



Gas phase amine depletion created by aerosol formation and growth



Hammad Majeed, Hanna Knuutila, Magne Hillestad, Hallvard F. Svendsen*

Norwegian University of Science Technology, Trondheim 7491, Norway

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ABSTRACT

Aerosols are systems of droplets or wet particles suspended in gases. In post combustion CO₂ absorption systems aerosols can be formed by spontaneous phase transitions in supersaturated gas phases or by droplets or particles entering the absorber with the gas to be treated. Micron and sub-micron mist droplets and fog formed in these processes cannot be removed by conventional demisting devices and because amine may be absorbed in the droplets this may increase dramatically the amine emissions from absorption columns as reported previously (Khakharia et al., 2015; Schaber et al., 2002). Thus, it is important to understand the mechanisms governing droplet growth and amine uptake through absorber as well as the effect large numbers of aerosol droplets can have on the bulk gas phase composition.

A model developed and implemented in Matlab, predicts how the gas phase composition and temperature change along the absorber taking into account mass and heat transfer to and from both the bulk liquid and the droplet phase. The objective of this work, compared to earlier work, Majeed et al. (2017), is to study the possible effect of gas phase component depletion on the droplet growth and droplet internal variable profiles and how this varies with initial droplet size and composition, droplet number concentration and amine volatility.

For MEA, as a relatively volatile solvent it is seen that gas phase depletion already takes place at number concentrations above 10⁵ droplets/cm³ with an initial droplet radius of 1.5 μ and 5 M MEA initial concentration. For initial droplet radius 0.15 μ and 0.0001 M MEA initial concentration, which may be a more realistic case, hardly any depletion effect is seen up to 10⁷ droplets/cm³. With change in amine volatility it is seen that the gas phase depletion effect is significantly stronger in the case of low volatility than for MEA at high droplet number concentrations. It is found that gas phase amine depletion has a strong effect on droplet growth.

1. Introduction

Very rapid growth in energy consumption was experienced in the 20th century and with an increasing world population and per capita use, it is expected that significantly more energy will be needed in the 21st century. Meeting these demands by the use of fossil fuels in power production and industrial processes is one main contribution to climate change by accumulation of greenhouse gases (GHGs) and in particular CO₂ in the atmosphere.

Post combustion CO₂ capture (PCCC) by chemical absorption is one of the most developed technologies for capturing CO₂ from low pressure emissions (Abu-Zahra, 2009; MacDowell et al., 2010; Rochelle, 2009; Sanchez-Fernandez et al., 2013). Solvent emission in the form of aerosols is one of the main challenges for widespread implementation of such plants (Schaber et al., 2002; Khakharia et al., 2015) as amine emission to air may impose a potential risk to human health and produce possible negative effects on the environment, as indicated by

studies reported in (Knudsen and Randall, 2009). In depth studies performed in the Gassnova CCM projects, see Fullskala Mongstad (2014), that possible consequences with full scale operation using MEA on environment will be very small.

Detailed experimental data on aerosol droplet growth and emissions do not yet exist. Analytical measurements techniques such as Fourier Transform Infrared Spectrometry (FTIR) and Phase Doppler interferometry (PDI) can be used to estimate overall emissions and particle size distributions (Fulk, 2016; Fulk and Rochelle, 2013). These methods give overall measurements but do not give insight into aerosol behavior inside the absorbers. Therefore, a detailed simulation tool is required that is able to predict aerosol formation and development as function of operational and chemical characteristics. Since the physical conditions in a gas–liquid contactor are very complex, it is necessary to use a somewhat simplified model to understand the behavior. A basic simulation tool for the description of a single aerosol droplet development in CO₂ absorption columns is already established and described in Majeed

* Corresponding author.

E-mail addresses: hammad.majeed@ntnu.no (H. Majeed), hanna.knuutila@ntnu.no (H. Knuutila), magne.hillestad@ntnu.no (M. Hillestad), hallvard.svendsen@ntnu.no (H.F. Svendsen).

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Notations		u_s	Superficial velocity ($m\ s^{-1}$)
C_x	Concentration of any component ($mol\ m^{-3}$)	v	Velocity ($m\ s^{-1}$)
C_p	Heat capacity ($kJ\ kg^{-1}\ K^{-1}$)	<i>Greek letters</i>	
D_x	Diffusivity of any component ($m^2\ s^{-1}$)	ρ_x	Density of any component ($kg\ m^{-3}$)
Keq	Equilibrium kinetic rate constant	μ	Viscosity ($kg\ m^{-1}\ s^{-1}$)
R	Gas constant ($m^3\ pa^{-1}\ mol^{-1}\ K^{-1}$)	<i>Indices</i>	
h	Heat transfer coefficient ($kW\ m^{-2}\ K^{-1}$)	MEA	Monoethanol amine
k_g	Over all mass transfer coefficient ($kmol\ m^{-2}\ s^{-1}\ kPa^{-1}$)	d	Droplet/aerosol
N_x	Flux of any component ($kmol\ m^{-2}\ s^{-1}$)	g	Gas
P	Partial pressure (kPa)	f	Film
R	Initial droplet radius (m)	l	Liquid
r_x	Rate of reaction ($kmol\ m^{-3}\ s^{-1}$)		
t	Time (s)		
T	Temperature (K)		

et al. (2017). This model gives examples as to how an entering droplet grows or shrinks through an absorber and how the internal composition changes with respect to position. The present work is an extension of this model in which the effect of amine depletion in the gas phase is taken into account. There are publications indicating that large particle number concentrations of aerosols will affect the bulk gas phase concentrations in absorber columns (Zhang et al., 2017). This change in bulk gas phase concentration levels may affect the overall efficiency of the absorption system and also the amine transfer to aerosol droplets, the droplet growth and thereby the amine emissions.

2. Modelling

The system of equations consists of differential balance equations for component mass and energy inside the aerosol droplets and similarly for the gas phase. These equations are coupled by a reaction rate model, an equilibrium model and models for heat and mass transfer between, on one hand the gas phase and liquid phase assuming that the liquid bulk remains unaffected by the aerosol phase, and secondly between the gas phase and aerosol phase. This is shown in Fig. 1.

2.1. Balance equations for aerosol phase

The combined transport and reaction equations for the aerosol phase are described in detail in Majeed et al. (2017) including boundary conditions and solution procedure and are not repeated here.

2.2. Balance equations for gas phase

If the number concentration c_N of aerosol droplets is large, the bulk gas phase composition can be altered. This possible change and its effect on droplet growth is the focus of the present work. There are experimental investigations that give number concentrations of aerosol

droplets in typical amine based PCCC plants (Khakharria et al., 2013; Mertens et al., 2014). The number concentration indicates how many droplets are present in a cm^3 of gas. The change in number of moles in the associated gas phase ($1\ cm^3$) will then be given by a dynamic balance between the inflow of a component from the bulk fluid minus the flow rate of the same component from gas to the droplet phase.

$$\frac{\partial n_x^g}{\partial t} = \dot{n}_x^f - \dot{n}_x^d \tag{1}$$

$$\dot{n}_x^f = a_f \cdot K_{g,x}^{overall} \cdot (P_x^* - P_x^g) \tag{2}$$

$$\dot{n}_x^d = N_x \cdot (c_N \cdot a_d) \tag{3}$$

Here n and \dot{n} are number of moles in the gas volume and the molar flows into and out of the gas volume. f and d indicate bulk fluid and droplet respectively while x denotes component. $K_{g,x}^{overall}$ is the overall mass transfer coefficient from bulk liquid to gas and P_x^* and P_x^g are respectively the liquid phase equilibrium pressure and gas phase pressure. a_f and a_d are interfacial area for bulk fluid and droplet respectively. N_x and c_N are molar flux and particle number concentration in droplets/ cm^3 of gas.

Similarly, the thermal balance equation for the gas volume is:

$$\rho_g \cdot V_g \cdot C_p \frac{\partial T_g}{\partial t} = h_{lg}^f (T_f - T_g) - h_g^d (T_g - T_d) \tag{4}$$

h_{lg}^f and h_g^d depict bulk liquid side and droplet side heat transfer coefficients. T_f , T_g and T_d are respectively the bulk fluid, gas and droplet surface temperatures.

2.3. Growth of droplet

The growth of the droplet is the rate of change in volume with respect to time (Majeed et al., 2017);

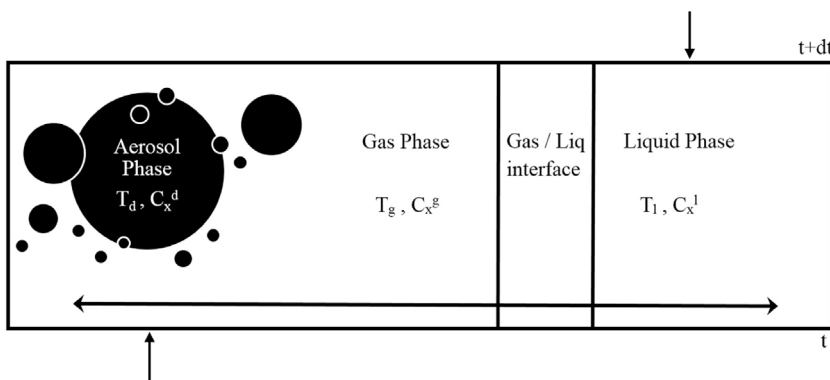


Fig. 1. Schematic illustration of heat and mass transfer between all the phases in a differential volume element in the absorber (T and C are temperature and composition while subscript x is for constituent, d , g , l are droplet, gas and liquid).

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