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Results from Process Modeling of the Mixed-salt Technology for CO₂ Capture from Post-combustion-related Applications

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Abstract

Mixed-salt technology, a solvent-based technology for removing CO₂ from flue gas streams offers a significant advantage over conventional amine-based CO₂ removal technologies (e.g., Fluor Econamine FG PlusSM technology). SRI International (SRI) is currently investigating the application of mixed-salt technology for pulverized coal combustion (PCC) power plant retrofit applications for removing >90% CO₂ at a cost not to exceed \$40/tonne of CO₂ captured. The research was performed at a large bench-scale level with funding from the United States Department of Energy (DOE), National Energy Technology Laboratory (NETL).

Very recently, a successful demonstration of mixed-salt technology at 0.25 tonne/day system was conducted in the USA, and the data obtained from the tests was used to develop a rate-based model to determine the mass and energy balance for a carbon dioxide recovery (CDR) removing 90% CO₂ from a 550-MW supercritical power plant. In this paper, we present the process modeling data including the preliminary techno-economic evaluation (TEA) of mixed-salt technology. CO₂ capture and CO₂ pipeline purity specifications were met in all the process configurations investigated in this study. SRI's mixed-salt process can strip CO₂ at high pressure as the stripper for rich-solvent regeneration is operated at higher pressure than the Fluor Econamine FG PlusSM process. Thus, the electrical power required for compressing CO₂ to delivery pressures (> 130 atm) is greatly reduced in the mixed-salt process compared to other solvent-based technologies operating with lower-pressure regenerations.

Ammonia-based technologies require absorber solvent cooling and treated gas washing to reduce ammonia emissions, and the raw water consumption of the process combines the water being used in the two water-wash sections. The Fluor Econamine FG PlusSM technology requires a large water recycle in the CDR unit for cooling purposes (1,173,350-1,286,900 lpm or 310,000-340,000 gpm), which greatly exceeds the PC plant cooling water requirement (643,450-757,000 lpm or 170,000-200,000 gpm).

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SRI's mixed-salt process requires a relatively smaller recycle for cooling purposes, and the overall cooling water recycled was 71% less in the mixed-salt process compared to the baseline case. As such, the auxiliary power required for mixed-salt process CDR unit was 60% less than the baseline case. The heat duty for the mixed-salt process was calculated to be 2.0 MJ/Kg of CO₂ recovered (in the stripper reboiler). This accounts for a 44% decrease in the heat duty requirement in the mixed-salt process compared to the baseline case.

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Keywords: CO₂ capture, post-combustion; mixed-salt; ammonia; potassium carbonate

1. Introduction

Cost-effective capture of CO₂ emissions from coal-powered plants is of critical strategic importance to further enable the use of coal, an abundant natural resource in the USA, without increasing greenhouse gas (GHG) emissions. SRI International (SRI) is currently developing a highly promising solvent-based CO₂ capture process called mixed-salt technology [1] with funding from the National Energy Technology Laboratory (NETL). The overall goal of the project is to test the technology at the large bench-scale level (0.25 to 1 tonne/day CO₂ capture – mini-pilot) to demonstrate the process can capture CO₂ at high efficiency (> 90%).

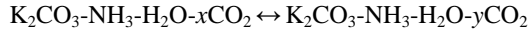
Mixed-salt technology capitalizes on the advantages of both ammonia-based and potassium carbonate-based technologies with improved reaction kinetics and reduced emissions [1, 2, 3]. This technology provides unique opportunities for better energy management and reduces the burden on the power plant steam cycle. It is suitable for capturing CO₂ from post-combustion, pre-combustion, and other industrial gas streams. Mixed-salt technology can strip CO₂ at high pressure, reducing the CO₂ compression costs, and requires no solvent chilling as in aqueous ammonia-based processes. It also has a low reboiler duty for regeneration compared to conventional amine-based processes.

Nomenclature

CDR	Carbon dioxide recovery
COE	Cost of electricity
DCC	Direct contact cooler
DOE	Department of Energy
FGD	Flue gas desulfurization
gpm	Gallons per minute
lpm	Liters per minute
MEA	Monoethanolamine
MSE	Mixed-Solvent Electrolyte
NETL	National Energy Technology Laboratory
PCC	Pulverized coal combustion
TEA	Techno-economic analysis

2. Mixed-Salt Process Description

In the mixed-salt technology, CO₂ capture is achieved by chemical absorption and the solvent regeneration by high-temperature and high-pressure stripping of CO₂ from the CO₂-rich solvent. The solvents used in chemical absorption processes are basic in nature, and they react readily with CO₂ by a series of chemical reactions and thereby absorb CO₂ from the flue gas stream. The reaction schemes related to aqueous-based, mixed-salt technology are ionic in nature. The mixed-salt process chemistry comprises rapid gas/liquid-phase mass transfer followed by chemical reactions in the liquid phase. The simplistic representation of the CO₂ absorption and removal can be:



where cyclic CO_2 is the numerical difference between y and x . The left and right side of the equilibrium represent the lean and rich solutions, respectively.

Speciation of the $\text{K}_2\text{CO}_3\text{-NH}_3\text{-CO}_2\text{-H}_2\text{O}$ system includes $\text{H}_2\text{O}(\text{l})$, $\text{CO}_2(\text{g})$, $\text{H}_2\text{O}(\text{g})$, $\text{NH}_3(\text{g})$, $\text{CO}_2(\text{aq})$, $\text{NH}_3(\text{aq})$, CO_3^{2-} , HCO_3^- , H^+ , K^+ , NH_2CO_2^- , NH_4^+ , OH^- , $\text{K}_2\text{CO}_3(\text{s})$, $\text{KHCO}_3(\text{s})$, $(\text{NH}_4)_2\text{CO}_3 \cdot 2\text{NH}_4\text{HCO}_3(\text{s})$, $\text{NH}_4\text{HCO}_3(\text{s})$, $\text{K}_2\text{CO}_3 \cdot 1.5\text{H}_2\text{O}$, and several other solid phases. Solid species will not be present in the mixed-salt system as the absorber is operated at a higher temperature, and below the solid-forming salt concentrations.

The chemical reactions in the mixed-salt process are all reversible, and their direction depends on pressure, temperature, and concentration in the system. At low temperature (e.g., 20°C), equilibrium moves from a left-to-right direction (exothermic process) and requires removal of heat from the process in order to maintain the desired absorption temperature. At high temperature (e.g., $>60^\circ\text{C}$), equilibrium shifts from right-to-left direction (endothermic process) that requires energy to release gaseous CO_2 . The heat of reaction for the process can be tuned (35-55 kJ/mol) depending on the composition of the mixture (potassium and ammonium salt ratio). The schematic of the mixed-salt CO_2 capture process is shown in Figure 1.

The mixed salt CO_2 capture system comprises a dual-stage isothermal absorber, a novel selective regenerator, and auxiliary equipment. The absorber consists of the first section (Absorber 1) with ammonia-rich solvent and a second section (Absorber 2) with potassium-rich solvent. The absorber can be assembled either as single absorber in two sections or as two separate absorbers in series (as shown). Flue gas from the power plant enters the Absorber 1 after passing through the flue gas desulfurization (FGD) unit and cooling to $20\text{-}40^\circ\text{C}$. Up to 80% of the CO_2 gets absorbed in the Absorber 1, and the remaining CO_2 in the flue gas is absorbed in Absorber 2.

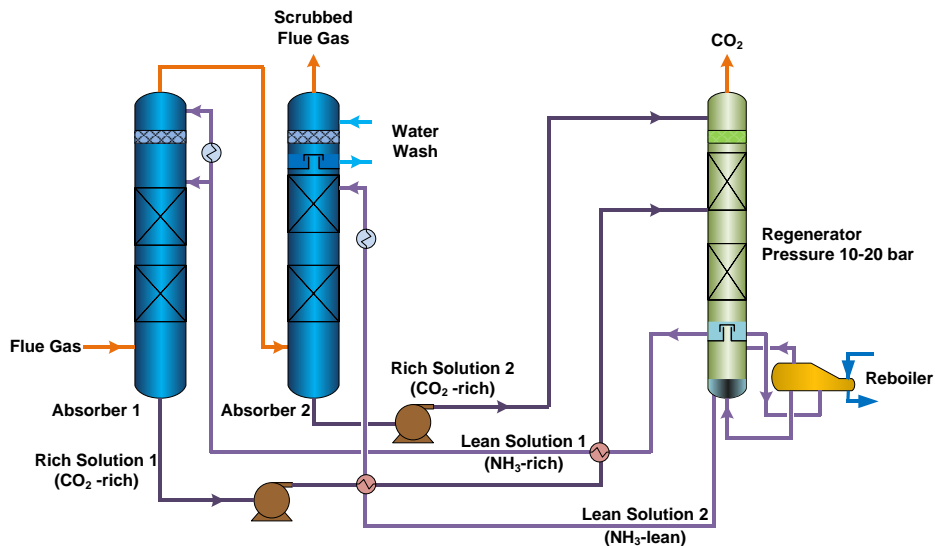


Fig. 1. Process flow diagram for the mixed-salt process.

The absorber operates with liquid recycle and cooling to keep the solution in the range of $20\text{-}40^\circ\text{C}$ range to maintain the absorber at relatively uniform temperature. In a two-stage absorber, the first stage operates with the highest CO_2 loading in the range of 0.6-0.7 mole of CO_2 per mole of ammonia and about 0.7-0.8 mole of CO_2 per mole of potassium. The heat of reaction is removed from the absorber using the heat exchanger in the recycle loop. The CO_2 -rich solution from the absorber is constantly removed from the bottom via a bleed stream on the recycle loop. The CO_2 -rich solution is sent to the regenerator for regeneration.

The mixed-salt process uses a selective regenerator. The key feature of the regenerator is the design that regenerates two CO₂-lean salt solution streams as follows: (1) an ammonia-rich mixed salt solution to the first stage of the absorber; and (2) an ammonia-lean mixed-salt solution stream to the second stage. The results from our recent testing runs with a 6-molal mixed-salt solution are shown in Figure 2. The key details of the process are described in our previously published paper from the GHGT-11 meeting [1].

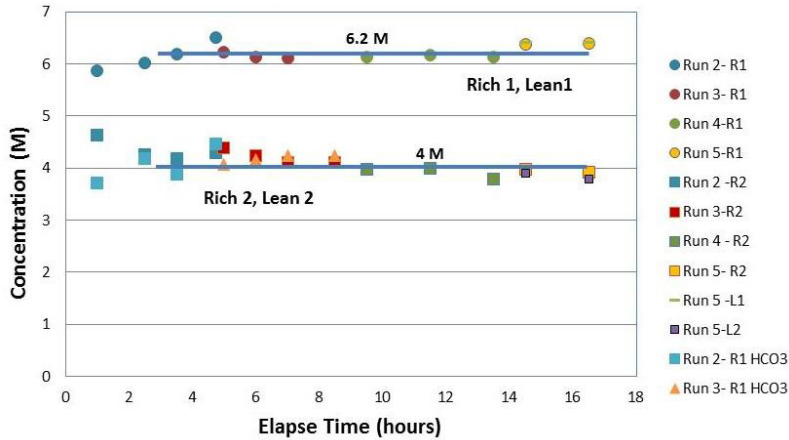


Fig. 2. Demonstration of the regeneration of lean solutions with two different ammonia concentrations (high and low N/K).

3. Process Modeling

OLI Systems, USA has developed a rate-based model with SRI test data that was used in the OLI Electrolyte Simulation Program (ESP) [4] for process modelling. For this purpose, the Mixed-Solvent Electrolyte (MSE) model was used to predict thermodynamic properties [5]. Using the updated rate-based model, OLI Systems has determined the complete mass and energy balance for a full-scale mixed-salt CO₂ capture system operating in a power plant. The technology was modelled for the carbon dioxide recovery (CDR) facility in which 90% of the CO₂ from the flue gas was captured from a supercritical pulverized coal (PC) plant with a nominal net output of 550 MW (DOE Case 11) [4, 6, 7]. The other fixed parameters were regeneration of high-pressure CO₂ at 99% purity and the ammonia release from the absorber to be less than 10 ppm. The mixed-salt technology was compared with Fluor Econamine FG PlusSM technology (DOE Case 12). The Econamine FG PlusSM process uses a formulation of monoethanolamine (MEA) and a proprietary corrosion inhibitor to recover CO₂ from the flue gas. The heat duty requirement at the reboiler stripper for the Fluor Econamine FG PlusSM technology was reported as 3,556 kJ/kg (1,530 Btu/lb) or 3.56 MJ/kg of CO₂ recovered. Table 1 summarizes the performance comparison of the mixed-salt technology (basic process option) with a DOE baseline case.

Table 1. Comparison between mixed-salt technology and DOE baseline case.

Performance Factors	Econamine Baseline	SRI's Mixed-Salt Technology*
CO ₂ capture, %	90.2	90.3
CO ₂ purity (before compression), %	99.61	> 99.0
Stripper pressure, atm	1.0	10.0
Raw water consumption, gpm	36	107
Raw water recycle, gpm	~325,000	<100,000
Auxiliary power, KWe	20,600	3,581
Heat duty, MJ/kg of CO ₂	3.56	2.0

3.1. Technology Review and Simulation Assumptions

The CDR facility removes 90% of the CO₂ in the flue gas exiting from the plant and purifies it. The striking advantages of this process include: high loading of CO₂, high-pressure regeneration of > 99% pure dry CO₂, low sensitivity to impurities, low process cost, use of a non-degradable low-cost solvent with a very low carbon footprint for its production, low emissions, reduced water use, and faster CO₂ absorption kinetics compared to the state-of-the-art ammonia-based technologies. The mixed-salt process is suitable for capturing CO₂ from flue gas streams from pulverized coal, natural gas, and other industrial sources. It does not produce any hazardous wastes.

SRI's mixed-salt technology is based on off-the-shelf, low-cost, industrially available chemicals (e.g., ammonium and potassium salts) that react readily with CO₂ at a near-ambient temperature by a series of chemical reactions and thereby absorb CO₂ from the flue gas stream. The absorbed CO₂ can be released as high-purity, high-pressure CO₂ ready for storage and reduces the CO₂ product compression stages. The mixed-salt ensemble has an inorganic moiety that has a higher diffusion coefficient that enhances the gas-liquid mass transport and reduces the activation energy of the reaction between solvent and CO₂. In addition, the process uses a selective regeneration scheme for utilizing the optimal characteristics of each salt.

3.2. Mixed-Salt CDR Facility

SRI's mixed-salt CDR facility is comprised of the direct contact cooler (DCC) for flue gas cooling, CO₂ absorption, solvent stripping, and water wash. A simplified flowsheet of the mixed-salt CDR process modeling is shown in Figure 3.

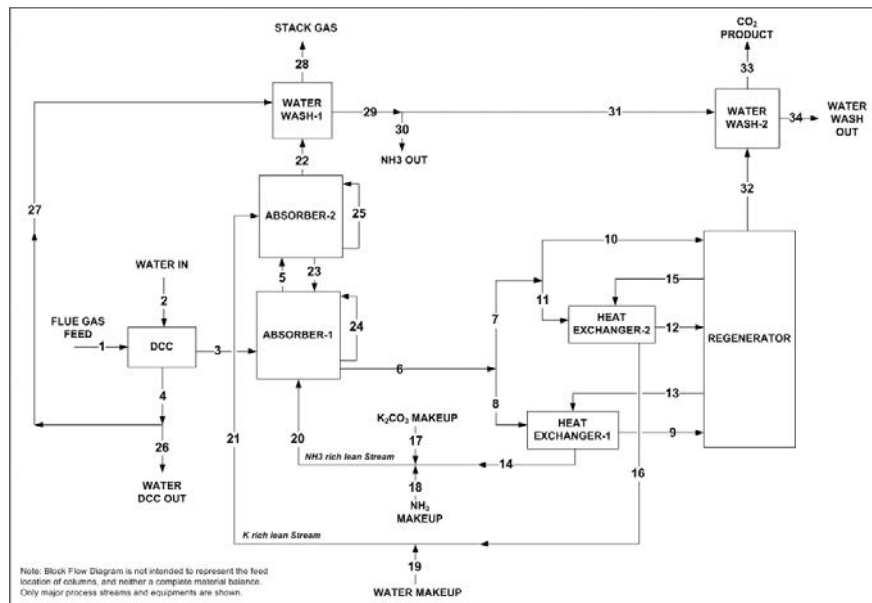


Fig. 3. Simplified process flowsheet block diagram for mixed-salt technology.

3.3. Process Description

Direct Contact Cooler: The DCC serves as a cooler for the flue gas. Cooling water from the plant is used to reduce the flue gas temperature to a level lower than the adiabatic steam saturation temperature, which results in a significant reduction of the flue gas moisture content. A small portion of the water stream from the DCC is cooled and used in the water-wash sections to remove residual ammonia from the stack gas and final CO₂ product to meet the required emission standards. The rest of the water stream from the DCC rejects heat to the plant cooling tower

and is recirculated. Approximately 5-10% of the water stream from the DCC is used in the water-wash sections. The flue gas is cooled to 20-25°C in the DCC.

CO₂ Absorption Columns: The absorbers can be assembled either as two separate absorbers in series (as shown in Fig. 2) or as a single absorber with two stacked-up sections. The Absorber 1 (or bottom section) circulates ammonia-rich solvent, and the Absorber 2 (or top section) circulates potassium-rich solvent. The cooled flue gas enters the bottom of Absorber 1 and flows up through the column counter-current to the stream of ammonia-rich, CO₂-lean solution. Approximately 80% of the CO₂ in the feed gas is absorbed into the lean solution, and the rest leaves the top of Absorber 1 and flows into Absorber 2. Absorber 2 operates with a potassium-rich, CO₂-lean solution flowing counter-current to the gas flow, and the remaining CO₂ in the flue gas is absorbed in Absorber 2. The typical temperature profile (column top to bottom) of the absorption columns is in the range of 20-45°C and 20-30°C for Absorber 1 and Absorber 2, respectively. The absorbers are operated at 1.0 bar. The rich solvent leaves the bottom of the absorbers with the absorbed CO₂. A portion of the rich solution leaving from the absorber bottom is cooled and recycled back to absorber top to keep the absorber top section temperature under control and to reduce ammonia emissions.

Rich/Lean Heat Exchange System: The rich solvent leaving Absorber 1 is at 35-40°C and is heated by the lean solvent coming from the stripper in a heat-exchange system. The rich solution from Absorber 1 is branched and routed to the bottom and mid-section of the stripper after heat exchange with the ammonia-rich, CO₂-lean solvent and potassium-rich, CO₂-lean solvent, respectively. Also, a small portion of rich solvent from Absorber 1 is routed to the top of the solvent stripper without any heat exchange or heat removal. The rich solution from Absorber 2 is routed to Absorber 1. The temperatures of the ammonia-rich, CO₂-lean solvent and potassium-rich, CO₂-lean solvent are about 125-135°C and 165-170°C, respectively. The flow arrangements are made to maximize the heat recovery in the heat exchange systems and to minimize ammonia emission.

Solvent Stripper: The purpose of the solvent stripper or regenerator is to separate the CO₂ from the rich solvent stream exiting the bottom of the CO₂ absorbers. The rich solvent is routed to three different feed locations on the solvent stripper where the rich solvent is heated by steam stripping the CO₂ from the solution. Steam is provided from the intermediate pressure (IP) section of the steam turbine at about 1.46 MPa (211.9 psia). Since the stripper operates at 12 bar, the CO₂-rich gas from the stripper exits at about 12 bar, which reduces the CO₂ product compressor stages significantly, saving compression energy cost. There are two CO₂-lean solvent product outlets from the stripper to use as feed streams in each of the absorbers (Absorber 1 and Absorber 2). The side draw is from the section of the column where ammonia is much more concentrated because a higher NH₃/K molal ratio is required in Absorber 1. The bottom draw has a lower NH₃/K molal ratio (K-rich) for use in Absorber 2. The CO₂ gas from the stripper enters the water-wash section to further clean any residual ammonia. The temperature at the reboiler is about 165-170°C, whereas the top section is at 50°-70°C.

Water Wash: The purpose of the water-wash section is to maintain emission standards in scrubbed flue gas and CO₂ product streams. The CO₂-scrubbed flue gas from Absorber 2 is contacted with a cooled, recirculating stream of water for the removal of residual ammonia in the Water-Wash-1 section. The scrubbed gases, along with unrecovered residual ammonia (less than 10 ppm) exit the top of the wash section for discharge to the atmosphere via the vent stack. A portion of the collected water from Water-Wash-1 section is recirculated in Water-Wash-2 section to remove the ammonia slip with the CO₂ product. The amount of water use in Water-Wash-2 section is controlled so that less than 10 ppm ammonia escapes with the high-pressure CO₂ product stream.

Circulating Water System: Cooling water is provided from the plant and returned to the plant cooling tower to reject the heat absorbed. The CDR facility requires cooling water for flue gas cooling, lean solvent cooling, rich solvent recirculation cooling, and water-wash cooling. The cooling water requirement for the CDR facility is about 80,569 gpm (305 m³/min) for a 550-MW plant.

4. Techno-Economic Analysis

The process economic analysis was conducted by Politecnico di Milano (POLIMI) based on a thermodynamic model developed by Aqueous Solutions ApS. In this analysis, a technology comparison was made using the economic results for mixed-salt process vs. the reference Case 12B proposed by NETL in the report “Cost and Performance Baseline for Fossil Energy Plants (July 6, 2015)” [6]. For this reason, the analyzed case adopts the

same technical specifications as Case 12B with exceptions made for the peculiarities introduced by the new post-combustion carbon capture technology. The methodology applied to the economic analysis strictly follows the NETL guidelines provided in the “Cost and Performance Baseline for Fossil Energy Plants” report [8, 9].

4.1. Methodology

Equilibrium Modeling: The commercial Aspen Plus code is employed for the simulation of the carbon capture plant. It can describe electrolyte solutions given that a proper thermodynamic model is utilized to account for the interactions between the species present in the solution, including strong and weak forces. The absorption and regeneration processes are simulated here by means of the Extended UNIQUAC thermodynamic model as proposed by Thomsen and Rasmussen [10]. In detail, the activity coefficients used for speciation, solid-liquid, and vapor-liquid equilibrium are calculated with the extended UNIQUAC model; the Soave-Redlich-Kwong equation of state is employed for computing the fugacities of the species in the gas phase, without requiring additional adjustable parameters. This model is not built into the Aspen Plus code, but it is defined in a user module as described by Darde *et al.* [11, 12] who performed an exhaustive validation of the thermodynamic model against experimental data available in literature. The model parameters for the CO₂-NH₃-H₂O system were updated and refined for application to carbon capture, focusing on the chilled ammonia process [11,12]. Model parameters were determined for the CO₂-NH₃-K₂CO₃-H₂O system based on a data compilation consisting of approximately 8000 experimental data points published in the open literature. These experimental data cover the temperature range from the freezing points of the solutions and up to 200°C and the pressure range up to 100 bar. These experimental data include measurements of vapor-liquid equilibrium, solid-liquid equilibrium, osmotic coefficients, speciation, heats of dilution, heats of solution, heats of absorption, and heat capacity for the system. The number of model parameters required by the model to describe this system is 46. In addition, standard-state thermodynamic properties of 15 solids potentially forming in the system were determined. Based on the parameters, the model was able to reproduce the experimental data with high accuracy. The 700 experimental vapor-liquid equilibrium data for the binary CO₂-H₂O system in the temperature range up to 200°C, and pressures up to 100 bar were reproduced with an average deviation of 4.9%. The corresponding average deviation for vapor pressures in the binary NH₃-H₂O system is 7.9% for the 1500 available data points in the same temperature and pressure range.

Cost Estimation: The methodology applied for the economic analysis strictly follows the NETL guidelines provided in the “Cost and Performance Baseline for Fossil Energy Plants” report [13]. Also in this study, we used the NETL dataset from the economic analysis in the “Quality Guidelines for Energy Systems Studies” [8]. Consequently, all the costs of the common parts of the plant are directly comparable with NETL report.

Regarding the specific mixed-salt technology section, a dedicated dataset is employed. Similarly, the NETL report does not provide any information about the costs of the components of the capture section but only the total equipment cost. Consequently, both a bottom-up method and in-house correlations are used to estimate the equipment costs within the carbon capture section. In order to conduct a comparison between two plants with two different post-combustion carbon capture technologies [Econamine FGSM and mixed-salt process], the technical specifications of the power plant must be the same. In this case, the capture plants treat the exhaust from a supercritical PC plant, so the simulations are conducted with the following technical conditions:

- PC steam cycle has the same characteristics as the power plant described in the baseline case 12B (superheated pressure, superheated temperature, re-superheated temperature, pressure of condensation, turbine efficiency, etc.);
- Carbon capture efficiency of the carbon capture plant is the same as Case 12B (90%);
- CO₂ outlet pressure at the end of the capture process is the same as Case 12B (152.7 bar);
- Net electric power is the same as Case 12B (550 MW).

The plant is assumed to be newly designed as opposed to retrofitted. Hence, the capture plant has the cooling system integrated with the cooling system of the power section. Therefore, the temperature of the cooling water is imposed by the performance of the same cooling tower as described in the NETL report. Finally, the technical assumptions for the capture plant are:

- Pump isentropic efficiency: 80%;
- Fan isentropic efficiency: 90%;
- Pinch-point temperature difference in the plate and frame heat exchangers: 5 K;
- Pinch-point temperature difference in the reboiler: 10 K;
- Compressor isentropic efficiency 85%;
- Total pressure loss of the exhaust: 0.1 bar.

Cost Methodology: The total equipment cost of the CO₂ removal systems is calculated as the sum of the equipment cost of each component. The cost estimation of each component follows a different approach, as explained below. For the calculation of the equipment cost of the columns, a bottom-up methodology is utilized. The columns (diameter and height) are dimensioned accordingly to Jilvero *et al.* [14], scaling them on the plant size analyzed in this report. The value of the height of the column remains the same as in Jilvero *et al.* because the kinetics of the reactions are assumed to be the same. The diameters of the direct contact cooler, Absorber 1, Absorber 2, and the water-wash column are scaled on the volumetric flow of the treated gas; the diameters of the regenerator are scaled on the volumetric flow of the rich solution inlet in the regenerator.

The cost of the vessel and the structured packing are derived from Peeters *et al.* [15] for an MEA carbon capture plant and corrected for the costs of the materials used in the mixed-salt technology: MEA requires vessels and packing made with a stainless steel with a strong resistance to corrosion. In contrast, the mixed-salt technology plant does not show particular issues of corrosion, so Absorber 1 and 2, the direct contact cooler, and the water-wash column as well as the vessel are made with lower-grade steel than required by the MEA technology. The packing of the mixed-salt columns is made of polypropylene because the absorption temperatures are low and the cost is ~80% lower than the stainless steel packing required by the MEA process. Due to the higher temperatures, the regenerator is assumed to be made of a higher-quality steel, and the packing is assumed to be equivalent to that used in the MEA case. The prices of the different kinds of steels were provided by Valenti *et al.* [16], and the packing prices were from the NETL Process Equipment Cost Estimation report [17].

Based on this economic evaluation, the cost of electricity (COE) presented by NETL for Case 12B is \$142.8/MWh vs. \$127.3/MWh described in the POLIMI analysis for the mixed-salt process capture plant. The mixed-salt technology shows a cost reduction of 10.8%. Table 2 shows the COE comparisons.

Table 2. Technology comparisons - COE.

Component, \$/MWh	NETL, Case 12 (2013)	NETL, Case 12B (2015)	SRI
	Econamine	Cansolv	Mixed Salt
Capital	66.4	72.2	57.1
Fixed	14.5	15.4	15.4
Variable	12.1	14.7	12.6
Fuel	35.3	30.9	32.3
Total (excluding T&S)	128.2	133.2	117.4
CO ₂ (T&S)	11.0	9.6	10.0
Total (including T&S)	139.2	142.8	127.3
Total Auxiliary Consumption (MWe)	112.8	91.0	72.0

5. Summary

CO₂ capture and CO₂ pipeline purity specifications were met in all the process configurations investigated in this study. SRI's mixed-salt process can strip CO₂ at high pressure, as the stripper for rich-solvent regeneration is operated at higher pressure than the Fluor Econamine FG PlusSM (Table 2). The electrical power required for compressing CO₂ to delivery pressures (> 130 atm) is greatly reduced in the mixed-salt process compared to other solvent-based technologies operating with lower-pressure regenerations. Ammonia-based technology requires absorber solvent cooling and treated gas washing to reduce ammonia emissions and the raw water consumption of

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In conclusion, SRI's mixed-salt technology can capture CO₂ at high pressure and can meet present DOE targets of CO₂ capture and pipeline purity requirements. The study shows the technology offers a much lower energy penalty than Fluor Econamine FG PlusSM technology and/or conventional MEA-based technology for post-combustion CO₂ capture. The technology can easily be scaled up with use of conventional process equipment.

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