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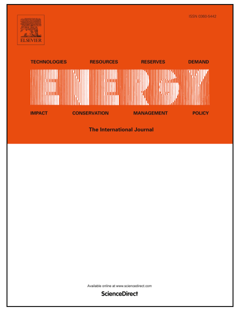

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Ultrasound Intensify CO_2 Desorption from Pressurized Loaded Monoethanolamine Solutions

II. Optimization and Cost Estimation

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ABSTRACT

Optimization and cost estimation were performed for the use of ultrasound for intensifying CO_2 reboiler stripping from lean monoethanolamine (MEA) aqueous solutions at 1.0 barg. This work was based on typical industrial reboiler operation conditions. Experiments were run by intermittently applying ultrasound for CO_2 stripping. A multi-variable data analysis was employed to explain the results and find the optimum for ultrasound operation. The results show that the CO_2 stripping rate by ultrasound is more than 3 times than heat only in the reboiler. A normalized specific energy consumption was defined based on the classic industrial case (4.2 kJ/kg CO_2), and the normalized specific energy consumption 3.6 MJ/kg CO_2 was deduced, showing 14% energy saving. Cost estimations have been conducted using Aspen Plus V9.0 and Aspen In-plant Cost Estimator for the industrial cases with/without the assistance of ultrasound. Total capture cost is 60.2 EUR/t CO_2 and cost saving is 19% when the CO_2 loading of the lean MEA solution can be further decreased to 0.20 mol/mol from 0.44 mol/mol assisted by ultrasound.

1. INTRODUCTION

In current amine-based CO_2 capture plants the operational cost is the major cost element representing approximately 2/3rds of the specific CO_2 capture cost, with the largest single contributor being the energy consumption in the desorber reboiler. There is a significant potential to reduce the total cost of CO_2 capture by improving the CO_2 stripping technology for this type of plant.

Ultrasound enhanced CO_2 stripping could unlock CO_2 capture cost savings. As a mature technology, ultrasound is widely used in other fields [1-3]. Recently, ultrasound was introduced in gas purification for stripping acid gas from loaded solutions, for absorbent regeneration, and reducing degradation of absorbents [4-7].

The major mechanisms for improving the mass transfer are the millions of cavitation bubbles created by ultrasound due to cavitation and nucleation effects. This results in increased interfacial area between gas and liquid. Bubbles form more easily and the activation energy for surface diffusion decreases [8]. The application of ultrasound intensifies the conversion of free CO_2 in the liquid phase to CO_2 (g). Thus, the chemical reaction freeing CO_2 is accelerated as the freed CO_2 is more efficiently removed from the liquid solution. The force driving CO_2 to the bubbles is great because the cavitation bubbles are in a state of vacuum at the beginning. Further effects of ultrasound that enhance mass transfer include formation of micro-streams and vortices, enhancement of bubble growth, acoustic streaming and rectified diffusion [9, 10]. Coalescence of small bubbles will further enhance the gas' ability to rise to the liquid surface, which can also be accelerated by ultrasound through Bjerknes forces [11].

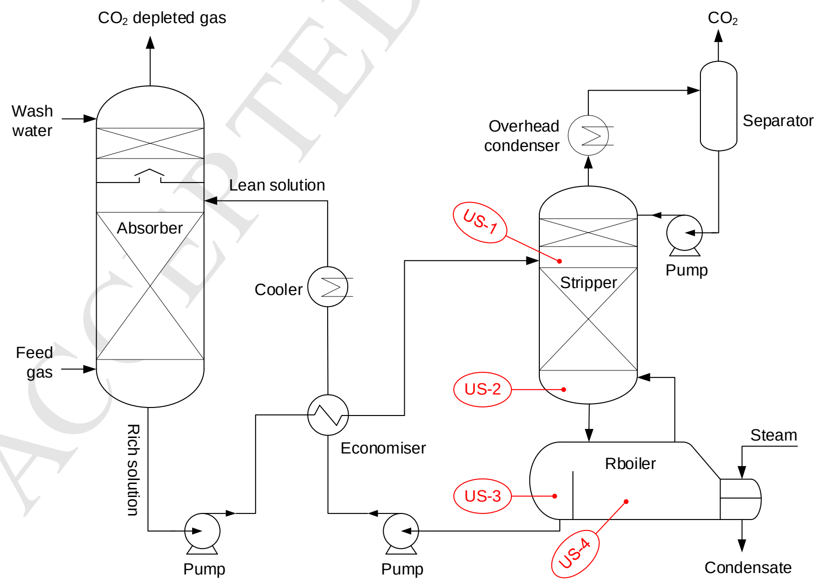

Figure 1. Four potential positions (highlighted in red) for installation of ultrasound to strip CO_2 in an amine-based CO_2 capture plant.

In a typical MEA-based CO_2 capture plant, as illustrated in Figure 1, there are four potential locations where ultrasound could be applied to enhance stripping of CO_2, these are indicated in the flowsheet by four ellipses. Placement in the reboiler (US-4 in Figure 1) is believed to be the most optimal for ultrasound. The reason being that it is in the reboilers that the absorbent is heated by steam to drive the reactions releasing the chemically bound CO_2. From our previous work [12-15] it was found that most ultrasound energy were used for heating rather than forming bubbles to drive free CO_2 (the CO_2 in liquid phase) off when the desorption temperature was low, and this resulted in using electrical energy to replace heat. The release of gas is one of the rate-limiting factors in the desorption process. The reboiler in particular, although there is significant bubbling already, contains liquid with a "rest content" of free CO_2 that struggles to reach the gas phase for reasons stated above. If ultrasound is applied efficiently, limited to enhance the release of CO_2 (aq.) in the absorbent into CO_2 (g) and not heat the solution, the total energy consumption in the reboiler should be reduced. Therefore, to save energy, the best position of ultrasound application in an industrial process could be the position US-4 in Figure 1. In this place, ultrasound could enhance CO_2 stripping from the lean MEA solutions, and achieve leaner MEA solutions than usual. For example, the loading could be reduced from 0.25 mol/mol down to 0.20 mol/mol assisted by ultrasound. This will result in an increased cycling capacity of the absorbent. Hence, the same CO_2 capture rate could be achieved with a lower circulation rate, resulting in a lower energy consumption, because the consumption of steam, energy for pumps and area of heat exchangers is reduced.

In the current work, in order to provide useful information for the industrial applications, the operating conditions of ultrasound intensification of CO_2 stripping was optimized for this lab-scale reboiler based on the results of previous parameter investigation [15], and a cost estimation was performed and compared to an industrial base case. To achieve this goal, an intermittent ultrasound operation was adopted in the experiments at varying pressures up to 1.5 barg at the boiling point of a lean MEA aqueous solution (0.25 mol/mol). Aspen Plus V9.0 and Aspen In-plant Cost Estimator were employed for the cost estimations, and an MEA-based CO_2 capture from a Combined Cycle Gas Turbine (CCGT) natural gas based power plant was chosen as the industrial base case for comparison.

2. EXPERIMENTAL SECTION

2.1. Reagents and Solutions Preparation

Reagent grade MEA with mass fraction purity ≥99.5% from Merck and was used without further purification. Deionized water (purified with an ELGA Purelab Prima 7, resistivity more than 0.05 MΩ-cm) and MEA were mixed in a tank to produce 0.02 m^3 of 30 wt% MEA aqueous solution. This solution was loaded by bubbling CO_2 (purity ≥ 99.995%, produced by AGA Gas, Norway) through 3 sinters in the bottom of the tank with a CO_2 flow rate of 3.3×10^{-5} m^3/s to prepare the desired loading. The CO_2 loading was determined by a density method [14] before the experiments. If the loading was not as expected, e.g. higher than 0.25 mol/mol, more unloaded fresh 30 wt% MEA solution was added and mixed in the solution to obtain the desired loading.

2.2. Experimental Equipment and Procedures

The test rig is same as the rig used in the previous parameter investigation work [15]. To avoid pressure surges in the reboiler when ultrasound was running, the pressure was controlled by a needle valve (8) instead of the on/off solenoid valve used in the previous work. During the experiments, once the pressure in the cell reached the desired pressure, the needle valve (8) was opened and adjusted manually to a stable gas flow. The modified flowsheet of the rig is given in Figure 2. More details of the operational procedure are given in our previous work [15].

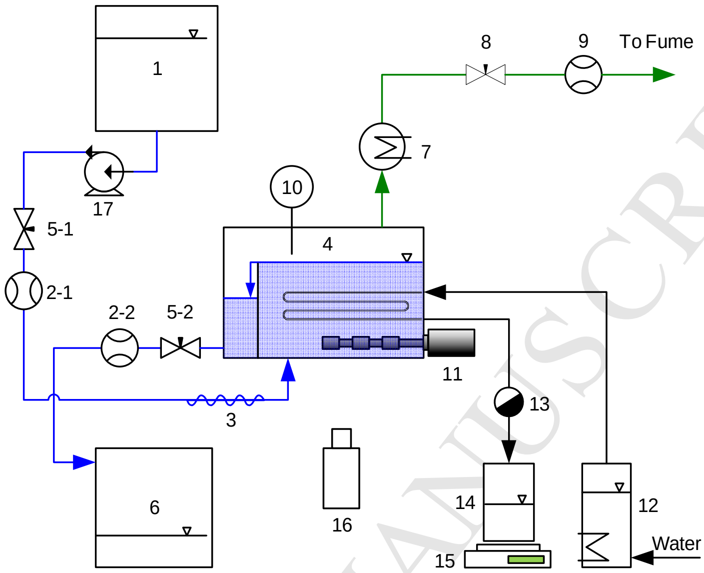

Figure 2. Schematic diagram of the rig used to study ultrasound-enhanced CO_2 stripping from amine solutions.
1, Rich amine feed tank; 2, Liquid flow meter; 3, Preheat unit; 4, Glass kettle reboiler; 5, Needle valve;
6, Lean amine receiving tank; 7, Gas-liquid separator; 8, Regulatable needle valve; 9, CO_2 flow meter;
10, Various sensors (P, T) in liquid and gas; 11, Ultrasound unit; 12, Electric steam generator; 13, Steam trap;
14, Condensed steam receiving tank; 15, balance; 16, High speed camera; 17, Pump.
(Blue lines are liquid phase and Green lines are gas phase)

2.3. Experimental Matrix Design

The influence of various parameters such as pressure, temperature, flow rate of liquid, CO_2 loading, frequency and intensity of ultrasound have been previously investigated and modelled [15]. The results suggested that higher CO_2 loading and frequency of ultrasound are strongly positive with regard to energy saving, and temperature (or pressure), liquid flow rate, intensity of ultrasound are weakly negative with respect to energy saving. Based on our previous investigation, a 28 kHz multi-surface sonotrode and 100 % intensity output of ultrasound were chosen and fixed in this work. The liquid flow rate was not a significant variable and was set to 1.7×10^{-5} m^3/s. CO_2 loading and pressure are very important variables. However, to keep the same conditions as a reboiler of a typical industrial MEA-based CO_2 capture process, CO_2 loading was fixed to 0.25 mol/mol, pressure was set to 1 barg and the steam temperature was set to 130 ºC in this work. The ultrasound running time has a significant effect on the CO_2 stripping rate and energy consumption, and it represents the variable to be optimized.

In this work, ultrasound application was intermittent with an on-time (t_{on}) and an off-time (t_{off}). These were varied for the purpose of energy optimization. The experimental matrix was constructed using a Central Composite Design [16] generated by Design Expert V.9.0.6.2 from Stat-Ease. The design matrix is shown in Table 1. For each variable, a low, a middle and a high level were decided on, and the experiments were carried out such that all variable combinations were systematically covered. Three duplicate experiments on average values of the variables (i.e. No. 3, 7 and 11) were used to quantify the random variation. In the design, the parameter, on-time fraction of ultrasound in a period (φ), was proposed to analyze the efficiency of ultrasound on CO_2 desorption.

Table 1. The designed test matrix for optimization

Run NO.	Factor 1	Factor 2	Ref. factor 1	Ref. factor 2
	t_{on} (s)	t_{off} (s)	$\varphi = t_{on}$/ period	Cycle period (s)
1	3	1	75%	4
2	5	5	50%	10
3	3	3	50%	6
4	5	1	83%	6
5	3	5	38%	8
6	1	1	50%	2
7	3	3	50%	6
8	1	3	25%	4
9	5	3	63%	8
10	1	5	17%	6
11	3	3	50%	6

The predicted optimum conditions can be obtained by both numerical and graphical evaluations, and verification experiments at the suggested optimum were made by three replicates to make a better verification.

3. RESULTS AND DISCUSSION

3.1. Definition of respondents / parameters

To explain the results clearly, three dependent variables related to specific energy consumption and one dependent variable with respect to CO_2 stripping kinetics are defined.

3.1.1. Specific energy consumption in reboiler

The first response used in this analysis, specific energy consumption, E_s, including two cases: $E_{s,H}$ for heat only and $E_{s,US}$ for heat + ultrasound assistance respectively. Both, in unit MJ/kg CO_2, are defined as

$$E_{s,H} = \frac{H_{st} \times \gamma - H_{cw}}{A_{CO2}} \quad \text{and} \quad E_{s,US} = \frac{H_{st} \times \gamma + H_{US} - H_{cw}}{A_{CO2}} \qquad (1)$$

where A_{CO2} is the total rate of CO_2 stripped (in unit mg/s), H_{st} is the energy input into the reboiler from steam, H_{US} is the ultrasound energy input, γ is the energy efficiency of steam heat input. A blank experiment (water in the reboiler) at room pressure when the steam temperature was set to 130 ºC, gave 88% energy efficiency.

3.1.2. Normalized Specific energy consumption

In this work, the vapor (mainly water) of the MEA solution from the reboiler was condensed and not taken to a desorption column for further CO_2 recovery from a richer solution as in an industrial setting. For this reason, the specific steam consumption (with respect to CO_2) observed in the experimental rig cannot be compared directly to the standard industrial yardstick of 4.2 MJ/kg CO_2, (which varies from 3.2 to 5.5 MJ/kg CO_2 reported by literature [17, 18]). To enable comparison to an industry case, a normalization calculation must be used. The hypothesis is as follows;

1. The vapor produced in the lab-scale reboiler could in principle be used for further CO_2 stripping in a desorption column (where most of the CO_2 stripping is normally accomplished). By definition the stripping rate of CO_2 by the vapor in the assumed desorption column is $A_{\mathrm{CO2,v,H}}$ or $A_{\mathrm{CO2,v,US}}$.
2. The energy consumption from pumps etc. is neglected. (This means that the proposed estimation is conservative because the power of the pumps and size of the exchangers can be reduced due to a leaner solution being produced by introducing ultrasound.)
3. The specific energy consumption in lab scale can be scaled up to industrial case linearly.

Based on these assumptions, the following calculation can be made. When the solution is treated by heat only, the rate of CO_2 stripping is $A_{\mathrm{CO2,v,H}}$, and this is defined by the equation:

$$4.2\ (\mathrm{MJ/kg\ CO_2}) = \frac{H_{\mathrm{st}} \times \gamma}{A_{\mathrm{CO2}} + A_{\mathrm{CO2,v,H}}} \tag{2}$$

where the value 4.2 MJ/kg CO_2 is the typical specific energy consumption in a CO_2 capture plant based on 30 wt% aqueous MEA solution, H_{st} is the energy input by steam heat, $A_{\mathrm{CO2,v,H}}$ is the assumed CO_2 stripping rate by the vapor (from the reboiler) into the assumed desorption column with heat treatment only, A_{CO2} is the CO_2 stripping rate observed in the reboiler rig, γ is the energy efficiency of steam heat input. $A_{\mathrm{CO2,v,H}}$ can be calculated by equation (2) when H_{st} and A_{CO2} are measured.

The CO_2 stripping rate in the assumed desorption column by vapor when ultrasound is introduced is defined as $A_{\mathrm{CO2,v,US}}$. In this case, vapor is produced by heat and ultrasound in the reboiler when ultrasound is applied. Assuming that the CO_2 stripping rate in the column is in proportion to the vapor flux, then $A_{\mathrm{CO2,v,US}}$ can be defined as follows (in unit mg/s),

$$A_{\mathrm{CO2,v,US}} = A_{\mathrm{CO2,v,H}} \times \frac{W_{v,\mathrm{US}}}{W_{v,\mathrm{H}}} \tag{3}$$

where $W_{v,H}$ is the weight of vapor from the reboiler produced by heat in unit time, and $W_{v,US}$ is the weight of vapor from the reboiler produced in unit time when steam heat and ultrasound are applied simultaneously.

The normalized specific energy consumptions, $\bar{E}_s$, including $\bar{E}_{s,H}$ or $\bar{E}_{s,US}$, can be calculated by equations (4) and (5).

For heat only,

$$\bar{E}_{s,H} = \frac{\text{Total energy input}}{\text{Total stripped CO}_2} = \frac{H_{st} \times \gamma}{A_{CO2} + A_{CO2,v,H}} \ , \quad (\text{MJ/kg CO}_2) \tag{4}$$

For ultrasound application (heat + ultrasound),

$$\bar{E}_{s,US} = \frac{\text{Total energy input}}{\text{Total stripped CO}_2} = \frac{H_{st} \times \gamma + H_{US}}{A_{CO2} + A_{CO2,v,US}} \ , \quad (\text{MJ/kg CO}_2) \tag{5}$$

3.1.3. Energy saving

The energy saving (η) due to using ultrasound in the process is defined as

$$\eta = \frac{E_{s,H} - E_{s,US}}{E_{s,H}} \times 100\% \tag{6}$$

The above defined energy saving is based on the reboiler as the control element. If based on the control element including a reboiler and a desorption column, the normalized energy saving is

$$\bar{\eta} = \frac{\bar{E}_{s,H} - \bar{E}_{s,US}}{\bar{E}_{s,H}} \times 100\% \tag{7}$$

Where $\bar{E}_{s,H}$ is the typical specific energy consumption of an MEA plant, i.e. 4.2 MJ/kg CO_2.

3.1.4. Improvement of CO_2 stripping rate

The improvement of CO_2 stripping rate by ultrasound is defined as

$$\lambda = \frac{A_{CO2} - A_{CO2,H}}{A_{CO2,H}} \times 100\% \tag{8}$$

Where $A_{CO2,H}$ is the CO_2 stripping rate by heat only (in unit mg/s).

3.2. Experimental Results

The results of the measurements are shown in Table 2, and illustrated in Figure 3 to Figure 6. The raw data of the measurements such as H_{st}, H_{US}, $W_{v,H}$ and $W_{v,US}$ are also listed in the table.

Table 2. The results of the test matrix for optimization

No.	Ultrasound t_{on}				Steam Mass		Condensed vapor $W_{v,US}$ (or $W_{v,H}$)	CO_2 stripping rate A_{CO2}			Specific Energy Consumption $E_{s,US}$ (or $E_{s,H}$)			
	t_{on}	t_{off}	φ	H_{US}	Mass	H_{st}	$W_{v,US}$ (or $W_{v,H}$)	A_{CO2}	$A_{CO2,v,H}$ (or $A_{CO2,v,US}$)	λ	$E_{s,US}$ (or $E_{s,H}$)	η	$\bar{E}_{s,US}$ (or $\bar{E}_{s,H}$)	$\bar{\eta}$
	s	s	%	J/s	g	J/s	mg/s	mg/s	mg/s	%	MJ/kg	%	MJ/kg	%
Heat	**0**	-	**0%**	**0.0**	**22**	**249.8**	**58.8**	**9.5**	**50.0**	-	**12.8**	-	**4.2**	-
1	3	1	75.0%	345.0	23.5	266.8	84.7	50.5	72.0	432%	8.5	34%	5	-19%
2	5	5	50.0%	230.0	23.3	263.0	84.2	54.0	71.5	468%	5.7	55%	3.9	7%
3	3	3	50.0%	230.0	23.2	262.0	83.5	37.0	71.0	289%	8.4	34%	4.6	-10%
4	5	1	83.3%	383.3	23.4	267.3	84.2	49.2	71.5	418%	9.5	26%	5.4	-29%
5	3	5	37.5%	173.3	22.4	254.3	79.3	24.8	67.5	161%	10.2	20%	4.6	-10%
6	1	1	50.0%	230.0	23.5	266.8	84.7	49.8	72.0	424%	6.3	51%	4.1	2%
7	3	3	50.0%	230.0	23.1	265.5	83.0	39.7	70.5	318%	7.9	38%	4.5	-7%
8	1	3	25.0%	115.0	22.1	251.0	78.2	26.2	66.5	176%	7.4	42%	3.9	7%
9	5	3	62.5%	288.3	23.3	264.7	83.5	43.2	71.0	355%	8.5	34%	4.8	-14%
10	1	5	16.7%	76.7	20.9	237.3	71.8	15.3	61.0	61%	10.2	20%	4.1	2%
11	3	3	50.0%	230.0	23.2	266.7	83.0	40.0	70.5	321%	7.8	39%	4.5	-7%
Opti.	***1***	***2***	**33.3%**	**153.3**	**23.3**	**264.7**	**84.2**	**43.2**	**71.5**	**355%**	**5.4**	**58%**	**3.6**	**14%**

Note: Optimized run (Opti.) done based on the optimization work runs no. 1 -11.

From Figure 3, it can be seen that the CO_2 stripping rate increases significantly when assisted by ultrasound. The improvement of CO_2 stripping rate by ultrasound treatment has been observed to reach 300% compared to heat treatment only. This is because ultrasound can produce millions of cavitation bubbles, which greatly increase the interface area of gas and liquid, and also the micro-streams and vortices produced by ultrasound can intensify the gas molecules' mass transfer in the liquid. It is found that the CO_2 stripping rate increases as the on-time fraction of ultrasound (φ) increases, indicating that a longer time of ultrasound treatment results in a higher CO_2 stripping rate. This is reasonable because longer action time of ultrasound would produce more cavitation bubbles and then desorb more CO_2 from the solution. However, the increasing tendency of the CO_2 stripping rate becomes weak and flattens out when φ > 50%. This is because the accumulated free CO_2 in the liquid decreases after ultrasound treatment, and the CO_2 production rate from carbamate is not fast enough, i.e., most of the free CO_2 is stripped by ultrasound at the early stage, and there is little free CO_2 in the solution to strip for the later stage. Free CO_2 tends to accumulate in the ultrasound off-time.

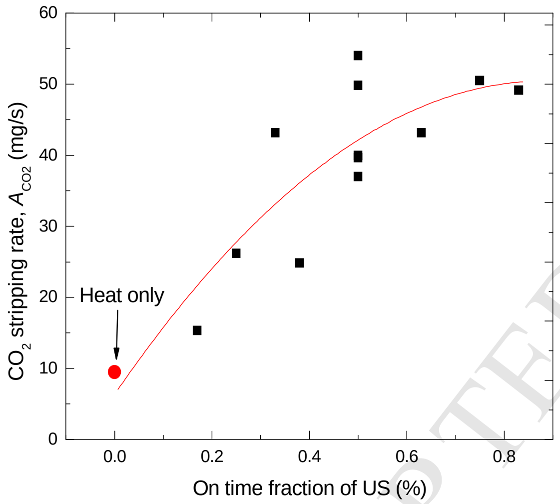

Figure 3. The CO_2 stripping rate (A_{CO2}) as a function of the on on-time fraction of ultrasound (φ)
(the red line is a 2nd order polynomial regression)

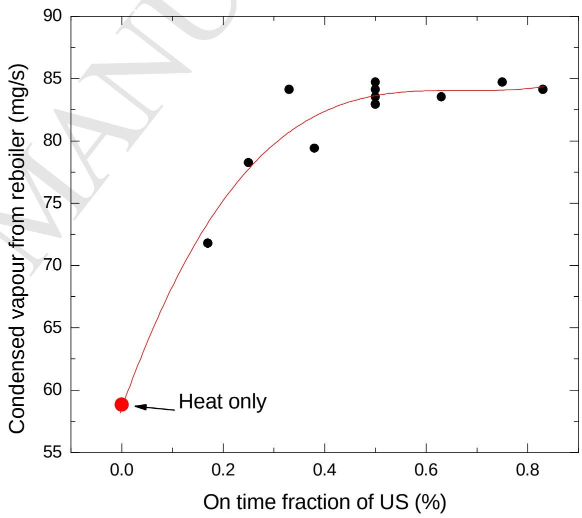

Figure 4. Condensed vapor production rate as a function of the on-time fraction of ultrasound (φ)
(red line is a 3rd order polynomial regression)

It is noted that the experimental data are not a good fit to the 2nd order polynomial curve, the determination coefficient (R^2) of the regression is only 0.77. For example, the dispersion (from 40 to 54 mg/s, the standard deviation = 6.7 mg/s) is high when φ = 50%. This is because the use of φ, which incorporates both t_{on} and t_{off}, as the independent variable in this plot.

The CO_2 hypothetically stripped in the assumed desorption column depends on the amount of vapor from the reboiler. In Figure 4, the flow rate of the vapor from the reboiler as a function of φ can be seen. Similar to the relationship of CO_2 stripping rate and φ, the vapor flow rate increases with an increase of t_{on}, the R^2 of the regression is 0.96, and dispersion of the data is lower. This is because the vapor produced depends on the numbers of growing cavitation bubbles, which are directly affected by t_{on}. However, because the accumulated free CO_2 in the liquid decreases along with the ultrasound application, especially when φ > 50%, lots of cavitation bubbles cannot grow but collapse, and then the increased trend of the condensed vapor produced rate becomes weak as t_{on} increases.

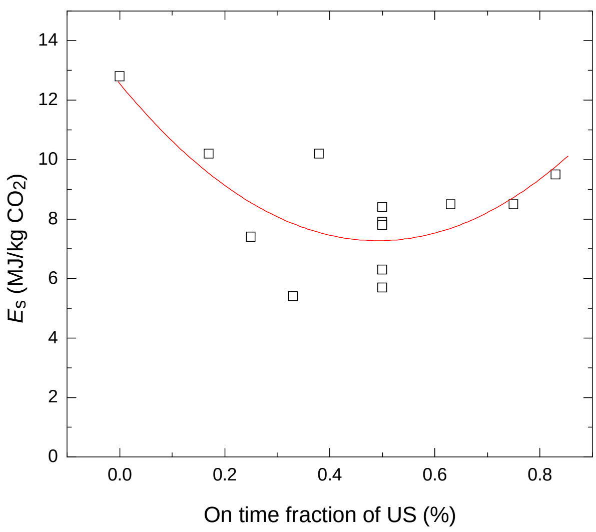

Figure 5. The specific energy consumption (E_s) as a function of on-time fraction of ultrasound (φ) (red line is a 2nd order polynomial regression)

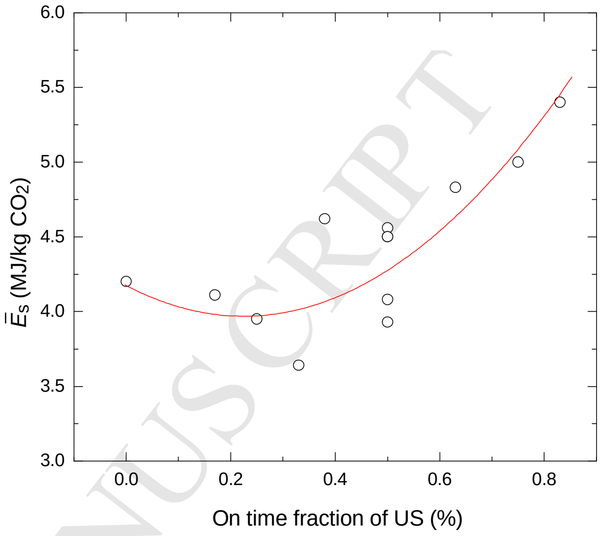

Figure 6. The specific energy consumption $\bar{E}_s$ as a function of on-time fraction of ultrasound (φ) (red line is a 2nd order polynomial regression)

Figure 5 shows the E_s as a function of φ. It can be seen that the E_s decreases significantly with the increase of φ at first, and after reaches minimum value (when φ is in the range 40% - 50%) before increasing again. This indicates that the optimum condition for maximum energy saving is when φ is in the range 40% - 50%. However, it is noted that the experimental data are not consistent when φ is 50%, the standard deviation is about 1.03 MJ/kg CO_2, implying that interaction between t_{on} and t_{off} cannot be neglected (this will be discussed in the section of data analysis). E_s decreases as the on-time of ultrasound application is lower than 40 % mainly because:

1. Cavitation and nucleation effects of ultrasound leads to lower energy consumption to form bubbles compared to heating only;
2. Micro-streams and vortices produced by ultrasound enhance the gas molecules mass transfer in the liquid, and they cause a lower concentration of free CO_2 in the liquid. This is beneficial for CO_2 conversion reaction from carbamate.

The specific energy consumption starts increasing after $\varphi \approx$ 40%. This is because the accumulated free CO_2 in the liquid decreases after ultrasound was applied for a few seconds, and the CO_2 production rate from carbamate is not fast enough. Hence many cavitation bubbles collapse and disappear, this observation manifests that a part of the ultrasound energy input is wasted /converted into other forms such as heating of the liquid.

The normalized specific energy consumption ($\bar{E}_s$) as a function of φ is shown in Figure 6. It can be seen that $\bar{E}_s$ decreases as φ increases of at beginning and reaches a minimum value when φ is in the range of 30% - 40% then starts increasing. The energy consumption increases fast after $\varphi \approx$ 50%. Because the CO_2 stripped in a desorption column depends on the amount of vapor produced from the reboiler, the produced vapor becomes less after $\varphi \approx$ 50% (see Figure 4), and the free CO_2 in the solution becomes less at the later stage.

3.3. Data Analysis and Optimization

Based on the results, a multivariable analysis for optimization was conducted. The respondent variable used in this optimization analysis is $\bar{E}_s$. The average $\bar{E}_s$ for the replicates (exp. 3, 7 and 11) are 4.53 MJ/kg CO_2 (95% confidence interval), and the relative standard deviation (RSD) is 1.3%, implying that the experimental work has a good quality proof. For comparison, the effect on $\bar{E}_s$ by varying t_{on} and t_{off} from 1 to 5 seconds, generates a variation in the $\bar{E}_s$ in the interval (3.9 – 5.4). Statistically speaking, the F-value [16] of the models is 4.8, and the probability is below 4 % that such high F-values can be caused by noise only.

The mathematical model derived from the regression analysis is,

$$\bar{E}_s = 3.332 + 0.517\times t_{on} + 0.175\times t_{off} - 0.106\times(t_{on}\times t_{off}) \quad (9)$$

R^2 of the multivariable regression is 0.8, a comparison of predicted and measured values is shown in Figure 7.

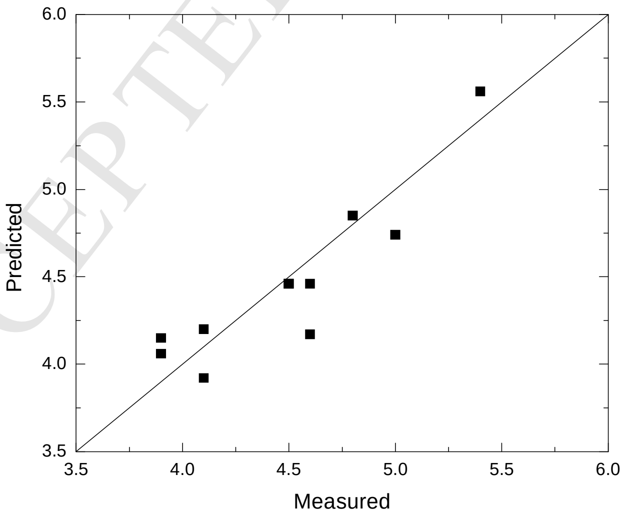

Figure 7. Comparison of predicted and measured values of the specific ultrasound energy consumption $\bar{E}_s$

Both non-linear effects and an interaction effect between the two independent variables are accounted for in this model. In the model, the t_{on} and the interaction term "$t_{on}\times t_{off}$" are statistically significant with p-values < 0.05. The term t_{off} is not statistically so significant in itself, but its interaction with the term t_{on} is. In practice, this implies that the effect of t_{on} on $\bar{E}_s$ is dependent on the level of t_{off}. This can be seen in Figure 8, which represents a contour plot showing the effect upon $\bar{E}_s$ from varying the t_{on} and t_{off} within their ranges from 1 to 5 seconds.

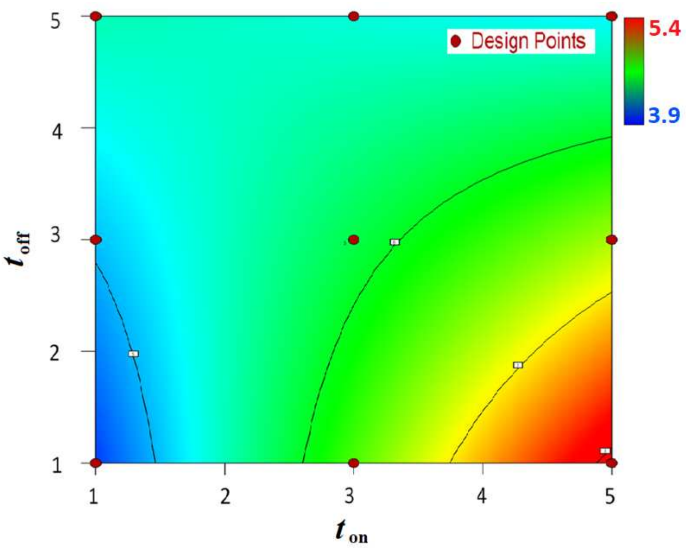

Figure 8. The contour plot showing the effect of t_{on} and t_{off} on the specific ultrasound energy consumption $\bar{E}_s$. (blue means lower $\bar{E}_s$, red means higher $\bar{E}_s$).

Figure 8 illustrates by colors that $\bar{E}_s$ depends on the variables t_{on} and t_{off}. In the figure, a darker blue means lower $\bar{E}_s$, more red means higher $\bar{E}_s$. It is seen that the left lower side area (dark blue) represents an area of optimum values for the independent variables. It shows that when t_{on} is high, increasing t_{off} become significant for decreasing $\bar{E}_s$. The data show a weak saddle structure where $\bar{E}_s$ have lower values both in the upper right and in the lower left. In the variable range investigated the lowest value range of $\bar{E}_s$ are found where t_{on} is 1 s and t_{off} is 1 to 2 s respectively. The structure of the values in Figure 8 is of such a nature that it would clearly be interesting to investigate even lower t_{on} than 1 s. This, however, could not be done at this stage because of the operational limitation of the ultrasound device and the experimental rig. Therefore, an optimum condition, t_{off} was set to 2 s and t_{on} is 1 s was predicted.

Using equation (9) it was predicted that an optimum condition is t_{on} = 1 s and t_{off} = 2 s where it is predicted that $\bar{E}_s$ = 3.99 MJ/kg CO_2 when CO_2 loading is 0.25 mol/mol. Experimental verification of this, using 3 parallels, gave $\bar{E}_s$ = 3.6 MJ/kg CO_2 which is better than predicted.

4. CO_2 MASS BALANCE AND ENERGY BALANCE ANALYSIS

4.1. CO_2 mass Balance

CO_2 mass balance calculation was conducted by comparing the amount of change of CO_2 in the liquid phase and the gas phase during the measuring time. The results are shown in Table 3 showing that the average deviation is 4%, and the maximum deviation is 9% (91% matched) in the measurements. The results manifest that the measurements error is in the acceptable range.

Table 3. CO_2 Mass Balance Calculation

No.	CO_2 in liquid side			CO_2 in gas side		Deviation %
	Rich loading mol/mol	Lean loading mol/mol	CO_2 stripped out g	A_{CO2} mg/s	CO_2 stripped in g	
Heat only	**0.25**	**0.248**	**1.64**	**9.5**	**1.61**	**1.3**
1	0.25	0.239	8.99	50.5	8.57	4.7
2	0.25	0.238	9.81	54.0	9.18	6.4
3	0.25	0.242	6.54	37.0	6.29	3.9
4	0.25	0.240	8.18	49.2	8.35	2.1
5	0.25	0.245	4.09	24.8	4.23	3.5
6	0.25	0.239	8.99	49.8	8.46	5.9
7	0.25	0.242	6.54	39.7	6.73	3.0
8	0.25	0.244	4.91	26.2	4.45	9.2
9	0.25	0.241	7.36	43.2	7.35	0.2
10	0.25	0.247	2.45	15.3	2.62	6.6
11	0.25	0.242	6.54	40.0	6.79	3.8
Opt.	**0.25**	**0.241**	**7.36**	**43.2**	**7.35**	**0.2**
					Average:	4%

Note: 1) Error cause from loading measurement, and CO_2 flow rate measurement by flow meter.
2) Lean solution samples were taken from the reboiler after treatment, not from the lean tank.
3) Total liquid inventory during the measurement was 3.8 kg.

4.2. Energy Balance

Because the liquid was pre-heated from 23 ºC to 110 ºC, and will be cooled from 121.4 ºC to 23 ºC after the measurement, this sensible heat (from 23 to 110 °C) will not be considered in the calculation. To simplify the problem, here the measurement treated by heat only was used as an example, assuming the reboiler as the control element, the energy input into the system H_{in} is

$$H_{\mathrm{in}} = H_{out} \tag{10}$$

When CO_2 stripping without ultrasound (heat treatment only)

$$H_{in} = H_{st} \tag{11}$$

and the energy output from the system H_{out} is

$$H_{\mathrm{out}} = E_{\mathrm{cw}} + E_{\mathrm{less\_preheat}} + E_{\mathrm{reb\_heat}} + E_{\mathrm{de\_CO2}} + E_{\mathrm{loss}} \tag{12}$$

where E_{cw} is the energy consumption of the condensation of the vapor produced in the reboiler, $E_{\mathrm{less\_preheat}}$ is the energy consumption due to the liquid temperature difference between in the reboiler and the inlet of liquid, $E_{\mathrm{reb\_heat}}$ is the energy consumption due to the temperature difference of the liquid in the reboiler before and after measurement, E_{de_CO2} is the energy consumption due to CO_2 desorption from liquid, the theoretical enthalpy of desorbed CO_2 from 0.25 loading 30% MEA solution at 120 °C is 2.6 MJ/kg CO_2 [19]. E_{loss} is the heat loss of the rig to environment.

The energy balance based on the reboiler as a control element show that the energy consumption is 82% of the energy input. Most of the energy losses were caused by the heat loss of the rig and error of condensed steam collection, considering these factors, 82% energy match is acceptable for this laboratory test. More detail of the calculation can be found in the supporting information.

5. COST ESTIMATION

An MEA-based CO_2 capture plant (see the Supporting information) is used as the basis of assessing the cost savings potential of ultrasound implementation. In this case, CO_2 is captured from the flue gas from a CCGT natural gas based power plant. Two cases, one with and one without ultrasound implementation are cost consistently estimated.

5.1. Assumptions and Basic Data Calculations

The following sections cover the assumptions used as basis for the cost estimation, both technical and economical.

5.1.1. Specific Energy Expected Because of Ultrasound

From Table 2 it is observed that the normalized specific energy for desorption of CO_2 by heat only obtained from measurements in the reboiler rig is reported [17, 18] as an average value 4.2 MJ/kg CO_2. Based on the value 4.2 MJ/kg CO_2, an optimized normalized specific energy for desorption of CO_2 by (ultrasound + heat) is 3.6 MJ/kgCO_2 in our lab test, and the energy saving is 14% compared to heat only treatment. This saving is conservative because this energy saving does not include the reduction of size of heat exchangers, reboilers and pumps with reduced energy due to the reduction of liquid flow, caused by a leaner regenerated absorbent.

In this case, the stripped CO_2 can be divided into two parts when ultrasound is applied:

- Part 1, in the assumed desorption column, the CO_2 stripping from rich loading to normal lean loading, e.g. from 0.44 to 0.25, $\Delta\alpha$=0.19, and the CO_2 is stripped by the vapors from the reboilers produced by steam heat and ultrasound. More vapors (mainly water) will be produced due to the assistance of ultrasound.
- Part 2, in the reboiler, the CO_2 stripping from normal lean loading to extra lean loading, i.e. from 0.25 to 0.20, $\Delta\alpha$=0.05, and the energy input is by steam and ultrasound. Assuming an optimum ultrasound application in the reboiler, and;
    - Based on the optimized experiment (in Table 2), 153.3 J/s from ultrasound input, 264.7 J/s from heat input, total stripped CO_2 is (43.2 + 71.5=) 114.7 mg/s.

- For heat treatment only, the stripped CO_2 is (9.5 + 50 =) 59.5 mg/s.
- Then the extra stripped CO_2 due to ultrasound is (114.7 – 59.5=) 55.2 mg/s.
- From the optimized experiment, the normalized total specific energy consumption is 3.6 MJ/kg CO_2. In this case, it was assumed that the specific energy consumption of theoretical desorption enthalpy (2.6 MJ/kg CO_2) [19] is provided by steam (H_{st}), and the rest energy consumption (3.6 - 2.6) MJ/kg CO_2 = 1000 kJ/kg CO_2 is contributed by ultrasound.
- A simulation results of the base case by Aspen Plus show that the CO_2 loading decrease is 0.05 before and after the reboiler (see the supporting information). In this work, the CO_2 stripping rate in the reboiler assisted by ultrasound (43.2 mg/s) is 4.5 times of heat treatment only (9.5 mg/s), indicating that it could achieve $\Delta\alpha$=0.05 from 0.25 to 0.20 when assisted by ultrasound in the industrial reboiler.

5.1.2. Implementation of Ultrasound for Cost Estimation Purposes

Possible locations for implementations of ultrasound in the process were described in the introduction. It was observed in the experiments that fewer bubbles are formed from the steam pipe for a while after ultrasound treatment. This means that the ultrasound affects the formation of bubbles on the surface of the steam pipe. Therefore, we suggest that the ultrasound sonotrode should be installed at some distance from the steam pipe.

Based on our experiments, we use the following implementation for cost estimation purposes: Sonotrodes mounted in the main section of the stripper reboiler as illustrated in Figure 9. The sonotrodes will be distributed evenly along the bottom of the reboiler to get maximum effective area for cavitation. The details of ultrasound equipment can be found in the Supporting information.

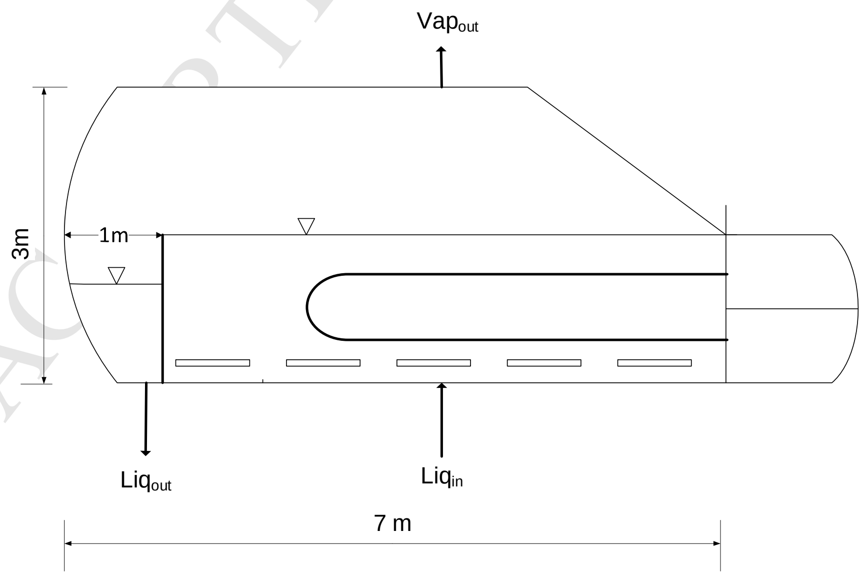

Figure 9. Illustration of an industrial kettle reboiler, with 5 ultrasound sonotrodes on the bottom.

5.1.3. Effect of CO_2 Loading

There is an increased cyclic capacity for the absorbent based on the reduced lean loading to 0.20 mol/mol from 0.44 mol/mol ($\Delta\alpha_{US}$=0.44 - 0.20=0.24), such that the flow rate can be reduced from the base case ($\Delta\alpha_{BC}$=0.44 - 0.25=0.19), as follows:

$$\frac{\Delta\alpha_{US} - \Delta\alpha_{BC}}{\Delta\alpha_{US}} = \frac{0.24 - 0.19}{0.24} = 0.21, \qquad i.e. 21\%\ reduced\ flow$$

This also implies that 21% of the CO_2 recovered in the process is the extra CO_2 desorbed caused by ultrasound. The solvent flow rate reduction will affect several of the components listed in Table 4. Some of the equipment sizes and energy consumptions are governed by the gas flow, while others are governed by the solvent (MEA) flow. It is the ones governed by the solvent flow that are affected and some more than others. Five components have been identified to undergo the most significant changes including the reboiler, and they are (basic case);

- H-3 Lean/rich solution heat exchanger
- H-4 Lean amine cooler
- Reboiler
- Pump, P-3, the rich solution pump
- Pump, P-4, the lean solution pump

The capacities and reductions of these units because of the reduction of absorbent are calculated as shown in Table 4.

5.1.4. Potential Steam Savings

With respect to CCGT, the base case is that there are 15 reboilers and 50.85 kg CO_2/s capacity in total. Based on the 21% reduction of absorbent flow, the number of reboilers can be reduced to 12. The main CO_2 stripping is from loading 0.44 to 0.25 (part 1), $\Delta\alpha = 0.19$ and main stripped by the solution vapor produced in the reboiler, and some extra vapor produced because of the application of ultrasound. In part 2, the loading is from 0.25 to 0.20, $\Delta\alpha = 0.05$.

- Part 1, because the ultrasound could increase the CO_2 stripping in this part, from the experiments ("opti.", "heat") in Table 2, the increase of CO_2 stripping rate is (71.5 - 50)/50=43% due to ultrasound applied, conservative assumption is 22% can be realized in an industrial unit (50% of experimental results). Then the total stripped CO_2 from Part 1 would be

$$50.85 \times \frac{0.19}{0.05 + 0.19} \times (1 + 22\%) = 49.11\,\frac{\mathrm{kgCO_2}}{\mathrm{s}}$$

Where 49.11 × 22% = 10.8 kg/CO_2 is contributed by introduction of ultrasound for the part 1.

- Part 2, the total CO_2 stripped is (50.85 - 49.11=) 1.74 kg CO_2/s
  - Specific energy consumption from ultrasound is 1000 kJ/kg CO_2
  - The total energy input from ultrasound equipment (of 12 reboilers) is

$$1000\,\frac{\mathrm{kJ}}{\mathrm{kg\ CO_2}} \times \left(1.74\,\frac{\mathrm{kg\ CO_2}}{s} \times \frac{55.2}{114.7}\right) = 838\ kW$$

Where 55.2 mg/s is the CO_2 stripping due to ultrasound, 114.7 mg/s is the total CO_2 stripping by heat and ultrasound in the reboiler.

Assuming that one ultrasound sonotrode unit can supply 16 kW, we need

$$\frac{838\ kW}{16\ kW \times 12} \approx 5\ (ultrasound\ units\ per\ reboiler)$$

Commercial ultrasound devices have been identified and available for industrial purposes on a medium and large scale.

It is assumed 2.6 MJ/kg CO_2 of the 3.6 MJ/kg CO_2 (the normalized total specific energy consumption) is the heat needed for theoretic desorption. The steam needed for the normal case is conservatively 4.2 MJ/kg CO_2. Because the heat transfer in the reboilers cannot be 100%, the steam reduction is

$$(4.2 - \frac{2.6}{88\%})/4.2\ = 30\%$$

Where 88% is the heat efficiency in our lab rig.

5.2. Changes in Equipment

In Table 4, the base case equipment list is shown, note that CO_2 compression is not included. The flowsheet is shown in Supporting information. The heat exchangers area, reboilers and pump sizes are reduced because of the reduction of absorbent circulation. Ultrasound equipment is added in the process.

Table 4. Base case equipment list, with changes due to ultrasound application.

Tag nr.	Description	Unit	Size	Amount	Change	New size	New amount
H-1	Cooling water cooler	m²	5 435	6			
H-2	Wash water cooler	m²	7 350	8			
H-3	Lean/rich solution HE	m²	33 865	34	7 044	26 821	27
H-4	Lean solution cooler	m²	1 520	2	316	1 204	2
H-5	Stripper condenser	m²	1 665	2			
H-6	Stripper reboiler	m²	14 160	15	2 945	11 215	12
V-1	DCC	m³	7 270	2			
(V-1)	Packing DCC	m³	1 450	2			
V-2	Absorber shell	m³	16 290	2			
(V-2)	Packing (absorber)	m³	6 790	2			
V-3	Water wash (absorber)	m³	1 250	2			
(V-3)	Packing (water wash)	m³	940	2			
V-4	Reclaimer	m²	550	1			
V-5	Stripper shell	m³	1 270	1			
(V-5)	Packing (stripper)	m³	510	1			
V-6	Separator	m³	16	1			
V-7	Lean solvent tank	m³	1 180	2			
V-8	Amine supply tank	m³	200	1			
V-9	Amine/chemicals mixing tank	m³	8	1			
V-10	Amine sump	m³	40	1			
P-1	DCC water pump	kW	320	1			
P-2	Wash water circ. pump	kW	450	1			
P-3	Rich solution pump	kW	890	2	185	705	2
P-4	Lean solution pump	kW	890	2	185	705	2
P-5	Condenser return pump	kW	22	1			
P-6	Amine storage tank pump	kW	22	1			
P-7	Water injection pump	kW	22	1			
P-8	Amine Fill pump	kW	12	1			
P-9	Water makeup pump	kW	22	1			
P-10	Condensate pump	kW	22	1			
P-11	Amine Sump pump	kW	22	1			
K-1	Flue gas fan	kW	5 075	3			
F-1	Filter package	-		1			
X-2	Soda ash package	-		1			
	ultrasound equipment	kW				838	60

5.3. Results of Cost Estimation

The cost estimates without (base case) and with the ultrasound have been done using the same flowsheet and equipment list, see Table 4. In the currently chosen configuration, five ultrasound sonotrodes at 16 kW is implemented in each kettle reboiler, as illustrated in Figure 9. The installation cost of the ultrasound sonotrodes was included in the kettle reboiler costs, resulting in an increased unit cost and installation factor compared to base case. The operational cost (electricity) was added separately.

The assumptions in the cost estimates were kept the same for both estimates. The two most important ones are related to energy cost; steam and electricity. When implementing ultrasound, a part of the steam consumption is replaced by electricity. Therefore, the cost of these elements is important, and in the current estimates their prices are as follows;

- Steam, 21.3 EUR/t
- Electricity, 0.05 EUR/kWh

The CAPEX of one ultrasound sonotrode unit was based on a quote from a supplier, 2200 EUR for one 2 kW sonotrode. The cost of one 16 kW sonotrode was estimated 8850 EUR ($=2200\times(16/2)^{0.65}$).

The cost of the ultrasound equipment is included in the reboiler cost at an increased unit cost and installation cost. This is likely to be the case for a new build where the ultrasound sonotrodes will be a highly integrated part of the reboiler and delivered as a package.

The results, capture cost only, CO_2 compression is not included, are shown in Table 5. The results are divided into CAPEX, OPEX and total capture cost, and the percentage improvement is included for each. The result showed a slight increase in CAPEX, with a more pronounced reduction in OPEX, with current assumptions.

Table 5. The results from the cost estimation, reference year 2018.

	CAPEX EUR/t CO_2	OPEX EUR/t CO_2	Total capture cost EUR/t CO_2
Base case	11.2	47.6	58.8
Process with ultrasound	10.8	36.6	47.4
Cost saving	4%	23%	19%

As briefly discussed above, the results are dependent on the cost of utilities. Low steam cost and high electricity costs will favor the base case, while the opposite will favor the ultrasound modified base case. It should also be mentioned that estimates for the modified base case are conservative, primarily regarding the electricity consumption, but also likely concerning ultrasound sonotrodes' CAPEX.

It is noted that the cost estimation is sensitive to the CO_2 loading of the lean solution, i.e., leaner solution leads to a higher capacity of the solution and then less MEA inventory.

6. CONCLUSIONS AND RECOMMENDATIONS

The use of ultrasound to improve the desorption of CO_2 from lean loaded solution was investigated covering a typical industrial case of a reboiler pressure of 1.0 barg. A test with varying ultrasound exposure times was performed from which the optimum times of t_{on} =1 s and t_{off} = 2 s was found in current lab-rig. The results show that the enhancement of CO_2 stripping by ultrasound is significant and a 300% improvement is obtained, indicating that the mass transfer can be intensified by ultrasound. This energy saving per kg of CO_2 is a direct consequence of the larger amount of CO_2 produced using ultrasound. To be able to compare with the typical MEA-based CO_2 plant, a normalized specific energy was defined based on industrial case (4.2 kJ/kg CO_2), it was deduced that the specific energy consumption (normalized) in an industrial reboiler is 3.6 MJ/kg CO_2, and the energy saving reached 14% when the extra vapor (mainly water) by ultrasound that enters an assumed desorption column for further CO_2 stripping is considered.

Cost estimations have been conducted by using Aspen Plus V9.0 and Aspen In-plant Cost Estimator for the industrial cases with/without the assistance of ultrasound. Total capture cost including CAPEX and OPEX is 60.2 EUR/t CO_2 and cost saving is 19% when the CO_2 loading in the MEA solution can be decreased to 0.20 mol/mol from 0.44 mol/mol assisted by ultrasound.

■ ASSOCIATED CONTENT

Supporting Information

The Energy balance calculations and the flow sheet of CO_2 capture plant for CCGT natural gas based power plant can be found in the supporting information.

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NOMENCLATURE

Abbreviations

MEA = monoethanolamine

CCGT= Combined Cycle Gas Turbine

Parameters and Variables

A_{CO2}, is the CO_2 stripping rate in the reboiler, mg/s

$A_{CO2,H}$, is the CO_2 stripping rate by heat only, mg/s

$A_{CO2,US}$, is the CO_2 stripping rate by heat + ultrasound, mg/s, $A_{CO2,US} = A_{CO2} - A_{CO2,H}$

$A_{CO2,v}$, is the CO_2 stripping rate of by the vapor in the assumed desorption column, mg/s

$A_{CO2,v,H}$, is the CO_2 stripping rate by the vapor in the assumed desorption column when heat treatment only, mg/s

$A_{CO2,v,US}$, is the stripping rate of CO_2 further stripped in the assumed desorption column by vapor when ultrasound is introduced, mg/s

E_{cw}, is the energy consumption of the vapor produced in the reboiler condensed to liquid state, MJ/min

E_{de_CO2}, is the energy consumption due to CO_2 desorption from liquid, MJ/min

$E_{less\_preheat}$, is the energy consumption due to the liquid temperature difference between in the reboiler and the inlet of liquid, MJ/min

E_{loss}, is the heat loss of the rig in the measurement, MJ/min

E_{reb_heat}, is the energy consumption due to the temperature difference of the liquid in the reboiler before and after measurement, MJ/min

H_{in}, is the energy input into the control element, MJ/min

H_{out}, is the energy output from the control element, MJ/min

H_{st}, is the energy input by steam heat, MJ/min

H_{US}, is the ultrasound energy input, MJ/min

E_s, is the specific energy consumption in reboiler, MJ/kg CO_2

$E_{s,H}$, is the specific energy consumption when CO_2 stripping only by heat in the experiment, MJ/kg CO_2

$E_{s,US}$, is the specific energy consumption when CO_2 stripping assisted by ultrasound in the experiment, MJ/kg CO_2

$\bar{E}_s$, is the normalized specific energy consumption, MJ/kg CO_2

$\bar{E}_{s,H}$, is the typical specific energy consumption of a MEA plant, i.e. 4.2 MJ/kg CO_2

$\bar{E}_{s,US}$, is the normalized specific energy consumption when CO_2 stripping assisted by ultrasound, MJ/kg CO_2

t_{on}, on-time of ultrasound in a period, s

t_{off}, off-time of ultrasound in a period, s

$W_{v,H}$, is the weight of vapor produced by heat only in unit time, mg/s

$W_{v,US}$, is the weight of vapor produced in unit time when ultrasound is introduced, mg/s

Greek Symbols

α, is the CO_2 loading in MEA aq. solution, mol CO_2/mol MEA

α_{rich}, is the CO_2 loading in rich MEA aq. solution, mol CO_2/mol MEA

α_{lean}, is the CO_2 loading in lean MEA aq. solution, mol CO_2/mol MEA

$\Delta\alpha_{BC}$, is the CO_2 loading change MEA aq. solution from rich to lean solution of the base case, mol CO_2/mol MEA

$\Delta\alpha_{US}$ is the CO_2 loading change MEA aq. solution from rich to lean solution of the base case when ultrasound applied, mol CO_2/mol MEA

γ, is the energy efficiency of steam heat input, 88% measured from a blank experiment (water in the reboiler) at room pressure and the steam temperature is set to 130 ºC

λ, is the improvement of CO_2 stripping rate, %

φ, is the “on” time fraction of ultrasound in a period, %

η, is the energy saving due to using ultrasound in the reboiler process, %

$\bar{\eta}$, is the normalized energy saving for a process, %

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Highlights:

1. Ultrasound is introduced to intensify CO_2 stripping from loaded amine solutions.
2. On/off time of ultrasound is optimized in simulated industrial conditions.
3. Energy saving 14% and cost saving 19% are achieved by ultrasonic assistance.