Characterization and modelling of aerosol droplet in absorption columns

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ABSTRACT

Formation of aerosols can cause serious complications in industrial exhaust gas cleaning processes. Small mist droplets and fog formed can normally not be removed in conventional demisting equipment because their submicron size allows the particles or droplets to follow the gas flow (Schaber et al., 2002). As a consequence of this aerosol based emissions in the order of grams per Nm^3 have been identified from PCCC plants (Khakharia et al., 2015).

In absorption processes aerosols can be created by spontaneous condensation or desublimation processes in supersaturated gas phases or as a result of droplets or particles entering with the exhaust gas. Undesired aerosol formation may lead to amine emissions many times larger than what would be encountered in a mist free gas phase in PCCC development. It is thus of crucial importance to understand the formation and build-up of these aerosols in order to mitigate the problem.

This paper presents a rigorous model of aerosol dynamics leading to a system of partial differential equations. In order to understand the changes taking place with a particle entering an absorber an implementation of the model is created in Matlab. The model predicts the development in droplet size, droplet internal variable profiles and the mass transfer fluxes as function of position in the absorber, and thus also at the outlet. The Matlab model is based on a subclass method of weighted residuals for boundary value problems named, the orthogonal collocation method.

The model comprises a set of mass transfer equations for transferring components and the necessary diffusion reaction equations to describe the droplet internal profiles for all relevant constituents. Also included is heat transfer across the interface and inside the droplet.

This paper presents results describing the basic simulation tool for the characterization of aerosols formed in CO_2 absorption columns and gives examples as to how various entering droplets grow or shrink through an absorber and how their composition changes with respect to position.

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1. Introduction

Global climate change is the most prominent environmental and energy policy issue of the current era. Burning of fossil fuels, several industrial processes, and various land use practices are contributing greatly to the accumulation of greenhouse gases (GHGs) and in particular CO_2 in the atmosphere.

Carbon capture and storage (CCS) is globally under immense research and development in order to reduce CO_2 emission to the atmosphere (Wall, 2007). Post Combustion CO_2 Capture (PCCC) by chemical absorption is one of the most developed and promising technologies for capturing the CO_2 emitted from large scale fossil fuel based power plants (Abu-Zahra, 2009; MacDowell et al., 2010; Rochelle, 2009; Sanchez-Fernandez et al., 2013)

Monoethanolamine (MEA, 30 wt% aqueous solution) has often been considered as the base solvent for CO_2 capture by absorption because of its relatively high capacity, absorption rate and low price (Abu-Zahra et al., 2007; Ahn et al., 2013; Puxty et al., 2009). The entire process is energy demanding and reduces the overall power plant efficiency up to 12% (Abu-Zahra, 2009). There has been an increasing concern about solvent emissions from absorption columns (Mertens et al., 2012). Prevention of amine emissions is also of high importance for the overall sustainability and performance of absorption based Post Combustion CO_2 Capture facilities. High amine emissions encountered have partly been attributed to aerosol formation in the absorption columns. The loss of amine solvent leads to higher operating costs as well as affects the envi-

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ronment (IEAGHG, 2010). In a typical PCCC pilot plant, 147 mg/Nm^3 of MEA emissions in the treated flue gas stream has been measured based on volatility of amine (Nguyen et al., 2010). In the presence of sulphuric acid aerosols these emissions can increase to 600–1100 mg/Nm^3 (Khakharia et al., 2013, 2015) and without water wash and demisting equipment, around 3000–3500 mg/Nm^3 MEA emissions have been reported (Mertens et al., 2014). BASF, Linde and RWE Power have tested BASF's innovative, amine-based capture technology at the post-combustion capture pilot plant at Niederaussem and have also tested an amine mitigation system using wet ESP. The campaign results are described in Moser et al. (2015, 2014).

Aerosols are systems of particles or droplets suspended in gases. Undesired aerosol formation is a well-known industrial phenomenon in gas–liquid contact devices in which absorption or condensation processes take place. It has been described and explained in various literatures (Ehrig et al., 2002; Neiburger, 1968; Schaber et al., 2002). In gas-liquid contact devices like absorbers, quench coolers, or condensers, aerosols can be formed by spontaneous condensation or desublimation in supersaturated gas-vapour mixtures. Supersaturation is an essential precondition of spontaneous phase transitions and aerosol formation. The degree of saturation S in a multicomponent mixture of an inert carrier gas and condensable vapour components K can be defined by Gretscher and Schaber (1999);

$$S = \frac{P_x\left(T, y_1^G, \ldots, y_K^G\right)}{P_{xs}\left(T, y_1^G, \ldots, y_K^G\right)}$$

Where P_x is the actual partial pressure of any vapour component ($x = 1, \ldots, K+1$, component $K+1$ being the inert gas) and P_{xs} is the equilibrium pressure at the dew point of the vapour mixture.

Supersaturation ($S > 1$) can appear in absorption and condensation processes by two different mechanisms:

- Chemical reactions in the gas phase followed by desublimation of the generated substances. For example the reaction of ammonia with hydrogen chloride can be mentioned.
- Simultaneous heat and mass transfer processes can cause an intersection of the phase equilibrium line by the process trajectory, describing the change of state of the gas phase.

The saturation has to surpass the critical barrier before nucleation and consequently aerosol formation can take place (Schaber et al., 2002).

These aerosol droplets may form before the gas enters the absorber column and may be of various compositions. In order to understand the formation and build-up of these aerosols, to compare with experimental data and to mitigate this problem by better design and operation of gas cleaning equipment, a detailed simulation tool is required that is able to predict aerosol droplet development as function of aerosol droplet characteristics and gas parameters.

This paper focuses on the behaviour of a single aerosol droplet entering an absorber. When modelling aerosol dynamics, governing equations for growth of aerosol droplets lead to a system of partial differential equations which has to be solved numerically. In order to predict the fate of a particle entering an absorber an implementation of the model is created in Matlab. The model predicts the size, the particle internal variable profiles and the mass and heat transfer fluxes as function of position in the absorber, and thus also at the outlet.

The model comprises a set of mass transfer equations for transferring components and the necessary diffusion reaction equations to describe the droplet internal variable profiles for all relevant con-
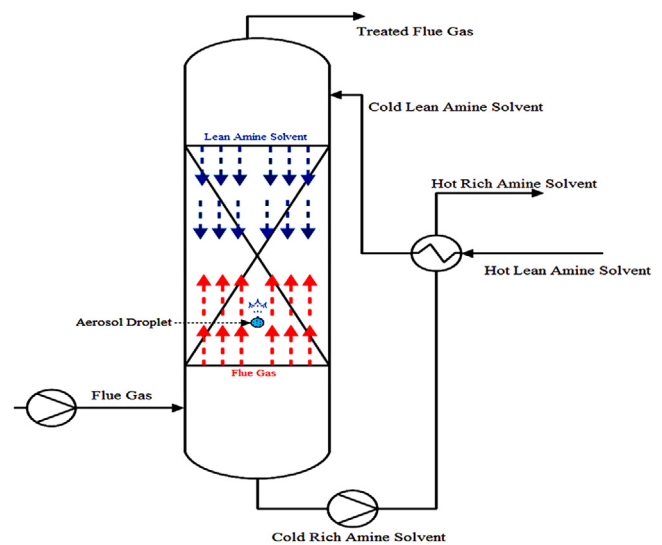

Fig. 1. Schematic illustration of aerosol phenomena in absorption column.

stituents. Also included is heat transfer across the interface and inside the droplet.

2. Modelling

In order to understand aerosol characteristics in absorber columns mechanistic model is required as physical scenarios in gas-liquid contact devices are very complex and detailed explanations are needed to understand results acquired on a more aggregate level.

In order to model the transfer processes between an aerosol and its surrounding gas and liquid phase. The work starts with a single aerosol droplet. In typical CO$_2$ capture absorption columns two streams are flowing counter-currently through the packing with a large interfacial area. The aerosol droplet is moving upwards with the flue gas in the column while amine solution is flowing oppositely as sketched in Fig. 1. In the current work it is assumed that mass and heat transfer to and from the aerosol phase does not influence the main gas and liquid flows in the column because of the small aerosol volume. In further development of the model, possible depletion of components in the gas phase will be taken into account. Variation in gas phase component partial pressure and temperature through the absorber leads to transport of components and energy between gas and droplet and also within the aerosol droplet. The modelling used is analogue to the membrane contactor models given in Hoff et al. (2004, 2002) and Hoff and Svendsen (2014) apart from the geometry which in the droplet case is spherical, and the fact that the droplet changes size. A model on a more aggregate level is presented by Schaber et al. (2002).

2.1. Mass balance

The general balance equation in terms of flux for spherical particles (Bird et al., 2002) can be written as;

$$\left(\frac{\partial C_x}{\partial t} + v_r\frac{\partial C_x}{\partial r} + \frac{v_\theta}{r}\frac{\partial C_x}{\partial \theta} + \frac{v_\phi}{r\sin\theta}\frac{\partial C_x}{\partial \phi}\right)$$
$$= D_x\left[\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial C_x}{\partial r}\right) + \frac{1}{r^2\sin\theta}\frac{\partial}{\partial \theta}\left(\sin\theta\frac{\partial C_x}{\partial \theta}\right) + \frac{1}{r^2\sin^2\theta}\frac{\partial^2 C_x}{\partial \phi^2}\right] + r_x \quad (1)$$

Where C_x indicates concentration of any component, D_x denotes diffusivity, r_x characterizes reaction rate and r, θ and ϕ represent radial, polar and azimuthal angles. Density and diffusivity have been assumed constant.