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Model Adaptation and Optimization for the Evaluation and Investigation of Novel Amine Blends in a Pilot-plant Scale CO_2 Capture Process under Industrial Conditions

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Carbon capture processes are highly energy intensive and the main driving force during the process design is the necessity to reduce the energy consumption for the solvent regeneration. The energy efficiency of absorption desorption processes is driven by the plant design and operation conditions, but also to a large extent by the choice of the scrubbing liquid (Wang et al., 2011). Absorbent screening for CO_2 capture is time-consuming and costly. Apart from the energy efficiency, aspects such as loading capacity and robustness towards industrial impurities and disturbances have to be investigated before designing a large-scale plant and optimally operating it. In this contribution, a systematic approach is presented to carry out evaluation tests for a novel solvent in an industrial pilot-plant and at the same time to determine an optimal operation point with maximum energy efficiency. The three-step approach is based on the assumption that for a novel absorbent little to no thermodynamic data is available. Hence, the investigation is solely based on simulation data from similar solvents and experimental data on the novel one. Monoethanolamine (MEA) still is widely used as reference absorbent for removing CO_2. Consequently, simulation data and the properties of MEA are taken as a baseline for the general performance and behaviour of amine-based absorbents. As a first step, data from rigorous simulations is used to develop a surrogate model describing the general behaviour of a carbon capture process for MEA. Subsequently, pilot-plant-scale experiments are carried out to investigate the application of MEA in practice. Secondly, the surrogate model is then updated to account for the plant characteristics as given by the experimental data for MEA. Finally, by means of the MEA-based data-driven model, the new solvent is experimentally investigated. By successive approaches the surrogate model's maximum in energy efficiency is identified and repeatedly updated for the novel solvent's experimental behaviour. In terms of the energy efficiency and based on this workflow the performance of the novel solvent is compared with MEA.

1. Introduction

Solvent based gas treating processes are widely used in the chemical industry, especially in oil and gas exploration / processing and for air pollution control applications. For CO_2 removal, the usage of reactive amine solutions is a promising technology and has already been commercially implemented in the industry for decades. The process has been well established for a long time and is used for retrofitting existing plants with additional CO_2 removal sections for emissions reductions (Wang et. al, 2011). The CO_2 removal processes with MEA still represent the reference for carbon capture processes. Nevertheless, MEA has also major disadvantages, e.g. a high sensitivity towards acidic components in flue gas and high energy requirements for regeneration. An investigation of the specific energy demand of MEA regarding varying operation conditions is shown in Lang et al. (2017). In order to improve the process performance and to extent the application for other types of gas streams beyond flue gas, novel solvents are being developed and uniquely adapted to each process and application. However, a mechanistic model of novel solvents is often very complex, e.g. due to thermodynamic properties. Previous studies used data-driven approaches to model physical phenomena

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based on experimental data. Dadhe et al. (2001) implemented neural networks to replace calculations for physical properties in reactive distillation. In the present paper a new approach for solvent characterisation and determination of optimal operating conditions based on simulations and pilot-plant data incorporated into data-driven surrogate models is introduced. Optimal operation points are characterized by a minimized specific energy demand for the regeneration in dependence of the gas and solvent flow rate. For this purpose, simulations are performed with ASPEN Plus® followed by a derived surrogate model, which is subsequently used for the optimization of the specific energy supply for each solvent. Practical experiments with MEA and the novel solvent are carried out using a pilot-plant and operated at the identified optimal operation points. The pilot-plant was designed and constructed at the Process Dynamics and Operations Group of Technische Universität Berlin. The plant is connected to a steel mill in Duisburg, Germany, and operates under real industrial conditions.

2. Methodology

The current work involves the experimental analysis and characterisation of a novel solvent for CO_2 absorption from an industrial gas and determination of optimal operation points using data-driven models. The methodology consists of a three-step approach: Initially, the general behaviour of an absorption process is mapped by extensive steady-state simulations of the pilot-plant of a known absorbent, in this case an aqueous solution of MEA. The simulation results are then used to construct a surrogate model, which is subsequently used to represent the physical trends of the amine systems. As a second step the surrogate model is adapted to pilot-plant´s experimental results with MEA to incorporate the real plant behaviour. As a final step the updated surrogate model is then fitted to experimental results for the novel absorbent applied in the pilot-plant. By an analytic analysis of the surrogate model an approximation to the globally optimal operation point is identified for each solvent, based on the lowest minimum of the model. In order to have a fair basis for comparison the optimal operation point of the novel absorbent is repeatedly updated for the solvent's experimental behaviour and compared to that of MEA. In the following, the experimental set-up as well as the model structure and application are detailed. In terms of process design of the pilot-plant it is necessary to enable a wide operation range for comprehensive investigations.

2.1 Process Design

A pilot-plant is constructed for the experimental investigation of novel solvents for the carbon capture. The pilot-plant is installed in bypass gas line to a steel mill in Duisburg, Germany. For the experimental setup blast furnace gas is used containing up to 26 vol.-% CO_2 as well as other components and impurities. The modular design of the plant enables an easy transport and operation at various industrial sites. The basic scheme of the process and a picture at the steel mill is shown in Figure 1.

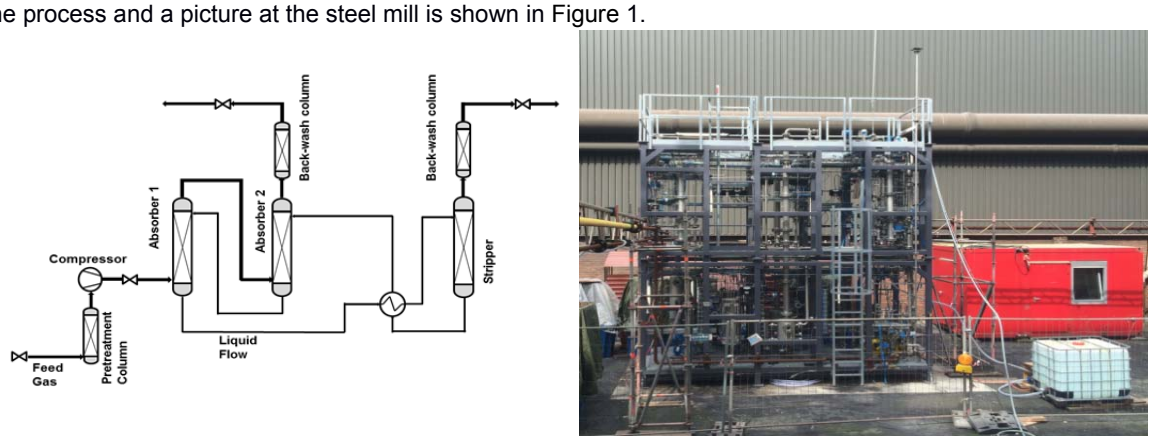

Figure 1: Basic flowsheet of the absorption desorption process (left) and a picture of the pilot-plant at the industrial site (right)

The installation of a compressor realizes a gas flow of 15 to 50 kg/h, whereas the liquid pumps reach a solvent flow rate of up to 300 kg/h. Two absorber columns are installed in sequence with a total packing height of 6 m and a diameter of 0.11 m (Montz Pak B1-350.60). It is possible to bypass one column to reduce the packing height to 3 m. The stripper contains 2.5 m packing with a diameter of 0.11 m (Montz Pak B1-350). At the gas outlets, back-washing columns are installed to achieve the required process conditions at the gas outlets and to reduce the loss of the amine solution. If necessary, a pre-treatment column is available for a caustic wash.

The pre-treatment and back-washing columns contain 1 m of structured packing with a diameter of 0.11 m each (Sulzer Mellapak 250.X). The experimental setup includes sampling of the gas flow at the inlet, outlet and in-between the two columns. Further details on the experimental setup can be found in Wilhelm et al., 2016.

2.2 Experimental Case Studies

The experimental characterisation of solvents and the verification of the surrogate model require systematic experimental case studies. The general approach for the experiments is the variation of important process variables within the technical boundaries, such as the gas and liquid load in the columns. In order to achieve constant conditions regarding the fluid dynamics, the gas and solvent load is kept constant for each operation point. The energy supply in the stripper is adapted to each operation point for an intended carbon capture rate of up to 90 vol.-%. At the bottom of the stripper the temperature is controlled via an electrical heater. In order to assess the performance of the stripper, the mass flow of desorbate is a meaningful indicator. A steady-state operation point is achieved when constant gas and liquid flows, as well as a constant CO_2 capture rate are established. One operation point needs to last for at least two hours to achieve one complete circulation of the amine solution. Liquid samples are taken to analyse the rich and lean loading. The CO_2 concentration is measured permanently with an infrared measurement device (ABB EL3020). The energy supply in the stripper determines the specific energy demand for the regeneration of the solvents and represents the benchmark of absorption desorption processes.

$$e_{CO_2} = \frac{\dot{W}_{Regeneration}}{\dot{m}_{CO_2}} \tag{1}$$

where e_{CO_2} is the specific energy demand, $\dot{W}_{Regeneration}$ represents the energy supply for regeneration considering plant characteristics, such as heat losses and efficiencies of heat exchangers, and $\dot{m}_{CO_2}$ is the captured mass of CO_2.

2.3 Approach for Solvent Characterisation

This work presents a three-step approach, which is taken for the experimental characterisation of a novel solvent under industrial operation conditions and the subsequent identification of the optimal operation point using a data-based model. The general workflow is shown in Figure 2.

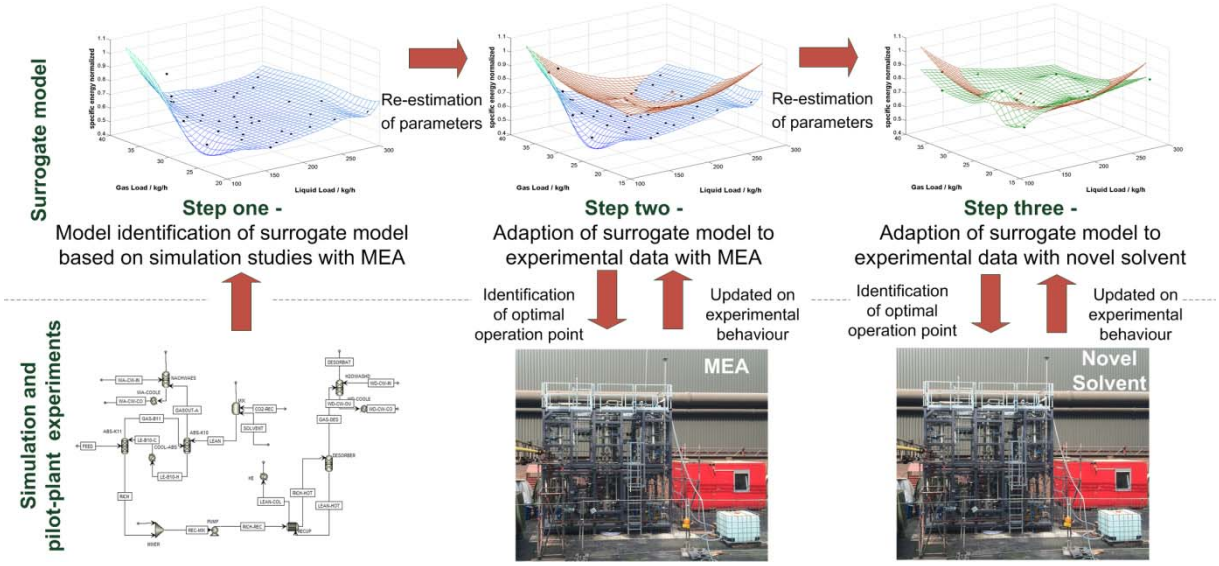

Figure 2: General three-step approach for the design of data-driven model and subsequent adaption to experimental results for the determination of optimal operation points for each solvent

As a first step, the structure of the surrogate model is determined by results of comprehensive simulations studies with ASPEN Plus® (Figure 2). According to Eq(1), the specific energy demand is determined in dependence of the gas and liquid flow. Due to little thermodynamic knowledge of the novel solvent the simulation are performed with MEA. Each simulation represents one operation point. The feasible region for the operation points is limited by the technical restrictions of the pilot-plant. The chosen operation points are

well-distributed and initialized by a Hammersley sequence to avoid clustering of the following data. Via statistical methods an approximate model is constructed based on gathered simulation data (Lophaven et al. 2012).

$$f_1(x_1, x_2, p) = e_{CO_2} \tag{2}$$

with f_1 as the structure of the data-driven model, x_1, x_2 for set values of gas, respectively solvent flow rate, p for the set of parameters within the model. This procedure enables the transfer of information from a physical model (ASPEN Plus®) to the data-driven model. Consequently, the identified structure of the model represents the theoretical behaviour of the required energy depending on the gas and solvent flow.
Secondly, plant experiments are carried out with MEA and the surrogate model is fitted to experimental data. For this purpose, the model structure remains unchanged, whereas the parameters within the surrogate model are updated. This leads to an adapted surrogate model based on experimental data with MEA, which is used to determine the minimal energy demand for regeneration by an optimization for e_{CO_2}.

$$f_2(x_1, x_2, p_2) = e_{CO_2} \tag{3}$$

Where p_2 is updated. The function f_2 represents the data-driven model adapted to experimental data. Due to the fitting to experimental data the surrogate model takes into account the plant characteristics as wells as industrial impurities and disturbances.
As a third step, the updated surrogate model with MEA is adapted to experimental results of the novel solvent to evaluate and investigate the novel solvent under experimental and industrial conditions. For this purpose the parameters are re-estimated once again. The model adaption results in a modified response surface for the novel solvents, which represents the energy requirements under industrial conditions. In terms of energy consumption the minimal e_{CO_2} is determined for the new solvent.
The construction of surrogate models is performed by applied statistical methods. Kriging is a method developed to construct an approximation model, which is fitted to data and used as a surrogate model. The approximated model can be used to predict output at untried inputs. In general Kriging is an optimal interpolation based on regression of observed values against surrounding data points (Oliver and Webster, 1990). The basic form is

$$Z^*(u) - m(u) = \sum_{\alpha=1}^{n(u)} \lambda_\alpha [Z(u_\alpha) - m(u_\alpha)] \tag{4}$$

with u, u_α as location vectors for estimation and one neighbouring data point, indexed by α, $n(u)$ with number of data points used for estimation, $m(u), m(u_\alpha)$ as expected values of $Z(u), Z(u_\alpha)$, λ_α as Kriging weight. The value $Z(u)$ represents the estimated value. The goal of this method is to determine the weight, λ_α, in order to minimize the variance of the estimator. The present application represents a regression problem, which is characterised by a strong dependence of the process variable (e_{CO_2}) against surrounding values. For the analysis of the process a regression of second order is chosen and a Gaussian correlation between the data points is applied.

3. Results

Data obtained from previous studies (Mangalapally et al., 2011) using pilot-plant experiments indicated a reduction of the regeneration energy by optimizing the operation conditions. According to Mangalapally et al., 2011 different solvent flow rates lead to a decrease of the specific energy demand for regeneration from 4.1 MJ/kg CO_2 to 3.8 MJ/kg CO_2 for MEA. In this contribution, the focus lies on the experimental characterisation of a novel solvent and comparison of the energy efficiency to MEA based on data-driven models.
According to step one of the workflow, the specific energy demand for MEA is determined in dependence of the gas and solvent flow rate using simulations. Figure 3 shows the results visualized by a response surface plot. According to Eq(5) the structure of the surrogate model is identified based on simulation data with MEA.

$$e_{CO_2} = A + B \cdot x_1 + C \cdot x_1 x_2 + D \cdot x_2 + E \cdot x_1^2 + F \cdot x_2^2 \tag{5}$$

wherein $A\ to\ F$ are regression parameters.
As can be seen in Figure 3, the specific energy shows a significant dependency of gas and solvent flow rate. An increasing solvent flow rate leads to a considerable decrease of energy requirements but is soon followed by smooth surface. This indicates a wide range of the operation window of the application with MEA. The phenomenon is consistent with results obtained in previous studies (Mangalapally et al., 2011), which proves that the identified surrogate model is well suited to describe correlation of energy requirements.

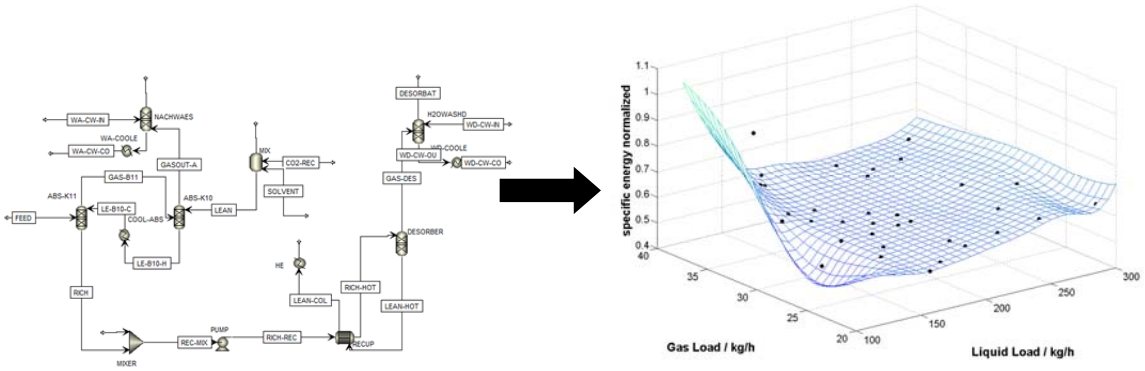

Figure 3: Computation of specific energy demand for regeneration in dependence of gas and solvent flow; left (simulation for carbon capture process), right (surrogate model based on process simulations for MEA)

As a second step, the identified model is adapted to experimental data with MEA. The structure of the model stays unchanged, whereas the parameters are updated using Eq(5). The gas and solvent flow rate are fixed set points within the automation system. The results for the data-driven model with experimental results for MEA are shown in Figure 4.

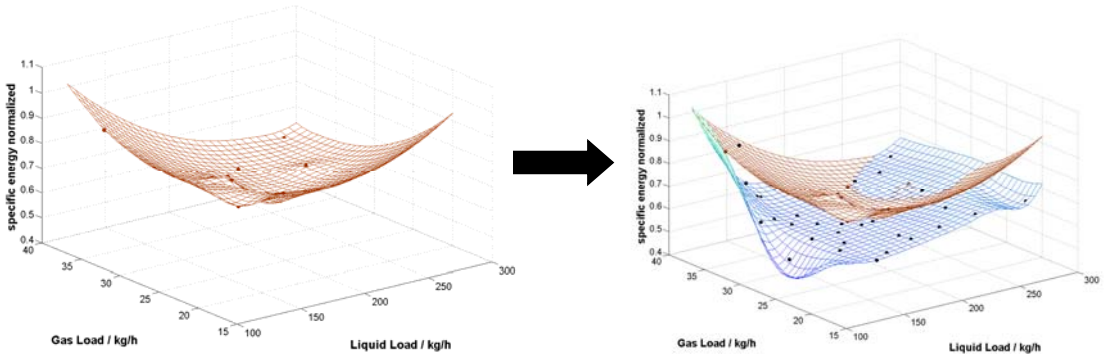

Figure 4: Response surface plot for the specific energy demand based on experimental results with MEA (left). Comparison of simulation (blue) and experimental data (red) with MEA (right)

Figure 4 shows that an increasing solvent flow causes a decrease of the specific energy demand. The offset between the theoretical and practical investigations can be explained by plant characteristics, such as heat losses and efficiencies of heat exchangers. Relative high heat losses are caused by significant ambient changes, such as wind and rain, and lead to higher energy requirements and deformation of the surface plot. By the use of the surrogate model the normalized optimal operation point for MEA is derived. Based on the identified minimum for the required energy, the plant is operated at this point in order to confirm the optimum with experimental data. The additional operation point confirms the energy minimum.
Finally, experiments are carried out with the novel solvent within the same technical specifications of the pilot-plant. The surrogate model from the simulations using MEA is applied and fitted to the experimental data with the novel solvent. Figure 5 shows the obtained results.
It can be observed that the characteristic decrease of the specific energy demand does not appear for the novel solvent. Furthermore, minima appear within the response surface plot, instead of a smooth surface (s. Figure 4 for MEA), what is identified as minimal specific energy. For validation purposes the pilot-plant is operated at this point. The experiment yields in a specific energy demand. Consequently, the surrogate model of the novel solvent is supplemented by this point. Based on the unique operation points of each solvent the minimal required energy demand of each solvent is compared. This method is only valid for novel solvents with similar characteristics to a known solvent.

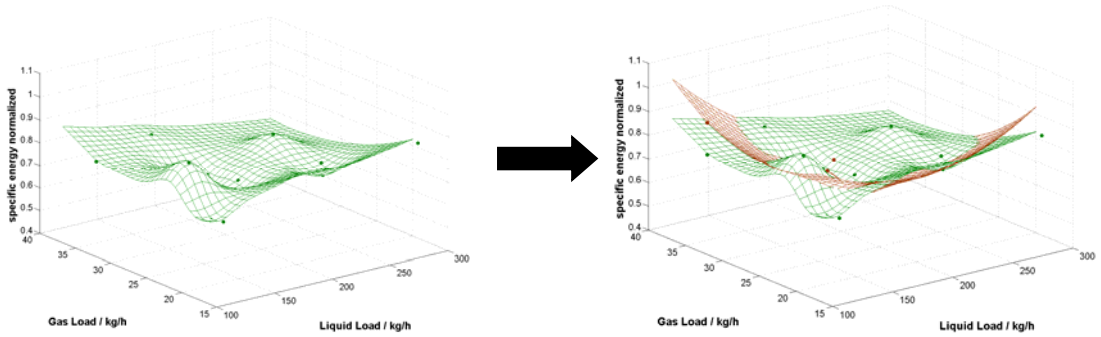

Figure 5: Response surface plot for specific energy demand based on experimental results with the novel solvent (green, left). Compared to experimental data with MEA (red, right)

4. Conclusions & Outlook

Prior research studies have pointed out the importance of reducing the energy requirements for the regeneration in CO_2 capture processes. Lang et al. (2017) reports that applications with novel solvents need to be taken into account in order to achieve the required energy reduction. However, these studies have focused on theoretical simulations based on complex mechanistic models for known amine solutions. This study deals with an experimental characterisation of a novel solvent and the identification of the optimal operation point using data-driven models. Therefore, a three-step approach is developed based on the assumptions that little thermodynamic knowledge of the novel solvent is available and the new scrubbing liquid indicates a similar behaviour as MEA. As a first step, simulation data with MEA is taken as baseline to characterise the general behaviour of a carbon capture process and for the identification of the surrogate model´s structure. The specific energy consumption e_{CO_2} classifies the performance of the process and is computed in dependence of gas and solvent flow. To establish comparability for the novel solvents experiments with MEA are carried out. The surrogate model is fitted to the experimental results with MEA to consider plant characteristics. By means of the adapted model, the optimal operation point for MEA is derived. As a final step, the surrogate model is adapted to experimental data of the novel solvent to evaluate and investigate the novel solvent in terms of energy consumption under experimental and industrial conditions. Using the adapted model the optimal operation point is derived for the novel solvent and compared to that of MEA. Based on the identified minimum, the pilot-plant is repeatedly operated at this point to confirm the minimal energy consumption for each solvent. Within the three-step approach different techniques for the modelling can be applied, such as neural nets. The choice of the model technique is of great importance and goes along with different advantages and disadvantages.

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