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Dynamic Modeling of Post-combustion CO_2 Capture Using Amines – A Review.

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Abstract

The recent years have seen growing attention towards the study of dynamic behavior in post-combustion CO_2 capture plants using amines, albeit, apparently contesting yet without comparison or critique. This paper reviews what has been reported in literature concerning issues pertinent to transient behavior of CO_2 capture including interaction with power plants. Details of models used, their validation and modeling tools as well as an attempt to piece-out convergent points from the various conclusions are given. Knowledge gaps and areas that still need more attention are emphasized.

Keywords: Dynamic modeling; post-combustion; flexibility; integration; review.

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1. **Introduction**

Humanity is facing an inherently paradoxical challenge: meeting the increasing global energy demand while simultaneously mitigating climate change. Consequently, there is growing concern across the world over rising concentrations of anthropogenic greenhouse gases in the atmosphere, mainly CO_2 which is considered to be the chief culprit propelling climate change. Fossil fueled power generation, apparently indispensable at the moment [5, 6], is the largest source of CO_2 emission.

Carbon capture and storage (CCS) is suggested as one of the main options for reducing global CO_2 emissions. However, the design of the existing fossil-fueled power plants was not meant to accommodate CCS. For this reason, post-combustion CO_2 capture has a significant edge over other alternatives because the technology can simply be implemented as an ‘end-of-the-pipe’ retrofit without the need for radical changes to existing power plants [7]. As such, the most mature, sufficiently studied and documented technology for post-combustion CO_2 separation is chemical absorption using amines. However, most of these amine-based CO_2 capture studies available in literature, be they experimental or validated theoretical simulations or modeling, are premised on steady state [8].

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Relative to steady state there is a modicum, nevertheless, growing attention towards the study of dynamic behavior in CO_2 capture over the recent years (being evidenced by an increasing amount of literature on the topic), apparently contesting yet without aggregational collation or critique. The aim of this paper is, therefore, to give a review of what has been reported in the literature concerning dynamic post-combustion CO_2 capture as well as pertinent studies on interactions between power plants and CO_2 capture plants. The main focus is on modeling of the capture plant and the interactions with the power plant are handled as external disturbances.

2. Motivation: Dynamic modeling is a practical necessity

It is becoming increasingly cogent (technically) that post-combustion CO_2 capture is heading towards full scale. Most probably, this will naturally be followed by subsequent commercialization. However, there is no real-world experience with large scale integration with power plants thus far. The extent to which the capture plant will affect the flexibility of the power plant is of great interest, especially during transient operation.

The absorber/stripper process is fairly complex, characterized by interactive interference between the columns. Moreover, incorporation of optimal design and operational improvements of the absorption process (e.g. intercooling, lean vapor recompression, multistage stripping) will contribute further to inherent complexity. In case of biofuels and coal-based power plants, the condition of the fuel might vary during operation implying varying flue gas composition. In addition, the path towards carbon neutral energy systems (with the expected growth of renewables and other ´green´ energy sources) will require an even higher degree of flexibility in fossil-fueled power generation. As an illustration, Figure 1 shows the expected dynamic interaction between conventional power generation (coal or gas) and the renewable alternatives. Of importance to note is that this prediction is based on realistic simulation of wind and tidal output data (from 2007) scaled-up to meet the anticipated goal of 38% from renewables in the UK by 2025 [1]. Obviously, the need for an in-depth grasp of the power/capture plant's operational flexibility is a palpable necessity before full-scale CO_2 capture can be realized.

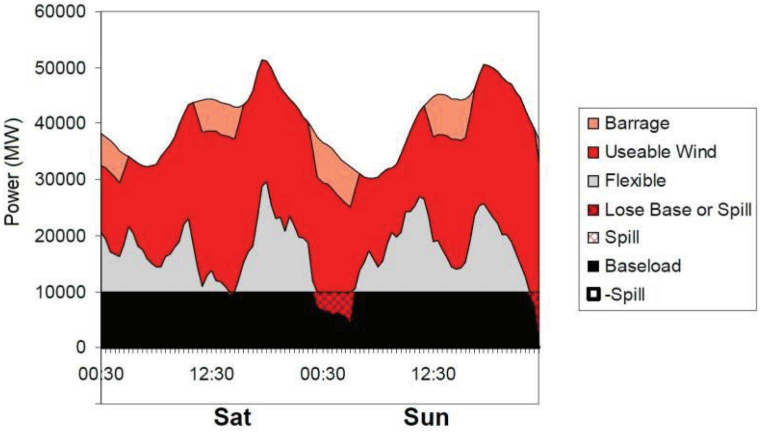

Figure 1: A schematic showing the expected dynamic interaction (speculative) between the green energy sources vs coal/gas power generation in 2025. Adapted from [1].

In order to get a full understanding of the transient characteristics of a power plant with CCS, both the power plant and capture process should be investigated in a combined dynamic model. The power plant steam cycle and CO_2 capture unit are integrated [9, 10] and therefore, highly dependent on each other's performance [11, 12]. The transient behavior of the power plants occurring during start-up, shut-down, and load variation, are well-known through operational experience. However, there is little knowledge of how the absorption process operates during these sequences. Due to chemical reactions occurring in the capture process, the dynamic behavior is more complicated compared to that of the power plant (mainly related to variations in mass and energy rates). Dynamic simulation will play a pivotal role in identifying any operational bottlenecks at transient conditions for the integrated power and CO_2 capture plants.

3. Modeling of post-combustion CO_2 capture using amines

The conventional CO_2 capture loop basically consists of two columns: an absorber and a stripper coupled via a heat exchanger, see Figure 4. In principle, the two columns require different conditions (tailored to favor either CO_2 absorption in the absorber or desorption in the stripper) for optimal operation. The chemical solvent is loaded with CO_2 in the absorber and then pumped to the desorber where the CO_2 will be stripped off. A condenser removes water from the gas exiting the stripper, leaving a CO_2 product of almost 99% purity which is then compressed for transportation. The lean solvent is circulated back to the absorber.

The importance and necessity to develop mathematical models that describe the absorption and desorption process as accurate as possible can not be overstated. For modeling fluid flow, simple plug flow models are widely used for both gas and liquid phase. However, describing mass and heat transfer is seen as a more challenging part of model development. Particularly two philosophies are popularly used in literature for modeling mass and heat transfer across the interphase, namely the two-film theory and the penetration theory. It seems the two-film theory is more popularly used compared to its counterpart. The concept is illustrated in Figure 2 and shows how CO_2 diffuses from the gas bulk phase through a gas film, before being absorbed at the interface and then diffuses through a liquid film to the liquid bulk phase. It is assumed that the resistance to mass transfer is concentrated entirely in the films adjacent to the interphase. In the fluid bulk phases outside the films, the level of mixing is assumed to be sufficiently high so that there is no composition gradient.

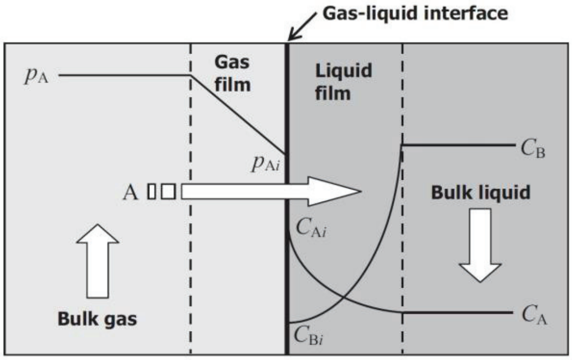

Figure 2: A schematic showing the concept of mass transfer model based on the two-film theory. Adapted from [3].

Absorption of CO_2 by amines involves chemical reactions, and in most cases the chemical reactions are fast enough to influence the rate of mass transfer in the films. Thus, mass transport and chemical reaction occurs simultaneously giving changes in the concentration gradients. This leads to enhancement of the mass transfer which must be described in the model.

Different modeling approaches are used to describe and model mass and heat transfer in the columns. Generally, two concepts are commonly used: the equilibrium and non-equilibrium stage models. The essence of the equilibrium approach is ideal for non-reactive systems and is based on theoretical segments (linked through mass and energy balance equations) in which the liquid and gas are assumed to attain equilibrium characterized by infinitely fast mass transport. The performance of the individual stages is then

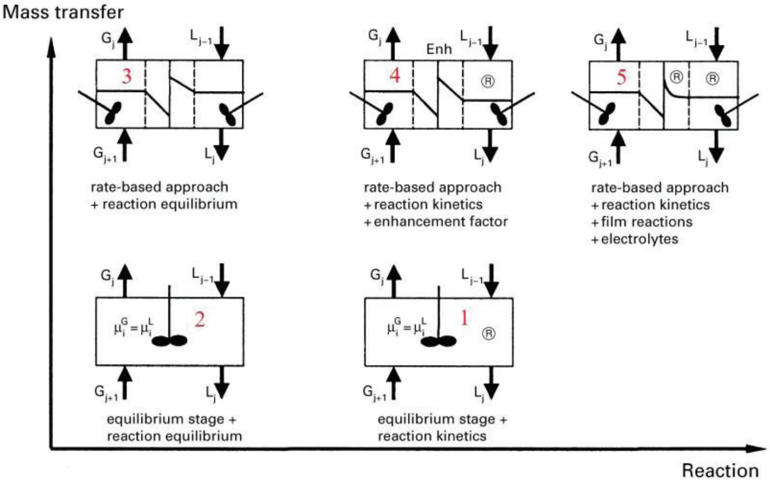

Figure 3: Theoretical representation of the different levels of model complexity for one segment. Adapted from [2]. The models are labeled 1-5 for convenience in reference, especially in Table 1.

adjusted by means of a tray efficiency factor. All the same, chemical reactions between CO_2 and amines imply that vapor-liquid equilibrium is rarely attainable in practice. This makes the non-equilibrium (i.e. rate-based) approach more appropriate. The gist of this concept takes into account the actual rates of mass and heat transfer including chemical reactions. Mass and heat transfer across the interphase can be described by the two-film model as explained above. The reaction depends on the kinetics regime, and can either be considered instantaneous (equilibrium can be assumed) or kinetically controlled.

Both approaches have varying degrees of complexity largely differentiated by intricacies as illustrated in Figure 3, in which the horizontal and vertical axes show increasing complexity for both reaction and mass transfer respectively. According to Baur et al [13], it is advisable to employ the rate-based approach when dealing with reactive columns. Nevertheless, rigorous dynamic descriptions of industrial gas-liquid contactors lead to extended systems of equations difficult to solve reliably and quickly enough. All the same, this seems to be less of a hindrance nowadays, thanks to the advances made to the processing speed of the modern computers.

4. Characterizing literature on dynamic modeling

4.1. General trends

Until only a few years ago, there has been some appreciable degree of inadvertence regarding dynamic modeling in the research of post-combustion CO_2 capture by chemical absorption. Generally, the main thrust has been directed towards steady state analysis of power plants operating at design conditions (full-load), and consequently, numerous publications on the topic exist. Nevertheless, steady state analysis does not correctly represent neither issues related to daily operations nor the transient behavior of these plants. All the same, there seems to be some kind of awakening towards dynamic modeling in recent years as depicted in Table 1. This tabulation attempts to categorize the issues addressed in literature regarding dynamic modeling. In this case, the main emphasis was directed towards technical aspects of the conventional post-combustion CO_2 capture by chemical solvents. It seems that there has not been so much effort towards dynamic modeling before 2008, albeit a dramatic increase on studies attempting to deal with the subject is noticeable.

Of importance to note is that quite a significant number of authors model either the absorber or the stripper only, although a greater majority attempts to include both columns in their models. Most of the models assume plug flow for both gas and liquid phase, and the modeling approach for describing mass and heat transfer as well as the effect of chemical reactions is indicated in the table according to Figure 3. Further more, it appears that almost all model validation is based on steady state, save a single attempt by Kvamsdal et al [14]. Even so, this effort does not give a complete picture because it deals with the absorber section only. Although there is a sizeable, growing and diversified list of tested and tried solvents, apparently all modelers have thus far chosen to use MEA for dynamic modeling on post-combustion CO_2 capture. Notwithstanding, the overall basis for evaluating dynamic behavior is quite broad, ranging from variations in power plant load to perturbations in reboiler duty including other important aspects like disturbances in flue gas composition, flow rates, rich/lean loadings, water balance, etc. It is also interesting to note that a variety of modeling tools (MATLAB, gPROMS, Modelica, Aspen Plus, etc) have been used for implementation of the developed models.

4.2. Summary of main results based on literature in Table 1

In this section we seek to discuss the major findings of the articles in Table 1. To start with, it is important to note that the basis on which dynamic behavior has been studied by the various researchers is quite diverse. In general, the majority of the articles show that there is a time lag between perturbations and system response, which needs to be taken care of as the system moves from one steady state to another [15, 16]. A cross-examination of the results, obtained by different researchers, concerning the capture system’s time response suggests that it is strongly influenced by local size and setup.

Table 1: **An epitomic overview of literature survey on dynamic modeling of post combustion CO_2 capture. All researchers in this table used MEA as the chemical solvent.**

Reference & (Year of publication)	**Modeled Section (Absorber or Desorber)**	**Data used for Validation**		**Level of Complexity & Modeling Tool**		**Basis for Evaluating Dynamic Behavior**
		Steady State	Dynamic State	Level (Figure 3)	Model Implementation Tool	
Noorlisa et al [8] (2010)	Both	x	-	4	gPROMS	– flue gas flow rate
Ziaii et al [20] (2009)	Both	-	-	5	MATLAB	– reboiler duty – rich solvent load
Ziaii et al [21] (2009)	desorber	-	-	5	Aspen Custom Modeller	– reboiler duty – rich stream flow rate
Ziaii et al [22] (2011)	Both	-	-	not specified	Aspen Custom Modeller	– partial boiler load – partial steam load
Sanoja et al [16] (2010)	absorber	x	-	4	MATLAB	– power plant load
Prölß et al [23] (2011)	Both	-	-	4	Modelica	– flue gas flow rate
Gáspár and Cormos [19] (2011)	Both	x	-	4	MATLAB & Simulink	– changing power plant load – decreasing rich stream temperature
Lawal et al [15] (2010)	Both	x	-	5	gPROMS	– water balance – flue gas flow rate – reboiler duty – flue gas composition
Lawal et al [24] (2011)	Both	x	-	not specified	gPROMS	– flue gas composition
Lawal et al [17] (2009)	Both	x	-	5	gPROMS	– power plant load – reboiler duty
Lawal et al [25] (2010)	Both	x	-	3	gPROMS	– power plant load – CO_2 capture level
Lawal et al [18] (2009)	absorber	x	-	2 & 5	Aspen Plus	– power plant load – lean loading
Greer et al [26] (2010)	desorber	x	-	not specified	MATLAB	– flue gas composition
Kvamsdal et al [27] (2009)	absorber	x	-	4	gPROMS	– L/G ratio
Kvamsdal et al [14] (2011)	absorber	x	X	4	MATLAB	– L/G ratio – flue gas composition
Greer, T [28] (2008)	Both	x	-	4	MATLAB	– perturbations in model parameters & inputs

Noorlisa et al [8] generally concluded that the partial reduction of the flue gas load significantly affects absorber/stripper performance. This is further supported by Lawal et al [17, 18], in which it is shown that absorber operation is more sensitive to perturbations in L/G ratio compared to individual liquid or gas flow rates while the regenerator performance is quite sensitive to disturbances in the reboiler duty.

However, findings by Gáspár and Cormos [19] suggest that the capture unit has even higher sensitivity to changes in the desorber feed stream temperature compared to the L/G ratio. Along the same line of thought, the studies by Ziaii et al [20, 21] suggest that the liquid residence time in the reboiler at the final steady state condition could be the dominant factor in the response time of the stripping section. In another study, Ziaii et al [22] found that for reboiler steam partial load, a linear relationship exists between optimum solvent rate and reboiler steam rate.

In a separate study, Lawal et al [25] extended their effort to include two dynamic case studies in which they surmise that the CO_2 capture section has a slower response compared to the power plant. It is further discussed how CO_2 capture level affects the power plant output together with the associated difficulties in achieving a steady power output quickly.

It can generally be said that the work done thus far regarding characterization of amine-based post-combustion of CO_2 capture in the dynamic mode is still largely an incongruous mixture of indeterminate conclusions. As such, it is still inherently difficult to affirmatively piece-out the puzzle regarding dynamic behavior of a CO_2 capture unit. Resultantly, clear strategy on how to handle the CO_2 capture unit in transient mode is still lacking although the technology seems to be moving towards full-scale. This is further compounded by the fact that the existing motley of conclusions is all essentially based on steady state validation. In other words, the present knowledge regarding dynamic behavior of CO_2 capture is incomplete both theoretically and in practice. The obvious implication is that this might slow down implementation of CCS if left unaddressed until full-scale.

4.3. Other issues discussed within dynamic behavior

Some of the works (in Table 1) extended their studies to include a variety of other issues. For example, Ziaii et al [21] attempted to address how flexibility in both power and CO_2 capture plants can improve operating profits by facilitating the operators to examine the balance between power output and pricing as determined by market conditions. In this regard, the authors essentially emphasize the fact that sound knowledge on the dynamic behavior is crucial in order to determine optimal loads as well as when and how to operate CO_2 capture lucratively on hourly basis. The success of such an approach is closely linked to a clear understanding of how dynamic optimal operation of the capture plant works.

At a technical level, Lawal et al [17] compare the equilibrium and rate-based approaches in modeling the absorber dynamically. As one would expect, it was concluded that the rate-based approach yields better predications compare to its equilibrium-based counter part. Besides discussing the technical dynamic behavior of the absorber section Kvamsdal et al [14] went further to investigate the effect on the overall performance of the model, by substituting different parameter-correlations available from various sources in literature. Important observations and indications are noted in their analysis; notwithstanding, the fact that this work was confined to the absorber section only makes the associated conclusions difficult to generalize.

5. Interaction between the power plants and CO_2-capture plants

CCS in general gives rise to an energy penalty which decreases the net efficiency of the power plant [29]. The sources leading to energy penalty are largely constituted by heat supply to the desorber reboiler (steam taken directly from the power cycle), shaft power to compression of CO_2 and other electric power consumers like pumps, blowers etc [10]. However, CO_2 separation (reboiler duty) is energy intensive, responsible for the largest (relative) energy penalty [7, 30]. A simplified diagram showing how the two plants interact with each other is given in Figure 4.

In comparative terms, efficiency has become the dominant issue when designing and selecting power plants with CO_2 capture. Other aspects, like reliability and operability, have been given less importance, if any at all, in literature. This section focuses on studies pertinent to the transient interaction between the power plant and the post-combustion CO_2 capture plant. As such, direct relevance to the integrated flexibility of both the power and capture plants has been of prime interest in this case. Due to the integration of power plant steam cycle with desorber reboiler in the capture plant, these units are highly dependent on each others performance and should be investigated in a fully combined dynamic model to identify any operational bottlenecks at transient condition. However, most of these studies assume steady state and full-load design, albeit, covering a diversity of investigations ranging from power plant types, usage of different grades and quality of fuels [5] to a variety of solvents. Resultantly, generalization and application of results is thus largely subject to considerable limitations.

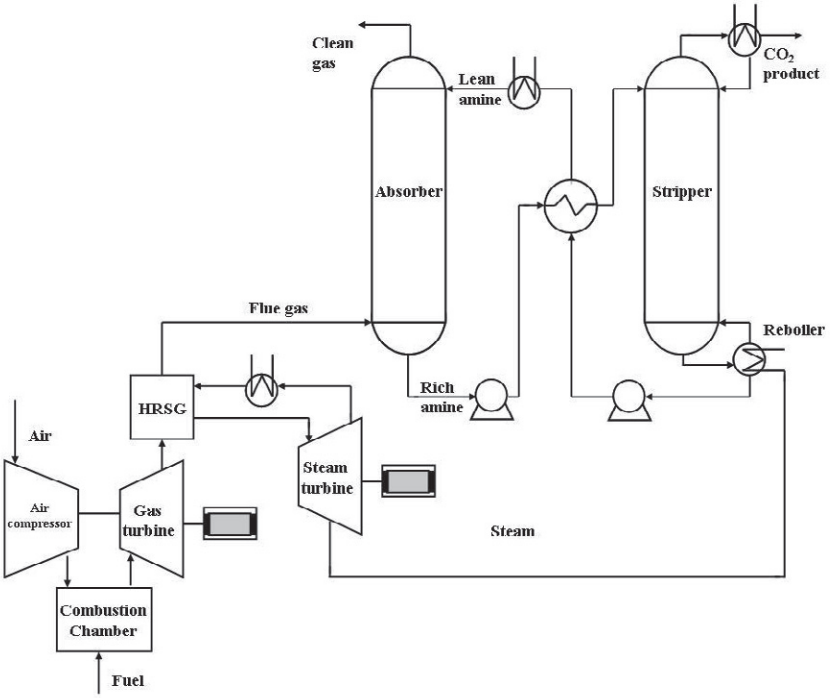

Figure 4: Simplified diagram to illustrate the integration of a natural gas power plant and a CO_2-capture plant. Adapted from [4] with modifications.

A study by Alie et al [31] focuses on the operability of power plants with CCS and highlights how flexibility is critical to integral operability. The article takes note of the inroad studies addressing issues related to flexibility, controllability, start-up, shut down and other aspects characteristic to the transient mode exist, albeit, techniques used are basically theoretical methodologies and experience-based approaches.

Sanpasertparnich et al [10] studied how various important parameters in the power plant, CO_2 capture plant and compression unit affects the coal-fired power plant performance. Focus is not only on full load, but also on the impact of part load. Their conclusions suggest that CO_2 capture efficiency yielding to optimal energy penalty is independent of type of coal studied and steam extraction location.

Generally, the closest various researchers have come to address the issue of integrated flexibility of power and capture plants is based on part load studies. Even so, steady state is assumed at partial load. Efforts to study what goes on as the system moves from one steady state to another seem to be lacking still. In this regard, Chalmers and Gibbins [11] highlights the need for some comprehensive understanding of the potential impacts of post-combustion capture on dynamic performance of the power plant to be able to optimize the process during varying operation. Identifying potential improvements to plant dynamic performance is important, since this may improve the power plant's economics [32].

6. Conclusions

There is in general a notable awakening and multifaceted activity towards dynamic modeling of post-combustion CO_2 capture using amines. However, one problem is visibly salient: lack of dynamic data to validate the developed models. As such, the majority of the models is validated against steady state data and based on one solvent, MEA. Attempts to assess flexible integration of power plants and CO_2 capture plants are basically based on part load, however, still assuming steady state.

Another outstanding feature that traverses literature analyzed in this work is that dynamically validated models are critical towards comprehension of the potential impacts of post-combustion capture on net flexible performance of integrated power/capture plants. Moreover, it is now technically cogent that post-combustion CO_2 capture is heading towards full scale and subsequent commercialization. However, the path towards synergistic and interactive hybridization of fossil-fueled power generation with intermittent 'green' energy sources will undoubtedly exert an even higher demand for dynamic flexibility on integrated power/CCS plants.

To the contrary, dynamic flexibility of integrated plants is still not sufficiently studied, since efforts on this topic (so far) are still largely theoretical and just based on experience to some extent. Moreover, the ability to predict accurately potential improvements and optimization of integrated dynamic performance can provide real economic benefits. Dynamic modeling naturally gives detailed foresight that is fundamental to the establishment of proper regulation as well as control and procedural strategies even as early as plant design stage.

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