

Energy Consumption. The reboiler and condenser heat duties were determined with Aspen HYSYS using the x_f and x_b organic solvent mole fraction values for each organic solvent in 2 M ammonium bicarbonate. The simulated feed solution contained the x_f organic solvent mole fraction and was distilled to form an outlet stream with the x_b organic solvent mole fraction. The simulated condenser was cooled using a 20 °C water stream. We note that acetaldehyde reboiler heat duty was 70% less than that of DMM and 64% less than that of acetone because acetaldehyde has a lower boiling point (20.2 °C)[32] and a greater x_f solvent mole fraction.

The reboiler energy and temperature requirements of our process were compared with those of current CO_2 capture processes, the chilled ammonia and MEA processes (Figure 3). Current CO_2 capture processes use energy intensive thermal desorption with costly high-temperature heat (>120 °C[22]) to generate pure CO_2. Our novel process requires no heat input during CO_2 desorption, instead generating pure CO_2 under RTP conditions through the addition of an organic solvent. The organic solvent is subsequently distilled using abundant low-temperature waste heat, resulting in recovery of the solvent and remaining aqueous ammonia/carbon dioxide solution. The energy requirement was calculated by dividing the energy to distill the organic solvent [16.7% (v/v)] added to a 2 M aqueous ammonium bicarbonate solution by the mass of pure CO_2 generated. We have used literature values for the energy and temperature requirements for the MEA[23,40] and chilled ammonia[22] processes.

As shown in Figure 3, the reboiler temperature requirements for acetone and DMM were 49 and 55 °C lower, respectively, than that of the MEA process and 30 and 36 °C lower, respectively, than that of the chilled ammonia process. The heat duty for acetaldehyde was 1.39 MJ/kg of CO_2, or less than one-quarter of the heat duty of a pilot chilled ammonia process. The reboiler temperature requirement for acetaldehyde was 68 °C, which is 72 and 53 °C lower, respectively, than the temperature requirements of the MEA and chilled ammonia processes. Reboiler temperature requirements for all three organic solvents investigated were significantly lower than those of current CO_2 capture technologies and within the temperature range of low-grade waste heat.[19]

The development of a low-cost CO_2 capture system is essential to the effective mitigation of climate change.[41] Our solvent addition carbon capture process is powered using entirely abundant low-temperature waste heat, dramatically reducing operational costs and eliminating the efficiency penalty of carbon capture on power generation. Capital costs are reduced by the utilization of low-cost, widely available reagents and operation at mild temperatures and pressures. Our process demonstrated temperature and energy requirements significantly lower than those of current carbon capture systems, the chilled ammonia and MEA processes. Further optimization will lead to improved performance. For example, the investigation of other solvents and solvent combinations will result in further reductions in temperature and energy requirements. Overall, our organic solvent addition carbon capture process represents a new approach for low-cost, efficient flue gas CO_2 capture powered entirely by low-temperature waste heat.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.estlett.6b00253.

Schematic diagram of the bench-scale on-line mass spectroscopy closed setup for continuous reading of gas stream components and their relative partial pressures (Figure S1), CO_2 release (moles) as a function of the final solvent mole fraction and solvent type for various ammonium bicarbonate concentrations (Figure S2), and distillation column reboiler and condenser energy loads per cubic meter of total solution following the addition of each organic solvent (Figure S3) (PDF)

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Notes

The process introduced in this research is patent pending.
The authors declare no competing financial interest.

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