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Economically efficient operation of CO_2 capturing process part I: Self-optimizing procedure for selecting the best controlled variables

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ABSTRACT

We study optimal operation of a post-combustion CO_2 capturing process where optimality is defined in terms of a cost function that includes energy consumption and penalty for released CO_2 to the air. Three different operational regions are considered.

In region I, with a nominal flue gas flowrate, there are two optimally unconstrained degrees of freedom (DOFs) and the corresponding best self-optimizing controlled variables (CVs) are found to be the CO_2 recovery in the absorber and the temperature at tray no. 16 in the stripper. In the region II, with an increased flue gas flowrate, the heat input is saturated and there is one unconstrained DOF left. The best CV is temperature at tray no. 13 in the stripper. In region III, when the flue gas flowrate is further increased, the process reaches the minimum allowable CO_2 recovery of 80% and there is no unconstrained DOF. We have then reached the bottleneck and a controller is needed to adjust the feed flowrate to avoid violating this minimum.

The exact local method and the maximum gain rule are applied to find the best CVs in each region.

1. Introduction

Fossil fuel power plants are one of the major sources of world energy. However, the combustion of fossil fuels invariably produces the greenhouse gas CO_2 that causes global warming. Due to the effect of CO_2 emissions on global warming, different countries are starting to impose taxes or regulations on the amount of CO_2 released to the air.

We consider an absorption/stripping amine process to remove most of the CO_2 from the combustion flue gas stream. Fig. 1 shows a typical CO_2 capturing process. In the first column (absorber), CO_2 in the flue gas is absorbed into a 30 wt% monoethanolamine (MEA) solution at atmospheric pressure.

The rich amine with about 5.3 mol% CO_2 is pumped to the second column (stripper) after preheating it by the recycled lean cooling amine solution which contains about 2.3 mol% CO_2. In the stripper, CO_2 is stripped off at a pressure lower than 2 bar.

The energy consumption in the reboiler of the stripper is very large; typically around 15–30% of the net power generated of a coal-fired power plant [1]. To operate the process optimally in presence of different disturbances, Skogestad [2] has developed a systematic procedure based on self-optimizing control to find the best controlled variables (CVs). The idea is that the process is indirectly kept close to its optimum when the CVs are held constant at their optimal nominal setpoints.

The proposed plantwide control procedure consists of two main parts:

I. A top-down analysis to optimize the process for various disturbances and identify primary self-optimizing controlled variables.
II. A bottom-up analysis to identify secondary controlled variables and find the structure of the control system (pairing).

In the present work, the top-down analysis is performed for three different operational regions for the CO_2 capturing process. An operation region is here defined as a region with a given set of active constraints. In general, we want to find good control policies for all active constraints regions.

Region I: the flowrate of the flue gas is at its nominal value and there are two unconstrained degrees of freedom.

Region II: the flowrate of the flue gas is increased so that the reboiler duty saturates and there is one unconstrained degree of freedom left.

Region III: the flowrate of the flue gas is increased further so that the minimum allowable CO_2 recovery is reached and the remaining unconstrained degree of freedom is saturated.

In a previous study [3], we designed the control structure using this method when a fixed amount of CO_2 (90%) was removed from the flue gas stream and the objective was to minimize the

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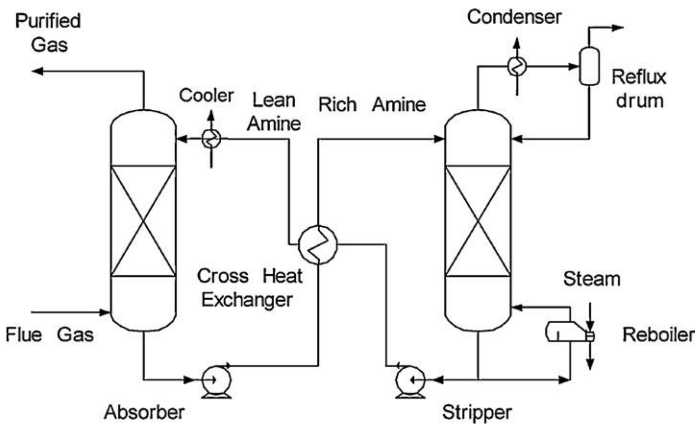

Fig. 1. Flowsheet of typical absorption/stripping CO_2 process [1].

energy usage. In this study, there is a tax on the CO_2 released to the air which makes it optimal to remove more of the CO_2 to get an optimal trade off between CO_2 removal and energy usage. The UniSim [4] process simulator with the Amine thermodynamic package is used for the simulations. The simulation is based on data from a pilot plant [5] that recovers around 1 ton/h CO_2, corresponds to that produced in a 1 MW coal-fired power plant. The nominal optimal data for the process are given in Fig. 2. The feed flue gas at 48 °C is assumed to be saturated with water.

2. Top down analysis: self-optimizing control of CO_2 capturing process

2.1. Region I: flowrate of flue gas is given

The steps of the top-down analysis are as follows.

2.1.1. Step 1. Define objective function and constraints

In general, the cost function should be the negative profit of the process. However in cases where we consider only part of the

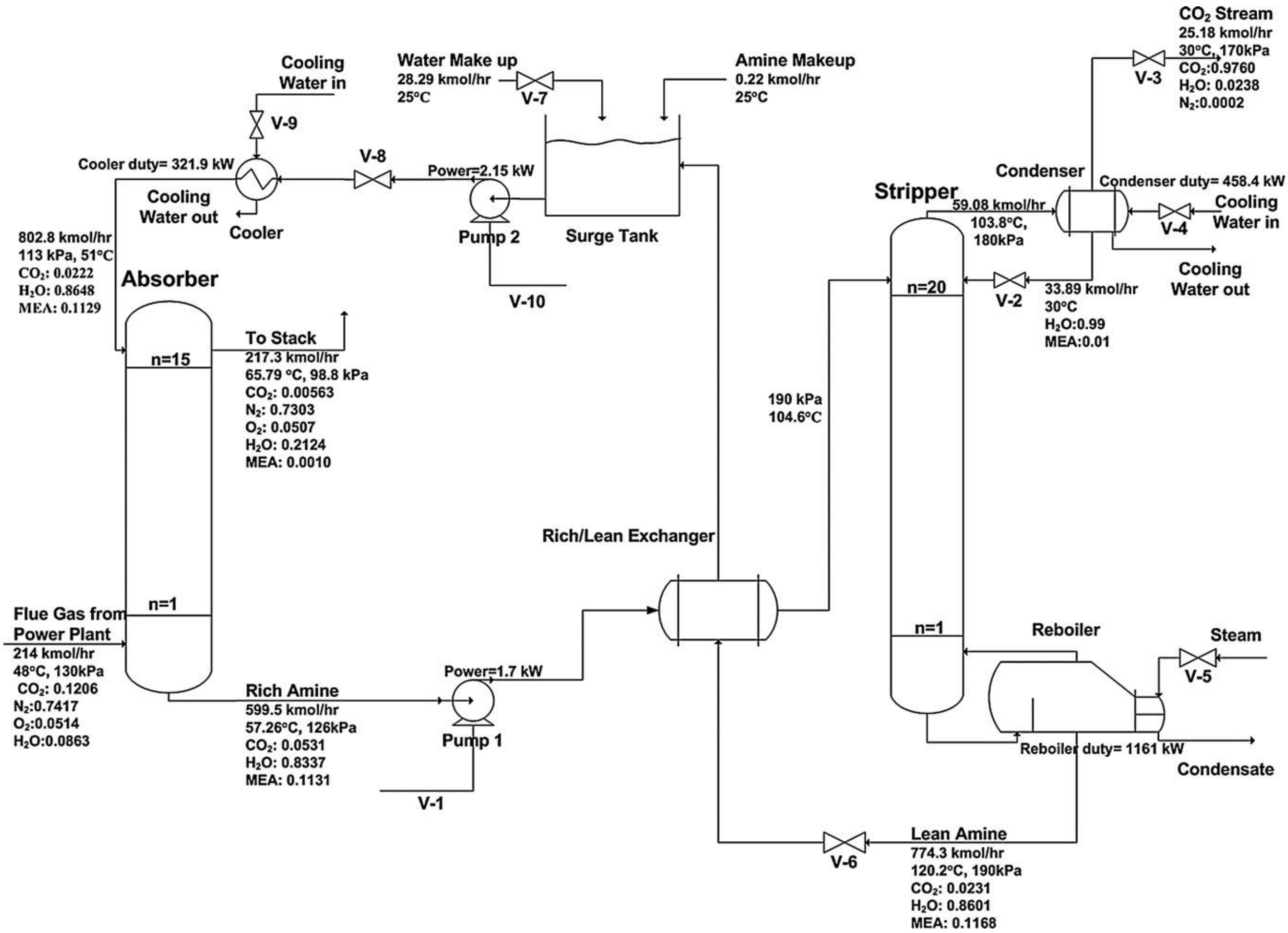

Fig. 2. Process with 10 dynamic DOFs (valves).

plant like a utility or an auxiliary unit, other cost functions may be more relevant. We assume there is a penalty of 50 USD/ton on CO_2 released to the environment or equivalently a price of 50 USD/ton CO_2 captured which must be traded against the energy required for CO_2 removal in the stripper and work for the pumps. The reboiler energy is converted to power via a factor f which assumes that the energy used in the reboiler could have been used to produce power [1]. The total required equivalent work of the reboiler and pumps is then:

$$W_{\text{total}} = Q_{\text{reboiler}} f + W_{\text{pumps}} \tag{1}$$

The pumps include pump 1 and pump 2 in Fig. 2 plus the pumps for cooling water in the condenser and cooler which are not shown in the flowsheet. The power price is assumed to be 0.063 USD/kWh [6] and the conversion factor is $f = (1 - (T_C/T_H)) \times \eta$, where $T_H = T_{\text{reboiler}} + 10$ [K] and $T_c = 313$ K. The efficiency η is assumed to be 75%. We have not included the feed flue gas blower, but this does not change the conditions, since the blower power is very weakly dependent on the operation of the absorber/stripper system.

The objective is to get an optimal trade off between the cost of CO_2 released to the air and the energy usage in the process to strip off the removed CO_2. Since we are considering an individual unit we divide the cost by the amount of flue gas to remove the effect of the feed rate. The cost to be minimized is then:

$$J[\text{USD/kg}_{\text{flue gas}}] = \frac{W_{\text{total}}(\text{kW}) \times 0.063\,(\text{USD/kWh}) + \dot{m}_{CO_2 \text{ in purified flue gas}}\,(\text{kg/h}) \times 0.05\,(\text{USD/kg})}{\dot{m}_{\text{flue gas}}\,\left(\frac{\text{kg}}{\text{h}}\right)} \tag{2}$$

We consider the following inequality constraint:

1. For environmental reasons at least 80% of the CO_2 must be removed,
2. The maximum reboiler duty is +20% compared to the nominal value.

and the following four equality constraints:

1. The temperature of the lean solution to the absorber is kept at 51 °C to get good operation in absorber; see also Section 3.
2. The stripper top pressure is kept at 1.8 bar to avoid MEA degradation. At higher pressures, the bottom temperature will be above 120 °C which will increase significantly amine degradation [7].
3. The stripper condenser temperature is kept at 30 °C, which is assumed to be the lowest achievable temperature. A low temperature is desired because it reduces the compression work for the captured CO_2 in the downstream process.
4. The outlet pressure of pump 2 should be 4 bar to transfer the recycle lean amine to the top of the absorber.

In addition, all flowrates must be non-negative and we assume maximum capacities compared to nominal values for cooler (+50%) and pumps (+40%) but these constraints are never encountered at steady-state for the assumed operating range.

2.1.2. Step 2. Identify DOFs for optimization [3]

We have 8 valves and 2 pumps (Fig. 2) which give 10 dynamic degrees of freedom. However, there are 4 levels (1 in absorber, 2 in stripper and 1 surge tank) that need to be controlled and since these levels have no steady state effect, the number of degrees of freedom (DOFs) for steady-state optimization is 6. Note that there is no bypass on the rich/lean heat exchanger because maximum heat exchange (zero bypass) is optimal. The small amine make up flowrate is not considered as a degree of freedom because it is assumed that it is adjusted to keep the amine concentration constant.

2.1.3. Step 3. Identification of important disturbances [3]

The main disturbances are the feed (flue gas) flow rate and its composition. In addition, the active constraints should generally be considered as disturbances. The stripper top pressure is included as a disturbance and this also takes into account changes in pressure drop due to load changes.

2.1.4. Step 4. Optimization (nominally and with disturbances)

To control the 4 equality constraints we need 4 DOFs and we need 4 DOFs to control the 4 levels which have no steady-state effect. There are then two degrees of freedom left for optimization,

$$N_{\text{opt.free}} = 10 - 4 - 4 = 2$$

These may be viewed as the CO_2 recovery ($u_1 \geq 80\,\%$) in the absorber and the CO_2 mole fraction at the bottom of stripper ($u_2 \geq 0$), but note that this choice is not unique and any independent set variables can be selected as "base" DOFs.

By optimization, we find for the nominal operating point (no disturbances) that the two remaining DOFs are unconstrained:

CO_2 recovery, $u_1 = 95.26\%$

CO_2 mole fraction at the bottom of stripper, $u_2 = 0.0231$

Optimal objective function = 2.526 USD/ton flue gas

2.1.5. Step 5. Identification of candidate controlled variables

To find the best set of two single measurements (CVs) for the two unconstrained DOFs, we consider 39 candidates measurements, including the two DOFs:

1. CO_2 recovery in the absorber, u_1, (y_1).
2. CO_2 mole fraction at the bottom of the stripper, u_2, (y_2).
3. Tray temperature of absorber column, 15 possible stages, (y_3–y_{17}).
4. Tray temperature of stripper column, 20 possible stages, (y_{18}–y_{37}).
5. Recycle lean amine flowrate (y_{38}).
6. Reboiler duty (y_{39}).

2.1.6. Step 6. Selection of CVs

One of the main assumptions in the methods used below is that the cost function has quadratic behavior around optimal point and is twice differentiable. Fig. 3 confirms this assumption in our case. To find the best set of two CVs, we apply the exact local method which gives the worst case loss $\bar{\sigma}(M)$ imposed by each candidate CV set [8]. The set with the minimum worst-case loss is the best.

$$\text{worst-case loss} = \frac{1}{2}\bar{\sigma}(M)^2 \tag{3}$$

$$M = J_{\text{uu}}^{1/2}(HG^y)^{-1}(H[FW_{\text{d}}\, W_{\text{n}}]) \tag{4}$$

$$F = G^y J_{\text{uu}}^{-1} J_{\text{ud}} - G_{\text{d}}^y \tag{5}$$

here H is the selection matrix ($c = Hy$), G^y is the gain of the selected measurements, J_{uu} is Hessian of the objective function with respect to unconstrained DOFs and J_{ud} is second derivative of objective function with respect to DOFs and disturbances. Alternatively, one could replace the singular value $\bar{\sigma}(M)$ by the Frobenius norm, $\left\|M\right\|_F$ which represents the average loss [9], but this happens to give the same optimal H [9]. Actually, we do not need the matrix J_{uu} for finding the optimal H, because we get the same optimal H as in Eqs.

Table 1
Expected magnitude of individual disturbances.

	d_1: Flowrate of flue gas	d_2: Composition of CO_2 in flue gas	d_3: Pressure of stripper
W_d	20%	10%	30 kPa

(3)–(5) by solving the following problem [10]:

$$\min_H \left\| M \right\|_F = \min_H \left\| H[FW_d \, W_n] \right\|_F \tag{6}$$

$$\text{subject to } HG^y = I \tag{7}$$

F is the optimal sensitivity of the measurements with respect to disturbances. It can be found either by Eq. (5), in which case one must also find gains, G_d^y from disturbances, d to measurements and J_{ud}, or numerically by reoptimization of the process in presence of different disturbances:

$$F = \frac{\Delta y^{\text{opt.}}}{\Delta d} \tag{8}$$

Based on our experience it is strongly recommended to find F numerically (from Eq. (8)) by reoptimization of the process rather than calculating it from Eq. (5) which needs J_{uu} and is sensitive to errors as it needs several matrices that may not be consistent. F is the slope of the optimal sensitivity of the measurements respect to disturbances and should be linear in different magnitudes of disturbances. We chose a magnitude of 5% for disturbances (Δd) and reoptimized the process to get the optimal sensitivity. In our case where we have 39 candidate measurements and 3 disturbances, the size of matrix F is 39×3. To see how the matrices are found, the reader is referred to Ref. [11].

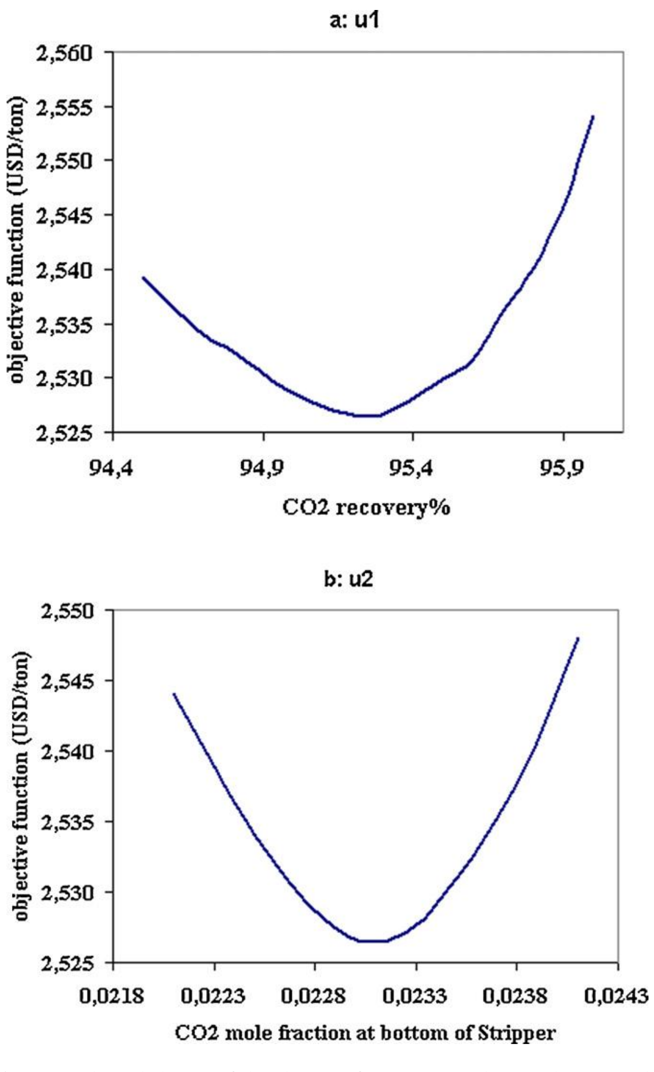

Fig. 3. Quadratic behavior of the objective function around optimal point in the region I. (a) CO_2 recovery and (b) CO_2 composition at the bottom of the stripper.

Finally, the expected magnitude of individual disturbances (W_d) and magnitude of the implementation error of CVs (W_n) must be specified. Tables 1 and 2 show these data for our case.

Kariwala and Cao [12] have developed a bidirectional branch and bound algorithm to find the optimal H using Eqs. (3)–(5). We applied this algorithm, except that we found F from Eq. (8), and the results are shown in Table 3. As mentioned, identical optimal measurements (H) were obtained using Eqs. (6 and 7). Note that although J_{uu} is not needed to find the optimal H, it is needed to compute the resulting worst-case loss in Eq. (3).

In the list of the best sets of CVs in Table 3, controlling the CO_2 recovery (y_1) is common in all six best sets. The best 2nd measurement is the temperature control of tray no. 16 (y_{33}) in the stripper. From the ranking, other good second stripper measurements are the temperatures of neighboring trays. Fig. 4 shows the proposed control structure in region I. Also, note that the losses are very small; about 0.01 USD/ton of flue gas treated. This means that there is little reason to consider measurement combinations as CVs.

2.2. Region II: large flowrates of flue gas (+30%)

The active constraints and thus the optimal control strategy will change as we increase the flue gas flowrate. To study this, we keep constant the best self-optimizing CVs found in region I and gradually increase flowrate of flue gas (Table 4). We find that the first constraint we encounter is the reboiler duty which saturates when the flue gas is increased by 19.35% and we are in region II. In general when the process reaches a new constraint region, we need to identify new self-optimizing CVs. In region II there is only one unconstrained degree of freedom which may be selected as the lean amine flowrate, so we need to find one CV.

We further increase the flowrate of the flue gas so that the total increase is +30% (278.2 kmol/h) and then reoptimize the process (Table 4). This point is selected as the optimal nominal point in this region. The same procedure as region I is repeated here. Note that one new active constraint (reboiler duty) has been added to the previous active constraints. We could use the exact local method but we choose to use the closely related maximum gain rule [13] to select the best CV which has the highest scaled gain. In the maximum gain rule the loss has inverse proportion to the square of the scaled gain: loss $\sim 1/(G_s)^2$. It is worth noting that since there is only one DOF left, the maximum gain rule and exact local method give the same result. To see how the scaled gains are calculated see [3].

There are 38 candidate measurements which are like before except for the reboiler duty which is saturated. Table 5 shows that temperature of tray no. 13 (y_{30}) in the stripper has the largest scaled gain and is the best CV to be controlled using recycle lean amine flowrate. Fig. 5 shows the resulting control structure in region II.

Table 2
Magnitude of the implementation error for CVs.

	CO_2 recovery	Temperature	Composition	Flowrate	Reboiler duty
W_n	0	1 °C	0.1%	10%	10%

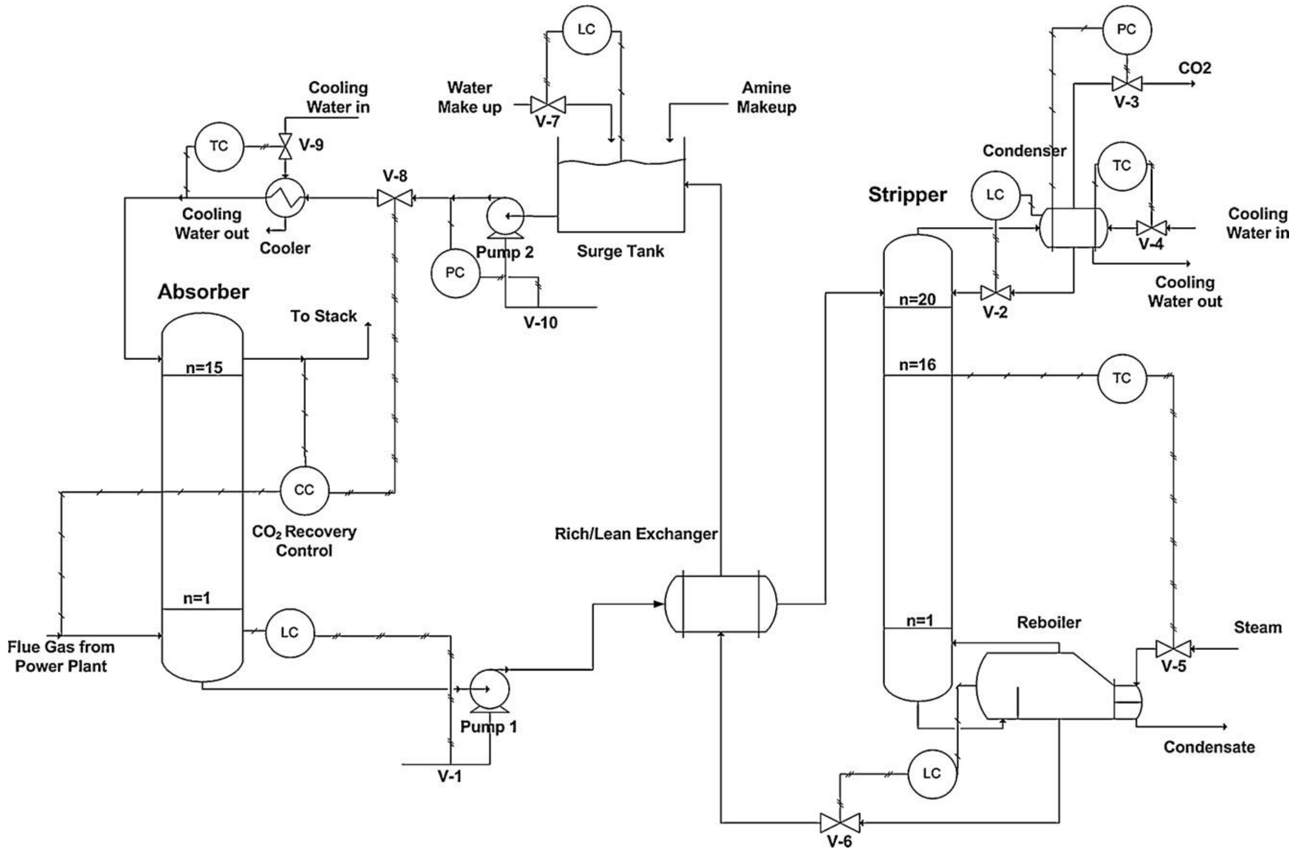

Fig. 4. Proposed control structure with given flue gas flowrate (region I).

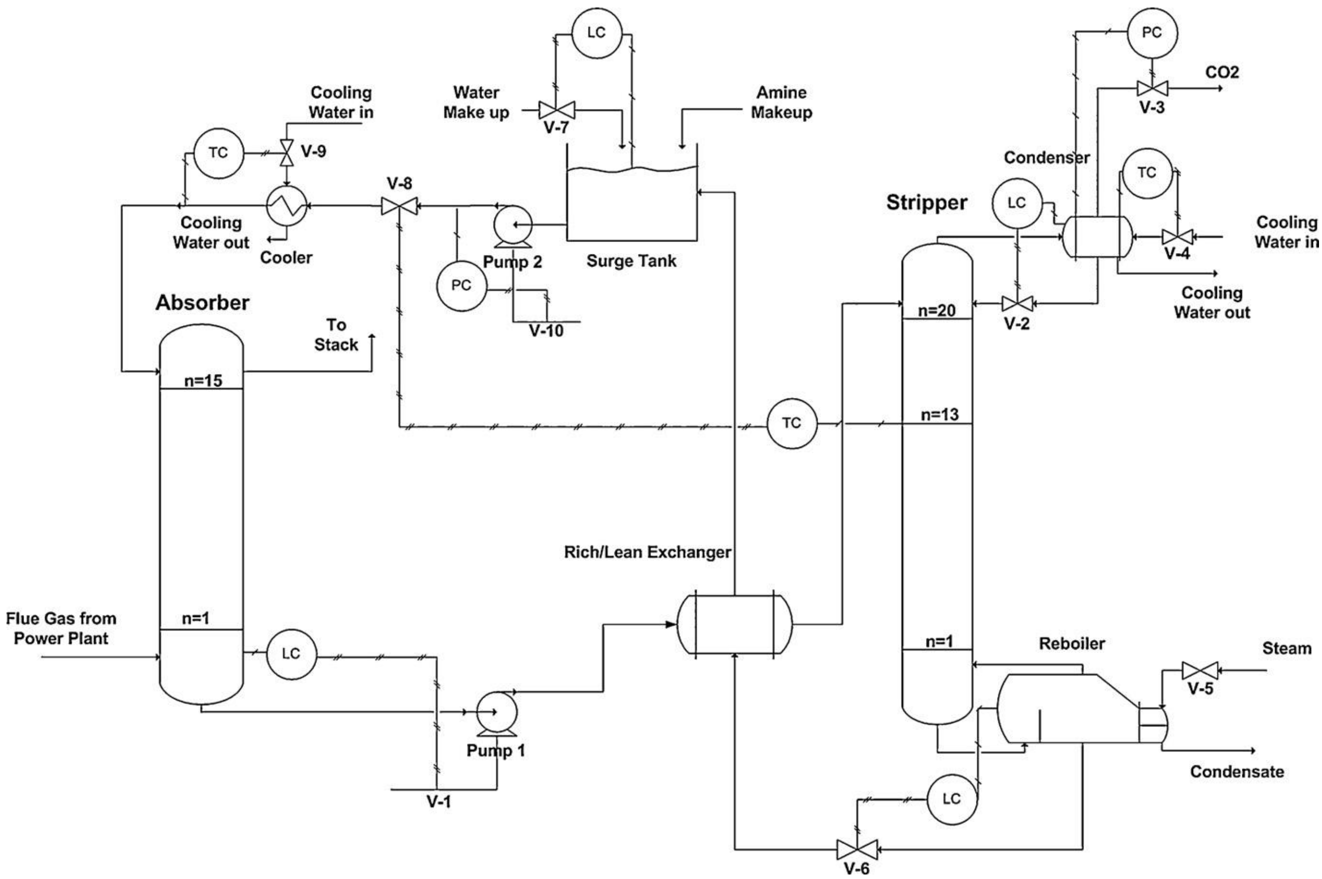

Fig. 5. Proposed control structure in presence of large flowrates of flue gas when reboiler is saturated (region II).

Table 3
The best candidate CV sets in region I.

Rank of sets	CV1	CV2	Worst case loss (USD/ton flue gas)
1	y_1: CO_2 recovery	y_{33}: Temperature on tray no. 16 in the stripper	0.0057
2	y_1: CO_2 recovery	y_{32}: Temperature on tray no. 15 in the stripper	0.0064
3	y_1: CO_2 recovery	y_{34}: Temperature on tray no. 17 in the stripper	0.0067
4	y_1: CO_2 recovery	y_{31}: Temperature on tray no. 14 in the stripper	0.0092
5	y_1: CO_2 recovery	y_{35}: Temperature on tray no. 18 in the stripper	0.0130
6	y_1: CO_2 recovery	y_{30}: Temperature on tray no. 13 in the stripper	0.0174
7	y_5: Temperature on tray no. 3 in the absorber	y_{33}: Temperature on tray no. 16 in the stripper	0.0198
8	y_5: Temperature on tray no. 3 in the absorber	y_{32}: Temperature on tray no. 15 in the stripper	0.0202
9	y_5: Temperature on tray no. 3 in the absorber	y_{34}: Temperature on tray no. 17 in the stripper	0.0206
10	y_5: Temperature on tray no. 3 in the absorber	y_{31}: Temperature on tray no. 14 in the stripper	0.0218

Table 4
Increasing the flowrate of flue gas with the control policy in region I; saturation of reboiler duty occurs when feed flowrate is +19.35%.

	Feedrate of flue gas (kmol/h)	Pump1 duty (kW)	Pump2 duty (kW)	Self-optimizing CVs in region I		Cooler duty (kW)	Reboiler duty (kW)	Objective function (USD/ton)
				CO_2 recovery (y_1), %	Temperature of tray no. 16 (y_{33}), °C			
Optimal nominal point	214	1.70	2.15	95.26	106.9	321.90	1161	2.53
+5% Feedrate	224.7	1.78	2.26	95.26	106.9	347.3	1222	2.53
+10% Feedrate	235.4	1.86	2.36	95.26	106.9	371.0	1279	2.53
+15% Feedrate	246.1	1.94	2.46	95.26	106.9	473.3	1339	2.53
+19.35%, when reboiler duty saturates	255.4	2.00 (+12.36%)	2.55 (12.83%)	95.26	106.9	419.4 (+30.29%)	**1393 (max) (+20%)**	2.54
+30% Feedrate (reoptimized)	278.2	2.03	2.58	91.60	103.3	359.3	**1393 (max)**	2.69

Active constraints are in boldface.

Tray no. 13 (y_{30}) is not the same as in region I (tray no. 16, y_{33}), which is a disadvantage because of the logic needed to reconfigure CVs as we switch between regions. However from Table 5 we see that y_{33} has the 7th largest scaled gain and is still a good CV in region II but it must have a new setpoint when we change regions. The change is from 106.9 °C to 103.3 °C which illustrates that is not the truly optimal controlled variable in both regions. This alternative structure will be discussed in part 2 of this article.

2.3. Region III: large flowrates of flue gas when process reaches minimum allowable CO_2 recovery

We keep y_{30} (the best CV in region II) constant and further increase the flowrate of flue gas. When the flowrate of flue gas reaches 326.9 kmol/h (+52.76%), the CO_2 recovery reaches its lower bound constraint of 80% and we have reached the bottleneck where no further increase is possible (Table 6). A controller or manual control is needed to set the feedrate of flue gas such that the recovery stays above 80%.

To validate the proposed control structures in different regions, dynamic simulation of the process is done. These results are presented in part 2 of this article.

Table 5
The best candidate CVs with the largest scaled gain in region II.

Rank	CV	100 × Scaled gain
1	y_{30}: Temperature on tray no. 13 in the stripper	6.03
2	y_{31}: Temperature on tray no. 14 in the stripper	5.77
3	y_{29}: Temperature on tray no. 12 in the stripper	5.63
4	y_{32}: Temperature on tray no. 15 in the stripper	4.95
5	y_{28}: Temperature on tray no. 11 in the stripper	4.81
6	y_{27}: Temperature on tray no. 10 in the stripper	3.89
7	y_{33}: Temperature on tray no. 16 in the stripper	3.88
27	y_1: CO_2 recovery	0.19

3. Discussion

The lean amine temperature to the absorber was assumed to be 51 °C. In practical CO_2 capturing processes using 30% MEA, lean amine is usually fed to the absorber at around 40 °C, which gives a good balance between the kinetics and thermodynamics for absorption reactions. The value of 40 °C is reported frequently in the literature when 90% recovery is the target [1]. However in our case we use a higher recovery because of a trade off between the cost of energy and the tax on the CO_2 released to the air, and this results in a higher optimal temperature, as shown in Fig. 6. We did not increase the temperature further above 51 °C, because this would increase amine losses and because we did not want to be too far from the current practical temperatures.

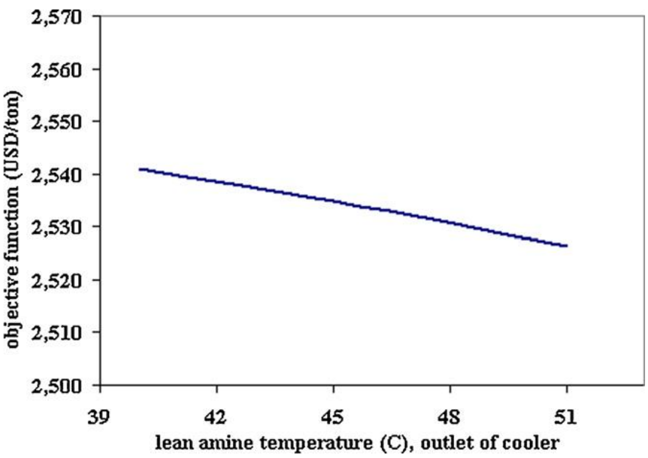

Fig. 6. Objective function with change in lean amine temperature.

Table 6
Increasing of the flowrate of flue gas with control policy in region II and reaching to the minimum allowable CO_2 recovery.

	Feedrate of fluegas (kmol/h)	Pumps duty (kW)	CO_2 recovery %	Self-optimizing CV in region II Temperature of tray 13 (y_{30}), °C	Cooler duty (kW)	Reboiler duty (kW)	Objective function (USD/ton)
Optimal nominal case in region II (+30% feedrate)	278.2	4.61	91.60	109	359.3	**1393 (max)**	2.69
+40% Feedrate	299.6	4.58	86.46	109	315.5	**1393 (max)**	3.01
+50% Feedrate	321	4.55	81.31	109	290.3	**1393 (max)**	3.36
+52.76% Feedrate, reach to minimum allowable CO_2 recovery	326.9	4.54	**80 (minimum)**	109	284.6	**1393 (max)**	3.45

Active constraints are in boldface.

In this article, we assumed that the stripper reboiler duty was the first capacity constraint to become active. Note here that a constraint on reboiler duty (vapor boilup) is almost equivalent to a constraint on gas capacity in the stripper (e.g., due to flooding). If instead the gas capacity of the absorber was the first capacity constraint to become active (e.g., due to flooding), then this would be a bottleneck and no more flue gas could be handled. Thus, in terms of control structure selection, there are no options except for reducing the feed flue gas flowrate. Here, we have considered the less obvious case which is saturation of reboiler duty where the feedrate can still be increased further (region II).

The price for power (electricity) and the CO_2 tax can vary widely and their ratio will determine the optimal amine recirculation and reboiler duty. However, we believe the structural issues regarding selecting good CVs will be less sensitive to this.

4. Conclusions

In this study a control structure is designed for a CO_2 capturing process with the aim to achieve optimal CO_2 removal. Self-optimizing method is used to select the best CVs in three different operational regions;

Region I: in low feedrates of flue gas and having two unconstrained degrees of freedom:

CV1 = y_1 = CO_2 recovery (95.26%).
CV2 = y_{33} = temperature of tray no. 16 in stripper (106.9 °C).

Region II: in intermediate feedrates of flue gas with saturation of heat input and having only one unconstrained degree of freedom:

CV1 = Max. reboiler duty (y_1 = CO_2 recovery is given up).
CV2 = y_{30} = temperature of tray no. 13 in stripper (109 °C).

However, we argued that an alternative with an only slightly larger loss would be to select CV2 = y_{33} also in region II.

Region III: in large feedrates of flue gas when minimum allowable CO_2 recovery (80%) meets, a controller is needed to set the flowrate of flue gas such that the minimum is satisfied.

To validate the proposed control structures, dynamic simulation of the process is needed which is considered in part 2 of this article. In part 2 we also further discuss the possibility of finding a single control structure that works in all regions.

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