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An Overview Of Dynamic Model For CO_2 Removal

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ABSTRACT

Background: This paper presents a general overview of available dynamic model for CO_2 capture. The dynamic model is the most essential step before any project can undergoes commissioning to observe the transient behavior of a system. The finding indicates that there is two models that represent the CO_2 absorption which is equilibrium based and rate based. This model include mass transfer model, reaction kinetics, vapor liquid equilibrium (VLE) and hydrodynamics. CO_2 removal using MEA as chemical solvent become basis of the most dynamic model study since MEA has been well-known for decade. Therefore, any rising attempt toward optimization will lead to enormous achievement. Thus, proper model selection and modifications are very important to minimize error between simulation results and experimental value. Accurate dynamic models can be used to enhance the understanding on absorption dynamic of CO_2 for optimal design and future control study in future.

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INTRODUCTION

CO_2 removal is important in any gas stream process to ensure high quality gas stream products and to fulfill the pipeline specifications, prevent corrosion of gas transport network and process equipment as well as optimize pipeline capacity (Safari, Ghanizadeh *et al*. 2009). The maximum level of CO_2 permitted in natural gas transmitted to customers by pipeline is typically less than 3% (Rufford, Smart *et al*. 2012). With regard to absorption process, packed column is viewed as the most efficient media for the removal of CO_2 since it offers superior contact between gases and liquid that promote high mass transfer rate (Aroonwilas, Chakma *et al*. 2003, Tan, Lau *et al*. 2012). The importance and necessity to develop mathematical models that describe the absorption process as accurate as possible cannot be overstated. The most common and established technology for CO_2 removal is from post combustion process using chemical solvent (Mandal, Guha *et al*. 2001, Godini and Mowla 2008, Rufford, Smart *et al*. 2012, Tan, Lau *et al*. 2012). The majority of their study is in the region of steady state condition but in the recent year, growing attention has been given to the dynamic topic (Chikukwa, Enaasen *et al*. 2012).

Obviously the need for in depth understanding is important to control the whole operation. The transient behavior of the power plant during start up, shut down, and load variation are well known through operational experience but little information to understand the process during this sequence (Chikukwa, Enaasen *et al*. 2012). Therefore, a study on a dynamic, non-equilibrium model for packed column using MEA has been conducted and validated through pilot plant data that are available in the literature (Chikukwa, Enaasen *et al*. 2012, Razi, Bolland *et al*. 2012, Mac Dowell, Samsatli *et al*. 2013). In (Mac Dowell, Samsatli *et al*. 2013), they attempt to look at the dynamic performance by varying the solvent conditions such as concentration and flow rate. A more detailed modeling effort has also been conducted (Cormos and Gaspar 2012) that takes into account the effect of kinetics reaction on the mass transfer, and hydraulic aspect of structured packing and pressure drop along the column as well as looking into various operating conditions.

2. Steady State Model:

Generally, in order to develop dynamic simulation model in simulation software, there are two basic stage models or methods. First is the equilibrium-based model that assumes that the liquid and vapor phases reach equilibrium at theoretical stages and perfect mixing occurs at each stage (Lawal, Wang *et al*. 2009). The rate of absorption and desorption in reactive distillation are determined

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by two main mechanisms, mass transfer and chemical reaction (Lawal, Wang *et al*. 2009). This assumption is combined with mass and energy balance equation to determine the concentration and temperature along the packed column. This approach is more suitable for non-reactive system (Chikukwa, Enaasen *et al*. 2012).

Another approach for description of separation units is the so called rate based approach. This method is more suitable for the reactive process such as CO_2 absorption in MEA. In rate-based modeling, the vapour–liquid equilibrium occurs at the phase interface. The mass transfer between the vapour and liquid phases is modeled based on the two-film theory and the Maxwell-Stefan formulation (Lawal, Wang *et al*. 2009, Bui, Gunawan *et al*. 2014). Some papers also used penetration theory to describe the mass transfer phenomena (Chikukwa, Enaasen *et al*. 2012). The rate-based approach is more rigorous, but the accuracy of the calculation is higher than equilibrium rate (Zhang and Chen 2013, Bui, Gunawan *et al*. 2014). The liquid film thickness is important parameter in the rate based model and usually it is estimated via empirical mass transfer correlation taking into account the type of column, hydraulics and transport properties (Kucka, Müller *et al*. 2003, Afkhamipour and Mofarahi 2014). It has direct link from the column internal and the concept is illustrated in the Figure 1.

Certainly a wide variety of models ranging from simplified equilibrium models to detailed rate based models have been developed in order to analyze the performance of amine based capture (Mores, Scenna *et al*. 2012). The availability of process simulators in recent years has allowed the process performance to be modeled and predicted within a relatively short time without the need to define the complex mathematical equations. However, the process is not a straightforward task since it demands a considerable amount of trial and error effort (Mores, Scenna *et al*. 2012). Table 1 give some review for model comparison, simulation tools, operating conditions considered and parameters studied that have been conducted by other researchers.

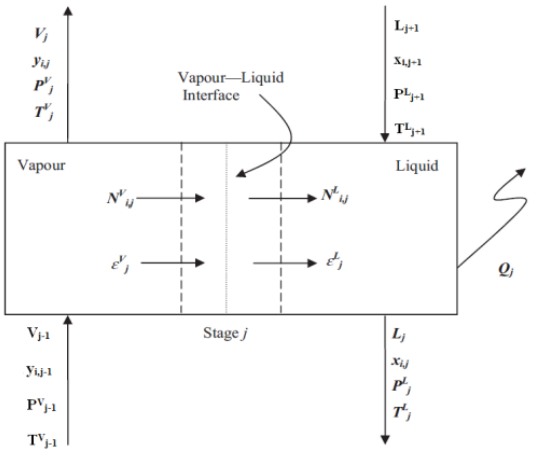

Fig. 1: Schematic diagram for non-equilibrium model, vapor and liquid bulk, film and interface. Adopted from (Mac Dowell, Samsatli *et al*. 2013)

3. Dynamic model:

It can be clearly observed from the Table 1 that most research works are conducted for CO_2 removal from post combustion processes operating at atmospheric pressure. Numerous simulators to emulate their current pilot plant or process have been used such as Matlab, gPROMS, Aspen, Hysis, Sradau, Fortran and etc. The majority of the works involved modeling the absorber and stripper but also include other parts such as reboiler and heat exchanger.

Interesting comparison of rate based simulation between Aspen Hysis and Aspen Plus has been conducted by (ErikØi 2012) to calculate the removal efficiency of CO_2 based on circulation rate, number of stages and inlet temperature. Only small deviation is found and therefore, generally both simulators will give more less the same result. Another comparison of simulators also conducted using different data of pilot plant and they also agreed that all simulators can give reasonable predictions of overall performance (Luo, Knudsen *et al*. 2009).

Researchers have highlighted that the main element in designing a model for absorption process in packed column is a deep understanding of gas solubility and the mass transfer. (von Harbou, Imle *et al*. 2014) has conducted numerous experimental works utilizing two pilot plants data with four different packing. The results shows that correlations such as effective interfacial area (a_{eff}) and gas solubility (Henry's constant) are the most sensitive parameters. They found many uncertainties between the prediction and experimental data is due to unreliable mass transfer correlation. This is also in the same agreement by (Tönnies, Mangalapally *et al*. 2011) as they conduct the sensivity analysis for

absorber and desorber. In their research, they found that influence of interfacial area is distinctly more important than mass transfer coefficient. Therefore, the improvement of effective interfacial area correlation is essential for reliable rate based simulation. However, there is no new correlation of interfacial area is proposed by the author.

Appreciation of dynamic model contribution can be given to (Kvamsdal, Jakobsen *et al*. 2009, Chikukwa, Enaasen *et al*. 2012, Mac Dowell, Samsatli *et al*. 2013) as they have been thoroughly studied dynamic condition in various part of CO_2 absorption process but still acknowledge that extensive work need to be conducted to get more reliable prediction for proper regulation, control strategies and real economic benefit. As the example, (Mac Dowell, Samsatli *et al*. 2013) had interesting query how of humidity and mass transfer zone shape might affect the overall process. Then, the operational challenges such as load variation and high degree of heat integration between power plant and the absorber/ stripper are also required. The other sub-process such as gas turbine, steam turbine, compressor or reboiler also needs to be designed before moving to full scale or commercialization.

A dynamic model of amine based CO_2 absorption that represents for major equipments also can be found in (Jayarathna, Lie *et al*. 2013). The dynamic simulation is performed to analyze the capability of the model to predict the effect on the plant operation caused by the disturbances from the upstream power plant and the operating conditions of the plants. The improvement of the model is presented using better interfacial mass transfer, physical properties of amine solution, and packing hydraulics (Jayarathna, Lie *et al*. 2013). The important of dynamic model for the development of control strategies is presented in (Harun, Nittaya *et al*. 2012). In their research, they found that the changes in gas flow rate and reboiler heat duty are key process variables that affect percentage of CO_2 removal. Besides, the lean loading stream and the energy performance of the process are directly affected by the operation of the process. Comprehensive dynamic model and validation using two film theory with rate based approach can be found in (Biliyok, Lawal *et al*. 2012). The absorber and generator were modeled and it was observed that the model is adequately predicts the pilot plant behavior under multiple process inputs and disturbances.

As dynamic model become more noticeable nowadays, the development of detailed mathematical correlations becomes necessary. In (Gaspar and Cormos 2012), several mass transfer and hydraulic correlations were investigated using four different types of alkanolamine. The study shows that the proper selection of mathematical model reveals a good quality of prediction for absorption process. Besides that, dynamic study also conducted by (Karimi, Hillestad *et al*. 2012) for two different configurations. Result shows that vapor recompression can handle disturbance better than split stream configuration. They intended to further the research for control structure and therefore can cover up for energy loss.

Another essential point to note down for dynamic model is the assumptions made by all the researchers. The assumptions made behind the mass transfer correlation and kinetic model can cause discrepancies between simulated profiles and experimental data (Afkhamipour and Mofarahi 2014). These assumptions applied to help the researchers simplify the difficulties of the model. Generally, researchers will assume that no material accumulation in liquid film, vapor film and bulk vapor (Biliyok, Lawal *et al*. 2012) and therefore the continuity equation can be eliminate. Another important assumption is made that phase equilibrium is attained at the vapor liquid interface and this will link to vapor fugacity and liquid fugacity (Biliyok, Lawal *et al*. 2012). The fugacity is closely related to compressibility and the function will be apparent as pressure become the control variable. The mass transfer is assumed like two film theory where resistance of mass transfer lies at both side (Kvamsdal, Jakobsen *et al*. 2009). Mass transport for other elements such as N_2 or O_2 are neglected since the solubility of those gases in MEA is very low resulting the contribution in dynamic performance to be very small (Jayarathna, Lie *et al*. 2013).

Table 1: Comparison of studied parameters and the simulation tools for dynamic model

Simulation Tools	Studied parameter	Operating Condition	Equipment	Reference
gPROMs / Microsoft Excel	Temperature profile/ CO_2 mass fraction/ Flue gas moisture content / Flow rate	T=37-120 ^{0}C, P= 1.05 bar	Absorber/ Regenerator	(Biliyok, Lawal *et al*. 2012)
SAFT-VR/ gPROMS	CO_2 concentration/ Heat transfer/ Mass transfer/ Solvent flow rate	T=20-77 ^{0}C, P= atmospheric	Absorber	(Mac Dowell, Samsatli *et al*. 2013)
gPROMS/ SRADAU solver	Temperature profiles/ Liquid flow rate/ Partial pressure	T=20-77 ^{0}C, P= atmospheric	Absorber/ Stripper	(Kvamsdal, Jakobsen *et al*. 2009)
Matlab	Temperature profile/ CO_2 removal efficiency/ Reboiler heat duty	T=39-83 ^{0}C, P= 0.1 -2.3 bar	Heat exchanger / Buffer tank / Absorption column	(Jayarathna, Lie *et al*. 2013)
Matlab	Flow temperature profile/ Packed Height/ CO2 removal efficiency	T=40-100 ^{0}C, P= atmospheric	Absorber/ Stripper	(Jayarathna, Lie *et al*. 2013)

gPROMS/ ASPEN	Temperature profiles/ Flue gas flow rate/ Reboiler heat duty	T=46.71 ^{0}C, P= atmospheric	Packed Column / Reboiler / Heat Exchanger/ Tank	(Harun, Nittaya *et al*. 2012)
Matlab/Simulink	CO_2 absorption based on different solvent / mass transfer / Hydraulic correlation	Not stated	Absorber	(Gaspar and Cormos 2012)
Unisim	Temperature profile / Reboiler duty	T=48 ^{0}C, P=1.1 bar	Absorber/ Stripper	(Karimi, Hillestad *et al*. 2012)

4. General Model Of CO_2 Removal:

4.1 Chemical reaction:

Generally, systems that engage with solvent have relatively rapid reaction kinetic. For example, MEA has become well known as basic solvent for the CO_2 absorption. It responds faster than those that have slower kinetics such as MDEA (Mac Dowell, Samsatli *et al*. 2013). The chemical reaction is important as it influences the mass transfer rate that takes into account the enhancement factor (Mores, Scenna *et al*. 2012). The absorption of CO_2 by aqueous MEA involves a complex system of weak electrolyte and reversible reaction (Kucka, Müller *et al*. 2003, Ghaemi, Shahhosseini *et al*. 2009). This reaction involved first and second order kinetics. Since CO_2 can absorb into water, the first reaction is CO_2 hydration and this process is very slow and called CO_2 hydration (Ghaemi, Shahhosseini *et al*. 2009).

$CO_2 + H_2O \leftrightarrow HCO_3^- + H^+$(*$CO_2$ Hydration*)

The primary and secondary ethanolamine reacts with CO_2 based on zwitterions mechanism with positive and negative charge (Bavbek and Alper 1999). The three reactions that appear to be of principle importance for the CO_2-MEA are as follows (Xie, Zhou *et al*. 2010, Mores, Scenna *et al*. 2012);

$CO_2 + 2RNH_2 \rightarrow RNHCOO^- + RNH_3^+$(*Carbamate Formation)*

$CO_2 + RNH_2 + H_2O \rightarrow HCO_3^- + RNH_3^+$(*Bicarbonate formation)*

$RNHCOO^- + CO_2 \rightarrow HCO_3^- + 2RNH_3^+$*(Carbamate reversion)*

4.2 Mass balance:

The accumulation of mass *(M)* based on one segment of packed column is explained below. The vapor (*V*) and liquid (*L*) is flow in countercurrent whereby gas coming from bottom while solvent come from the top and meet at the section or stage *j* in packed column (Mac Dowell, Samsatli *et al*. 2013).

The mass balances for liquid and vapor phases are:

- $\frac{dM_{ij}^L}{dt} = L_{j-1}x_{ij-1} - L_j x_{ij} + N_{ij}^L$

i= 1,….., *c* *j*= 1,….., *NS*

- $\frac{dM_{ij}^V}{dt} = V_{j+1}y_{ij+1} - V_j - N_{ij}^V$

i= 1,….., *c* *j* = 1,….., *NS*

Mole fraction in each stream leaving the stream must sum to unity.

- $\sum_{i=1}^{c} x_{ij} = \sum_{i=1}^{c} y_{ij} = 1$ *j*= 1,…..,NS

No accumulation of mass at interphase.

- $N_{ij}^V - N_{ij}^L = 0$*i*= 1,...,*c* *j*= 1,…..,NS

Where *i* is the component and *j* is the number of stages.

4.3 Heat balance:

The energy balances for liquid and vapor phases are:

- $\frac{dU_j^L}{dt} = L_{j-1}h_{j-1}^L - L_j h_j^L + \varepsilon_j^L - Q_j$

j= 1,…..,NS

- $\frac{dU_j^V}{dt} = V_{j+1}h_{j+1}^V - V_j h_j^V - \varepsilon_j^V$

j = 1,…..,NS

4.4 Reaction Kinetic:

This is the most important and challenging part of the model development. This is because the model development for kinetics reaction will be different for different solvent used. Without a good understanding and detail parameters, an accurate model could not be obtained (Afkhamipour and Mofarahi 2014). The enhancement factor, *E* is one of the complicated characteristics of modeling gas absorption with chemical reaction. It represents the effect of chemical reaction of CO_2 based on the solvent used (Cormos and Gaspar 2012). *E* is defined as the ratio between mass transfer coefficients with chemical reaction, k_L to the mass transfer coefficient for purely physical absorption, k_L^o (Khan and Mahmud). It requires extensive and reliable experimental data especially for novel or mixture solvent (Mac Dowell, Samsatli *et al*. 2013). Table 2 presents the enhancement factor that has been designed for chemical reaction between CO_2 with various solvents.

The enhancement factor given by (Kvamsdal, Jakobsen *et al*. 2009) is strictly valid for pseudo first order regime, It require large physical mass transfer coefficient to determine the rate of chemical reaction in the liquid film. This is applicable for the separation of CO_2 in packed column using alkanolamine because of effective liquid mixing. The

pseudo first order reaction is supported by the combination of low CO_2 partial pressure, high reactant concentration and short contact time.

In (Mandal, Guha *et al*. 2001), the investigation for CO_2 removal into aqueous blend has been developed based on Higbie's penetration theory. Good agreement have been found between the models predicted rates and enhancement factor with experimental results. This indicates that proper combination of mass transfer, reaction kinetics and equilibrium model can represents CO_2 mass transfer for the aqueous amine blends effectively. The effect of chemical reaction that using similar enhancement factor from Higbie's penetration theory also applied in (Sema, Naami *et al*. 2012). In the sensivity analysis study using rate based approach, (Afkhamipour and Mofarahi 2014) had informed that the performance of model is strongly depend on the selection of model parameter such as mass transfer correlation, transport properties, kinetic model and physical properties. In their research, mass transfer correlation was combined was combined with kinetic model. Since, the reaction of CO_2 and amine is considered very fast and instantaneous, thus based on two film theory, explicit expression of enhancement factor is derived as instantaneous reaction, E_∞ and Hatta number, Ha.

Conclusion:

There is a notable development and comprehensive activity toward dynamic model but majority of the dynamic modeling studies are found for the post combustion process operate at atmospheric pressure. The result from this study is expected to open up the possibility to explore the availability of utilizing natural gas with high CO_2 content at elevated pressure. Another key point to highlight is that the application of standard absorption models (e.g. either rate based or equilibrium based) for CO_2 removal is not a straightforward task The successful of the model can be exploited to imitate the treatment of CO_2 removal at any conditions with various type of situation or disturbance. Certain studies had start to evoke the important of dynamic model but the validation is not yet enough. Therefore, more research work need to be conducted to understand the fundamental phenomenon occurring in packed column particularly at high pressure before designing the simulation model.

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