An Experiment on Flashing-Spray Jet Characteristics of Supercritical CO₂ from Various Orifice Geometries

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Supercritical CO₂ pipelines usually are used to link the CO₂ capture system to the geological storage. There are severe hazards once the asphyxiating gas leaks from the long-distance pipeline. The uncertainty of near-field jet characteristics results in imprecise consequences assessment of accidental release of supercritical CO₂. To improve the prediction of consequences of accidental release accuracy, the near-field mechanisms of flashing-spray jet was investigated. In this work, an experimental setup with multiple measurement instruments was developed to impose controllable CO₂ release from a high-pressure vessel. The flashing-spray jet structures of supercritical CO₂ from circular and rectangular orifices were recorded by a high-speed camera. Results indicate that the near-field structures of supercritical CO₂ jet from circular and rectangular orifices are totally different, which causes the different dispersion consequences. The jet angle and shock waves were analyzed quantitatively. Lastly, the models of flashing-spray based on the two different phenomena from rectangular and circular orifices were discussed. The combination of macroscopic and microscopic data in the jet can help to understand the complex physics and improve discharge and dispersion model. This work provides a fundamental data to consequences assessment of accidental release of supercritical CO₂.

Keywords: supercritical CO₂, release, near-filed structure, flashing-spray model, Mach disc

INTRODUCTION

Carbon dioxide (CO₂) as a major greenhouse gas (GHG) has increased significantly impacts on the earth owing to human activities such as burning of oil and gas and the discharge of exhaust gases. The Intergovernmental Panel on Climate Change (IPCC) reported that Carbon Dioxide Capture and Storage (CCS) can eliminate 20–40% of global carbon emissions (Metz et al., 2005). Carbon dioxide usually would be captured at a large point emission source (e.g., power plants) and be transported via long pipelines to another spot for use [e.g., oil field for enhanced oil recovery (EOR)] (Ziabakhsh-Ganji and Kooi, 2014). Currently more than 50 million tons of CO₂ is transported by over 6,400 km of pipelines in the United States (Metz et al., 2005). And the most pipelines are under supercritical/
some experiments related to the supercritical CO₂ release almost focused on the macroscopic parameters, such as pressure, temperature, concentration, and velocity. However, few studies focused on microscopic parameters, such as the structure of shock waves, the evolution of solid CO₂, and the expanded angle. (Ahmad et al., 2013) carried out a controlled CO₂ release experiment from various circular orifices to obtain the thermo-hydraulic data of CO₂. A superheated jet was founded during the releases. In our previous work (Wang et al., 2019), effects of impurity concentration, initial inner pressure, and temperature on dispersion behavior were studied. (Guo et al., 2016) designed and built a large-scale supercritical experimental pipeline with a total length of 258 m and an inner diameter of 233 mm. The under-expanded jet flow structure and phase transitions in the near-field were studied for supercritical CO₂ released through different orifice diameters. Relating to the small-scale experiments focused on the near-field jet of CO₂. (Wareing et al., 2014) measured dry ice particles distribution along the jets in liquid CO₂ release and found that the sizes of particles are around 0.1–100 μm. And the study presented Mach disc in these releases is at a distance of around seven nozzle diameters along the centerline from the nozzle and the particles are likely to be close to equilibrium after Mach disc. The effect of superheat on flashing atomization characteristics and on the snow formation of liquid CO₂ has been investigated (Lin et al., 2013). Results show that the spray pattern transfers from jet spray to cone spray, and then to a bowl spray configuration with the increase of superheat. As mentioned above, a lack of near-field experiments data limits the development of models of discharge and dispersion of CO₂. In addition, CO₂ was released from a circular orifice or nozzle in almost current release experiments of CO₂ pipeline. However, the cracks in the damaged pipeline usually are not circular. The difference in orifice pattern may results in different consequence in an accident release.

In this paper, an experimental setup with a high-speed camera system was designed and constructed to study the near-field characteristics of flashing-spray jet of supercritical CO₂ from various orifice geometries. The work focused on (1) the near-field structures during CO₂ released from different orifice geometries; (2) the evolution of jet angle which can affect the dispersion region; (3) the shock waves system in the release from orifices with different geometries; (4) finally models of the flashing-spray jet of supercritical CO₂.

BACKGROUND OF HIGHLY UNDER-EXPANDED JET

The jet from a high-pressure CO₂ vessel is different from general gas jet due to the phase transition and multiphase flow and must be considered. In view of general gas jet, the jet zone is divided into three sub-zones: flow establishment zone, transition zone, and established flow zone, as shown in Figure 1. The fluid jet from the orifice to the ambient, which causes discontinuous velocity,
further causes turbulence. As shown in Figure 1, extending the upper and lower boundaries of the jet to intersect at one point (Point O), and O is the virtual origin of the jet. And then θ is the jet angle.

A schematic of the highly under-expanded jet is shown in Figure 2. An expansion fan generates at the nozzle lip as the flow expands into the atmosphere. The pressure ratio \( P_0/P_\infty \) is an important parameter to describe the expansion level, where \( P_0 \) is a stagnation pressure in the vessel and \( P_\infty \) is the ambient pressure. When \( P_0/P_\infty > 15 \), the complicated shock waves system will forms. Except for the intercepting shock in the interior of the jet, the Mack disc normal to the flow is unique for under-expanded jet. The flow front the Mach disc is supersonic, whereas the flow behind the Mach disc is obviously subsonic. The temperature will rise sharply near the Mach disc. However, the shock-wave structure in these jets also depends on a geometry of the nozzle and the property of gas (Velikorodny and Kudriakov, 2012). Some researchers also reported that the Mack disc cannot be observed when the flow jets from an elliptic nozzle (Menon and Skews, 2010).

For non-condensing gas jet, a theoretical analysis has been developed to predict the Mach disc location,

\[
\frac{X_m}{d_c} = 0.5 \sqrt{\frac{P_\infty}{P_0}} \times \left( \frac{\gamma + 1}{\gamma - 1} \right)^{0.25} 
\]

where \( X_m \) is the location of the Mach disc, \( P_0 \) is the static pressure at the exit section. It should be noted that in the fact the Mach disc location weakly depends on \( \gamma \), and it can be approximated by a commonly used experimental correlation of (Ashkenas and Sherman, 1966):

\[
\frac{X_m}{d_c} = 0.67 \times \sqrt{\frac{P_0}{P_\infty}} 
\]

EXPERIMENTAL DETAILS

In order to study the near-field structure of supercritical CO₂ released from the pressure pipe, a new experimental device was
designed and built, as shown in Figure 3. The experimental apparatus consists of high-pressure vessel, gas source, CO2 pump, refrigerating unit, thermostatic water bath, nozzle, and high-speed camera system. The rated pressure of the vessel with a volume 6 L is 15.0 MPa, and the material is 316L stainless steel. The container is filled with liquid CO2 cooled by a refrigeration unit and controlled by a constant temperature water bath. To study the influence of different orifice diameters and orifice patterns (circular and rectangular) on near-field structure and dispersion of supercritical CO2, six different orifices were used, as shown in Figure 3. The supercritical CO2 near-field jet structure was observed with a single-lens reflector camera with a maximum frame rate of 200,000 FPS (frames per second). In this experiment, the frame rate of the high-speed camera is set at 3000 FPS.

It is very necessary to carry out the experiment under the premise of ensuring the safety of the experiment. In the process of supercritical CO2 injection, steel frame is adopted to prevent the generation of reaction force, and the noise level is controlled in an acceptable range. To ensure the stable experimental conditions, the experiments were carried out indoor to avoid the impact of atmospheric turbulence. According to the actual transportation conditions of CO2 pipeline, most of the initial conditions in the experiment are in the supercritical region. The main steps list as follows: (1) Before work, check whether the connection of the experimental device is loose and whether the container is damaged to ensure the normal operation of the equipment; (2) Open the cleaning mode to empty the air in the container to remove impurities; (3) The liquid carbon dioxide cooled by the refrigerator is fed into the container by means of a CO2 pump; (4) When the appropriate amount of CO2 is injected into the container, all valves are closed and a water bath heating sleeve is used to control the temperature in the container; (5) When the experimental conditions in the container reach the design conditions, the pneumatic valve in the pipeline will be opened quickly; (6) Record the experimental process with high speed camera.

RESULTS AND DISCUSSIONS

The Near-Field Structure of the Supercritical Jet

Figure 4 presents that the near-field structure of the supercritical jet from a circular orifice and a rectangular orifice, and the experimental conditions are also presented. Same as the non-condensing gas jet, the jet region of supercritical CO2 is divided into three zones. As shown in Figure 4, in core zone, the concentration of CO2 is 100% and the constituent is gas-solid CO2 (Teng et al., 2018). With increase of jet distance, the air entrainment caused by turbulence resulted in the decrease of CO2 concentration. There is a difference between circular orifice and rectangular orifice. It can be obviously seen that the jet angle of rectangular orifice is much larger than that of circular orifice. But the jet distance of circular orifice is further than that of rectangular orifice. The jet structure in near-field caused the difference. Figure 4A shows that the release from circular hole was typical highly under-expanded jet. The shape of jet is a barrel configuration. The Mack disc appeared in this process. When supercritical CO2 is released from the rectangular orifice, the
shockwave system has a fan-shaped structure, as shown in Figure 4B. In addition, the fan-shaped region in near field was brighter than other regions, because a greater concentration of solid CO₂ caused a stronger reflex of light. The Mach disc was unobserved and the intercepting shock can be observed. The CO₂ jet flows radially after shock wave.

**FIGURE 4** | The patterns of jet of supercritical CO₂ for circular (A) and rectangular (B) orifices.
In the process of release, the air entrainment occurred in a broader region when supercritical CO\(_2\) released from a rectangular ori
ifice. This phenomenon may affect the component of heavy gas cloud in the dispersion.

Some researchers show that the Mach disc may not be observed in rectangular jet, even though the pressure ratio is ca. 5.60 (Li et al., 2017). In our research, the Mach disc cannot be observed for rectangular jet. However, the change process of the intercepting shock was recorded by high-speed camera, as shown in Supplementary Video S1. The pressure ratios in the experiments are 70–100. It can be seen from Figure 4 that the brighter region where plenty of micron-level dry ice particles generated has stronger reflectivity. Across the shock system, lots of dry ice particles sublimate due to the temperature rose sharply. Thus, lower concentration of dry ice cause weaker reflectivity.

To understand the overall supercritical CO\(_2\) jet, the configuration of jet of supercritical CO\(_2\) released from a rectangular ori
ifice (5 \times 1 mm) was analyzed, as shown in Figure 5. Overall, the shape in the major axis plane is fan-shaped, and the photo from the high-speed camera was presented in Figure 4B. The shape in the minor axis plane is conical and the photo from the high-speed camera was presented in Figure 5. It may be interesting to note that the results show in the minor axis plane, the fluid expands firstly and then shrinks, and finally appears core-shaped. It can be called “Expansion-Contraction-Expansion Configuration (ECEC).” It can be obviously seen from Figure 4, Figure 5 that the shape of jet of supercritical CO\(_2\) released from a rectangular orifice is very different from that release from a circular orifice, which may cause the difference of dispersion region.

**The Characteristics of the Jet Angle and Shock Waves**

**Expanded Angle**

The jet angle and shock waves have important impact on the near-field characteristics, which is related to the accuracy of source term model. In order to investigate the effect of orifice pattern on jet angle, a characteristic model of crack was developed to analyze the angle evolution. We assumed that the crack of the pipeline is elliptic. The rectangular orifice was used to characterize the crack. As shown in Figure 6, the length of the minor axis (b) of the elliptic and the width of the rectangular orifice is the major axis (a) of the ellipse. Thus, the elliptic equation is \( \frac{x^2}{a^2} + \frac{y^2}{b^2} = 1 \).

If \( a \neq b \), the variation of the jet angle with \( b/a \) was presented, and if \( a = b \), the variation of the jet angle with diameter was presented in Figure 6.
Figure 7 presents the correlation between the peak value of $\theta$ (=$\theta_{\text{max}}$) and the characteristic parameters ($b/a$ and $d$) for different pressure. Figure 7 shows the jet angle increases linearly with increased diameter of leakage orifice in circular release. And the jet angle decreases with increase of $b/a$. It indicates that the longer and narrower crack may cause broader dispersion region. Generally, the maximum jet angle can reach to 180°.

Comparing with the circular release with the similar area ($d = 2$ mm), the jet angle of the rectangular release ($3 \times 1$ mm) is six times larger than that. Figure 7 also indicates that the transportation pressure has a relatively small impact on the jet angle during supercritical CO$_2$ release. Thus, the peak jet angle ($\theta$) can be given as an empirical equation,

$$\theta = -115.18 \left(\frac{b}{a}\right) + 159.89 \quad R^2 = 0.98, \quad b/a \neq 1 \quad (3)$$
\[ \theta = 3.91d + 9.19 R^2 = 0.99, \frac{b}{a} = 1 \]  

(4)

Generally, the current \( CO_2 \) transportation is under supercritical or dense state (Teng et al., 2016a). But some short-distance \( CO_2 \) pipeline is under gas state (Teng et al., 2016b). To evaluate the effect of phase states on jet angle, the discharge experiments from a rectangular orifice \((5 \times 1 \text{ mm})\) for supercritical, dense and gas \( CO_2 \) were carried out. Figure 8 shows the variation of jet angle with time for three phase states. It can be seen that the effects of different phase states on jet angle are different. The rate of change of jet angle for supercritical \( CO_2 \) release is nearly constant. In other words, the jet angle increases linearly with time. However, the rate of change of jet angle for dense and gas \( CO_2 \) release is constant firstly and then increasing. And the peak jet angle of dense \( CO_2 \) is slightly bigger than that of gas \( CO_2 \). Overall, it can infer that the jet angles are affected by expanded process and phase transition process inside the nozzles.

Shock Waves

The data points and the error bars denote the averaged value of experimental data and the standard deviations, respectively, as shown in Figure 9. The Mach disc is a feature of under-expanded jet and the temperature changes dramatically across the Mach disc. Many researchers investigated the Mach disc (Abbett, 1971; Veser et al., 2011; Mitchell et al., 2013; Zhou et al., 2018). The pressure ratio in most of the studies is below 20 and the multiphase flow and phase transition was not involved (Otobe et al., 2008). Generally, the location of Mach disc away from the orifice is increasing with the pressure ratio. The condition is a supercritical state in our research, which indicates that the pressure ratio can be over 70. Figure 9 shows the variation of the Mach disc location with pressure ratio when supercritical \( CO_2 \) released from a circular orifice. It reveals that the Mach disc location \( X_m \) increases linearly with the pressure ratio.

It can be seen from Figure 9 that Eq. 2 tends to under-predict the Mach disc location. It indicates that the theoretical equation may be not applicable to the jet of supercritical \( CO_2 \) because the phase transition and multiphase flow appear in this process.

Figure 10 shows the peak intercepting shock locations in different size rectangular orifices. It indicates that the intercepting shock location increases approximately linearly with the equivalent diameter \( d_e \). The aspect ratio has little impact on the intercepting shock location.

Modeling the Flashing-Spray of Supercritical \( CO_2 \)

Note that it is difficult to observe or measure directly the internal situation of multiphase jet in such a supercritical release because of the complex phase transition and multiphase flow. The mechanisms of multiphase jet of supercritical \( CO_2 \) can be drawn from the present experimental results including the near-field jet structure, expanded angle, and shock waves. Figure 11 presents the schematic of the model of multiphase jet of supercritical \( CO_2 \) released from different pattern orifices. Overall, the multiphase jet appears due to the joint effect of internal and external transition. The supercritical \( CO_2 \) in the chamber is transformed to metastable state (supersaturated state) due to the rapid pressure drop. The extremely rapid nucleation of vapor bubble in superheated state would occur with further pressure drop. This process can be supposed to homogeneous nucleation. Meanwhile, the droplets appear in the process of homogeneous nucleation. The classical nucleation theory (CNT) (Zel'dovich, 1961) for the spontaneous nucleation is
\[ J = \left( \frac{\rho_f}{\rho} \right) \sqrt{2\alpha/\pi m^3} \cdot e^{-\Delta G/(k_b T)} \]  

where \( J \) is nucleation rate, \( k_b \) is Boltzmann constant, \( m \) is single molecular mass, \( \Delta G \) is the free energy barrier, \( \rho \) is density, \( \alpha \) is surface tension, and \( T \) is temperature. Subsequently, the radius of bubbles grows extremely. The critical radius for CO\(_2\) bubbles is a constant for a given nozzle geometry and \( p \) is pressure. Outside the chamber, the temperature reduces below triple point, which caused by Joule-Thomson effect, and then plenty of dry ice particles jet fast. As shown in near-cylinder structure, the process is a highly under-expanded jet. The intercepting shock location in a rectangular jet increases approximately linearly with the equivalent diameter \( d_e \).

\[ r_c = \frac{2av_l}{RT \ln \frac{\rho_l}{\rho_g}} \]  

Where \( r_c \) is the critical radius, \( v \) is specific volume, \( R \) is universal gas constant, and \( p \) is pressure.

The original contributions presented in the study are included in the article/Supplementary Material, further inquiries can be directed to the corresponding author.

**DATA AVAILABILITY STATEMENT**

The present work is supported by the Research Foundation of Fuzhou University (Grant No. GXRC-20041) and the Natural Science Foundation of Chongqing (Grant No. CYY202010102001).

**AUTHOR CONTRIBUTIONS**

LT and JB contributed to conception and design of the study. LT, JB, YL, and CW wrote sections of the manuscript. All authors contributed to manuscript revision, read, and approved the submitted version.

**SUPPLEMENTARY MATERIAL**

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/fenrg.2021.697031/full#supplementary-material
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