Investigation of Droplet Nucleation Inside A Beryllium Injector Using Small Angle X-Ray Scattering (SAXS) Technique

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ABSTRACT

The structures of condensed supercritical ethylene jets inside an axisymmetric beryllium injector were explored using the small-angle X-ray scattering (SAXS) technique. The experiment was conducted at the 12-ID-B beamline at the Argonne National Laboratory. Beryllium was selected as the injector material for its high X-ray transmittance. Scattering intensity was measured both inside and outside the injector at three injection temperatures. Based on the detected scattering intensity profiles, size and population of scatterers were modeled. Evolution of droplet nucleation and droplet growth processes along the injector axis were investigated. The feasibility of using the SAXS technique to study the properties of condensed phase inside the beryllium injector was successfully demonstrated. Small scatterers on the order of 10-30 Å (1-3 nm) were observed within the injector. The creation processes and identity of these small scatterers could not be confirmed in this study. Scatterers bigger than 200 Å (20 nm), up to the detection limit of the present SAXS setup, were observed mainly outside the injector, but also within the final passage region. These scatterers are believed to be ethylene droplets. The presence of big scatterers within the final passage region at a high injection temperature was observed. Injection temperatures close to the critical temperature were found to promote early occurrence of droplet nucleation, which allows a longer residence time for droplets to grow. For the present injector contour, abundant big scatterers appear at the entrance of final passage at an injection temperature near the critical point, and persist downstream of the nozzle exit, where the discharged ethylene jets undergo the greatest expansion process, for all three injection temperatures. Several questions raised by the present study must still be answered to confidently advance the understanding of droplet nucleation and growth in supercritical ethylene jets.

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INTRODUCTION

Injection of supercritical fuel has become an increasingly important research area in the development of advanced air-breathing propulsion systems [1]. In some applications, the use of endothermic hydrocarbon fuels as the primary coolant around airframe and combustor components inevitably creates thermally cracked hydrocarbon mixtures at supercritical conditions [2]. The injection of this supercritical fluid may, however, impact the combustion behavior of the engine, due to liquid generation through homogeneous nucleation processes [3].

Early studies on the structures of supercritical jets injected into a quiescent environment mainly focused on the global appearance of the discharged plumes, using shadowgraph images and Raman scattering [3-5]. Figure 1 shows supercritical ethylene jets injected into a quiescent chamber. The global jet appearance changes from somewhat transparent at a high injection temperature, with less condensed phase (Fig. 1(a)), to highly opaque at a temperature close to the critical temperature, with high concentration of condensed phase (Fig. 1(c)).

Properties of the condensed phase within the discharged supercritical ethylene jets, including droplet size and liquid volume fraction, were first explored by Lin et al. [6], using the small angle X-ray scattering (SAXS) technique available at the Argonne National Laboratory. It was found that the SAXS technique is capable of measuring droplet size and liquid content inside a highly dynamic condensed jet. The droplet size inside a condensed supercritical ethylene jet is on the order of 500-2000 Å (50-200 nm), which is smaller than that generated from industrial atomizers.

Having visually observed the occurrence of condensed phase within the supercritical ethylene fluid inside a rectangular injector (see Fig. 2, [3]), Lin et al. later applied the SAXS technique to identify the onset of nucleation processes and to characterize droplet properties and liquid content inside a rectangular injector equipped with diamond windows for X-ray access [7]. This study demonstrated, for the first time, the capability to explore a highly dynamic supercritical ethylene jet inside an injector. Droplet size in the range of 30-100 Å (3-10 nm) inside the rectangular injector was observed. In addition, it was observed that large droplets are preferentially distributed along the injector sidewall. Results from this study, however, may have been compromised by hardware issues, including undesired flow leakage and uneven junctions between the metal injector body and the diamond windows. Nonetheless, this study led to the development of a conventional axisymmetric injector out of beryllium, which allows high X-ray transmittance, for the study of both internal and external flow structures.

The objectives of the present study, therefore, are to assess the feasibility of using a beryllium injector for SAXS measurement, to identify the onset of the nucleation process, to measure the size of incipient droplets, and to characterize droplet size evolution inside the injector. A beryllium injector with an exit orifice diameter of 0.5 mm was designed and fabricated for SAXS measurements.

EXPERIMENTAL METHODS

Apparatus

The experiment was conducted at the 12-ID-B beamline at the Argonne National Laboratory. Pure ethylene (99.5%) at the desired temperature and pressure was injected into a chamber filled with nitrogen. The apparatus consists of an accumulator, a heating/chilling unit, a section of heat exchange tube, a solenoid valve, an injector, and the injection chamber. The chamber is equipped with two mica windows opposite to each other to provide high x-ray transmittance and low scattering, and two conventional optical windows to allow for visual observation of injection. Each mica window has a thickness of 100 µm. A heating/chilling unit controlled the temperature of ethylene glycol, used as the heat exchange medium, which flowed inside the annular passage of the heat exchanger.

The injection chamber is connected to a large injection tank. The combined total internal volume is 122 liters. The injection tank is then connected to the exhaust system through a venturi vacuum pump, which is used to quickly minimize the pressure rise inside the injection chamber. An increase in chamber pressure of less than 10%, however, was observed over the course of injection. Since the ethylene jet is expected to be choked at the injector exit, the pressure increase in the injection chamber does not affect the flow structure inside the injector. The effect of pressure rise in the chamber on the structures of the discharged ethylene plumes is also negligible, due to the large ratio between injection pressure and chamber pressure. Figure 3 shows the integration of the injection chamber setup with the X-ray beamline and detector. The injection chamber was rigidly mounted on a translation stage to provide the desired movement perpendicular to the X-ray for SAXS measurements.

Injection Nozzle

An axisymmetric injector made out of S65C beryllium was used in the present study. Beryllium was selected as the injector material due to its high X-ray transmittance. With a carefully designed wall thickness to safely withstand the anticipated injection pressure and to minimize scattering from the injector itself, flow structures both inside and outside the injector can be probed by the present X-ray diagnostics. Figure 4
shows a photo and the internal geometries of the beryllium injector. The injector begins with a 7.0 mm diameter passage, followed by a short 43.6° converging section, before reaching a 5.0 mm diameter passage. A second converging section with a converging angle of 60 degrees and a length of 4.0 mm leads to the final passage. The final passage has a length of 2.0 mm and a small converging angle of 2 degrees, in order to ensure that the choke point can only occur at the nozzle exit plane. The discharge orifice diameter (d) is 0.5 mm. The injector wall thickness is between 0.50 and 0.58 mm, which provides a factor of safety of 10 or higher with respect to the selected injection pressure and still allows a significant amount of photons to pass through. This beryllium injector was fabricated via a powder metallurgy process with a specified final surface roughness of 50 µm. All transition corners are contoured surfaces to avoid any undesired flow separation. The beryllium injector was placed inside a specially designed protective sleeve, which accommodates three small openings for X-ray access and ethylene discharge. The protective sleeve was installed to minimize the probability of breakage of the mica windows by beryllium fragments in the event of injector failure.

Injection Procedures

In test preparations, the well-purged and evacuated accumulator was first filled with ethylene. The filled accumulator was then pressurized using nitrogen to 5.11 MPa (744 psia), which is greater than the critical pressure of pure ethylene, P_c = 5.04 MPa (733.54 psia). Ethylene was then introduced into the heat exchanger via a powder metallurgy process with a specified final surface roughness of 50 µm. All transition corners are contoured surfaces to avoid any undesired flow separation. The beryllium injector was placed inside a specially designed protective sleeve, which accommodates three small openings for X-ray access and ethylene discharge. The protective sleeve was installed to minimize the probability of breakage of the mica windows by beryllium fragments in the event of injector failure.

Instrumentation

A monochromatic, 0.10 (W) × 0.07 (H) mm² X-ray beam having a photon flux of ~10¹¹ s⁻¹ in the 12-ID-B beamline was directed through the injection chamber. The distance between the injector axis and the detector was maintained at 2.0 meters. A photon-counting 2-D array X-ray detector (PILATUS 2M detector) was utilized to capture the forward-scattered X-ray photons. The PILATUS detector has 24 detector modules, which were arranged in an 8 (V) × 3 (H) format, to provide a total of 2.48 mega pixels. A total of 14 images were obtained for each injection condition. Each image has an integration time of 1.0 second to capture a sufficient amount of photons scattered from the medium of interest. In the present study, droplets and possibly precursors of droplet nuclei within the ethylene flow are the medium of interest. Scattering signals from the beryllium injector, mica window, and ambient gases were subtracted out, with the assumption of negligible X-ray multiple scattering.

Figure 5 illustrates the X-ray probing regions and the specific sub-regions associated with specific flow structures and droplet dynamics. To explore droplet nucleation and growth processes, the X-ray was directed through the centerline inside the beryllium injector. In the present study, Zone 1 was designated as the region upstream of the final passage of the nozzle, i.e. x<-2.0 mm, where x is the axial distance from the injector exit plane with x<0 inside the injector. For the injection conditions with a large ratio between injection pressure and ambient pressure, flow within this zone undergoes and initial expansion process and may potentially create suitable local conditions for droplet nucleation. Zone 2 is the region within the final passage, i.e., -2 mm <x<0. Zone 3 is the region outside the injector, i.e., x>0, where the discharged flow expands significantly. In the present study, the region between x=-3.0 mm and x=1.5 mm was probed. With the focus of the present study on droplet nucleation and growth processes, there was no attempt to probe the discharged plumes at locations further downstream.

Data Reduction

Details on data reduction can be found in Ref. [6]; the system is summarized here. The procedure begins with the processing of raw images obtained from the X-ray detector. Figure 6 shows the typical small-angle scattering raw image. Creation of the radial scattering profile and particle size distributions from the raw scattering images were accomplished using Igor Pro commercial software, along with a software package developed by Ilavsky and Jemian [8]. First, the center of the X-ray beam, which was blocked by a beam stop to protect the detector, was located. Second, a mask was created to carve out the areas covered by the beam stop and the gaps between the detector modules, since
scattered photons reaching these areas could not be counted. All pixels in the image were then mapped into a scattering angle by considering the geometry of the experiment (detector size and the distance between detector and sample of interest). The average zenith angular scattering profile could then be calculated from a field of view encompassing a large azimuthal angle.

The scattering profile intensity were plotted with respect to the scattering vector \( Q = (4\pi/\lambda)\sin(\theta/2) \), where \( \theta \) is the scattering angle and \( \lambda \) is the wavelength. The \( Q \) range for the present study is 0.004 – 0.4. The maximum particle size to which these measurements are sensitive depends on the minimum \( Q \) value with the relationship \( D_{\text{max}} = 2\pi/Q_{\text{min}} \). Based on this relationship, the upper detection limit for the maximum diameter is 1570 Å (157 nm) for the present SAXS setup.

To obtain particle size distributions from the small angle scattering profiles, the scattering profile was modeled from assumed particle size distributions to match the measured intensity profile within a prescribed tolerance. Figure 7 shows the droplet size distributions at \( x = 2.0 \) mm for the supercritical ethylene jet injected at \( T_{\text{inj}} = 286.6 \) K. Two ranges of measured scattering intensity profiles were selected for fitting to illustrate the challenges in size modeling. Measured and fitted scattering intensity profiles are marked with green and blue lines in Fig. 7, respectively. The blue line is overlapped by the green line in Fig. 7, indicating that fairly good size modeling has been achieved. The calculated droplet size distribution and the calculation residual are marked with a green bar with red border (appear as red column) and red dots, respectively. For the result with a \( Q \) range of 0.0045-0.25 selected for modeling in Fig. 7(a), a large number of small scatterers in the size range of 10-15 Å (1-1.5 nm) was obtained. Bigger scatterers with a diameter in the range of 60-1000 Å (60-100 nm), however, could not be observed unless the intensity profile with \( \text{wavy} \) distribution characteristics within the \( Q \) range of 0.0045-0.017 was selected for size modeling, as illustrated in Fig. 7(b). There are relatively few of these big scatterers, so they do not show up in Fig. 7(a). The big scatterers, however, dominate the scattering intensity in the overall intensity profile. In order to avoid any misinterpretation of the SAXS measurement, the scattering intensity profile will first be examined to determine the populations of large and small scatterers. According to the \( D = 2\pi/Q \) relation, the presence of a low \( Q \) intensity profile indicates the existence of large scatterers, which are presumed to be droplets in the present study.

With this fitting method and the assumption of the same scattering contrast and spherical shape for all sizes of scatterers, the scatterer volume distribution, total volume fraction, and volume-weighted scatterer size could be obtained. The average volume fraction of condensed droplets across the ethylene jet could be computed by integrating the droplet volume distribution with local densities. These densities, however, were not measured in the present study and change throughout the injection processes. Consequently, the reported liquid volume fraction (or particle volume distribution in most of the figures) was not accurately quantified and was used as only as an indicator of relative population in the present study.

RESULTS AND DISCUSSION

Evolution Along Injector Axis:

a) Case #1, \( T_{\text{inj}} = 293.2 \) K:

Figure 8 shows the intensity profiles at various axial locations for the ethylene jet injected at a temperature of 293.2 K, which corresponds to a reduced temperature of 1.04. This is the high end of the temperature range explored here; the corresponding jet appearance outside the injector is shown in Fig. 1(a), and exhibits a fairly low concentration of condensed phase. Figure 9 illustrates the modeled particle size distribution profiles at selected axial locations for each zone (see Fig. 5(b) and Fig. 8). Fig. 8 shows no smooth scattering profiles from big scatterers in the low \( Q \) range in Zone 1 (upstream of the final passage). The modeled scatterer size is between 10 Å (1 nm) and 20 Å (2 nm), as shown in the size distribution profile at \( x = 2.5 \) mm in Fig. 9(a). It is unknown whether the small scatterers are aggregates of ethylene molecules at supercritical state, nuclei of ethylene droplets, or incipient ethylene droplets at this stage. The non-constant distribution of scattering intensity over the detected \( Q \) range indicates that the scatterers are not in the gaseous state.

As the probing stations move through Zone 2, more scattering signal from big scatterers can be observed in Fig. 8. The scattering intensity in the range 0.004< \( Q <0.3 \), however, is not smooth, indicating a low number density for the big scatterers. Consequently, the size distribution could only be modeled within 0.06< \( Q <0.14 \), to give small scatterers with a size range of 10-20 Å (1-2 nm) in Fig. 9(b). The scattering intensity profile begins to show the \( \text{wavy} \) characteristics of big droplets, similar to that seen in Fig. 7(b), at the nozzle exit plane (\( x = 0 \)) in Fig. 9(c). Still, the size distribution in the low \( Q \) range could not be modeled, due to the low number density of big droplets. Once the ethylene enters Zone 3, the smooth scattering intensity profiles in the low \( Q \) range (\( Q <0.04 \)) indicate the presence of abundant big droplets within the ethylene plume, as demonstrated in Fig. 8. The modeled size distribution at the \( x = 1.4 \) mm location is shown in Fig. 9(d). Droplets between 200 Å (20 nm) and 700 Å (70 nm) were modeled with a high confidence level. The droplet size of 670 Å (67 nm) has the highest volume distribution.
Based on the observations from Figs. 8 and 9, significant droplet nucleation mainly takes place within Zone 3 (after the nozzle exit) of the ethylene jet injected at 293.2 K. A limited number of big scatterers can be found within Zone 2. The creation processes and identity of these big scatterers could not be clearly defined. Similarly, the creation processes and identity of smaller scatterers on the order of 10-20 Å (1-2 nm) in Zones 1 and 2 should also be further explored.

If the appearance of big scatterers is used as the criterion to define the occurrence of droplet nucleation, droplet nucleation probably takes place at a low rate or preferentially along the side wall within Zone 2 and at a high rate immediately downstream of the nozzle exit plane for injection at $T_{\text{inj}}=293.2$ K. The study of Lin et al. [7], using a rectangular injector equipped with diamond windows, observed the preferential distribution of big droplets along the sidewall. Results from SAXS measurements in the radial direction, in an attempt to resolve the preferential size distribution within the small axisymmetric flow passage of the beryllium injector, however, are thus far inconclusive. The possibility that the presence of big scatterers within the final passage is caused by injector vibration during the injection process will be discussed later.

Based on these observations, the feasibility of applying the SAXS technique to explore the properties of condensed phase inside the beryllium injector has been shown.

b) Case #2, $T_{\text{inj}}=286.6$ K:

Scattering intensity profiles at various axial locations and size distributions modeled at selected locations for the injection condition with $T_{\text{inj}}=286.6$ K ($T_r=1.01$) are shown in Figs. 10 and 11, respectively. More liquid condensation is expected for this injection condition, as illustrated in Fig. 1(b). The scattering intensity profiles and modeled size distributions were reasonably modeled. The size distribution in Fig. 13(b) shows that big scatterers, presumably droplets, on the order of 200-1570 Å (20-157 nm), with low volume distributions were reasonably modeled. The droplet size of 670 Å (67 nm) has the highest volume distribution. Please note that the detection limit of the present SAXS measurement setup is 1570 Å (157 nm). It is very likely that even bigger droplets are present in the flowpath for this injection condition but cannot be adequately resolved. A similar scattering intensity profile and size distribution can be observed at the nozzle exit plane ($x=0$), indicating that no significant droplet growth occurs within the final passage region.

As the fluid enters Zone 3, another significant change in the scattering intensity profiles can be observed in Fig. 12. X-ray scattering from small

Slightly larger droplets were modeled at the $x=1.4$ mm location. Droplet populations between 600 Å (60 nm) and 1000 Å (100 nm) were derived. The highest droplet volume distribution is for the droplet size of 920 Å (92 nm). With an early occurrence of droplet nucleation for this injection condition, droplets can potentially grow to a bigger size.

c) Case #3, $T_{\text{inj}}=280.3$ K:

As the injection temperature is further reduced, more liquid condensation is anticipated, as illustrated in Fig. 1(c). Figure 12 shows that the scattering intensity profiles exhibit higher intensity values in the high Q range, with no sign of big scatterers within Zone 1. The corresponding size modeling shows a dominant size population around 30 Å (3 nm) and a second size population around 75 Å (7.5 nm) in Fig. 13(a). The reasons for the increase in the droplet size of the dominant size population from 10-20 Å (1-2 nm) to 30 Å (3 nm) as the injection temperature approaches the critical temperature should be explored in the future. The presence of the second size population was identified by modeling the intensity profile at a Q value as low as 0.02. Creation mechanisms and properties of these 75 Å (7.5 nm) scatterers should be further examined.

At this injection temperature, X-ray scattering from big scatterers generates a significant amount of photons to create a smooth scattering intensity profile in Zone 2, as illustrated in Fig. 12. Judging from the well-defined and significantly different intensity profiles for Zone 1 and Zone 2, a high-rate of droplet nucleation probably takes place at the entrance of the Zone 2. Two ranges of Q values were selected for size modeling within the final passage region. The modeling for a broad range of Q (0.0042<Q<0.2) gives a size distribution similar to the one shown in Fig. 13(a). Only small scatterers of 30 Å (3 nm) and 75 Å (7.5 nm) were clearly identified. In order to identify the size of big scatterers, a narrow Q range (0.0042<Q<0.018) was specifically selected for size modeling. The size distribution in Fig. 13(b) shows that big scatterers, presumably droplets, on the order of 200-1570 Å (20-157 nm), with low volume distributions were reasonably modeled. The droplet size of 670 Å (67 nm) has the highest volume distribution.
scatterers becomes less dominant. Droplet size modeling in the low Q range, shown in Fig. 13(d), however, gives unreliable size distributions, probably due to the detection limit of the present SAXS setup. Possible wavy distribution characteristics at Q<0.0042 are left out.

To summarize, locations with a sudden change in flow passage contour, which promote the fluid expansion process, can initiate the droplet nucleation processes. For the injector contour specified in Fig. 4, with the present SAXS probing range, the contour transition from Zone 1 to Zone 2 creates a small number of big droplets at a high injection temperature. Lowering the injection temperature to close to the critical temperature can significantly increase the number of big droplets, probably through a higher nucleation rate. Variations in scattering distribution profiles within Zone 2 are relatively small, indicating limited droplet nucleation and growth within the final passage of the nozzle. The contour transition from Zone 2 to Zone 3 promotes the generation of big droplets at a higher rate for all three injection temperatures. At x=1.4 mm from the nozzle exit, droplet size varies from 200-700 Å (20-70 nm) at Tinj=293.2 K to a large value beyond the detection limit of the present SAXS setup at Tinj=280.3 K.

**Effects of Injection Temperature:**

The effects of injection temperature on liquid condensation are further illustrated in Figs. 14-16 with a representative axial location for each zone. At x=-3.0 mm (Zone 1), both detected scattering intensity and its associated Q range increase as the injection temperature approaches the critical temperature in Fig. 14(a). The corresponding size distributions show that the size of the small scatterers increases slightly as the injection temperature decreases in Fig. 14(b). Similar to the observation in Fig. 13(b), a second size population with a scatterer size of 60-75 Å (6-7.5 nm) was obtained for the injection condition with Tinj=280.3 K. It is unclear whether the second size population is associated with the preferential droplet nucleation along the injector wall.

At x=-1.0 mm, in Zone 2, scattering at a low Q range (Q<0.02) appears for all three injection temperatures, indicating the presence of big scatterers within the final passage. The injection condition with Tinj=280.3 K generates a smooth scattering profile with a higher intensity level in Fig. 15(a), indicating a higher number for the big scatterers. It seems that the injection conditions with both Tinj=286.6 K and 293.2 K generate a comparable amount of big scatterers, whose sizes, however, could not be reasonably modeled. The corresponding size distributions in Fig. 15(b), using the maximum range of Q for size modeling, are very similar to those in Fig. 14(b). With lower intensity levels in the high Q range, however, the modeled volume distributions in Fig. 15(b) are lower than those in Fig. 14(b) for small scatterers. It is believed that the reduction in volume distribution for small scatterers is compensated for by the presence of big scatterers through the droplet nucleation and growth processes. The modeled size distribution within the low Q range at x= -1.0 mm for the injection condition with Tinj=280.3 K is very similar to those depicted in Figs. 13(b) (x=-1.5 mm) and 13(c) (x=0) and is not shown here.

The scattering profile for the injection condition with Tinj=280.3 K has the lowest intensity values at x=1.0 mm in Zone 3, as seen in Fig. 16(a). This observation contradicts the visual observation of the highly opaque ethylene plume in Fig. 1(c). It is possible that the majority of the scattering signal for this injection condition lies outside the detection limit of the present SAXS setup. Big droplets scatter X-ray photons at small angles that may be blocked by the physical beam stop. The resulting size modeling is, therefore, unreliable. Nonetheless, the characteristic wavy intensity profiles in the low Q range for both Tinj=286.6 K and 293.2 K in Fig. 16(a) can be modeled to resolve the sizes of big scatterers in Fig 16(b). Droplet size at the peak volume distribution varies from 640 Å (64 nm) at Tinj=293.2 K to 820 Å (82 nm) at Tinj=286.6 K. It is likely that the SAXS measurement for the Tinj=286.6 K condition may also be affected by the detection limit of the present SAXS setup.

To reinforce previous observations, Figs. 14-16 show that an injection temperature approaching the critical temperature can initiate the early appearance of big droplets through droplet nucleation processes and can also generate bigger droplets.

**Comparison with Non-Condensing Nitrogen Jet**

In an attempt to resolve the creation processes and identity of the small scatterers with a size on the order of 10-30 Å (1-3 nm), SAXS measurements were made inside the injector for both nitrogen and ethylene jets injected at a temperature of 290.4 K and a pressure of 5.11 MPa. The scattering intensity profiles are shown in Fig. 17. With the critical temperature and critical pressure of 126.3 K and 3.39 MPa for nitrogen, respectively, the injection condition for the nitrogen jet corresponds to a reduced temperature, T_r, of 2.30 and a reduced pressure, P_r, of 1.51. No condensed phase is expected within the injector for this nitrogen jet. For the ethylene jet, the injection condition corresponds to T_r =1.03 and P_r=1.01. Moderate droplet nucleation is expected to occur for this ethylene jet with a jet appearance resembling somewhere between Figs. 1(a) and 1(b).

The scattering intensity profile at x=3.0 mm for the nitrogen jet in Fig. 17(a) exhibits a constant intensity value within the Q range of 0.25-0.03. The
constant intensity profile, which represents a lack of scattering from interfaces, indicates the presence of gaseous molecules. The constant scattering intensity within the high Q range of the scattering profile can also be observed for the ethylene jets outside the injector in Figs. 8, 10, 12, and 17(b), where big scatterers are surrounded by gaseous molecules inside the injection chamber. For the ethylene jets, the non-constant scattering intensity at the high Q range inside the injector points to the existence of small scatterers, which exhibit interfaces with the surrounding medium. Further efforts to identify whether these small scatterers are aggregates of supercritical ethylene molecules, nuclei of ethylene droplets, or incipient ethylene droplets should be carried out in the future.

The source of the detected scattering profile in the low Q range, which is typically contributed by big scatterers, within the final passage of the injector for the nitrogen jet in Fig. 17(a), could not be explained. Condensed phase is not anticipated for this nitrogen jet. Within the final passage of the beryllium injector, the wall thickness is at least twice the diameter of the nitrogen passage within the line of sight of the X-ray beam. Vibration of the beryllium injector during the injection process may shift the compressed and fused S65C beryllium to create certain scattering patterns. The subsequent data reduction may not subtract all undesired background scattering, resulting in scattering intensity profiles not originating from the scatterers in the flow passage. If the hypothesized injector vibration is actually taking place, then the observations that few big scatterers are present within the final passage of the ethylene jets in Cases 1 and 2 may not be valid. This uncertainty highlights the challenges of the SAXS measurement technique. This issue should be further examined in future work.

**SUMMARY**

The structures of condensed supercritical ethylene jets inside an axisymmetric beryllium injector were explored, using the small-angle X-ray scattering (SAXS) technique. The experiment was conducted at the 12-ID-B beamline at the Argonne National Laboratory. Beryllium was selected as the injector material, due to its high X-ray transmittance. Scattering intensity was measured both inside and outside the injector at three injection temperatures. Based on the detected scattering intensity profiles, the size and population of scatterers were modeled. Evolution of droplet nucleation/growth processes along the injector axis was investigated. Findings and questions raised by the present study are summarized below:

1. The feasibility of applying the SAXS technique to explore the properties of condensed phase inside the beryllium injector has been successfully demonstrated.
2. Small scatterers on the order of 10-30 Å (1-3 nm) were observed within the injector. These small scatterers could be aggregates of ethylene molecules at supercritical state, nuclei of ethylene droplets, or incipient ethylene droplets. Creation mechanisms for and identity of these small scatterers could not be confirmed in this study.
3. Scatterers larger than 200 Å (20 nm) and up to 1570 Å (157 nm), which is the detection limit of the present SAXS setup, were observed within the final passage region and mainly outside the injector. These scatterers are believed to be ethylene droplets.
4. The existence of big scatterers within the final passage region at a high injection temperature may be the result of a low-rate of droplet nucleation, preferential nucleation near the injector wall, or injector vibration.
5. The condition with an injection temperature close to the critical temperature promotes the early (upstream) occurrence of droplet nucleation, which allows a longer residence time for droplets to grow. For the present injector contour, big scatterers appear at the entrance of the final passage for an injection temperature near the critical point.
6. For the present injection conditions, big droplets always exist downstream of the nozzle exit, where the discharged ethylene jets undergo the greatest expansion process.

Answers to several open questions are still needed to advance the understanding of droplet nucleation during the injection process. Efforts to address these questions will be carried out in the future. In addition, selection of future test conditions should be based on the detection limit of the SAXS setup. The present study indicates that injection with \( T_{\text{inj}} \) slightly lower than 286.6 K can fully utilize the detection limit of the SAXS setup in 12-ID-B beamline at the Argonne National Laboratory. Efforts to quantitatively resolve droplet volume fraction should also be carried out in the future.

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Figure 1 Photos of supercritical ethylene jets injected from the beryllium injector into a quiescent chamber at various temperatures to illustrate the degree of condensation within the discharged plumes. (a) $T_{\text{inj}}=293.2$ K, (b) $T_{\text{inj}}=286.6$ K, (c) $T_{\text{inj}}=280.3$ K. $P_{\text{inj}}=5.11$ MPa. $P_{\text{chm}}=137$ kPa.

Figure 2 Shadowgraph images of ethylene jets in a rectangular injector equipped with quartz windows. $D=1.0$ mm, $P_{\text{inj}}=5.14$ MPa, $P_{\text{chm}}=0.14$ MPa. Images are from Ref. [3].
Figure 3 Photos of experimental setup in 12-ID-B beamline. (a) Setup around the injection chamber, (b) Detector setup. X-ray is from right to left.

Figure 4 (a) Photo and (b) schematic to illustrate the details of the axisymmetric beryllium injector.

Figure 5 Schematics to illustrate (a) X-ray probing regions and (b) specific regions of interest associated with flow structures and droplet dynamics. x<-2.0 mm for Zone 1; -2.0 mm < x < 0 for Zone 2; x>0 for Zone 3. x=0 at nozzle exit plane.
Figure 6  Typical small-angle scattering image for highly-condensed supercritical ethylene jet.

Figure 7  Measured (green line) and fitted (blue line, overlapped beneath the green line) scattering intensity profile, calculated droplet size distribution (green bar with red border, appearing as red column), and the calculation residual (red dots) at x= 2.0 mm for the supercritical ethylene jet injected at T_{ij}=286.6 K. (a) A broad range of scattering intensity profile selected for droplet modeling, (b) a narrow range of scattering intensity profile with wavy scattering characteristics of big droplets selected for droplet modeling.
Figure 8  Scattering intensity profiles for ethylene jet at various axial locations. $T_{ij}=293.2$ K, $P_{ij}=5.11$ MPa, $r=0$.

Figure 9  Modeled size distributions for ethylene jet at various axial locations. $T_{ij}=293.2$ K, $P_{ij}=5.11$ MPa, $r=0$. (a) Zone 1, $x= -2.5$ mm, (b) Zone 2, $x=-1.5$ mm, (c) nozzle exit, $x=0$, (d) Zone 3, $x=1.4$ mm.
Figure 10  Scattering intensity profiles for ethylene jet at various axial locations. $T_{\text{inj}}=286.6$ K, $P_{\text{inj}}=5.11$ MPa, $r=0$.

Figure 11  Modeled size distributions for ethylene jet at various axial locations. $T_{\text{inj}}=286.6$ K, $P_{\text{inj}}=5.11$ MPa, $r=0$. (a) Zone 1, $x=-2.5$ mm, (b) Zone 2, $x=-1.5$ mm, (c) nozzle exit, $x=0$, (d) Zone 3, $x=1.4$ mm.
Figure 12  Scattering intensity profiles for ethylene jet at various axial locations. $T_{\text{inj}}=280.3 \text{ K}$, $P_{\text{inj}}=5.11 \text{ MPa}$, $r=0$.

Figure 13  Modeled size distributions for ethylene jet at various axial locations. $T_{\text{inj}}=280.3 \text{ K}$, $P_{\text{inj}}=5.11 \text{ MPa}$, $r=0$. 
(a) Zone 1, $x= -2.5 \text{ mm}$, (b) Zone 2, $x=-1.5 \text{ mm}$, (c) nozzle exit, $x=0$, (d) Zone 3, $x=1.4 \text{ mm}$.
Figure 14  (a) Scattering intensity profiles and (b) modeled size distribution for ethylene jets injected at three temperatures. $x= -3.0$ mm, $P_{inj}=5.11$ MPa.

Figure 15  (a) Scattering intensity profiles and (b) modeled size distribution for ethylene jets injected at three temperatures. $x= -1.0$ mm, $P_{inj}=5.11$ MPa.
Figure 16  (a) Scattering intensity profiles and (b) modeled size distribution for ethylene jets injected at three temperatures. $x=1.0$ mm, $P_{inj}=5.11$ MPa.

Figure 17  Scattering intensity profiles for (a) nitrogen and (b) ethylene at four axial locations inside the injector. $T_{inj}=290.4$ K, $P_{inj}=5.11$ MPa, $r=0$. 