Supersonic Expansion of Liquid and Gaseous Carbon Dioxide

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Abstract

Differ is involved in fundamental research on energy transition from fossil fuels. The climatic changes due to greenhouse gases from fossil fuels have increased the demand for sustainable energy sources. In the solar fuel division, work related to carbon dioxide neutrality is investigated. The Plasma Solar Fuel Devices group investigates and develops reactors which convert CO_2 to solar fuel to complete the neutrality cycle. Thus various techniques and methods are researched involved in capturing carbon dioxide from the atmosphere and reusing it, therefore forming a carbon dioxide neutral cycle. As research on carbon dioxide is being developed, it's implementations in various diverse fields is being explored. The industrial application of using liquid carbon dioxide jet for cleaning is a well-known and applied process because of the unique property of carbon dioxide which exists in solid-gas phase. This jet can go to low temperature of about -73^0C hence it can be used for cooling purposes in devices. This property of the jet is investigated in this study.

The flow of liquid and gaseous carbon dioxide via small diameter nozzle is studied in atmospheric and vacuum operating conditions. Experiments are conducted to characterize the flow from the nozzle by obtaining parameters like pressure, temperature and mass flow rate. The effect of dry ice formation and supersonic expansion stability in each operating condition is studied. Along with the experiment, simulations are also done using the ANSYS fluent software, these simulations are used to verify the assumption and previously performed similar experiments. The simulation results are compared with the nozzle results obtained from this study as well. Mathematical modeling is done to calculate the cooling power of the system.

Contents

Introduction

The Dutch institute for Fundamental Energy Research (DIFFER) involves fundamental scientific research into new and improved energy technology. The research is carried out in two major focus groups: Fusion energy and Solar Fuels. Fusion energy division researches: The transition of energy from fossil fuels to sustainable energy which requires large quantities of clean and reliable energy from compact power stations.

Solar fuel division investigates methods to effectively obtain, store and transport solar fuels. The Plasma Solar Fuel Devices group (PSFD) majorly researches in techniques to split carbon dioxide to carbon monoxide and oxygen as an initiating point for carbon dioxide neutral fuels. This split is done in devices with very high energies like plasma reactors. From various studies the microwave plasma reactor has shown to be a suitable method for efficient CO_2 dissociation. However the maximum efficiency which can be achieve is 52% as the process is limited from thermal conversion which means while cooling the dissociated CO and O_2 from high temperature of around $\sim 3000^0$C, burning occurs which converts the CO and O_2 to CO_2. The enhancement of the conversion efficient can be obtained by cooling the CO_2 before it enters the plasma[1].

On this line of research since CO_2 is utilized in various diverse applications. Amongst them, CO_2 jet is most commonly used in an industrial technique for surface cleaning because this jet contains a specific characteristic of low temperature solid-gas two phase flow. This application is widely researched and applied in various industrial sectors like electronics, automotive, food and pharmaceutical. In this study the behavior of the CO_2 jet is explored for its cooling abilities. These jets can be implemented in plasma microwave reactors to improve the efficiency of splitting of Carbon dioxide molecules. From literature[1] these jets can achieve temperatures of -78^0C and -40^0C in case of liquid and gas expansion. The feature of the two phase gas-solid flow is investigated to understand how icing effects the flow dynamics of the jet. The primary aim of this study is to understand the cooling properties of the CO_2 jet and the influencing parameters to implement it in a plasma microwave reactor.

The objective of this study is to characterize the gaseous and liquid Carbon dioxide expansion under controlled conditions outside of the plasma reactor. The flow rates of the nozzles are chosen to be compatible for the plasma process. Hence a test rig is built where several diagnostics like pressure, temperature sensors, mass flow meters are used to characterize the performance for different nozzle designs in vacuum and atmospheric conditions. Performing the experiments in vacuum and atmosphere can give a good understanding of the role of air and carbon dioxide interactions in the expansion regime and icing effects in vacuum. Computational Fluid Dynamics (CFD) simulations are carried out to give more insight into the parameters by eliminating the physical constraints of the experiment. These results are used to assess the quality of the nozzles and compare with the literature and simulation.

Review of Previous Studies

Many studies are present to understand the accidental release of CO_2 and evaluate the risk in carbon dioxide storage (CCS) facilities in cases of leak. These studies can shed light on the nozzle designs, key parameters and their measurement techniques. As mentioned earlier the industrial applications of using CO_2 jets for cleaning is very efficient because CO_2 can co-exist as solid gas two phase. The study[4] compared the orifice nozzle and CD nozzle to understand how the variation in design will affect the solid production. The CD nozzle enhances the solid ice production which increasing the efficiency of the cleaning process. In this study nozzle with relatively less solid ice formation is required hence the orifice nozzle is considered.

The liquid and gaseous flow of carbon dioxide from high pressure to atmospheric pressure through an orifice nozzle gives a supersonic expansion. Hence this nature of these expansion are been studied to ensure safety in CO_2 storage units[3].
The physical process of the supersonic nature of the jet leads to unavoidable system of shocks at the exit of the nozzle. This region is called as the Near field region.
Near Field Region: The sudden change in pressure from a high pressure region to low pressure region leads to formation of shocks. It is this pressure drop which constrains the expansion to a supersonic expansion. These shock waves cause drastic changes in pressure, temperature and density. Taking measurements in this region is challenging because inserting probes in this area will affect the shock waves and also cause additional reflected shock waves, which would lead to change in properties as every type of shock wave have different effects. For instance if the flow passes through oblique shock wave: pressure and temperature increase where as in under expanded shock wave the pressure and temperature will decrease. Therefore optical techniques is be used to study the behaviour of the expansion. Amongst the optical techniques, shadowgraphy is used in studies[2] to observe the nearfield flow properties. The visualization for gaseous CO_2 expansion is successful. For liquid CO_2 expansion on the other hand the shock structure is unclear because high concentration of liquid and solid CO_2 and the presence of water droplets from the humid air obstruct the view.

The parameter measurement:
The mass flow rate is measured using weighing scales. The weight of the CO_2 bottle is measured before and after the release of carbon dioxide with the scale and stop watch can be used to note the time. Hence the mass flow rate can be computed. But this method requires weighing scales with precision of 0.1gms. The orifice acts as throat hence the assumption of chocked flow is considered and the expected mass flow rate using isentropic equations is computed. The ratio in the values of measured mass flow rate and calculated mass flow rate gives the discharge ratio of the orifice. The key parameters that influence the mass flow rate is the diameter of the orifice, temperature and pressure. The temperature and pressure is controlled by a heating jacket around the liquid CO_2 liquid. Since liquid carbon dioxide is in a state of equilibrium at its saturated condition in the bottle. Increase in the bottle wall temperature leads to increase in pressure and vice versa. The orifice diameter is controlled by varying the compression

fitting at the nozzle exit. The temperature is be measured using the K-type thermocouple[3]. The formation of dry ice is also a major aspect that has to be kept in mind. The formations of these particles are majorly influenced on the temperature and velocity of the jet. However, this study shows by implementing a tube around the nozzle the solid formation can be enhanced. The measurement of the particle size and formation rate requires more complex techniques like laser diffraction. The diameter of the tube is critical to study expansion in vacuum which causes increase in agglomeration particle size and decrease in particle velocity [6].

Simulation:

The expansion and diameter of the solid formation can also be modelled with the combination of 3D CFD modelling and mathematical modelling .In the CFD modelling- pressure based solver and carbon dioxide with ideal gas properties is used. The energy equation, realizable K-e are activated to count for energy and turbulence. The solid formation can be assumed as negligible considering the volume of the domain hence no phase models are required. The temperature and velocity profiles are recorded from the simulation. The parameters are substituted in the Mathematical model which has three stages: i. Aerodynamic break up, ii. Thermodynamic break up iii. Evaporation solidification and sublimation. The obtained particle diameter is compared to diameter of solid from experiment [5] and is in good agreement[3]. The methodology of the simulation can be implemented to obtained the expansion of this study.

Methodology

This chapter explains the procedure which is adopted to characterise the expansion from the nozzle. The explanation of the all the measurements, calculations and simulation are present in this section.

The investigations on the expansion of liquid carbon dioxide in this report are divided into three subdivisions:

Experiment

Experimental study of nozzle performance is done. Here the parameters like mass flow rate, temperature and pressure are measured. Schlieren imaging measurements are used to visualize the flow at the outlet of the nozzle. Due to the restrictions that shielded expansion experiments pose on for instance optical access, two configurations are studied

1. Free expansion of carbon dioxide in open air at 1 atmosphere
2. Expansion of carbon dioxide in an vacuum conditions

CFD Modelling of Flow

The commercial ANSYS Fluent solver is used to model the expansion in both liquid and gaseous. The model is validated using the experiment. This simulation can give an insight into the parameter in the near-field region which can't be measured using probes.
The optimum nozzle required for a cooling system can be simulated in the software and then given for manufacturing thereby reducing the time in trial runs as well as cost.

Calculation of Cooling power

The cooling power of the expansion is assessed using a thermodynamic and heat transfer equations. Some additional isentropic calculations are done to compare the results obtained from the simulation and experiments.

Experiment

Apparatus & Safety Precautions

The experiment consists of a liquid carbon dioxide bottle, nozzle, quartz tube to create vacuum. The basic

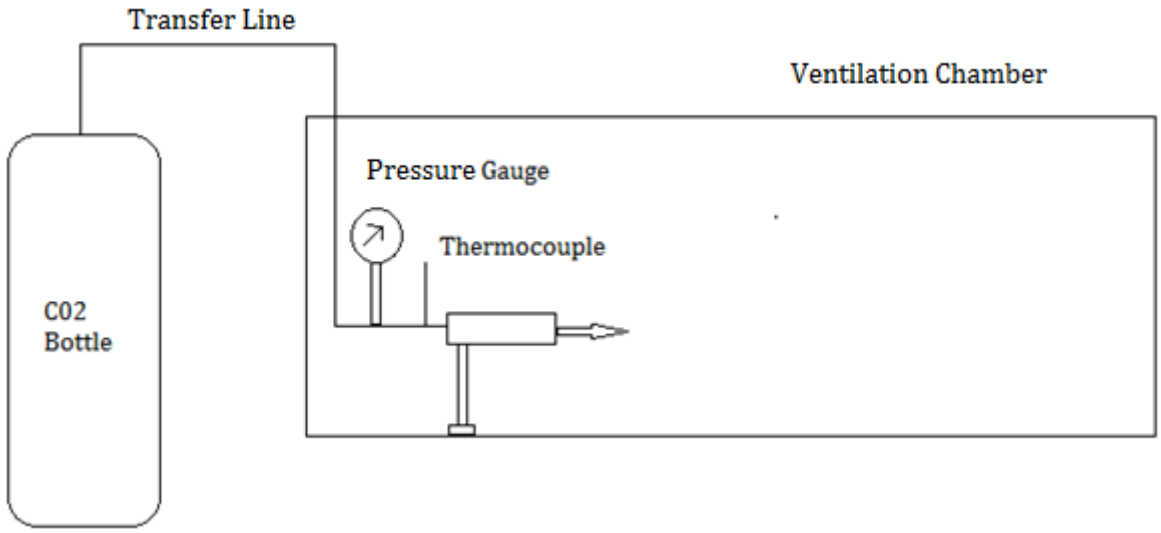

FIGURE 1: CO_2 EXPANSION SETUP – BASIC CONFIGURATION

Several configurations of the experimental setup, depicted in Figure 1 **,** are used to study the various aspect of the CO_2 expansion.

The free liquid expansion: In this setup, the carbon dioxide bottle is directly connected to the nozzle. A pressure gauge and thermocouple are installed before the nozzle. The setup consists of a removable tube which is used for mass flow measurements. A rail is also fixed after the nozzle exit which contains a moveable thermocouple for temperature measurements.

The liquid expansion in vacuum: This setup is similar to the free liquid expansion case except some additional installations are incorporated. A brass nozzle holder[Appendix 1.1] with inserts to attach the quartz tube is installed. The other end of the tube is connected to a vacuum pump which creates the vacuum. The function of the brass holder is to behave like an attachment which holds the nozzle and quartz tube from one end thus making the system air tight.

The free gas expansion: This setup is similar to the free liquid expansion case but a buffer volume is installed between the liquid CO_2 source and the nozzle to act as an expansion volume. Liquid CO_2 is expanded to the gas phase before it reaches the nozzle.

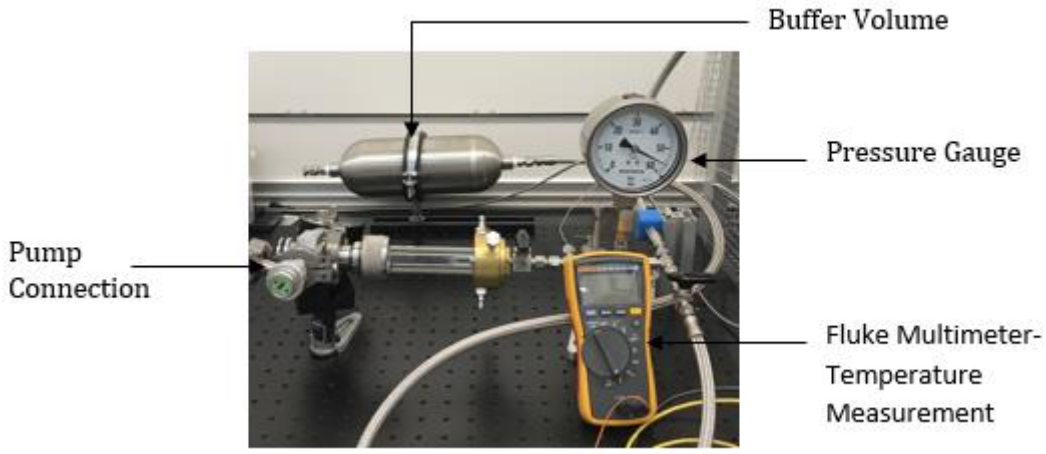

FIGURE 2:VACUUM SETUP

Some of the safety considerations taken because of working fluid and high pressures are: Care is taken that the ventilation chamber is on, googles and ear muffs have to be worn to protect the eyes and ears.

Nozzle Design

These nozzles are designed at the Engineering Design faculty and manufactured at the Mechanical Workshop in Differ. In the workshop the base structure of the nozzle is constructed.

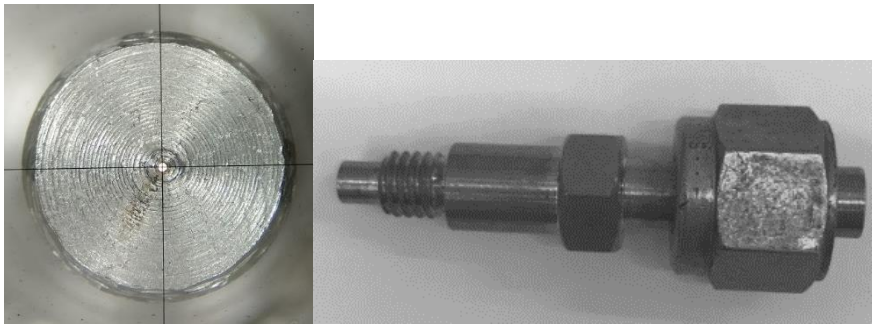

FIGURE 3:NOZZLE

The orifice diameters which will be used in this study are 0.1mm,0.2mm and 0.3mm. Stainless steel is used for the 0.1 mm and 0.2mm diameter nozzle because the orifice diameters are small, laser drilling is adopted. Laser drilling technique is used to drill lesser than 0.3mm diameter holes with is obtain uniformity throughout the full depth. The nozzle with the diameter of 0.3mm is made from brass and a conventional drilling machine is used to make the orifice. The flow is driven because of the pressure difference; the amount of flow through the nozzle can be controlled by the diameter of the nozzle or by regulating the pressure. These diameter are chosen from making an estimate with mass flow rate present in papers[1][6].

Depending on the mass flow rates of the current nozzles, an estimate of the suitable diameter can be made for the nozzle in the plasma reactor. The parameters are measured and depending on the temperature attained more complex design of the nozzle for the plasma reactor can be considered based on the requirement.

Pressure

The pressure is recorded via a pressure gauge before CO_2 enters the nozzle. This measurement can give us an estimate in the pressure in the bottle as well as the input for the simulation.

This is the only parameter which can be controlled during the gas expansion. The pressure in the buffer volume is regulated using the valve from the CO_2 bottle. Thus converting the carbon dioxide from liquid into gaseous phase.

Temperature

A K-type thermocouple is used to measure the temperature. Like pressure the temperature is recorded before the nozzle. Another thermocouple is used to measure the temperature in/after the expansion; here the temperature is noted from 0.5 to 10cm/ 20cmfrom the nozzle for gaseous/liquid. expansion The second measurement is done only in case of free expansion.

Calibration of Temperature:

This calibration is done to acquire accurate temperature measurements. As it is known that the k-thermocouple can be used for a wide range of temperature measurements (-200^{0}C to 1260^{0}C). In this calibration two kinds of probes with variation in design and two multimeters are used. The first type of probe has a stainless steel braided cable whilst the second probe's wire junction(tip) is exposed. The difference in the two multimeters is the RS 1315 Thermometer is built to exclusively measure temperature while the second multimeter is a fluke general purpose multimeter.

Three temperatures are measured with the thermocouple and multimeter setup:

1. Liquid nitrogen: At atmospheric pressure the expected value is -196^{0}C.
2. Ice water bath: At atmospheric pressure the expected value is0^{0}C.
3. Boiling Water: At atmospheric pressure the expected value is 100^{0}C.

These temperatures are chosen because they cover the full range of the k type thermocouple's measurement ability. The calibration tests the performance of the multimeter and thermocouple system.

Calibration:

The following combinations of the system used are:

Type 1: Stainless steel braided cable thermocouple with RS1315 Thermometer.

Type 2: Tip exposed thermocouple with RS1315 Thermometer.

Type 3: Tip exposed thermocouple with Fluke Multimeter.

Material	Type 1	Type 2	Type 3
Ice Water Bath	0.4	0.1	0
Boiling Water	99	99.3	98.9
Liquid Nitrogen	-188.9	-196.8	NA

TABLE1: TEMPERATURE CALIBRATION

Looking at the results obtained in the above table Type 2 system is used to measure the temperature downstream from the expansion jet. And the stainless steel braided probe with fluke multimeter is used to measure the temperature of CO_2 before entering the nozzle.

While performing the temperature measurement downstream of the nozzle the reproducibility error is has an uncertainty of +/-2^0C which is greater than the error obtained from the temperature system. The minimum temperature in the expansion is -78^0C which is half of the calibration minimum temperature and maximum temperature is room temperature. So no correction for the read out results is implemented [Appendix 3.1].

Flow Rate

The flowrate of the nozzles is measured downstream in the expanded region. Two flow meters are used depending on the flow rate: a rotameter for high flow and gilibrator which is accurate at low flow rates. Since the flow at the orifice is assumed to be chocked, the downstream pressure will not affect or cause change to the mass flow rate. Hence the effect of the downstream pressure on the mass flow rate is assumed to be negligible.

Rotameter is a device which measures the volumetric flow rate of the fluid in a closed tube. It consists of a vertical conical measuring tube and float. Due to gravity the float is always present at the bottom of the measuring tube thus making it as the reference position.An orifice ring is an imaginary ring at the position float in the measuring tube. The diameter of this orifice ring keeps increase as the float moves upwards thus making it a variable diameter ring orifice. The three forces acting in this system are:

- Gravity (W) or the weight of the float which acts in the downward direction. This force resets the float back to its initial position. The force is constant as the weight of float doesn't change.
- Buoyancy this force acts in the upward direction. Thus lifting the float. The buoyancy force acting on the float is also a constant force.
- Drag this force also act is the upward direction also lifting the float. This is the only force which is variable as it depends on fluid velocity. The velocity also depend on the temperature and density of the working fluid.

At the reference position the drag, buoyancy and gravity are in equilibrium. When the setup is connect to system whose flow has to be measure the float rises as the velocity increases as the float travels upwards the ring orifice diameter increases thereby reducing the velocity and balancing the forces. The reading corresponding this point is noted.

The measurement for the free expansion case is performed by using a tube with a sufficiently large diameter of 10cm and length 20 inches with rubber plugs on its ends, no dry ice formation is observed. This case is similar to what happens in free expansion and can be seen that the air present the tube is sufficient to provide the same conditions as in the absence of the tube.

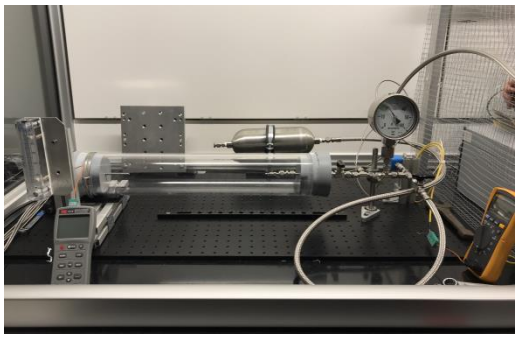

FIGURE 4:MASS FLOW RATE MEASUREMENT SETUP

In case of dry ice formation, the gas flow is measured and subtracted from the flow rate in the free liquid expansion case. The difference amounts to the mass flow rate of solid formation and can be used to calculate the solid formation fraction.

Calibration of Flow Rate:

The rotameter flow meter is calibrated using the Gilibrator[Appendix 2.1] as the calibration device. Air as the working fluid is used for the calibration.

Flow rates for CO_2 are obtained by correcting for specific density using the following formula:

$$q^0{}_G = q^0{}_A \sqrt{\frac{\rho^0{}_A}{\rho^0{}_G}} \qquad 1$$

The above formula[1]$q^0{}_G$and $q^0{}_A$represents the flow rate of gas G and A respectively. The equation[1]corrects for density ratio between the calibration gas A and working gas G. The temperature correction also can be included by taking the density of the respective gases at measured temperature from ideal gas equation.

The flowrate of air is measured using the Gilibrator and rotameter simultaneously. The obtained results are plotted and linear fit is adopted to correct for the error in the flowmeter result.

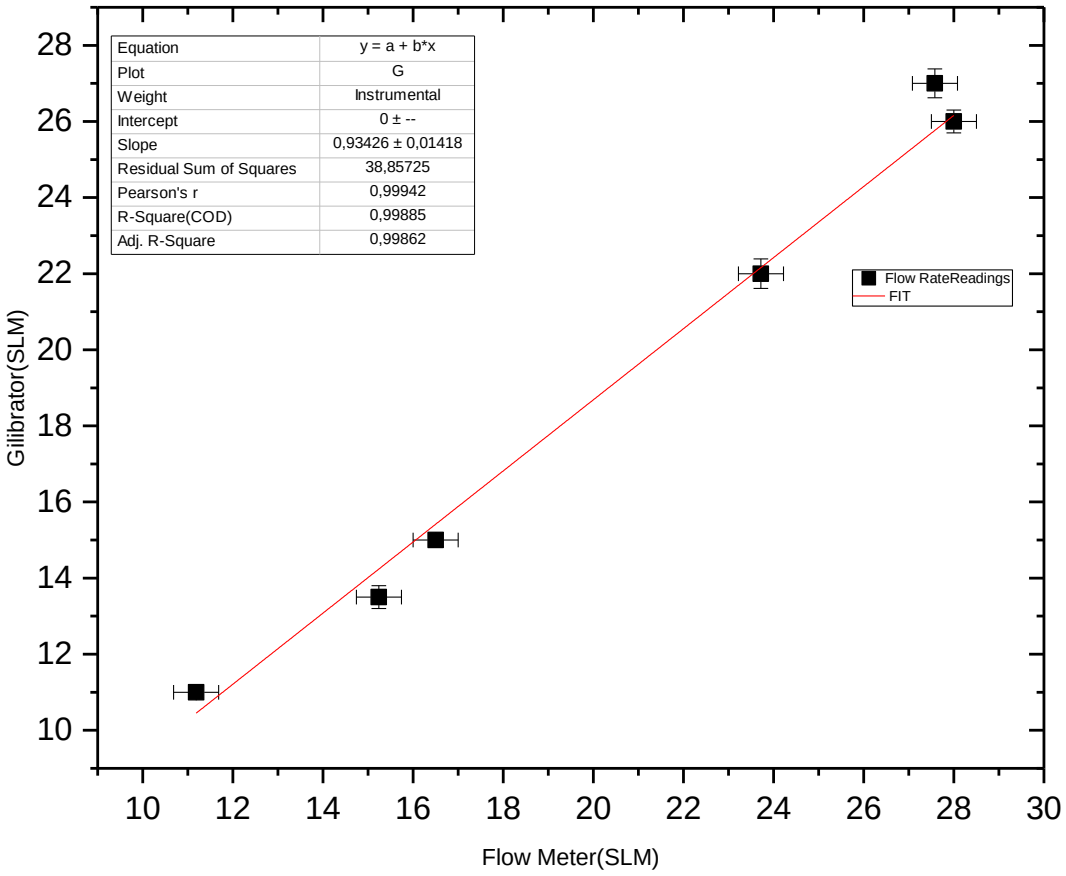

FIGURE 5: FLOW RATE CALIBRATION

For flow rates below 15 slm the rotameter readout is not accurate so the Gilibrator is used directly for the measurements.

Expansion Visualization

A Schlieren imaging technique[Reference] is constructed to visualize the expansion and the shock wave pattern at the exit of the nozzle. With Schlieren imaging the density variation in a transparent medium can be visualized.

Working Principle:

The Schlieren setup consist of a point source, knife edge, spherical mirror and camera. Schlieren imaging uses the phenomena of refraction of light to characterize the flow. The light passes through the expansion region and the rays undergo refraction hence, there is a shift in the angle of incidence. The shift can be positive or negative depending on the density at the region. As seen in the figure, the rays which don't have any change in angle go directly to the camera. Depending on the knife position, the rays bending in the direction of the knife edge are cut off and the remainder of the rays reach the iris of the camera. This causes a contrast thereby providing a crisp image of the flow behaviour.

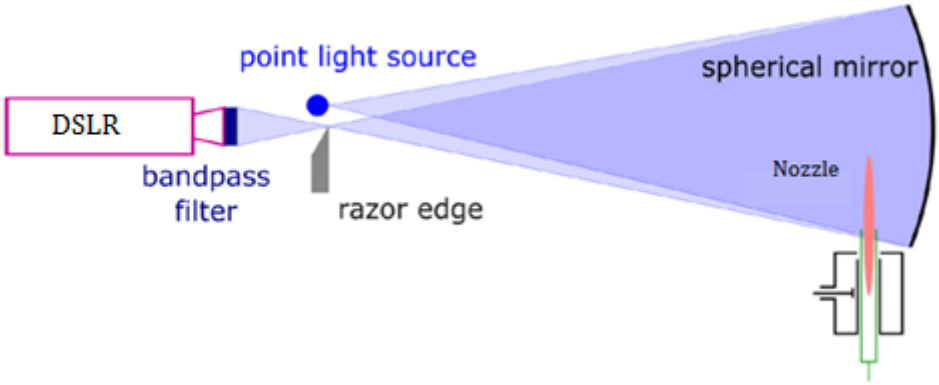

FIGURE 6: SCHEMATIC SCHLIEREN SETUP

Schlieren Setup Specifications and Construction:

Point source: Is created by coupling a 3 Watt LED into an optic fibre. A convex lens is used to converge the light into the optic fibre with a diameter of 400µm.

Spherical mirror: The mirror used here has a focal length of 1.3m.

Tele lens: A lens with high magnification of 1800mm is used to zoom into the region of interest is approximately ~> 5 mm just after the nozzle exit.

The mirror is placed behind the object whose flow is to be captured. The point source is placed at 2 times the focal length of the mirror (f=1.3m). The knife edge is placed next to the point source; behind it the camera is located. A tele lens with high magnification (1800mm) is used to zoom into the region of interest is approximately ~ 5 cm just after the nozzle exit.

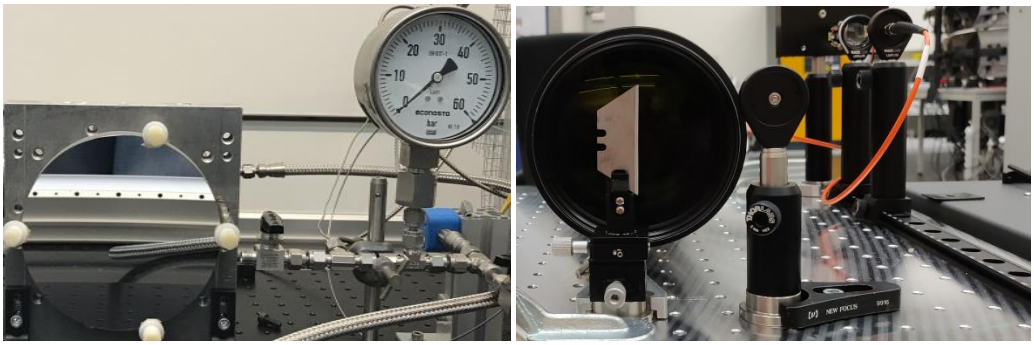

FIGURE 7 :SCHLIEREN IMAGING SETUP

The functioning of the setup is first demonstrated with a candle flame. The heat waves above the flame are clearly visualized. The knife edge distance is varied and the contrast of the image is observed as shown in the figures below:

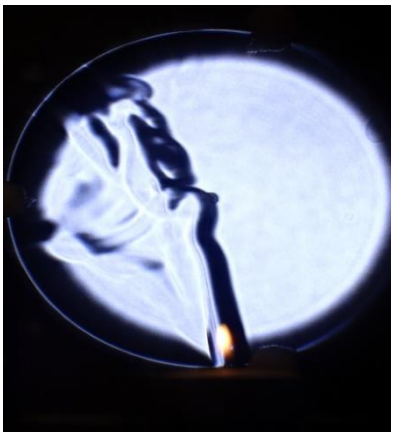

FIGURE 8: THE KNIFE EDGE BARELY TOUCHING THE REFLECTED POINT SOURCE

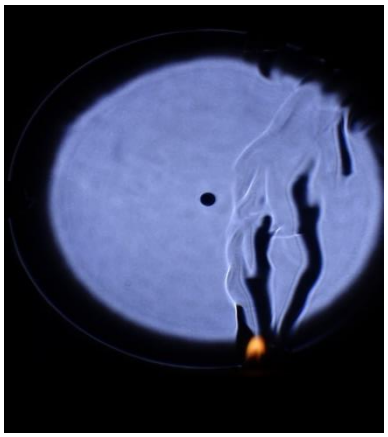

FIGURE 9: THE KNIFE EDGE TOUCHING 2/3RD OF THE REFLECTED POINT SOURCE

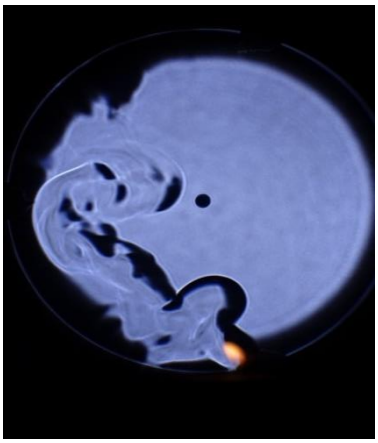

FIGURE 10: THE KNIFE EDGE CUTTING EXACTLY HALF REFLECTED POINT SOURCE

It can be clearly seen that the position of the knife edge can vary the quality of the image. As explained earlier the Schlieren image portrays the density gradients. The position of the knife edge is crucial to determine how the density gradient is varying. For instance in the Figure[10] the mirror where the influence of the candle is absent is the bright and has a constant low density region and the light waves all the reach the camera. The darker regions are the light waves which got cut off, this region is where the density is high. The graph below gives a clear picture:

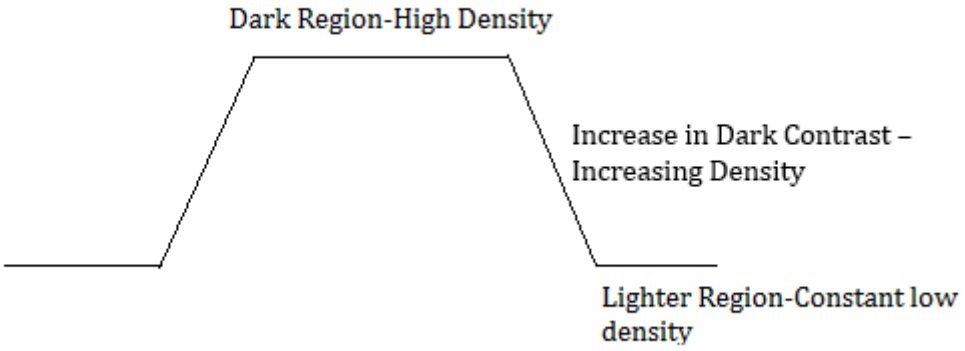

FIGURE 11: REPRESENTATION OF DENSITY GRADIENT

This setup can't be used in the case of expansion in vacuum as the quartz tube will obstruct the image.

CFD Modelling of Flow

A CFD model is developed using the Fluent 19 student version. The domain of interest is from the outlet of the nozzle to the expansion region. Hence the flow inside the nozzle is neglected for the simulations. Isentropic equations are used to calculate the pressure and temperature at the outlet of the nozzle. The velocity of the flow is calculated by using speed of sound relations.

$$v = \sqrt{\gamma R T} \qquad 2$$

$$\frac{P^*}{P_o} = \left(\frac{2}{\gamma + 1}\right)^{\frac{\gamma}{\gamma - 1}} \qquad 3$$

$$\frac{T^*}{T_o} = \frac{2}{\gamma + 1} \qquad 4$$

The material properties for carbon dioxide are chosen as ideal gas with density based solver. The simulations are done in a single phase assuming that the ideal gases properties will compensate for both the liquid and gas phase[2]. The energy and turbulence is captured using the built in energy equation and LES equation. The flow is simulated from high pressure to atmospheric pressure hence additional parameters are given to the solver as the flow is supersonic. The convective interaction of air molecules with the carbon dioxide is also neglected. The solid phase is not considered because the volume fraction of the solid formation is very small compared to the expansion domain.

Calculation of Cooling Power

The cooling power is calculated for the volumetric flow rate to be 10slm. This flow rate is considered because it is the flow rate which operates in a plasma microwave reactor.

The cooling power of the expansion is calculated in two ways:

1.Estimate

If the system is observed it can be seen that the initial temperature and the final temperature are the same. This behaviour is similar to an isothermal system. Using this observation a crude estimate of the cooling power can be calculated by using the isothermal relation:

$$W = d(pv) \qquad 5$$

In the above equation(4) represents the work done equal to the change in pressure and volume.

By applying the mass conversation and dividing the equation(4) by time. The volumetric flow rate ($\dot{v}$) remains constant and the parameter which changes is pressure.

$$\frac{W}{t} = \frac{v}{t} d(p) \qquad 6$$

$$Power(P) = \dot{v} dp \qquad 7$$

2.Detailed Calculation:

A detailed calculation is made by using the assumption that the flow is isentropic and at the orifice the flow is chocked [1].

Here the calculation is divided into three parts:

1. Nozzle: The flow from the inlet to outlet nozzle is assumed to be adiabatic. Therefore the Adiabatic relation used here:

$$P = \dot{m} c_p dt \qquad 8$$

2. The expansion region: The phase change from liquid to gas and liquid to solid is accounted here. The phase change from liquid to gas absorbs heat whereas from liquid to solid gives out heat.

3. Outlet: Here the pressure change that occurs from the orifice exit to atmospheric conditions is considered. As explained the expansion is isenthalpic which means the work done is only because of the pressure difference.

$$W = \dot{v} dp \qquad 9$$

Theoretical Mass Flow

The mass flow rate($\dot{m}$) at the orifice/outlet of nozzle is calculated using the following equation:

$$\dot{m} = \frac{AP_t}{\sqrt{T_t}}\sqrt{\frac{\gamma}{R}}\left(1+\frac{\gamma-1}{2}\right)^{-\frac{\gamma+1}{2(\gamma-1)}} \quad 10$$

The calculations done using this equation is used in the mass flow rate comparison studies against the experiment and simulation.

Results and Discussion

Qualitative characterization

Liquid CO_2 Expansion

The Schlieren figures below represent the liquid expansion from a pressure of 55bar, it can be seen that the shock waves at the exit of the nozzle is not visible.

Some observations and discussions from the images:

The darker portion of the image can is the higher density regions but here it is of the condensed water vapour in the air making the region opaque.

In the diameter 0.3mm some lighter regions can be seen on the tip. Performing the density gradient analysis: There is a high density region hence the temperature at that region is cold. An estimate can be made that an expanded shock wave is present.

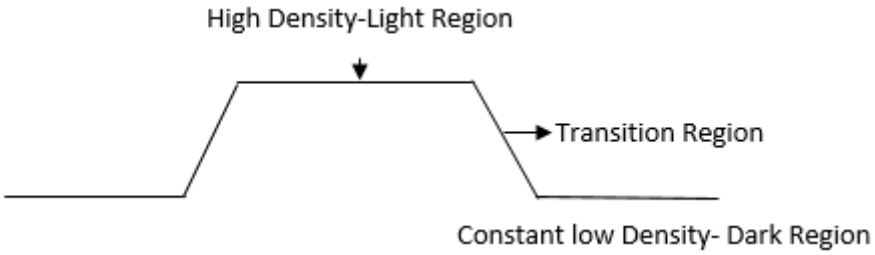

FIGURE 12: REPRESENTATION OF DENSITY GRADIENT FOR THE 0.3MM NOZZLE

The same effect can be in the 0.1mm nozzle with a reverse density gradient.

The nozzle with 0.2mm diameter does not have straight expansion. It can inferred that the nozzle has a manufacturing defect.

FIGURE13:NOZZLE DIAMETER-0.3MM

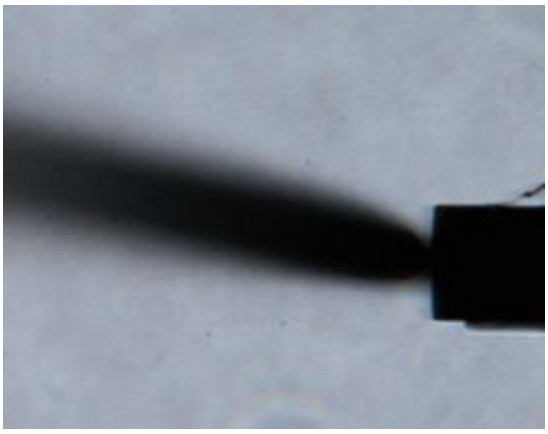

FIGURE14:NOZZLE DIAMETER- 0.2MM

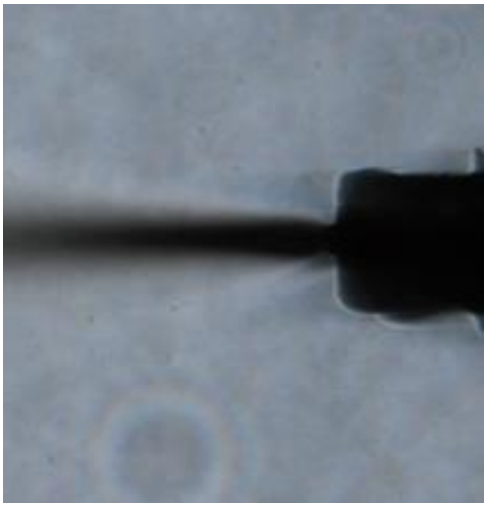

FIGURE15: NOZZLE DIAMTER-0.1MM

FIGURE 16: NOZZLE DIAMETER-0.3MM EXPANSION

Gaseous CO_2 Expansion

The below figures represent the gas expansion from a pressure of 30bar, it can be seen that the shock waves at the exit is visible.

The pattern of the shock wave at the exit is similar to the expected pattern.

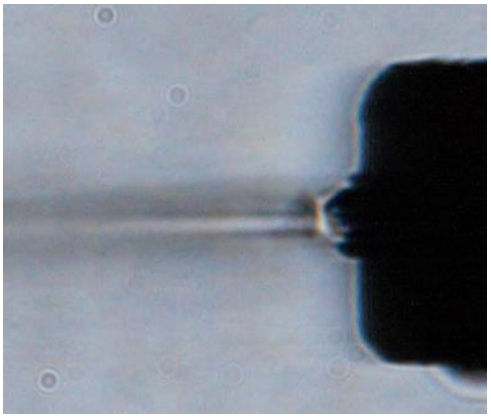

FIGURE17: NOZZLE DIAMETER- 0.3MM

FIGURE18 :NOZZLE DIAMETER-0.2MM

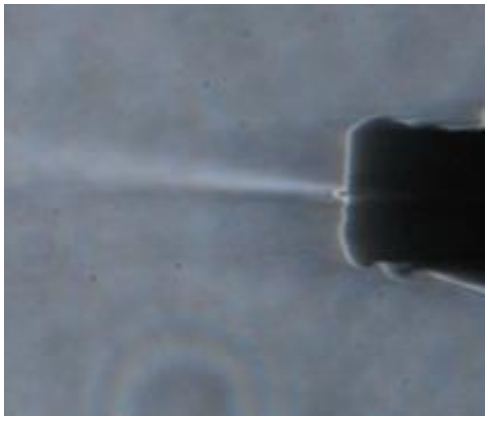

FIGURE 19: NOZZLE DIAMETE- 0.1MM

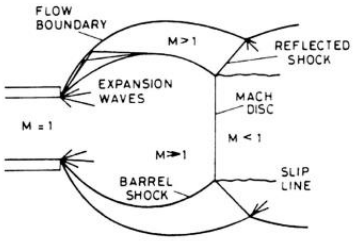

FIGURE 20: EXPECTED SHOCK WAVE PATTERN[2]

Images of gaseous expansion at lower pressures:

In the below figures it can be seen that a diamond shock wave pattern is observed. The pattern was seen at pressures:

Nozzle diameter-0.3mm for 40-35bar

Nozzle diameter-0.2mm for 40-30bar

Nozzle diameter- 0.1mm for 20-10bar

The presence of a diamond shock shows symmetric in the expansion.

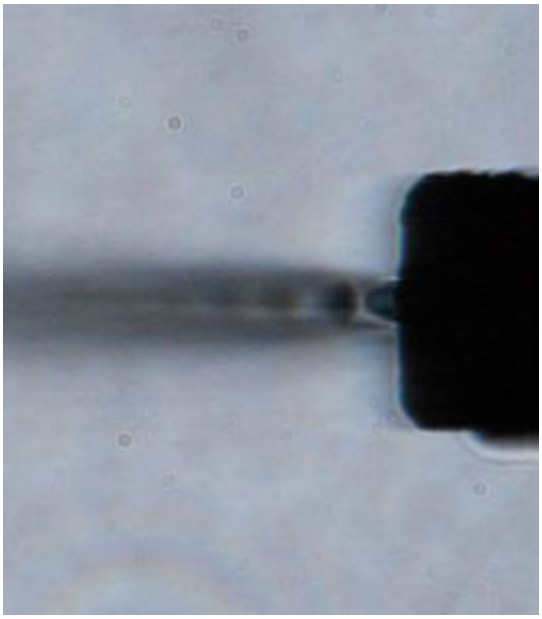

FIGURE21: NOZZLE DIAMETER 0.3MM

FIGURE22:NOZZLE DIAMETER 0.2MM

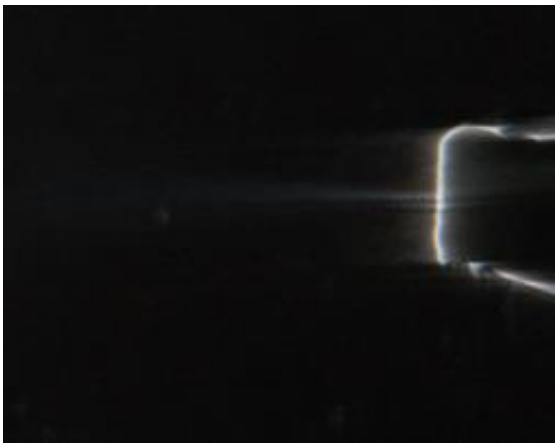

FIGURE 23:NOZZLE DIAMETER 0.1MM

The Schlieren images capture the shock waves with significant detail for gaseous expansion. For liquid expansion the shock waves are not visible. This occurs because the water vapour in the air condenses thus making that region opaque. However the images from the gas expansion can give a good insight about the shock wave pattern and can also be used to explain the exit shock pattern of liquid expansion as well. The diamond shock observed gives a new insight to this study. However the effects of these diamond shocks are not consider as important for this study because the temperature of the gas expansion don't have significant low temperatures.

Flow rates

Simulations

The simulation is run to compare the results obtained from the literature [2] using the approach from paper [3]. The nozzle of 2 and 4mm are constructed and input values from the literature[2] are used. While performing this simulation there were divergence issues. Then the values for the input of the simulation are calculated using mathematical formula which led a convergence in the results. Also the values given in the paper [2] didn't match the calculation even though they were both calculated in the same fashion [Appendix 4.1].

So the calculated values in the paper [2] were completely neglected. Considering the assumption of chocked flow the velocity, pressure and temperature were calculated [Appendix 4.2]. The mass flow rate is calculated and the mass flow rate from the simulation is taken and compared. This is a preliminary validation to check the simulation.

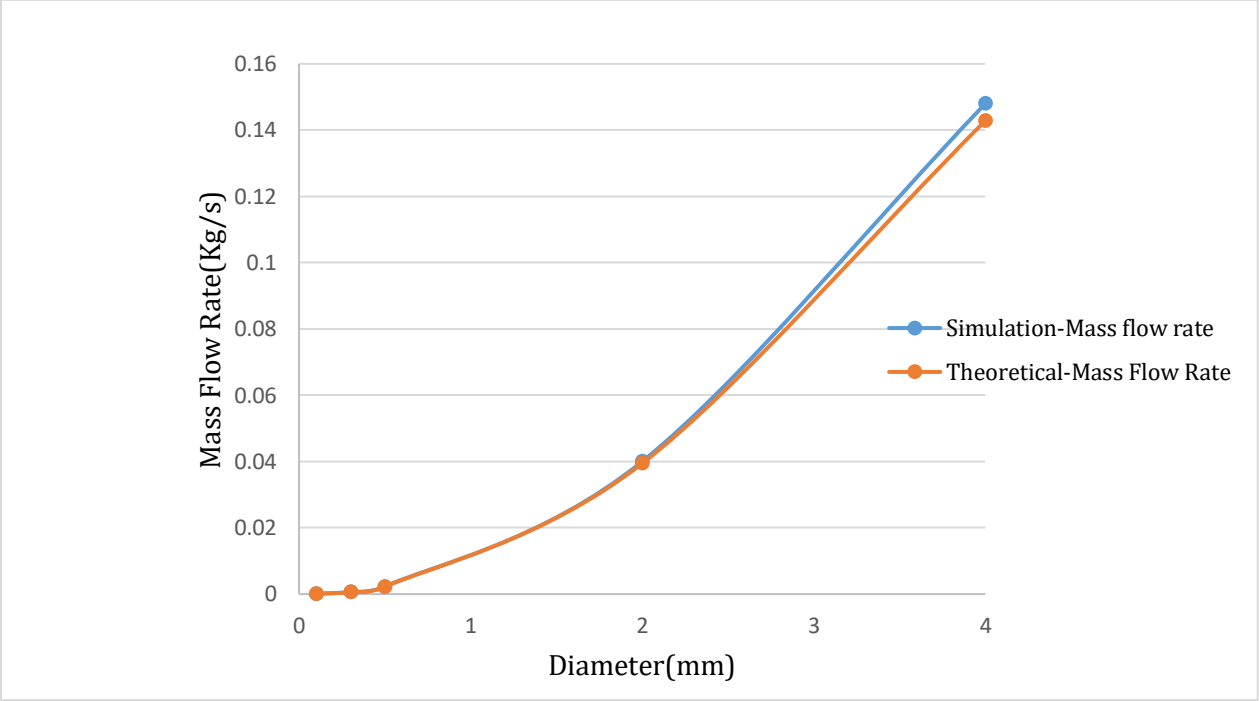

FIGURE 24: COMPARISON OF ISENTROPIC MASS FLOW RATE TO SIMULATION MASS FLOW RATE

Experimental Gaseous CO_2 Expansion

The flowrate measurement for the nozzle with 0.1, 0.2 and 0.2mm orifice as function of inlet pressure is measured [Appendix 2.2]. The table below present the mass flow rate at 35bar pressure for 0.1mm, 0.2mm and 37bar for 0.3mm.

Diameter(mm)	Mass Flow Rate				C_d
	Theoretical		Experiment		
	Kg/s	slm	Kg/s	slm	
0,1	0,000122	3,66	0,000104	3,4	0,85
0,2	0,000489	14,67	0,000368	11,95	0,75
0,3	0,00110	33	0,00037	12,12	0,34

TABLE 2: DISCHARGE COEFFICIENT

Since an orifice nozzle is used the difference in the theoretical and experiment mass flow rate can be because the discharge ratio is not considered. The discharge ratio can be considered as the ratio between the theoretical and experiment mass flow [2]. The discharge co-efficient for the 0.3mm nozzle is low the reason for this might be because

the orifice was made using a drill bit so the hole diameter might not be very uniform throughout the depth.

Experimental Liquid CO_2 Expansion

The flowrate for liquid expansion is measured for the nozzles with 0.1, 0.2 and 0.3 mm orifice. The results obtained are compared with theoretical and experiment results [6].

Diameter(mm)	Inlet Pressure(Bar)	Inlet Temperature (^{0}C)	Mass Flow rate			
			Experiment		Literatureexperimental[6]	
			Kg/s	slm	Kg/s	slm
0.1	55	16	0,00036	12.36	0,0002	6
0.2	52	16,6	0,000901	27.03	0,0005	15
0.3	53	16,5	0,001206	36.18	NA	NA

TABLE 3:MASS FLOW RATE MEASUREMENTS

In the above table it can be seen that mass flow rate obtained in this study for the diameter 0.1 and 0.2 mm is almost twice the mass flow rate obtained in the literature[6]. Some of the reasons which might have led the deviation is the nozzles in this study have a depth of 1mm while in the literature[6] capillary tubes are used which are 8mm long. The literature doesn't explain how the mass flow rates are being measured.

Operation condition: Expansion in vacuum

The mass flow rate measurement is performed for vacuum condition using the 0.3mm diameter nozzle. This nozzle is chosen because the maximum solid formation can be observed compared to the other nozzles with smaller diameter.

The mass flow rate of solid is computed as explained in the methodology.

Downstream pressure(mbar)	Experiment mass flow rate		Mass flow rate of solid		Solid fraction(%)
	kg/s	slm	kg/s	slm	
200	0.00096	28.8	0.000246	7.38	20

TABLE 4: MASS FLOW RATE-VACUUM MEASUREMENT

While performing this measurement there were few observations made:
The pressure in the bottle was not stable. There is pressure drop in the tube and after a time period the tube pressure becomes stable as well the bottle pressure. At that instance the mass flow rate of the gas is stable and this is the value represented in the above table.
The experiment was performed for time period of less than 2 minutes so the transient behaviour of solid formation was not recorded.

Temperature profiles

The temperatures are measured along the centreline of the expansion. The nozzle with 0.2mm diameter is neglected for this measurement expansion because the expansion is not along the centreline but inclined away from it.

The paper [2] was concluded by stating that the temperature of -78^{0}C is reached below 100mm from the jet release through the nozzle which matches the experimental case [6]. A more reliable validation is performed by measuring temperature from 0.5 to 10/20cm from the nozzle release and comparing it with the experiment performed in this studied.

The pressure is maintained at 30 and 55bar respectively for the gas and liquid expansion. The temperature is noted before the CO_2 enters the nozzle. These same conditions are used in the simulation [Appendix 4.3].

Comparing the release temperatures for gaseous carbon dioxide is shown in the figure below:

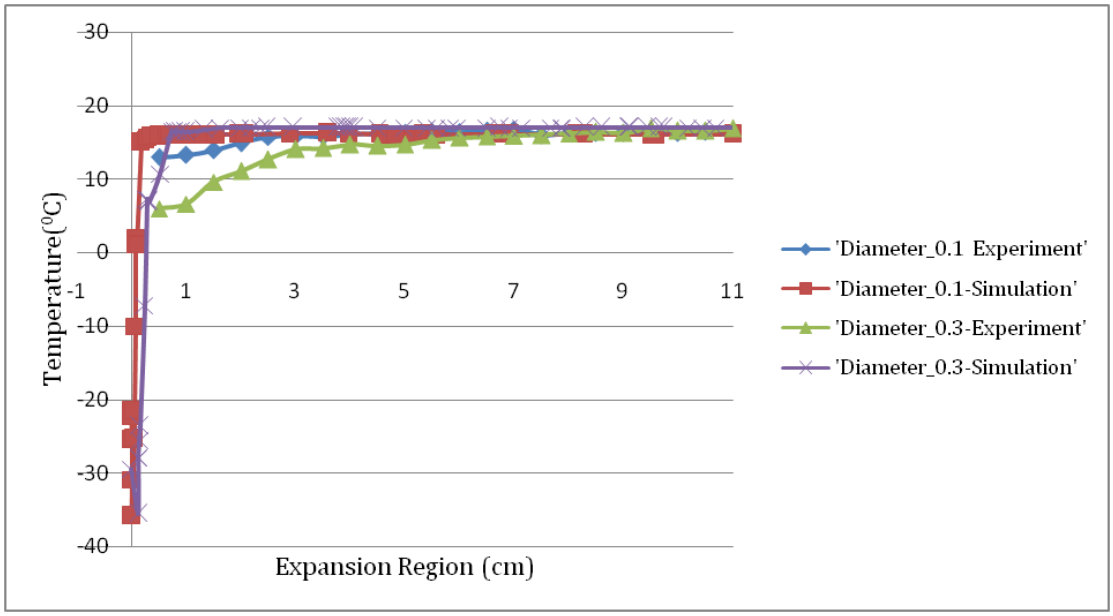

FIGURE 25: GASEOUS EXPANSION-TEMPERATURE PROFILE

Comparing the release for liquid carbon dioxide from the figure shown below :

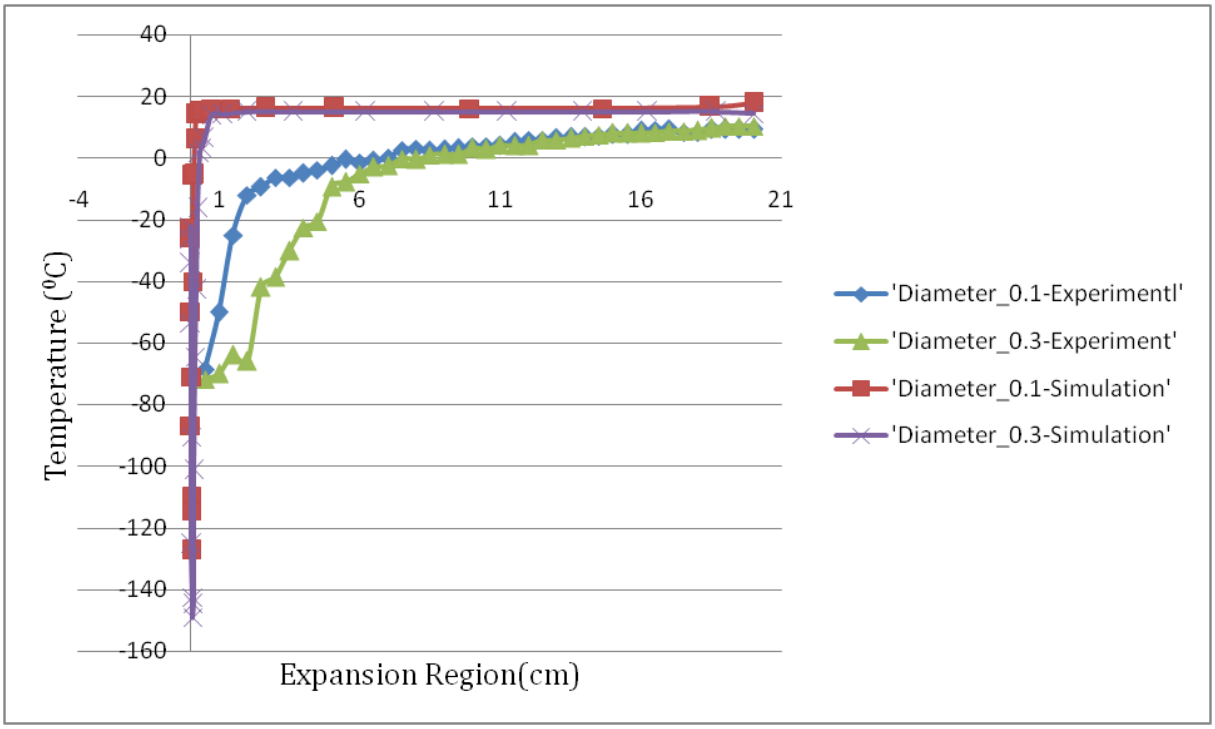

FIGURE 26: LIQUID EXPANSION-TEMPERATURE PROFILE

The temperature in the figure[26] is very low because of lack of phase transition models. When phase change occurs from liquid to solid the process is exothermic so heat is given out. Therefore in reality temperature below -80^{0}C is not measure.

The rise is temperature because of the interaction of air molecules and connective heat transfer. If an insulated tube is inserted around the nozzle with no solid ice formation the exit temperatures of the gas theoretical will be around ~-65^{0}C (+/- 5^{0}C may be lost in cooling the pipe). However in vacuum condition the expansion length will increase and the exit temperature of the jet will bellow 0^{0}C. This low temperature was measured during the mass flow experiments at the exit of the flow meters.

Cooling power

1. Estimate:

The system is an isothermal system because the inlet and outlet temperature are the same. Using this observation the cooling power can be calculated:

We know the volumetric flow is constant as mass is always conserved and 10slm is chosen because the microwave reactor has the working fluid entering at 10slm.

$$Power = \dot{v}(p_2 - p_1)$$

$$P = \frac{0.01(100000 - 5500000)}{60}$$

$$\boxed{P = -900W}$$

2. Detailed Calculation:

A detailed calculation is made by using the following assumption:

- The flow is isentropic
- At the orifice the flow is chocked [1].
- Mass is conserved.
- At the exit of the complete phase change occurs no liquid phase of CO_2 is present it either gaseous or solid.

The computation is divided in the following steps:

The flow from the bottle to the nozzle exit is computed using the adiabatic relation as explained in the methodology.

The inlet conditions are the conditions of the bottle which are 288K and 55Bar temperature and pressure respectively while the outlet of the nozzle is at chocked conditions which are at 250K and 30bar.

At the nozzle exit is where the phase change occurs:

Firstly it is assumed that the liquid CO_2 changes completely to gas. The conversion from liquid phase to gas absorbs heat which means this is maximum cooling power:

$$\boxed{P = -1089W}$$

From the experiments it is observed that solid CO_2 is formed at vacuum. A generic assumption of 5% solid formation is chosen. This value is chosen because it is the maximum solid formation observed in the literature[1]. The phase change mechanism from liquid to solid gives out heat. So this case is more close to reality the cooling power is computed as:

$$\boxed{P = -1083W}$$

From the calculation of the estimate and detailed calculation it can be seen that the cooling power difference is 189W.

Conclusion

- From this study it can be concluded that the simple orifice nozzle of ~0.1mm diameter is ideal for the implementation in the plasma reactor for the following reasons:
  - ✓ The operating volumetric flow rate of the plasma reactor is ~10slm.
  - ✓ The low temperature of -68^{0}C is achieved.
- The cooling power achieved by the liquid expansion is sufficient to cool the plasma because 1000Watt is used for the generating of plasma with 10slm of input flow and from the mathematical calculation a cooling power of 1089Watt (free expansion) is obtained.
- The solid ice formation seen in the vacuum tube will enhanced the cooling properties of the expansion jet as the plasma reactor is at extremely high temperatures ~3000^{0}C which will cause the solid to evaporate.
- The simulation for gaseous expansion matches for the temperature profile validation while for liquid expansion the simulation requires phase changing models and the material properties to be defined for carbon dioxide as supercritical fluid. The approach used the paper[3] is not valid for the liquid CO_2 expansion for temperature validation.

Future Studies

- The mass flow rate measurements must be repeated with nozzle of significantly varying depths. This study can give an insight if the carbon dioxide is liquid at the exit or supercritical (gas-liquid phase).
- In future studies the cooling power of the nozzle has to be verified by conducting heat transfer experiments. This can give a concrete validation for the mathematical model.

Outlook

➔ Access non-equilibrium regime in a vibrationally excited CO_2 discharge by adiabatic expansion cooling of the reactant - Is the plasma-chemical dissociation efficiency enhanced by cooling the CO_2 significantly before the discharge?

- Investigation of the theoretical and practical aspects in fluid dynamics of liquid CO_2 expansion in a plasma reactor
- Reconsidering the design nozzle with gas/liquid expansion depending on the effect observed with the simple orifice nozzle.

Symbols

A- Area(m^2)

C_d-Discharge

C_p-Heat Capacity(J/K)

CCS-Carbon Dioxide Storage

$\dot{m}$-Mass flow rate(Kg/s)

NA-Not Available

P-Power(Watt)

P_t-Total/Stagnation Pressure(Pa)

P^*-Critical Pressure(Pa)

R- Gas Constant (J/KgK)

slm-Liters/min

T_t- Total/Stagnation Temperature(K)

T^*- Critical Temperature(K)

t- Time (second)

$q^0{}_G$- Flow rate of carbon dioxide gas (m^3/s)

$q^0{}_A$- Flow rate of air (m^3/s)

$\rho^0{}_A$-Density of air (Kg/m^3)

$\rho^0{}_G$- Density of carbon dioxide(Kg/m^3)

$\dot{v}$-Volumetric Flow rate(m^3/s)

W- Work done(J)

γ-Specific heat ratio-1.3

Reference

[1] Fridman Alexdman (2008) Plasma Chemistry,Cambridge University Press 261.

[2]Pursell Mark (2012) "Experimental Investigation of High Pressure Liquid CO_2 Release Behaviour"158

[3]Risza Rusli, Eddie J.T. Change, Hoang H.P.L Phame and Azmi M. Shariff,2014,Solid Carbon Dioxide formation from Rapid Fluid Expansion using Integration of Computation Fluid Dynamics and Mathematical Modelling, Chemical Engineering Transtions,36,607-612 DOI:10.3303/CET1436102

[4]Sherman Robert, 2007, Carbon Dioxide Snow Cleaning, Particulate Science and Technology, 25:1, 37-57, DOI: 10.1080/02726350601146424

[5]Yi-Hung Liu, Graham Calvert, Colin Hare, Mojtaba Ghadira, Shuji Matsusaka,2012.Size measurement of dry ice particles produced from liquid carbon dioxide, Aerosol Science 48 1-9,DOI:10.1016/j.jaerosci2012.01.007

[6] Yi-Hung Liu, Hiroyuki Maruyama, Shuji Matsusaka,2010Agglomeration process of dry ice particles produced by expanding liquid carbon dioxide, Advanced Powder Technology 21, 652-657,DOI:10.1016/j.apt.2010.07.009

[7]Wolgang Merzkirch,2012, Flow Visualization,Universitat Essen, Federal Republic of Germany

Appendix

1. Apparatus

1.1 Brass Nozzle Holder:

This attachment was designed at Engineering Design Faculty and manufactured at the Mechanical Workshop in Differ. It consists of a conical nozzle holder; a plate and the brass cover which holds the quartz tube. The conical holder contains the nozzle within is inserted in the brass holder and the plate is fixed. Thus making the system air tight from one end.

FIGURE 27: BRASS NOZZLE HOLDER

2. Mass flow rate calibration:

2.1 Gilibrator

The gilibrator is a calibration system which can be used as a primary standard for the calibration of sampling air equipment. The system consists of two elements Wet flow cell and Control Unit base.

Wet flow cell: The wet flow cell has the following components:

1. Bubble Generator: The bubble generator consists of :
   i. Soap: A special soap is used for the bubble generation. This soap is designed to provide high strength to the film and is compatible with the materials in the flow cell.
   ii. Pulsation damper: This damper is built-in to reduce the pulsation in the airflow thereby reducing the oscillation of the bubble.
   iii. Bubble Initiation Button: This is a manual push button as the name suggests it is provided on the flow cell assembly to start the bubble film. When the button is pressed it lowers a ring into the soap solution and while the button is released the ring lifts out of the soap. A soap film is generated at the opening of the tube.
   iv. Bubble breaker: Is located at the upper section of the flow cell. It provides the travelling bubble with room to expand which leads to breakage to the bubble.
   v. Storage tubing: A storage tube connects the upper and lower cell chambers and this avoids the soap from evaporating which might cause changes in the concentration.
2. Sensor Block: This block includes the infrared sensors which are located above the ring used to generator the bubble and below the bubble breaker.

Control Unit base: This accommodates a crystal controlled microprocessor with inbuilt software to obtain the time interval for the calculation of flow rate parameters. This unit also contains a liquid crystal display system.

Working Principle:

The flowrate is measured by dividing the volume with the time interval. The Bubble initiation button is pushed generating a bubble which is carried above the wet cell tube by the air whose flow is being measured. The bubble passes the two infrared sensor of the sensor block and reaches the bubble breaker. The volume is measured as the space between the two infrared sensor. The time taken is the interval needed for the soap film bubble to travel between the two sensor within the volume. The timing information is sent to control base unit and is displayed. This crystal microprocessor makes the system very accurate thus making the gilibrator to be an eligible system for calibration of other flow rate devices. Hence a gilibrator can be used as calibrator.

2.2 Gas Mass flow rate:

The mass flow measurement was done for varying inlet pressure. The mass flow rate increases as the inlet pressure is increased.

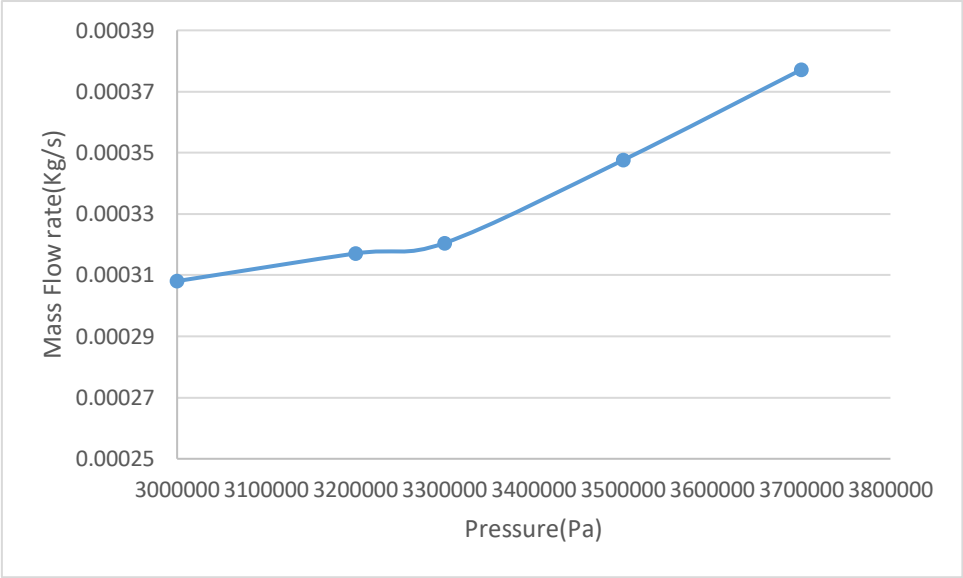

FIGURE 28: DIAMETER 0.3MM- MASS FLOW RATE VS PRESSURE

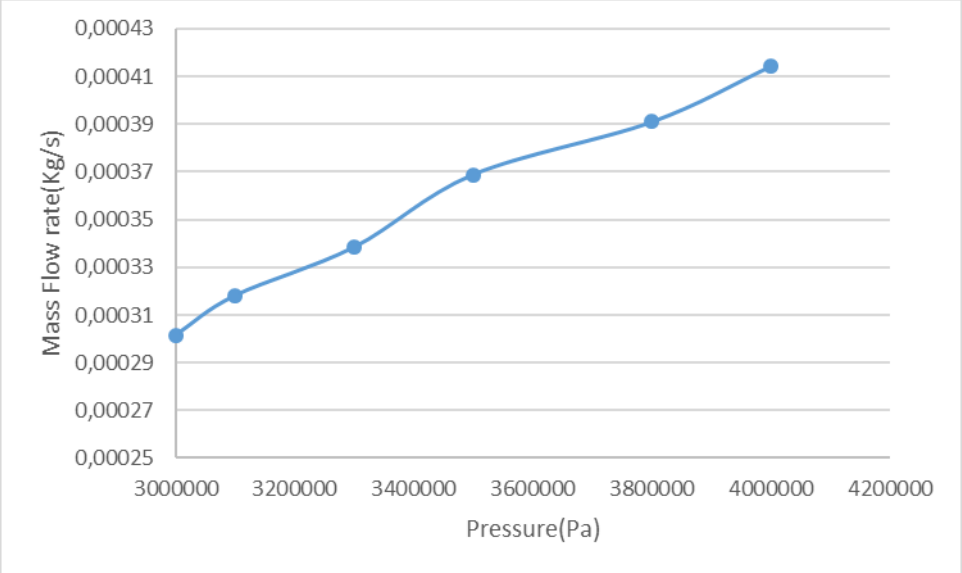

FIGURE 29: DIAMETER 0.2MM- MASS FLOW RATE VS PRESSURE

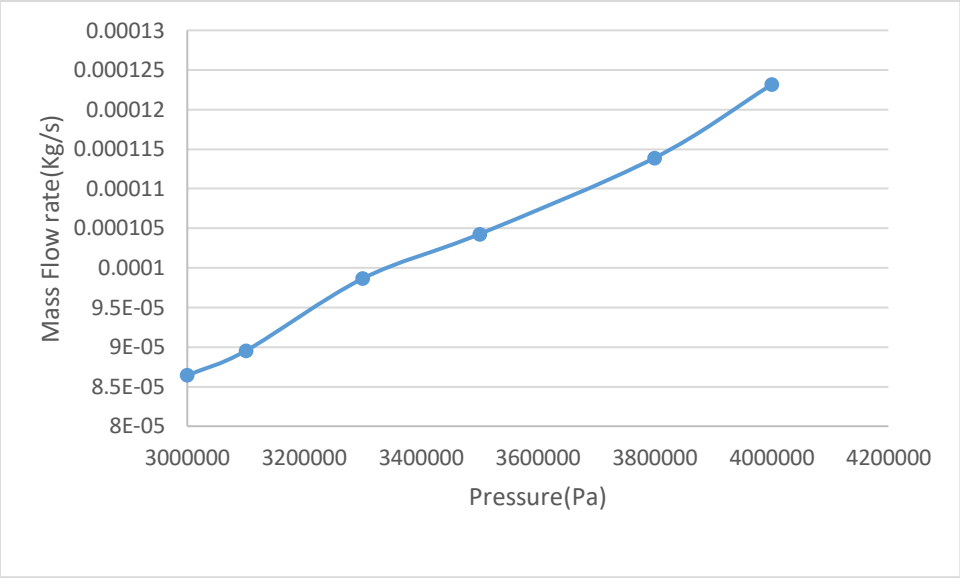

FIGURE 30: DIAMETER 0.1MM- MASS FLOW RATE VS PRESSURE

3.Experiment:

The experiments are conducted using different nozzles in two operating conditions. While performing the experiments for gaseous expansion the only parameter which can be controlled is the pressure whilst the temperature of the carbon dioxide can't be controlled. It is noticed that the temperature of gas keeps changing in the buffer volume hence while performing experiments it is quite difficult to reproduce the same results as the temperature is inconsistent. Hence the simulations are done with the temperature and pressure values while performing the temperature profile measurement. The mass flow rate obtained is compared with theoretical calculation at that operating pressure and temperature condition. As shown in the result, the mass flow rate obtained in the simulation matches theoretical calculation hence comparison between mass flow rate with the simulation is not repeated after the preliminary validation.

For the liquid expansion case, a pressure drop can be observed of around 5bar and temperature keeps fluctuating within +/-5^{0}C hence, the results are reasonably reproducible by switching the system on and off and recording the reading in pulses with a difference +/-2^{0}C in temperature profile measurements.

For the mass flow rate measurements a rise or drop of +/-5^{0}C will not make a large difference since the measurement is done in a short time period so this result is accurate and can be reproduced.

4. Simulation

4.1 Discrepancy in the values

In the table below it can be seen that the temperature values match the paper[2] exactly while the pressure values are different. The simulation depends on the input value given to get a good convergence. The discrepancy in the pressure values led to divergence in the simulation.

Diameter(mm)	Critical Pressure(Pa)-Paper[2]	Critical Temperature(K) -Paper[2]	Critical Pressure(Pa)	Critical Temperature(K)
2	2680000	263.47	2444860	263.47
4	2420000	259.86	2210197	259.86

TABLE 5: COMPARISON OF THE THEORETICAL VALUES OF THE PAPER[2] AND THIS STUDY

4.2 Prelimary mass flow validation:

The pressure and temperature values are chosen randomly and the orifice pressure and temperature are calculated. These values are feed in the simulation. This is just a prelimary validation of the simulation.

Diameter(mm)	Pressure(bar)	Temperature(K)	Critical Temperature(K)	Critical Pressure(Pa)	Velocity (m/s)	Mass Flow rate (Kg/s)	
						Simulation	Theoretical
0,1	35	298	259,1304	1910047	270,51	8,06E-05	7,727E-05
0,5	40	298	259,1304	2182911	252,26	0,00229	0,0022
2	44.8	303	263,4783	2444860	254,36	0,04	0,0392
4	40.5	298,84	259,867	2210197	252,61	0,148	0,143

TABLE 6: MASS FLOW RATE- INPUT FOR THE SIMULATION

4.3 Temperature Profile Validation:

The input for the simulation is given by the using the pressure and temperature at the inlet of the nozzle and computing the orifice pressure and temperature using these values.

Diameter (mm)	Pressure(bar)	Temperature (k)	Orifice Temperature(K)	Orifice Pressure (Pa)	Velocity(m/s)
0.1	30	289	253,913	1637183,2	266,4
0.2	30	288	250,43	1637183,2	265,94
0.3	30	280	243,48	1637183,2	262,22

TABLE 7: GASEOUS EXPANSION-INPUT FOR THE SIMULATION

Diameter (mm)	Pressure(bar)	Temperature (k)	Orifice Temperature(K)	Orifice Pressure (Pa)	Velocity(m/s)
0.1&0.3	50	288	250.4	3001503	265.9

TABLE 8: LIQUID EXPANSION-INPUT FOR THE SIMULATION