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Solid Carbon Dioxide Formation from Rapid Fluid Expansion using Integration of Computational Fluid Dynamics and Mathematical Modelling

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A model was developed to investigate carbon dioxide droplet size distribution and the size of solid particles formed during horizontal rapid fluid expansion. These two parameters are crucial for risk assessment when constructing carbon capture and storage (CCS) facilities. The model was an integration of two sub-models: a CFD model to obtain temperature and velocity profiles, and a mathematical model to calculate droplet and particle sizes. The model was validated using experimental data of CO_2 expansion, and was able to describe the formation of solid CO_2 particles with sufficient accuracy. The model also found that when rapid CO_2 expansion occurred under supercritical storage conditions, the solid particles formed were too small to develop a rainout pool.

1. Introduction

One of the important aspects in carbon sequestration lies with the transportation of CO_2 to a storage site after the capture process. Transporting CO_2 at a high-pressure, supercritical phase is considered to be the most economically feasible. Nevertheless, CO_2 transport pipelines can be susceptible to fractures and leakages, which will cause accidental releases of carbon dioxide to the atmosphere, a scenario hazardous to people and the environment.

Apart from dispersing toxic vapour cloud, this adiabatic expansion of supercritical CO_2 from leakage orifice will also produce solid CO_2 micro-particles, which at sizes larger than 100 microns can rain out to form a solid pool. This rainout pool will eventually sublimate to the atmosphere, and contribute significantly to the concentration of toxic vapour cloud. However, most of these studies focus only on vapour cloud dispersion; formations of solid micro-particles and the resultant rainout pool are often not taken into consideration in most available software (Vianello, et al., 2012). If the occurrence of rainout pool is neglected when modelling accidental releases, the safety distance calculated has the potential to be inaccurate. Therefore, the main objective of this paper is to construct and validate a consequence model that can describe a horizontal rapid CO_2 expansion through an orifice at supercritical storage conditions. The results of this study will be able to provide insight on the occurrence of solid CO_2 rainout pool, which is a vital piece of information required to perform a reliable risk assessment on CO_2 transport pipelines. An accurate safety distance of CCS facilities can be obtained to ensure the well-being of humans, animals and other living things.

1.1 Review of Previous Studies

A number of studies have taken to model CO_2 accidental releases in order to evaluate risks and establish a safety distance when building CCS facilities. In order to accurately calculate the concentration and dispersion of toxic carbon dioxide vapour cloud, rainout of CO_2 following an accidental release must be taken into consideration. Mazzoldi et al. (2009) presumed that generally no rainout will occur in the course of a horizontal CO_2 release. However, the droplet size distribution and size of solid particles formed are neither measured nor calculated in these publications hence, the occurrence of CO_2 rainout remains as postulation and devoid of scientific and quantitative evidence. Hulsbosch-Dam et al. (2012) described the formation of solid CO_2 micro-particles using the Joule-Thompson effect (Figure 1). The authors agreed that

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rainout is only likely in the case of larger solid CO_2 particles of 100 – 200 μm. The proposed mathematical model is proven to correspond well with high-pressure releases of several fluids such as propane, butane and water. It is the interest of this study to validate the model with the experimental data of CO_2 releases obtained by Liu et al. (2012) in measuring the size of dry ice particles produced from expansion nozzle with varying diameters.

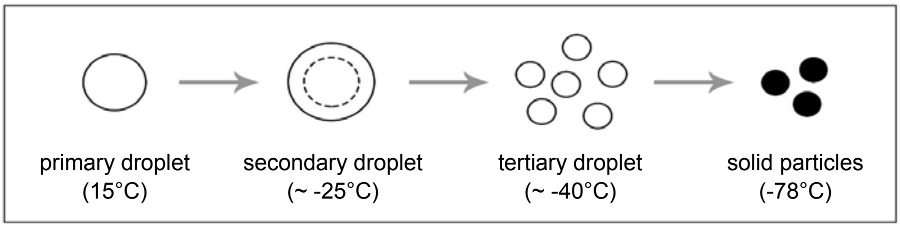

Figure 1: Solid particles formation from rapid expansion

With regard to the relationship between particle size distribution and orifice size, Liu et al. (2010) found that with increasing size of the orifice, the velocity of particles decreases, while the size of the particles increases. Liu et al. (2012) found that the process of droplet sublimation into smaller solid particles happens more quickly in smaller orifices than in bigger ones. Thus, it is proven that the nozzle diameter plays an important role in the velocity and size of solid CO_2 particles. This is in accordance with the observations of Koornneef et al. (2010).

2. Methodology

To ensure the feasibility of the study, the model is constructed by integrating two sub-models: (i) a 3-dimensional Computational Fluid Dynamics (CFD) model, and (ii) a mathematical model published by Hulsbosch-Dam et al. (2012). The CFD model functions to obtain temperature and velocity profiles of rapid fluid expansion, while the mathematical model calculates the droplet size distribution from the point of release and size of final solid particles formed. By splitting the model into two major parts, the computational time can be greatly reduced, ensuring prompt completion of the research study.

2.1 CFD modelling

A CFD model was developed using the software FLUENT 14.0. A pressure-based solver and ideal gas properties were used. The physical geometry and boundary conditions of the model were firstly constructed to emulate the horizontal releases from Liu et al.'s experiment. It was only tailored for present study and other parametric investigations after validation. The energy equations and realizable k-ε turbulence model were used for the simulation. Mazzoldi et al. (2011) proposed the use of such models to simulate high-pressure CO_2 discharge with solid particles formation. No volume of fluid method was needed for this calculation as the volume fraction of non-vapour (solid particles) would be very small. From the CFD model, temperature and velocity profiles of CO_2 releases could be obtained for the calculation of droplet and particle size in the next section.

2.2 Mathematical Modelling

The model was complemented by the results obtained from CFD modelling. It could be broken down into three major stages: (i) aerodynamic break-up, (ii) thermodynamic break-up, and (iii) evaporation, solidification and sublimation.

i) Aerodynamic break-up: this process is isothermal, and can happen within the first few microseconds of the jet release. The correlation between droplet velocity and initial jet velocity with time, t proposed by Pilch & Erdman (1987) is applied,

$$\frac{u_d}{u} = \frac{\sqrt{\rho_{vap}}}{\sqrt{\rho_{liq}}}\left(0.5*\frac{3}{4}*t+3*0.0758*t^2\right) \tag{1}$$

Where u_d is the droplet velocity when the break-up process is ended, u is the relative velocity between the droplet and the air, ρ_{vap} is the vapour density, ρ_{liq} is the liquid density. The time taken for the aerodynamic break-up was determined as the period where there was no temperature increment in the jet.

This information could be extracted from the CFD simulation of the temperature profile. The droplet diameter was assumed to be a log-normal distribution (Razzaghi, 1989) and proposed a correlation of average droplet diameter D_{av}, critical Weber number, We_{crit}, surface tension, σ, air density, ρ_{air} and velocity, u as in Eq(2). The average diameter obtained was then used in the second part of the mathematical modelling.

$$D_{av} = \sqrt{\frac{We_{crit}}{\left(1-\frac{u_d}{u}\right)^2}\frac{\sigma}{\rho_{air}u^2}} \tag{2}$$

ii) Thermodynamic break-up: this includes nucleation, bubble growth and blasting. The calculations could be divided into three main steps in a cycle. First, it was investigated whether the droplets formed during aerodynamic break-up were susceptible to boiling Eq(3). If the bulk temperature of the jet was higher than minimum boiling temperature of droplet ($T_{boil} > T_{min}$), the droplets would boil.

$$T_{min} = T_{boil}\left(1+\frac{1}{L_v M\rho_{vap}}\frac{4\sigma}{D}\right) \tag{3}$$

Where T_{boil} is the bulk boiling temperature, M is the molar weight, L_v is the heat of vaporization, D is the droplet diameter. At this stage, the nucleation and bubble growth ensue, where bubble forms in the droplets, increasing their sizes. The Hertz-Knudsen equation is applied to calculate maximum evaporation flux $\dot{Q}$,

$$\dot{Q} = P_{sat}^{*}\sqrt{\frac{M}{2\pi\Re T_{sat}}} \tag{4}$$

Where P_{sat}^{*} is the vapour pressure, T_{sat} is the equilibrium temperature with the outside pressure and R is the gas constant. The droplet outside radius growth rate was based on an equation by Shusser and Weihs (1999),

$$\frac{dR}{dt} = \frac{2}{3}\frac{\dot{Q}}{\rho_{liq}}\frac{R}{M}T_{sat} \tag{5}$$

Once the radial growth rate was obtained, the time taken for the primary droplet to grow until twice its original size was calculated. Distance of this secondary droplet from the point of release could then be calculated based on the velocity profile obtained from the CFD simulation. This information was recorded as part of the particle size distribution data.

Blasting or bursting occurs when the outside radius of the droplet reaches two to five times of its original size (Vandroux-Koening and Berhoud, 1997). The value 2 was used. When the primary droplet grows twice its size (value 2 is reached) into secondary droplet, it will blast into several pieces, forming equal-volume tertiary droplets. The number of resulting droplets can be anything between 1 and 10. A random number generator was used to generate 1,000 random numbers manually, and the median was used for calculation. This is a significant modification to the Monte-Carlo approach used by Hulsbosch-Dam et al. (2012), as it is not implemented directly to the model, but calculated as a separate entity.

The ability of these tertiary droplets to boil was tested again using Equation 3. If $T_{boil} < T_{min}$, the droplets would proceed to the third and final stage of evaporation, solidification and sublimation. Otherwise, the abovementioned steps were repeated.

iii) Evaporation, solidification and sublimation: the final diameter of solid CO_2 particles D_f after the effect of evaporation, solidification and sublimation could be calculated as follow,

$$D_f = \left[D_{in}^3\frac{\rho_l(T_{in})}{\rho_s(T_{boil})}\frac{\left[C_{p,L}(T_{in}-T_{tp})-L_v(T_{tp})-C_{p,v}(T_{boil}-T_{tp})\right]}{\left[(C_{p,s}-C_{p,v})(T_{boil}-T_{tp})-L_s(T_{tp})-L_v(T_{tp})\right]}\right]^{1/3} \tag{6}$$

where L_s is the latent heat of solidification, T_{tp} is the triple point temperature of CO_2 (-56.6 ^{0}C), T_{in} is initial temperature.

3. Result and discussion

3.1 Model Validation

The model was validated using the experimental data obtained by Liu et al. (2012). The release conditions, experimental results and simulation results were tabulated in Table 1. It shows that the model could accurately describe rapid CO_2 expansion in terms of solid particle formation. (Note: storage pressure and temperature are 55 bar and 288 K respectively).

Table 1: Validation of model against Liu et al. (2012)

Nozzle diameter (mm)	Inlet mass flow rate (g/s)	Exp. particle size (μm)	Sim. particle size (μm)	Error (%)
0.5	2.9	1.000	1.048	4.80
0.2	0.5	0.950	0.936	1.52
0.1	0.2	0.900	0.891	1.04

The experiment conducted by Liu et al. (2010) provided the temperature profile of CO_2 release from a 0.2 mm diameter nozzle. The temperature profile was matched with the one obtained using the present study model, as shown in Figure 2. Based on the model, during a 0.2 mm diameter nozzle release, solidification of CO_2 droplets occurred approximately 7.4 mm away from the point of release (temperature at (-)78 °C or 195 K). This finding corresponded well with the experimentally obtained temperature profile, whereby the freezing point of CO_2 was within the first 10 mm from the point of release. This was again indicative that the model was accurate.

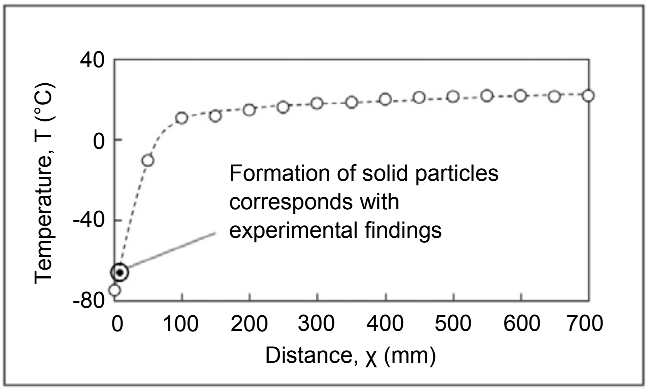

Figure 2: Validation of model against Liu et al. (2010).

3.2 Parametric Study

The model was used to investigate the effect of nozzle diameter on droplet size distribution and particle size formation. From Figure 3, it could be seen that the carbon dioxide droplets reduced in size log-normally as they travelled away from the point of release.

It was also observed that cooling, evaporation and sublimation occurred more quickly in smaller orifices than in bigger one. It required a longer time for the droplets to reach terminal small sizes (~1 micron) when carbon dioxide was expanded from a bigger orifice compared to a smaller one. Figure 4 showed that the final solid particles formed to increase in size as the orifice size increased.

The above mentioned trends were all in accordance with the research conducted by Liu et al. (2012). This warranted the relevance, reliability and applicability of the model.

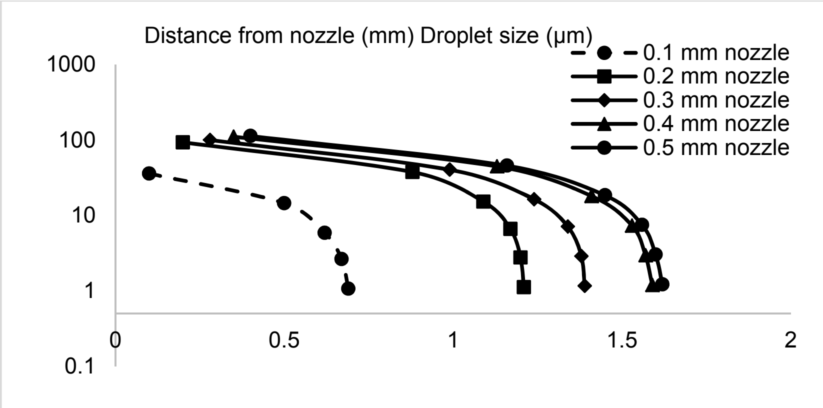

Figure 3: Effect of nozzle diameter on droplet size distribution.

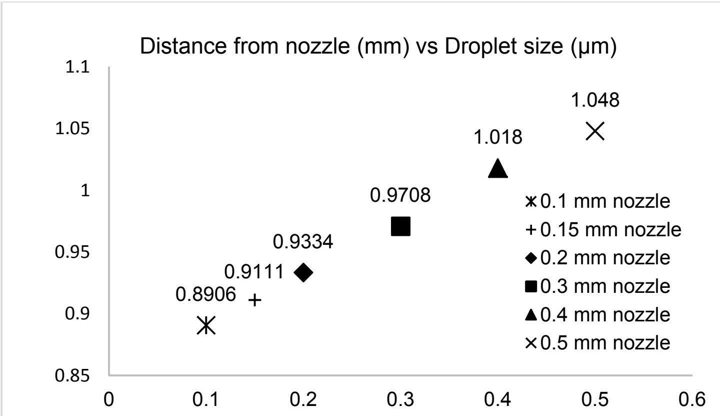

Figure 4: Effect of nozzle diameter in size of particles formed.

3.3 Modelling Supercritical Release

In carbon capture and storage (CCS) technology, carbon dioxide is usually transported at supercritical conditions for higher efficiency. Therefore, it is of interest to investigate the fluid expansion phenomenon in the case of a pipeline leakage at supercritical carbon dioxide transport pipeline.

A trial run was conducted at 310 K, 150 bar. Figure 5 shows that the range of solid particles formed were between 0.6 and 0.8 µm. They were smaller in size when the storage pressure was higher (150 bar) compared to the case when the storage pressure was lower (55 bar). This finding corresponded to the study by Witlox et al. (2009), which demonstrated that smaller solid particles were formed when the storage pressure was higher.

It was also of high interest to assess whether or not solid particles formed during supercritical releases would subsequently rainout. As shown in Figure 5, the increment in solid particle size could be seen as logarithmic when nozzle diameter increased. With a regression coefficient, R^2 of 0.9595, the relationship could be equated as follow,

$$y = 0.0638\ln(x) + 0.7497 \tag{7}$$

Using Eq. 7, rudimentarily, it was found that in order for the solid particles to reach the size of 100 µm, the diameter of the leakage (orifice) would have to be infinite. To further visualize, for a big leakage of 20 mm in diameter, the solid particles formed would only assume a diameter of 0.94 µm, not significantly big enough to cause a rainout pool formation. Therefore, for horizontal CO_2 releases with supercritical storage condition (specifically at 150 bar), there would be no rainout.

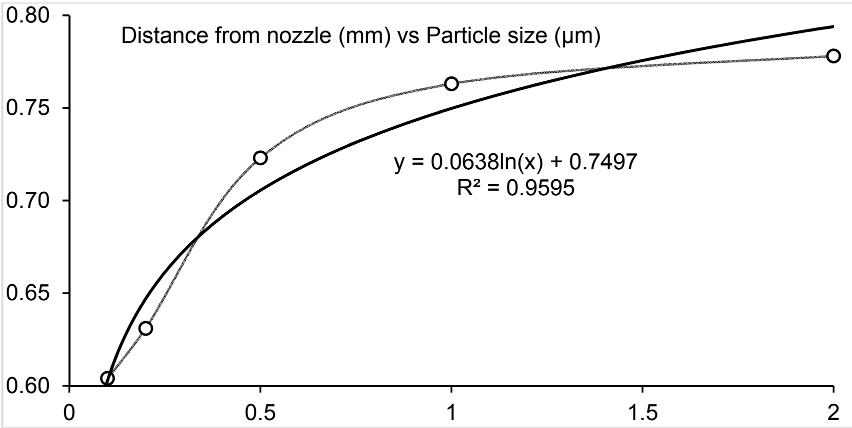

Figure 5: Size of particles formed for supercritical storage conditions.

4. Conclusion

A model was successfully constructed to investigate the horizontal rapid expansion of carbon dioxide through a nozzle. It was able to accurately predict the CO_2 droplet size distribution from point of release, as well as the size of solid particle formed. The model was also validated by several sources of literature in terms of results (direct calculation) and parametric trends. Finally, horizontal CO_2 expansion was simulated at supercritical storage conditions (specifically at 310 K and 150 bar) to observe droplet size distribution and size of solid particles formed. It was conclusive that in the course of a horizontal release, particles will only assume the size of less than 1 micron, which makes the formation of a solid rainout pool impossible. This model can also be used for other supercritical release conditions for a more accurate calculation of vapour dispersion. For future expansion work, it was recommended that the model be validated using experimental data for supercritical CO_2 releases to confirm its legitimacy.

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