RATE BASED MODEL AND TECHNO-ECONOMIC ASSESSMENT OF A POST-COMBUSTION CO₂ CAPTURE UNIT OPERATING WITH POTASSIUM LYSINATE FOR NGCC DECARBONISATION

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Abstract

The present work reports the results of process design and techno-economic assessment carried out on a CO_2 capture unit operating with an aqueous solution of potassium Lysinate (LysK) at 43.7%$_{w/w}$, in post-combustion arrangement for natural gas combined cycle (NGCC) decarbonisation. A dedicated rate-based model of the absorption unit has been defined and programmed in Matlab®. The techno-economic assessment of the reference natural gas combined cycle (frame-F gas turbine technology, 829.9 MWe without capture unit) coupled with a CO_2 capture unit with the low-maturity LysK-based solvent has been carried out in order to estimate the energy penalty due to capture, the specific primary energy consumption per unit of CO_2 avoided as well as economic indicators, such as overall capital costs, cost of electricity and the cost of CO_2 avoided. The results are compared against a case study envisaging the same reference NGCC coupled with a commercial post-combustion capture solution working with 30%$_{w/w}$ aqueous monoethanolamine (MEA).

Keywords: amino acid salts, post-combustion capture, natural gas combined cycle (NGCC), potassium lysinate

1. Introduction

Greenhouse gas (GHG) emissions associated to human activity have reached 49 GtCO₂-eq/y in 2015[1], with 36 GtCO₂-eq/y CO_2 from fossil fuel combustion and industrial processes remaining the main responsible for global warming[1]. Within this context, a leading position is covered by the power industry, whose amount has reached 13.6 GtCO₂ in 2017[2] caused by the elevated energy demand and the still intensive use of fossil fuels in electricity and heat generation. In order to reduce the carbon emissions associated to power production from natural gas combined cycles (NGCC), solvent absorption processes in post-combustion arrangement are usually identified as the most mature and reference technology. In fact, amine-based solutions are considered effective for CCS (Carbon Capture and Storage) applications, and the specific case of aqueous MEA (monoethanolamine and its possible water-based solutions) has been taken as a benchmark in several European initiatives such as CaESAR [3] and CESAR [4] projects, while analogous amine-based solvents are applied in commercial scale CCUS facilities (e.g. Boundary Dam).

However, Monoethanolamine scrubbing shows many drawbacks that include high-energy consumption for regeneration, limited CO_2 loading capacity, equipment corrosion, generation of toxic compounds and amine volatility, hence leaving the field to a great margin of improvements. New studies on alternative solvents are, therefore, ongoing to answer the still open items on absorption technologies, moving towards the identification of more effective, energy saving and greener solvents for a low-carbon energy production industry. Among these, recent research activities carried out from University of Texas consider piperazine (PZ) as a new potential baseline for amine-based carbon dioxide absorption, due to its higher operating temperature range and expected lower specific thermal duty [kJ/kg of CO_2 captured] for regeneration. The direct major effect of this newly identified secondary amine-based solvent is to allow stripper operations at higher pressure, with a sensitive reduction of the energy requirement for the CO_2 compression unit. In the field of amine-free solvents, amino acid salts (AAS) solutions are one of the most recent options under investigation. They are considered environmental friendly and, some of their formulationsdemonstrate fast reaction kinetics, lower regeneration energy and good CO_2 loading capacity; moreover, they are non-volatile and stable to oxidative degradation.

Despite their advantages, little knowledge on AAS absorption is available, as a consequence of limited experimental data (e.g.: heat capacity, vapor-liquid equilibria, heat of reaction, etc.), scarce pilot plant campaigns data and lack of comprehensive process engineering studies. With these regards, a first attempt at considering amino acids for CO_2 absorption has already been proposed in Decab, Decab Plus and Casper processes [5], however a limited range of solvents has been analyzed. Moreover, they envisage solid precipitation with a significant complication of the contacting equipment, which has discouraged technology spreading and application at industrial level.

For the above-mentioned reasons, a thorough understanding of the real potential of AAS-based non-precipitating solutions still requires further evaluations, and it has inspired the present research.

The final goal of this work is to assess the viability of non-precipitating AAS solutions applied to CO_2 absorption systems (DCC + absorber + stripper + heat exchanger network + CO_2 compression unit) treating post combustion gases from a natural gas combined cycle (NGCC, 829.9 MWe w/o capture).

Due to the high cost share of the absorber on the overall investment cost of a post-combustion unit, a proper design and sizing of this column is required ahead of costing. For this reason, the first step of this investigation consists in the definition of a rate-based absorber model developed in Matlab® with in-house codes. Moreover, the study is finalized by proposing a very first comprehensive techno-economic assessment on AAS based-post combustion unit coupled with NGCC, to be compared against the results of dedicated techno-economic assessments on MEA and other solvents within a consistent and comparable framework.

The target AAS molecule is potassium lysinate (LysK) at 43.7%w/w. The solution has been identified based on literature reviews and dedicated experimental tests carried out during previous works [6], [7].

The technical baseline for the study is be the European best practice guidelines for assessment of CO_2 capture technology (released by the European Benchmarking Task Force-EBTF), which makes reference to a natural gas fired power plant of 829.9 MWel without capture and outlines the performance of the same NGCC coupled with a CO_2 capture unit operating with 30%$_{w/w}$ monoethanolamine solution.

2. Context, Approach and Methodology

In order to identify suitable alternative solvents for post combustion CO_2 capture, a bibliographic review has been run, taking into account relevant solvent properties such as operating temperatures and pressure, physical and chemical quantities (viscosity, solubility, etc.), absorption capacity and enthalpy of reaction with CO_2. The review has highlighted that amino acid salts constitute a solvent family of interest for CCS compared to traditional molecules: AAS-solutions envisage chemical reaction (chemical absorption process), they can reach high CO_2 loading capacity, fast reaction rates, and they may require less regeneration energy with reference to commercial amines. Besides, these solvents are biodegradable and environmental friendly, they present low volatility and ecotoxicity, stability against oxidation, negligible corrosion effects; hence, they look appealing in the perspective of industrial scale applications [8].

From a preliminary literature review [9]–[11], lysine-based salts (e.g.: LysK) stand out among the most noteworthy molecules for further analysis. In a recommended concentration range of 3.5-4.5 m, LysK exhibits capacities in line with a 7m MEA without

showing precipitation. Reference values are 2.66 mol$_{CO2}$/kg$_{LysK Solution}$ for a 3.5 m solution of lysinate[10] compared to 2.64 mol$_{CO2}$/kg$_{MEA Solution}$ for a 7 m solution of MEA [10] (at 297K and partial pressure of CO_2 equal to 9 kPa). Overall, the few data available from the literature on the enthalpy of absorption of aqueous LysK seems to be in line with the MEA benchmark, meanwhile higher reaction kinetics has been recorded for LyK, with consequent potential reduction of the absorber height, packing volume and costs.

In spite of their potential, no complete and comprehensive techno-economic assessment on AAS applications to post-combustion CO_2 capture analyzing both solvent effectiveness and process engineering repercussions is available. The workflow followed by the authors aims at overcoming the state-of-the-art by filling the aforementioned literature gaps:

1. Experimental tests previously run [6], [7] have been produced to assess the performance of the amino acids selected from the literature review via an initial experimental screening at Sotacarbo research campus;

2. LysK solutions have shown the highest performance. CO_2 absorption capacity data have been employed to define a target concentration of LysK aqueous solution (i.e., 43.7%$_{w/w}$) which guarantees the same capacity of the reference 30%$_{w/w}$ (7m) MEA;

3. A suitable thermodynamic model based on Deshmukh and Mather approach has been reproduced for the MEA/CO_2/H_2O. The same approach has been followed in order to describe vapour-liquid equilibria and CO_2 solubility of the LysK/CO_2/H_2O system;

4. A dedicated rate-based model and preliminary design of the absorber operating with aqueous LysK has been defined via in-house coding developed in Matlab;

5. Based on the newly defined thermodynamic model, the regeneration energy for CO_2 rich LysK solutions has been estimated for the very first time, adopting a well-established literature approach [13], [14];

6. Preliminary design of the CO_2 capture unit including direct contact cooler, absorber, stripper, heat exchanger network and CO_2 capture unit has been carried out;

7. Evaluation of the solvent regeneration duty and energy penalty over the combined cycle have been necessary for a preliminary techno-economic assessment of the investigated system. Therefore, suitable energy and economic indicators such as the specific primary energy consumption per unit of CO_2 avoided (SPECCA), cost of electricity (COE) and cost of CO_2 avoided (CCA) have been calculated for LysK-based absorption and compared against the outcomes of MEA[3] scrubbing. The three techno-economic assessments provide a quantitative overview on the viability of AAS solutions for CO_2 capture from power plants.

This article presents the key outcome of steps 4 to 7.

3. Material and Methods

3.1 Thermodynamic model

During the research activities, the MEA/CO_2/H_2O system has been modelled according to Deshmukh-Mather formulation and following the framework of Weiland et al.[15]. The MEA model has set the ground for LysK analysis, it has been calibrated with MEA experimental data from Aronu et al.[16], and validated against MEA experimental results from Li and Shen[17].

The thermodynamic framework established from the MEA case has been transferred to describe the LysK system, introducing the following methodology:

- Given the scarce amount of vapor-liquid equilibrium data, the most suitable data-set (41.2%w/w LysK solution) has been selected from Shen et al.[18] to calibrate the model with experimental points from tests that are as close as possible to the identified LysK concentration (43.7w/w LysK solution);
- A Kent-Eisenberg-like approach has been used, enhancing Kent-Eisenberg model with Debye-Hückel activity coefficients in order to provide a synthetic data-set for LysK-Deshmukh-Mather model to be developed in the next step. The enhanced Kent-Eisenberg is able to mathematically fit Shen's experimental data, simultaneously estimating the coefficients of unknown equilibrium constants;
- Exploiting the estimation of the equilibrium constants and an extended pseudo data-set (i.e.: synthetic data) produced from Kent-Eisenberg after Shen's data fitting, a LysK Deshmukh-Mather model has been developed. Both Kent-Eisenberg and Deshmukh-Mather models have been extended to loading values lower than 0.8. Deshmukh-Mather can be used to evaluate vapor-liquid equilibria for the targeted 43.7%w/w LysK solution.

The Deshmukh-Mather model for LysK can be considered predictive and it can be used to evaluate the vapor-liquid equilibria for the selected aqueous solution concentration. Thus, the here mentioned thermodynamic model has been integrated in a proprietary rate-based model to design the absorber column working with 43.7%w/w aqueous LysK.

Beside playing a key role in the design of absorption/stripping units, the thermodynamic model has been relevant to estimate the solvent regeneration duty required from the stripping section. In fact, the solvent regeneration duty is composed of 3 contributions: enthalpy of vaporization, sensible heat and heat of absorption; the Deshmukh-Mather and Van't Hoff equation have been adopted to investigate the contribution provided by the heat of reaction.

More details on the thermodynamic model can be found in the literature produced by the authors[19].

3.2 Rate-based model

Rate-based models for MEA and LysK absorption columns have been developed in Matlab® and are important to draw suitable techno-economic evaluations of the capture technology. Validation of the rate-based model has been carried out applying the model to absorption processes working with aqueous MEA at pilot scale in order to match the experimental outcomes in terms of CO_2 molar fraction in the gas phase, solvent loading as well as solvent and gas temperature profiles.

The validated absorber rate-based model evaluates mass and energy balance of the absorption unit and is inspired by the extensive literature consulted during the workflow [20]–[27]. The adopted approach follows the here-reported steps:

- A model representative of CO_2 and water mass transfer in a gas-liquid counterflow contacting reactor has been developed. The model benefits from Deshmukh-Mather thermodynamic function representative of vapour-liquid equilibria. The rate-based model runs a step-wise calculation of the heat and mass transfer envisaging chemical reaction according to the two-film theory approach;
- The model has been developed both for MEA and LysK absorption systems;
- To validate the approach, pilot plant literature data representative of a CO_2 absorption process with MEA have been reproduced in case of random and structured packing. The validation process has consisted in adapting the column's model to reproduce selected pilot set-ups; specifically, experimental data from Tontiwachwuthikul et al. [21] have been used to assess the simulation results for an absorber operating with random packing, meanwhile test results from Aroonwilas and co-workers [28] have been employed in case structured packing is required.
- A particular point of relevance is that the response of the model with MEA to replicate pilot plant data is based on purely theoretical literature mass and energy transfer correlation (Onda et al., Bravo-Rocha), hence no corrective coefficients have been used (i.e.: IAF-interfacial area factor = 1).

In the present work, a rate-based model has been developed in Matlab® using the two-film theory, and solving energy and mass balance drawn over the infinitesimal control volume reported in Figure 1. The system of differential equation has been solved by implementing a finite difference forward Euler approach.

Design choices and encompassed assumptions are the following:

- the chemical reaction occurs only in the liquid phase, and the bulk is at equilibrium;
- the absorption column is an adiabatic vessel;
- gas/liquid temperature at the interface is equal to the temperature in the liquid bulk;
- Axial dispersion is not accounted for;
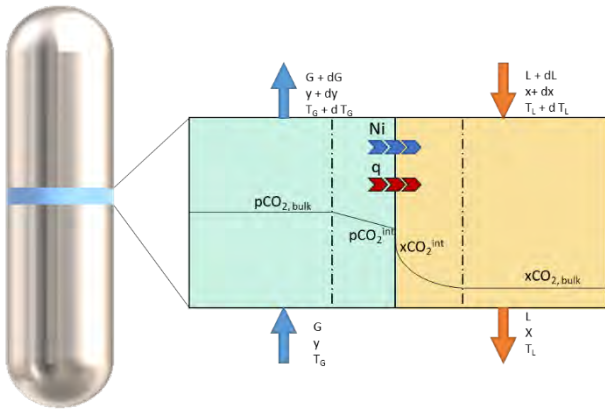- heat and mass transfer interfacial surface areas are equal.

Figure 1: infinitesimal element considered to draw energy and mass balance.

(b)

Figure 2: **(a)** simplified block flow diagram of the NGCC+CO₂ capture unit. The absorption and stripping units are highlighted with a dashed green square; **(b)** focus on the CO₂ absorption/stripping system.

3.3 Conceptual process design

The overall CO_2 conventional capture process reported in Figure 2 has been considered for a complete performance assessment: the full-scale absorption unit treating flue gas from the EBTF-NGCC combined cycle has been designed proposing two rate-based models for the aforementioned cases such as 30%w/w MEA and 43.7%w/w LysK scrubbing. The two columns have been sized and compared to evaluate eventual LysK benefits (e.g: lower required packing volume) with respect to MEA. The estimation of the absorber packing volume has to be carried out in comparative and not absolute terms as no interfacial area factors have been calibrated against pilot-plant data to support the design. In order to investigate the effect of the most relevant operating conditions, a sensitivity analysis has been run over the LysK CO_2 absorption rate-based model. More specifically, the height of the vessel has been calculated as a function of liquid-to-gas ratio (L/G) and lean loading. Reduced column heights which result into lower investment cost for the capture unit have been recorded for low values of lean loading and high L/G, which favor higher driving force. However, lower liquid-to-gas ratios imply savings in terms of circulating solvent (i.e.: lower energy penalty for pumping and regeneration). For this reason, selected L/G and loading operating window have been identified with an iterative approach, preserving an absorber packed volume equivalent to the MEA case and minimizing the reboiler duty.
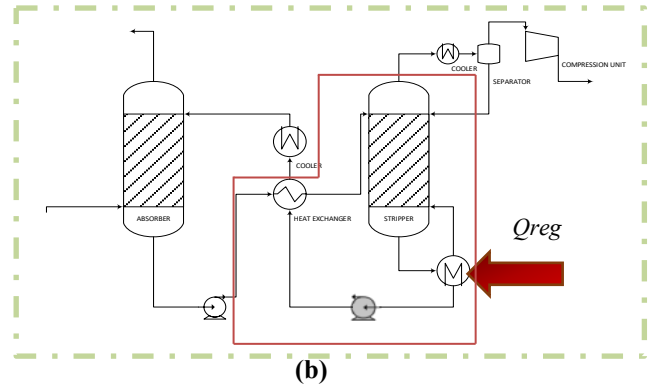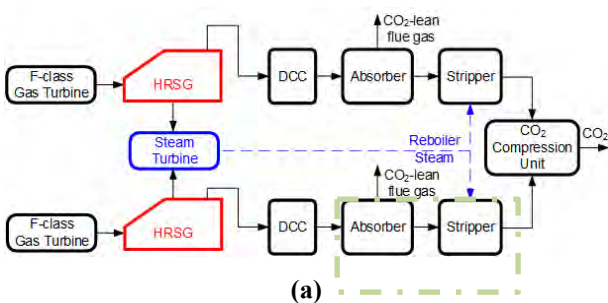

(a)

4. Results and discussion

The results of the absorber unit (2 trains) design for LysK and MEA cases are reported in Table 1.

Table 1: Design of the LysK vs. MEA CO₂ capture unit.

		MEA-EBTF	LysK-Optimized	Unit
SET-UP	Power plant size (no CCS)	829.9	829.9	MWe - net
	Flue Gas Flowrate per train	665	665	kg/s
	Flue Gas Temperature	48.7	48.7	°C
	Flue Gas Pressure	1.06	1.06	bar
	Absorber Packing	Mellapak 250Y	Mellapak 250Y	-
	Absorber Area	242.37	252.0	m2 80%fl.
	CO₂ in Flue Gas	4.02	4.02	mol%
	Solvent concentration	30	43.7	%w/w
	CO₂ capture rate	90	90	%
	Lean Loading	0.257	0.495	mol/mol
	Rich loading	0.466	0.861	mol/mol
	L/G	1.5	1.4	mol/mol
RESULTS	Absorber Packed Height	20.4	19.5	m - from correlation
	Packing volume	4944	4914	m3
	Reboiler Duty	3.96	4.58	MJ/kgCO₂

A performance analysis of the NGCC power plant coupled with the investigated capture systems working with LysK has been carried out and compared against the MEA benchmark. Results are reported in Table 2 and highlight a SPECCA value for LysK based absorption,

which is ~15% higher than the MEA reference case. This energy penalty mainly depends on the higher reboiler thermal duty for solvent regeneration, resulting in a lower net power output from the steam cycle.

Power consumption for the CCS-auxiliaries is higher than the MEA one due to the higher solvent circulating mass flow rate, higher energy penalty for heat rejection and different DCC configuration selected (upstream position of the fan in the LysK case with respect to the direct contact cooler against the configuration of the MEA EBTF benchmark where the fan follows the DCC).

Table 3: Economic indicators for NGCC+LysK and NGCC+MEA.

Indicator	Unit	LysK	MEA
Cost of Electricity	[$/MWh]	72.91	71.54
Cost of CO_2 Avoided	[$/t$CO_2$]	90.16	84.92

Table 2: LysK vs. MEA CO_2 capture unit - performance analysis.

Process section	Quantity	N. Trains (Full Plant)	Units	LysK Value	MEA Value
EBTF Combined Cycle	Fuel input	-	MWLHV	1422.6	1422.6
	Gas Turbine Power Output (2 units)	2	MWe	544.2	544.2
	Steam Cycle Net Power Output w/o CCS	1	MWe	285.7	285.7
	Net Power Output w/o CCS	-	MWe	829.9	829.9
	Steam Cycle Net Power Output with CCS	-	MWe	194.4	207.9
	Gross Power Output with CCS	-	MWe	744.8	752.1
Advanced CCS process	Total Power CO_2 Capture Auxiliaries	-	MWe	26.6	19.9
	Captured CO_2	2	kg/s	73.4	73.4
	Reboiler Thermal duty	2	MWth	336.0	290.7
	Specific thermal consumption for solvent regeneration	-	MJth/kgCO_2	4.58	3.96
	Total Power CO_2 Compression	-	MWe	22.60	22.6
Overall plant performance	Net Power Output with CCS	-	MWe	695.59	709.7
	Power Plant Net Electric Efficiency with CCS	-	%	49%	49.9%
	CO_2 emissions w/o CCS	-	kgCO_2/MWh	351.80	351.8
	CO_2 emissions with CCS	-	kgCO_2/s	8.16	7.87
	CO_2 emissions with CCS	-	kgCO_2/MWh	42.22	39.30
	EBTF Plant Net Electric Efficiency w/o CCS	-	%	58.3%	58.3%
	SPECCA	-	GJLHV/tCO_2	3.84	3.34

Process design and pricing has allowed the evaluation of relevant economic indicators such as cost of electricity (COE) and cost of CO_2 avoided (CCA), which have been calculated with a consistent approach for the MEA benchmark and LysK case. The calculation is based on a factorial costing methodology for equipment pricing as from previous literature works [29].

The economic evaluation of MEA has been carried out with reference to literature information and it has been calculated to be consistent with the LysK case. The results of the techno-economic evaluation reported in Table 3 compare the LysK case against MEA. Standing out, the slightly higher cost of CO_2 avoided of the LysK case with respect to MEA (~90 vs. 85 $/t$CO_2$ respectively). This is mostly related to the higher energy penalty calculated for the amino acid salt case (SPECCA index of the LysK case is ~15% higher compared to the SPECCA index of the benchmark from Table 2).

5. Conclusions

A new techno economic assessment of a CO_2 absorption process working with 43.7%w/w aqueous LysK solution coupled with a reference NGCC (830 MWel without capture) has been carried out. Solvent selection is based on previous experimental campaigns and literature data, and it is justified by the higher environmental compatibility, promising capacity and fast kinetics of the chosen amino acid salt.

The present work has required the definition of a detailed thermodynamic model of the LysK/CO_2/H_2O system and the definition of a proprietary in-house rate-based model of the absorption unit. Moreover, a preliminary evaluation of the solvent regeneration duty based on Van't Hoff equation has been carried out in order to estimate the energy penalty of the capture unit over the combined cycle. Performance results and process cost have been calculated within a dedicated techno-economic assessment, which has provided suitable indicators.

Standing out, the higher cost of CO_2 avoided of the LysK case with respect to MEA, which is mostly related to the

higher energy penalty calculated for the amino acid salt case. Specifically, the SPECCA index of the LysK case is ~15% higher compared to the SPECCA index of the benchmark.

Cost of electricity (72.91 \$/MWh) and CO_2 avoided (90.16 \$/tCO₂) for the alternative case are higher with respect to the benchmark capture unit with MEA (i.e., 71.54 \$/MWh and 84.92 \$/tCO₂).

The evaluation of the energy penalty of the LysK case still requires further experimental evidences. Specifically, additional tests on vapor-liquid equilibria for the low loading region together with calorimeter analysis of the heat of reaction, as well as heat capacity and viscosity measurements of the loaded solvent are recommended.

Moreover, cyclic absorption/regeneration tests and stress testing of the solvent at different temperature and flue-gas composition would allow a better quantification of the degradation effects, consequent required solvent make-up rate and operating cost aimed at performing a more accurate techno-economic assessment of AAS-based capture technology applied to post-combustion CO_2 capture for decarbonized power production.

Acknowledgment

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