



IEAGHG 8th Post Combustion Capture Conference

16th to 18th September 2025 Marseille, France

Supersonic Point Source Capture

David Chartrand, Aurélie Grebe, Gaku Fukushima, Martin Brouillette*

Université de Sherbrooke, 2500 boulevard de l'université, J1K 2R1, QC, Canada

Abstract

Reducing carbon emissions globally is crucial to mitigating the effects of human-driven climate change [1]. A significant portion of these reductions can be achieved by capturing emissions directly at their source, especially in industries where effective mitigation strategies have yet to be identified, such as cement, steel, and fertilizer production. These major industrial emitters are actively seeking solutions for capturing their CO₂ emissions, but viable options remain limited. Existing carbon capture technologies are primarily adapted from industrial anti-pollution systems, with some utilizing solid sorbent particulates [2]; liquid sorbent systems have also gained popularity due to their ability to handle large volumes of gas [3]. However, both approaches face challenges when scaling up, as solid particulate systems are prone to fouling, complicating thermal management, while liquid sorbent systems require energy-intensive high-temperature regeneration. Cryogenic CO₂ separation is also employed but is most effective only when the CO₂ concentration in the effluent exceeds 60% [4]. A promising alternative involves expanding CO₂-laden gases in a supersonic flow, which lowers the temperature enough to induce CO₂ condensation, allowing for its physical separation from the gas stream. This method eliminates the need for specific absorbing or adsorbing materials, thereby avoiding operational issues associated with material degradation. This concept, already used in natural gas drying [5], has also been proposed for industrial carbon capture [6]. However, previous studies faced challenges due to inadequate particle sizes, hindering effective CO₂ condensation, separation, and collection [7].

The system proposed in Fig. 1 [8] aims to address these limitations. In the first step of the process, gaseous effluents are compressed and cooled to ambient temperatures. In step 2, inert solid particles of desired size are injected into the compressed flow. The gas then passes through a converging-diverging nozzle in step 3, where the flow is accelerated to supersonic velocities and the temperature is lowered, triggering CO₂ condensation onto the inert particles. The appropriate flow conditions are maintained for a sufficient length of the nozzle to ensure that a large fraction of the CO₂ condenses. At step 4, a lateral acceleration field is generated in the flow, directing the particles to one side of the stream. This is achieved using an abrupt convex corner, creating a centred expansion wave that further cools the flow, preventing the premature evaporation of CO₂. Finally, in step 5, the particle-laden flow is separated from the main stream, with both flows passing through diffusers to reduce flow velocities and recover flow pressure. As the flow is slowed down, the CO₂ on the particles re-evaporates, forming a CO₂-rich gas stream, while the principal flow is significantly depleted of CO₂.

* Corresponding author. Martin Brouillette Tel.: 819-821-7144, E-mail address: martin.brouillette@usherbrooke.ca

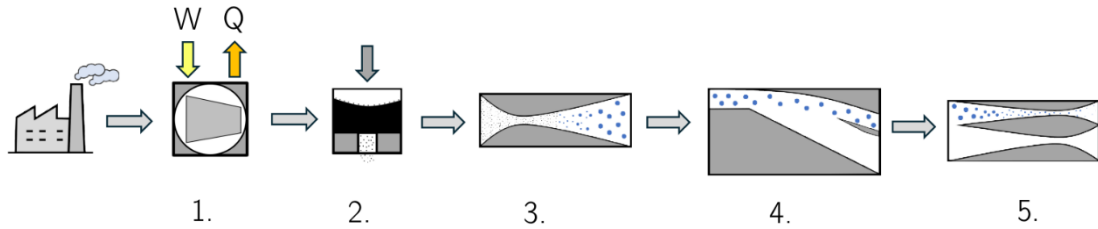


Figure 1: System overview

To assist with the design of an industrial-scale system, the relevant physical phenomena within the condensation and separation nozzles are simulated using Quasi 1-D and 2-D numerical models, validated using relevant experimental data. The Quasi 1-D model simulates the two-way coupled gas-particle flow including the condensation of gaseous CO_2 onto solid particles within a supersonic flow-stream. This model accounts for wall friction, drag and heat transfer between the gas and the particles. The governing differential flow equations are adapted from Igra and Ben-Dor [9]. In addition, the growth model provided by Kulmala [10] is used to predict CO_2 condensation. The numerical solution is explicitly calculated by integrating steady state equations along the length of the nozzle using a backward finite difference scheme with boundary conditions determined at the subsonic inlet.

The 2-D model is developed to better comprehend the flow fields in the nozzle of condensation and separation. The governing equations for this model are 2-D two-way coupled gas-particle flow described in Igra and Ben-Dor [9]. For gas phase, the compressible viscous Navier-Stokes equations are used, and particle flow is modelled considering conservation of mass, momentum, and energy. These individual equation systems are coupled by drag force and heat transfer using source terms. Therefore, the interaction effects between gas and particles are fully coupled. To close the Navier-Stokes equations taking into account turbulence effects, the Baldwin-Lomax model, which is the fundamental zero-equation turbulence model [11], is employed. The equations are discretised by a finite volume method. The accuracy is the second-order in both space and time by using the monotonic upstream-centred scheme for conservation laws (MUSCL) interpolation and the two-step Runge-Kutta method. For condensation flow, the effect of heterogeneous condensation of CO_2 on particles is modelled using the same method as the Quasi 1-D model. The mass, momentum transport, and emission of latent heat are added to the corresponding source term.

The initial results from laboratory-scale experiments on this system, along with comparisons with the models, were presented in a previous paper [12]. The test bench used in these experiments is a blow-down wind-tunnel, where a pressure vessel containing a gas mixture is connected with valves to a particle injector placed before a supersonic nozzle and the flow empties into a vacuum vessel CO_2 -containing. This configuration allows a steady flow to develop in less than 1 second and is maintained for 5 to 60 seconds depending of the test conditions. For the condensation experiments, the supersonic nozzle presented at Fig. 2a is a 3-D printed axisymmetric nozzle with a 1 m diverging section. For the separation experiments, the supersonic nozzle presented at Fig. 2b is a 2-D nozzle made of aluminum walls and acrylic windows. Pressure sensors are placed on top for both nozzles at regular interval to capture the static pressure distribution of the flow during test.

The flow models and experimental results can be compared based on the fraction of condensed CO_2 . One method used to calculate CO_2 concentration within the experimental nozzle is the Pressure Trace Measurement (PTM) method [13]. This method works by integrating the measured static pressure trace taken from various axial positions along the nozzle to infer the heat released during CO_2 condensation, and then comparing it to an experiment without CO_2 . Initially, the pressure traces from both experiments are identical, but as the pressure increases at the nucleation point, condensation begins. An example of PTM result is shown in Fig. 3.

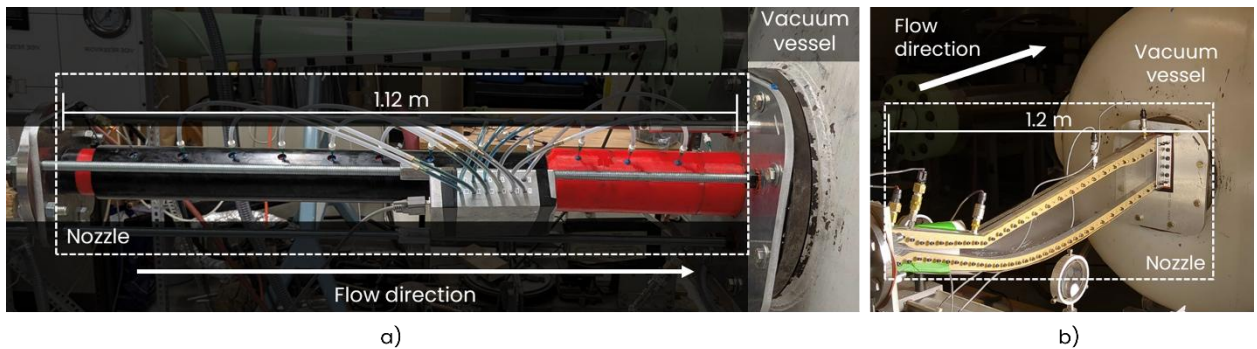


Figure 2: a) Condensation nozzle / b) Separation nozzle

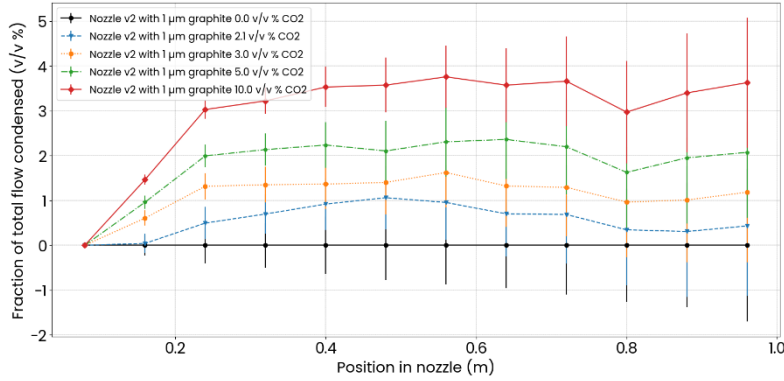


Figure 3: CO_2 condensation measurement from PTM (Condensation nozzle at total pressure of 7 bar for 0 to 10% v/v CO_2 with 10% m/m 1 μm graphite particles)

In this graph, the black line is the base line test with no CO_2 that the others tests used to compare the difference in static pressure. For the others lines, we observe a rapid increase in condensation near the beginning of the nozzle until 0.3 m.

Results from the Quasi 1-D and 2-D models in Fig. 4 show a similar amount of condensation at the beginning, compared to PTM. However, as the flow progresses through the nozzle, the models predict a higher amount of CO_2 condensation than PTM.

To improve the accuracy of CO_2 condensation data in the nozzle and provide a second measurement method, a Tunable Diode Laser Absorption Spectroscopy (TDLAS) system has been integrated into the condensation nozzle. This system uses wavelength modulation to measure the local concentration of CO_2 while correcting for variation in the optical depth of the system due to particles in the flow or fouling of the windows [14]. Through multiple tests at different positions all of the nozzle is mapped, and the results are used to compare with the previous PTM data. Additionally, the TDLAS results enable a more rigorous validation of the numerical simulations.

In addition, an improved nozzle combining both condensation and separation sequentially has been developed to better simulate an industrial-scale system to experiment with. This nozzle allows for the observation of the transition of particles from a 3-D axisymmetric condensation nozzle into a 2-D separator. In the separator, a second channel is added near the distal end to isolate the clustered particles with condensed CO_2 from the rest of the main flow. At the exit of both channels, a pitot tube is positioned to collect a small amount of gas during the test, which is transferred into a 500 ml syringe connected outside of the nozzle. After the experiment, the gas in the syringe is allowed to return to atmospheric pressure, and the composition of both syringes are measured using a gas chromatograph (Agilent 3000A Micro GC). The chromatograph analysis of the particles channel reveals the original amount of CO_2 injected into the system due to the sublimation of CO_2 from the particles as they enter the pitot tube. For the main flow channel, the results help confirm the condensation efficiency further down the nozzle and enable comparison with the TDLAS data. Imaging of particle separation in the second channel also validates the 2-D coupled model at the interaction of the flow with the start of the wall.

The focus of the full paper will first be to validate the condensation models (Quasi 1-D & 2-D) using new TDLAS measurement results and gas chromatography data in supersonic flow with CO_2 and particle injections. Secondly, it will present experimental results on particle separation in an integrated condensation-separation nozzle with an isolation channel, aimed at validating the 2-D coupled model.

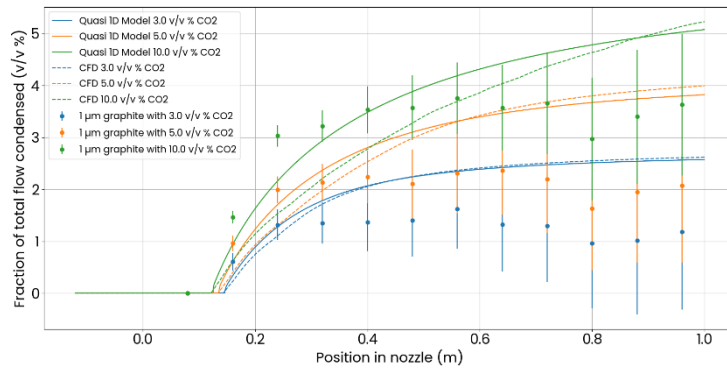


Figure 4: CO_2 condensation measurement from PTM compared with results from predictive models (condensation nozzle at total pressure of 7 bar for 0 to 10% v/v CO_2 with 10% m/m $1\mu\text{m}$ graphite particles)

Keywords: Supersonic flows; industrial effluents; CO_2 capture; TDLAS; Condensation.

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