



Non-intrusive characterization of heat transfer fluid aerosol sprays released from an orifice

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Abstract

Organic heat transfer fluids (HTFs) are widely used because of their low vapor pressures and wide ranges of working temperatures. It is a common misconception that operations involving HTFs below their flash point temperatures do not represent a hazard, but aerosols of combustible liquids at temperatures below their flashpoints have been recognized for their potential to cause fires and explosions. The hazard is further exacerbated by the paucity of experimental information about the formation and behavior of aerosols from HTF releases. This article presents droplet sizes, spray development, and aerosol formation distances for an alkylated aromatic HTF aerosol generated mechanically by means of a leak from a pressurized container into ambient air as functions of HTF temperature, pressure, and orifice size. A Malvern Laser Diffraction Particle Analyzer was used to measure aerosol droplet sizes along the centerline of the liquid sprays, and high-speed photography was used for spray visualization and interpretation of stream atomization. Effects of pressure, temperature, and orifice diameter confirm critical Weber Number theory. © 2001 Elsevier Science Ltd. All rights reserved.

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1. Introduction

Heat transfer fluids (HTFs) are synthetic hydrocarbon liquids with high boiling points and high molecular weights. Extensively used in the process industries and available in a wide range of properties, they are generally assumed to be safe for use in the plant, because they are very stable at room temperature and usually have high flash point temperatures. However, Factory Mutual Engineering and Research has reported 54 fires and explosions and \$150 million losses due to fires involving HTFs during a recent 10-year period (Febo & Valiulis, 1995). Because of their high boiling and flash points, industrial personnel often assume that HTFs are non-flammable and non-combustible. In fact, the vapors of

these fluids are flammable above their flash points and can cause fires and explosions.

To prevent fires and explosions due to releases of vapor from HTFs used at high temperatures, the hot HTFs are operated at elevated pressures to increase the boiling temperatures. If loss of containment does occur, however, the liquid will then leak under pressure and may disperse as an aerosol, accumulate in the plant, encounter an ignition source, and explode. The potential hazards of combustible fluid aerosols have been recognized as early as 1955 by Jacob Eichhorn, who reports earlier research on the flammability of combustible liquids (Eichhorn, 1955). Aerosol explosion hazards are discussed also by other researchers (Febo & Valiulis, 1995; Vincent & Howard, 1976; Bowen & Shirvill, 1994; Richer, Swithenbank, & Wedd, 1994).

Although it has been recognized that aerosols can explode, research is critically needed to measure their behavior and relate this behavior to their flammability. This article reports research on the spray development

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and droplet sizes of HTF aerosols from process leaks. Measurement techniques used in this research were based on methodology of earlier experiments on fuel sprays for automotive research (Kihm, Terracina, Payne, & Caton, 1994).

2. Experimental details

2.1. Samples

The HTF used in this research was an alkylated aromatic mixture with an average molecular weight of 320, a normal boiling point of 351°C, and a flash point of approximately 175°C. This HTF was used as received, without purification or analysis, from an industrial source.

2.2. Apparatus

The experimental system, as shown in Fig. 1, consists of a HTF cell, positioning system, exhaust system, and a Malvern Laser Diffraction Particle Analyzer, Model 2600 (Malvern), which is based on the Fraunhofer diffraction principle with the diffraction angle inversely proportional to the diameter of a spherical droplet (Barth, 1984), as shown schematically in Fig. 2. The HTF cell is made of stainless steel and has a capacity of 1000 ml with inside dimensions of 10 cm diameter and 13.3 cm depth. Orifices to simulate process leaks were prepared by drilling brass plugs and measuring with a microscope. The orifice diameters for the data reported here are 0.21 ± 0.01 mm and 0.38 ± 0.02 mm.

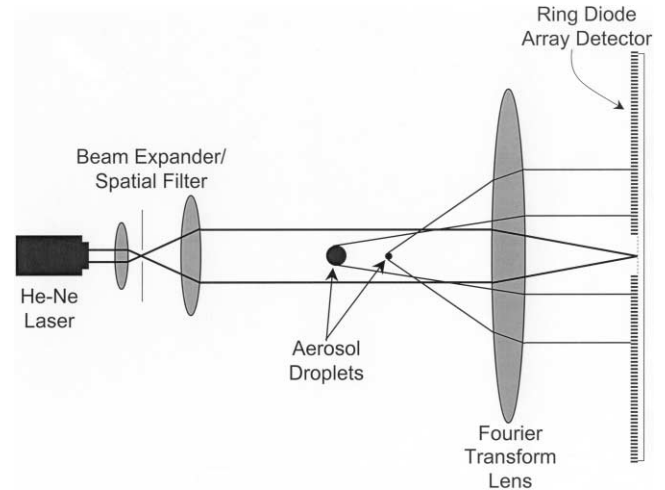


Fig. 2. Laser diffraction to measure spherical droplet sizes.

Because the Malvern apparatus is fixed, the orifice is moved for measurements along the horizontal line (x direction, the spray axis) using the positioning system, which can move the HTF cell in two orthogonal directions, x and y . Direction x allows measurements of droplet sizes at distances horizontally from the orifice, and direction y allows measurements of droplet sizes in the radial direction from the horizontal or the centerline of the liquid sprays. Only measurements along x are reported here.

The exhaust system consists of a grounded collection chamber and a mist separator, which removes the vapor from the liquid to a particle size of 5 micron. To prevent accumulation in the laboratory, the vapor and aerosol phases are pulled through a polyester felt filter by an

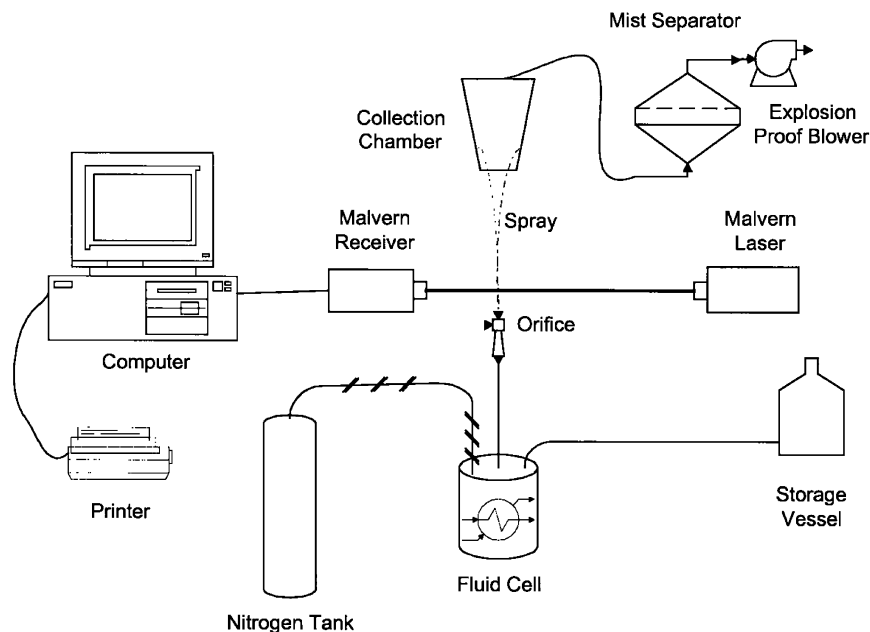


Fig. 1. Experimental aerosol apparatus.

explosion proof blower, which is located outside the building.

The Malvern includes a detector and a 2 mW Helium–Neon laser tube, which generates a collimated and monochromatic beam of red light, centered at 632.8 nm, and a series of optics produces an expanded beam of 5.0 mm diameter. When drops pass through this beam, they diffract or scatter the laser light with the scattering angle inversely proportional to their size. The diffracted light falls on a series of 31 ring diodes, each ring detects a specific range of droplet sizes, and a portion of the light that is not diffracted passes through the center pinhole, where a diode detects the measurement obscuration. The diode signals from the scattered laser light intensities are converted by a computer into drop-size data. A schematic of laser light diffraction is shown in Fig. 2. Detailed Malvern measurement techniques and their applications are discussed elsewhere (Kihm & Chigier, 1991).

2.3. Experimental procedure

The HTF cell contains and controls the temperature of the test HTF, which is pressurized by nitrogen gas and propelled through an orifice. A calibrated, strain gauge pressure transducer measures the pressure at the orifice with an overall uncertainty of ± 12 kPa. In addition, the liquid temperature at the orifice is measured by a calibrated thermocouple with an overall uncertainty of $\pm 1.9^\circ\text{C}$. The pressure transducer and thermocouple are installed as closely as possible to the orifice for accurate measurements of the liquid properties entering the orifice.

Each sample measurement is based on averaged data from 500 sweeps of diode measurements per test run. The data are analyzed by the Malvern software using the Rosin–Rammler model based on the Sauter Mean Diameter (SMD), which is the ratio of the total volume to the total surface area of the sampled drops (Kihm & Chigier, 1991; Lefebvre, 1989; Richer et al., 1994). SMD data are used because they are needed for aerosol dispersion models and provide important information for droplet evaporation, condensation, or heat transfer among droplets and ambient media.

The uncertainty of the SMD measurements of droplet sizes for this study is estimated to be ± 5 micron, assuming spherical drops. The uncertainty of particle sizes in the transitional regions that include liquid ligaments and drops can be much higher, because the Malvern operating principle performs most accurately for spherical drops. To help interpret the diffraction laser measurements and visualize the formation of aerosols from the initial breakup of the liquid streams to the fully developed regions, the sprays were photographed and compared with the Malvern size-distribution data. Some of the spray photographs are shown in Figures 3–6. More

detail about the experimental procedure is available elsewhere (Sukmarg, 2000).

3. Experimental results

3.1. Spray development

This research investigated aerosol formation from an orifice that simulates a leak in a pressurized container and the formation of a liquid stream or jet into air at ambient temperature. Encountered are several stages of jet disintegration (Elkotb, 1982) and atomization to form aerosols (Lefebvre, 1989). The jet is influenced by outside disturbances such as the friction between the jet surface and the surrounding air causing instability of the jet. In its initial stage, the jet oscillates and later disintegrates into ligaments and/or large drops, because the energy from the disturbances is greater than the surface energy of the jet. Under certain conditions, the ligaments and large drops break up further until they reach their stable sizes of fully developed aerosols. Ohnesorge's research on atomization regimes showed, for example, that higher jet velocities result in smaller aerosol droplet sizes (Ohnesorge, 1936).

The atomization regimes therefore consist of the liquid stream regime, the transitional regime, and the developed spray regime, as shown in the photographs of Figures 3–6. For increasing pressures from Fig. 3 to Fig. 4, the liquid jets have higher velocities, which increase the friction between the liquid surface and the surrounding air, resulting in greater instability and more effective stream break up into aerosol droplets and smaller SMDs. With increasing liquid temperatures from Fig. 4 to Fig. 5, the breakup into droplets is more rapid and the droplet sizes are smaller because of reduced liquid surface tension and viscosity. With the condition of both higher pressures and liquid temperatures from Fig. 5 to Fig. 6, the breakup is highly effective to generate fine droplets, and this high level of atomization results in the greatest amounts of aerosols.

Stream atomization can be characterized by the dependence of droplet diameter on the droplet Weber number, We_D , which relates the shear forces that contribute to the stream breakup to the surface tension forces holding the stream together. $We_D = [(\rho_g V^2 D) / \sigma]$, where ρ_g is the air density, V is the jet velocity, D is the droplet diameter (SMD in this work), and σ is the fluid surface tension. High Weber numbers indicate that shear forces dominate surface tension forces and the stream is unstable and is breaking up. Low Weber numbers indicate that surface tension forces dominate, and the stream is stable because either the velocity is too low to induce breakup or much of the stream has already atomized into stable droplets. The critical Weber number range between the stable stream or droplet region and the atomization region was

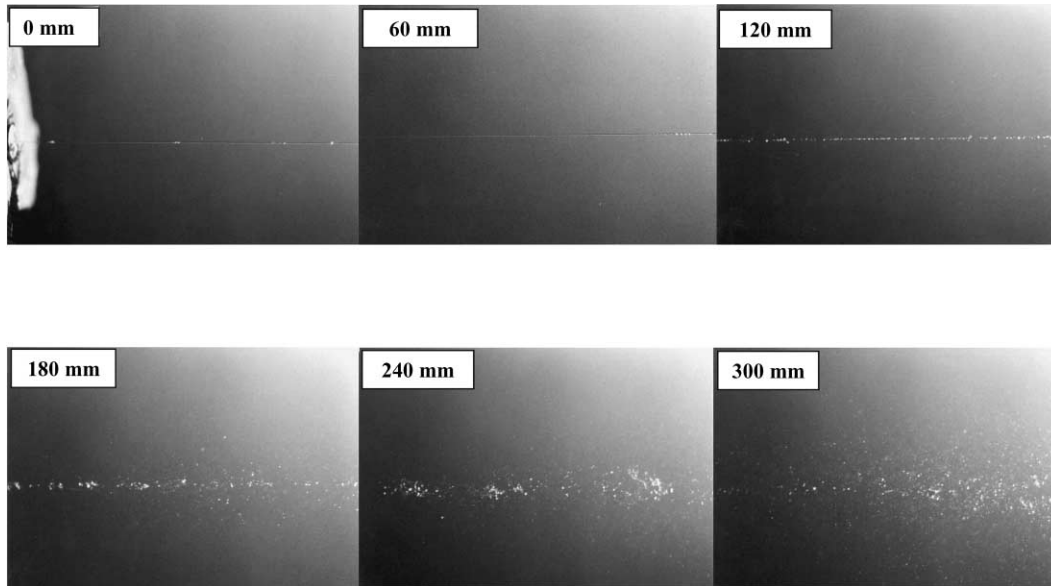


Fig. 3. Photographs of an alkylated aromatic HTF spray at various distances from the orifice at 1,135 kPa (150 psig), 80°C; 0.21 mm orifice.

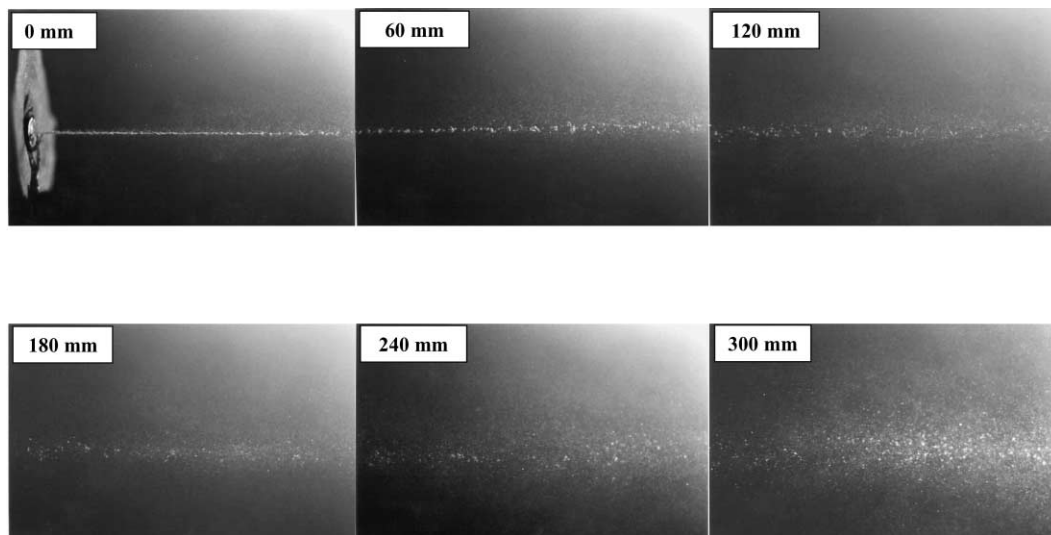


Fig. 4. Photographs of an alkylated aromatic HTF spray at various distances from the orifice at 2,170 kPa (300 psig), 80°C; 0.21 mm orifice.

estimated to be 12 to 22 by Johnson and Woodward (1998).

The droplet Weber number as functions of droplet SMD for various conditions of pressure and temperature is shown in Fig. 7 for an orifice diameter of 0.21 mm and in Fig. 8 for an orifice diameter of 0.38 mm. The Weber number values in Fig. 7 and Fig. 8 were determined using injection velocities calculated from Bernoulli's equation and the coefficient of discharge for each orifice. Three regions of critical Weber number ranges, spanning from 8–15 for the reported data, are outlined in each figure by dashed lines and designated by A, B, and C. The difference between these Weber values and those reported by Johnson and Woodward (1998) may be due in part to the differences between the calculated

or estimated droplet velocities and actual injection velocities.

The critical Weber number ranges with the corresponding aerosol formation distances are listed in Table 1. Region A has low Weber numbers and represents a relatively stable stream formed at low injection pressures, which did not effectively atomize the jet. Region B has higher Weber numbers and represents an unstable stream that is being atomized to smaller ligaments and droplets. Region C has low Weber numbers and represents the fully developed aerosol with relatively stable droplets.

Characterization of these jet regimes in this research was based on a composite analysis of the spray SMD trends, spray photographs, and the droplet sizes obtained

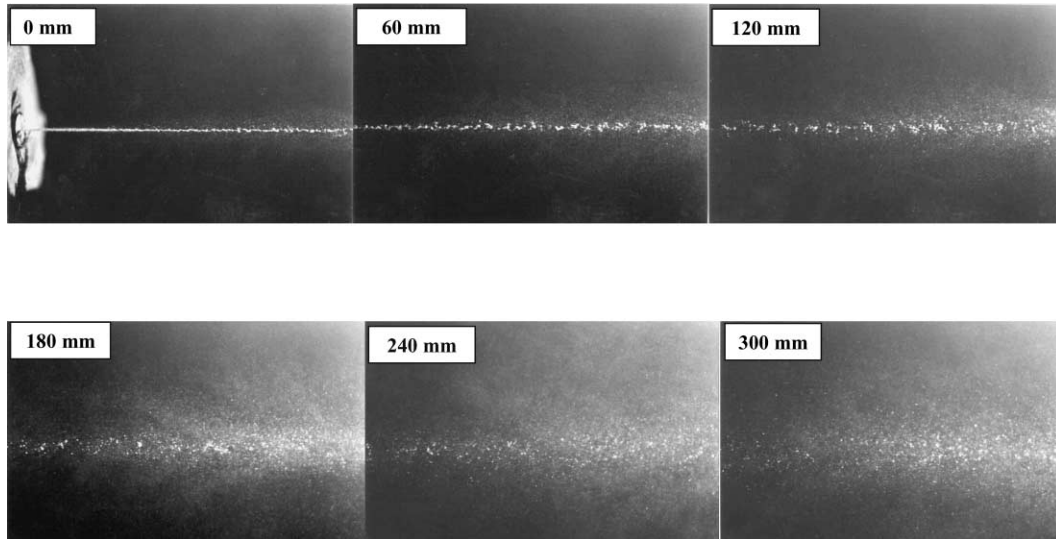


Fig. 5. Photographs of an alkylated aromatic HTF spray at various distances from the orifice at 2,170 kPa (300 psig), 120°C; 0.21 mm orifice.

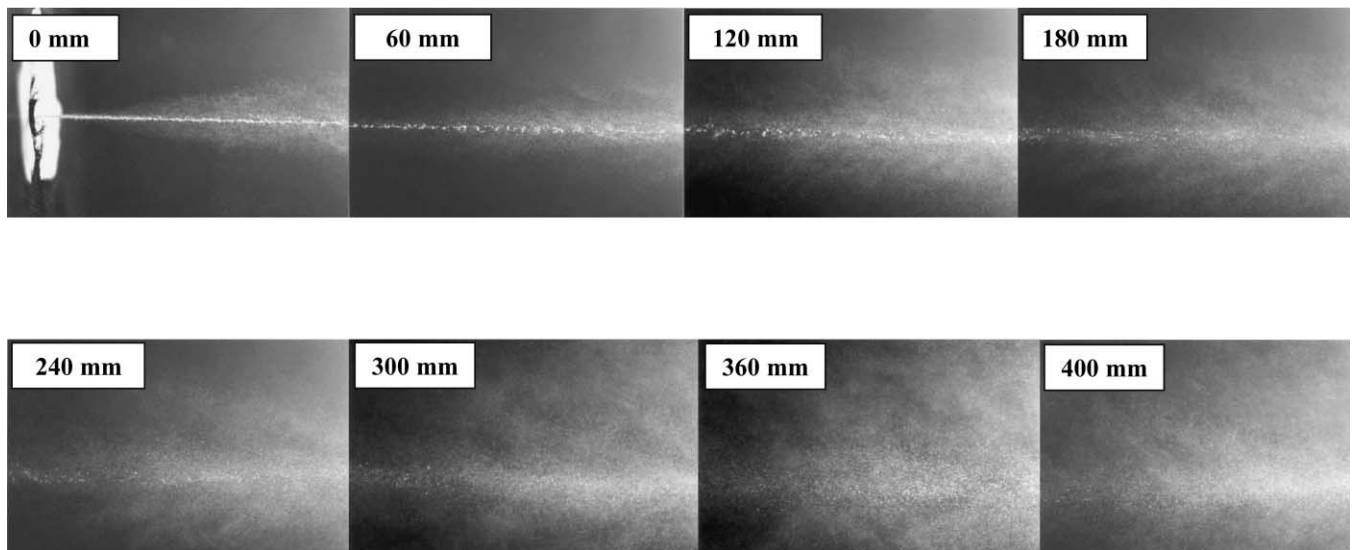


Fig. 6. Photographs of an alkylated aromatic HTF spray at various distances from the orifice at 3,550 kPa (500 psig), 150°C; 0.21 mm orifice.

from the Malvern measurements using the Rosen–Rammler distribution. The presence of ligaments and large drops in the developing region biases the measurements, and the peak of the Rosen–Rammler distribution often exceeds the available dynamic range. In addition to the Weber number, an indication of the developed spray region is that the spray SMD remains nearly constant. Also the resulting SMD spectra exhibit a Rosen–Rammler distribution with its peak included within the dynamic range of the instrument (5.4–540 μm). The developed spray regime represents the region where the Malvern measurements of spherical drops are considered accurate to within ± 5 micron.

3.2. Droplet size measurements

To measure aerosol formation behavior, experiments for an alkylated aromatic heat transfer fluid were conducted at three pressures (1,135, 2,170, and 3,550 kPa), at liquid temperatures ranging from 80 to 190°C, and with two orifice sizes of 0.21 and 0.38 mm in diameter. Table 1 summarizes the tested atomization (or leaking) conditions where the spray SMD measurements were made along the horizontal spray axis at distances up to about 450 mm from the orifice. This table also presents the distance to a developed spray region, with a critical Weber number range for the adjacent transition region.

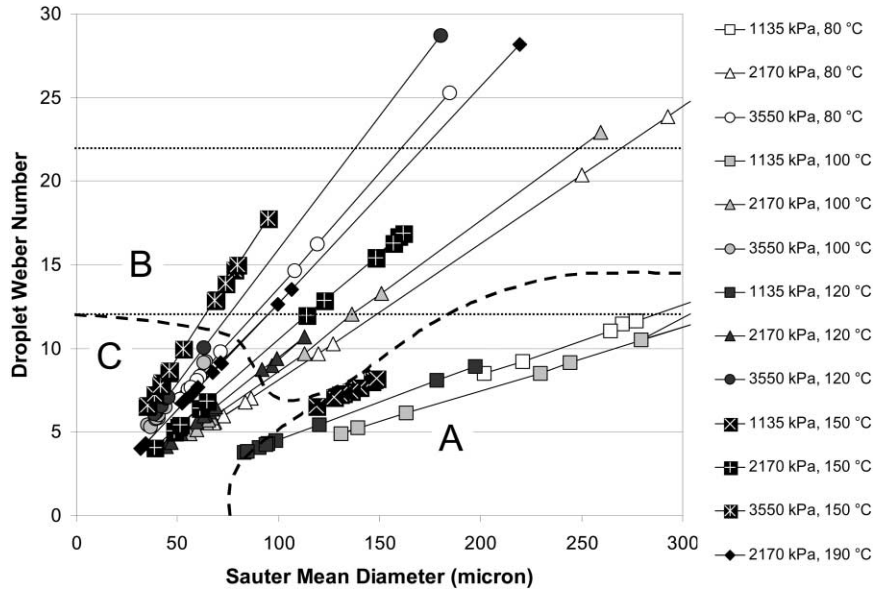


Fig. 7. Weber Number as a function of SMD; 0.21 mm orifice.

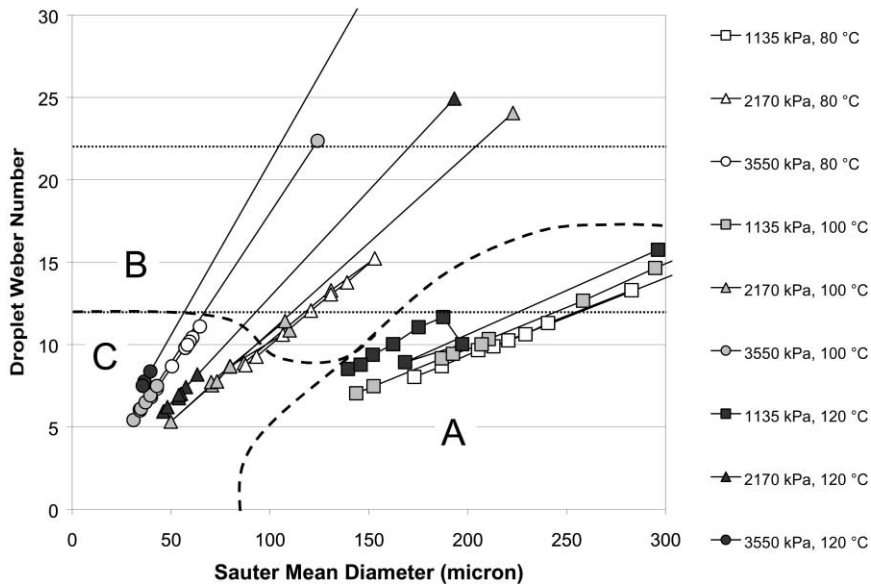


Fig. 8. Weber Number as a function of SMD; 0.38 mm orifice.

The developed aerosol region was determined using high-speed visualization of sprays and the Malvern droplet size data analysis, as discussed in the previous section.

The developed spray formation distances from the 0.21 mm orifice decreased from greater than 450 mm to 150 mm as liquid temperature and injection pressure were increased. The dependence of developed spray distance on the orifice size shows increasing distances for the low pressure injections of 1,135 kPa but somewhat decreasing distances for the high pressure injections of 3,550 kPa. Further detailed measurements will be necessary to examine the dependence of the fully-developed

spray distances on the orifice size. Mean droplet size data were not accurately achieved for non-developed sprays consisting of liquid streams and nonspherical ligaments produced at the lowest pressure (1,135 kPa) and 80°C with the 0.21 orifice and for the lowest pressure and all three temperatures with the 0.38 mm orifice.

The experiments demonstrated that the lowest pressure of 1,135 kPa at either orifice size produced the smallest amounts of aerosol, as shown in Fig. 3. Because the low pressure sprays are composed primarily of nonspherical ligaments, the Malvern data analysis, which is based on spherical drops, does not yield accurate droplet sizes or SMDs. The experiments conducted at higher

Table 1
Aerosol formation distance from the orifice

| Orifice size (mm) | Injection pressure (kPa) | Temperature (°C) | Distance ^a (mm) | Weber number ^c |
|-------------------|--------------------------|------------------|----------------------------|---------------------------|
| 0.21 | 1,135 | 80 | >450 | ^d |
| | | 100 | >450 | ^d |
| | | 120 | 250 | 8–10 |
| | | 150 | 200 | 8–12 |
| | 2,170 | 80 | 350 | 9–11 |
| | | 100 | 300 | 9–12 |
| | | 120 | 250 | 9–11 |
| | | 150 | 200 | 12–15 |
| | | 190 | 250 ^b | 12–15 |
| | 3,550 | 80 | 300 | 10–14 |
| | | 100 | 200 | 9–10 |
| | | 120 | 200 | 10–12 |
| 150 | | 150 | 10–14 | |
| 190 | | 100 | 10–14 | |
| 0.38 | 1,135 | 80 | >450 | ^d |
| | | 100 | >450 | ^d |
| | | 120 | >450 | ^d |
| | 2,170 | 80 | 350 | 9–11 |
| | | 100 | 250 | 12–15 |
| | | 120 | 150 | 9–15 |
| | 3,550 | 80 | 100 | 12–15 |
| | | 100 | 100 | 9–15 |
| | | 120 | 100 | 9–15 |

^a Distance from the orifice to the developed aerosol region.

^b Bias to larger drop sizes due to the laser beam steering effect through HTF vapor.

^c Range of critical Weber number.

^d Developed aerosol range is beyond the measured range.

pressures, 2,170 and 3,550 kPa, demonstrated spray regions with larger amounts of aerosol for each orifice size at various distances from the orifice, as shown in Figures 4 to 6.

Figure 9 shows the effect of liquid temperature on liquid stream atomization at 1,135 kPa for an orifice size of 0.21 mm. As mentioned above, the droplet size distribution at this low pressure is not fully developed, particularly at low temperatures, so the trend of size measurement using the Malvern with an increase in temperature only approximately follows the expected trend of a decrease in the liquid particle sizes with reduced liquid surface tension and viscosity. Also, there is a general

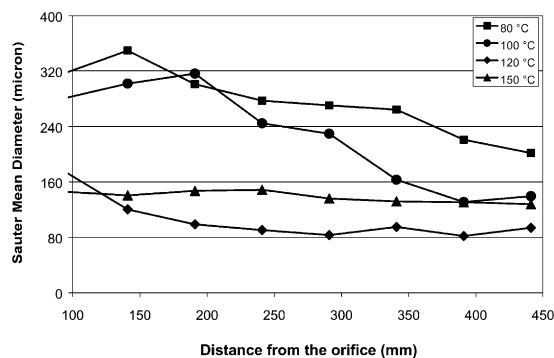


Fig. 9. Effect of liquid temperature on atomization of an alkylated aromatic HTF at 1,135 kPa; 0.21 mm orifice.

trend toward increased stream breakup and smaller particle sizes with larger distances from the orifice. Developed aerosol formation distances varied from >450 mm at 80°C to 200 mm at 150°C. Stream breakup at greater distances from the orifice may be considered less hazardous, because a leaking stream could collect or be deflected into pools and not be so widely distributed as an aerosol.

Higher pressures produce higher liquid velocities with increased shear forces between the liquid surface and the surrounding air and consequently increased atomization to aerosols. The experiments at the higher pressures confirmed the formation of aerosols for the orifice size of 0.21 mm, as shown in Fig. 10. (The lines in this figure are to aid the eye, not fits to the data.) This figure shows also the trend of smaller droplet sizes with increasing liquid temperature, increasing pressure, and increasing distance from the orifice.

With respect to the trends with liquid temperature of droplet size and formation distance, the 2,170 kPa spray at 190°C exhibited larger SMDs and larger formation distance than expected, because at this high temperature, compared to the flash temperature for this HTF of ~173°C, large amounts of vapor with a significant change in the refractive index compared to air cause a shift in the laser beam and a bias (or “beam-steering” bias) in the measured droplet sizes. Though this bias is noticeable, it is not large with respect to the overall SMD

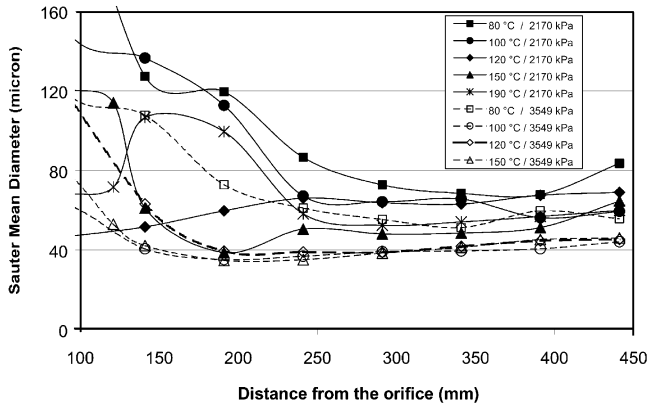


Fig. 10. Effect of liquid temperature on atomization of an alkylated aromatic HTF at 2,170 and 3,550 kPa; 0.21 mm orifice.

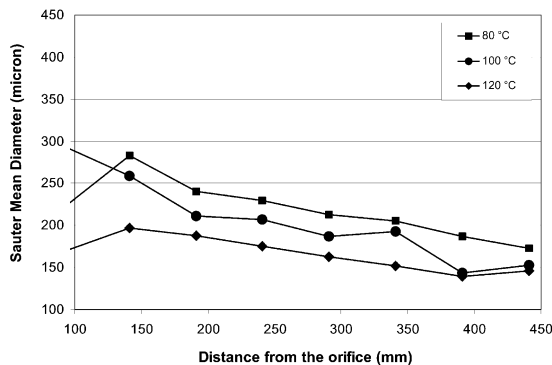


Fig. 11. Effect of liquid temperature on atomization of an alkylated aromatic HTF at 1,135 kPa; 0.38 mm orifice.

measurement uncertainty of ± 5 micron. The SMD values in the developed aerosol region for these conditions are mostly within 30 to 70 micron.

With the larger orifice size, 0.38 mm, larger aerosol droplet sizes were measured, but the trends over pressure and liquid temperature were similar to the trends with the 0.21 mm orifice size, as shown in Fig. 11 and Fig. 12 and summarized in Table 1. Developed aerosol formation distances for the 0.38 mm orifice at 1,135 kPa

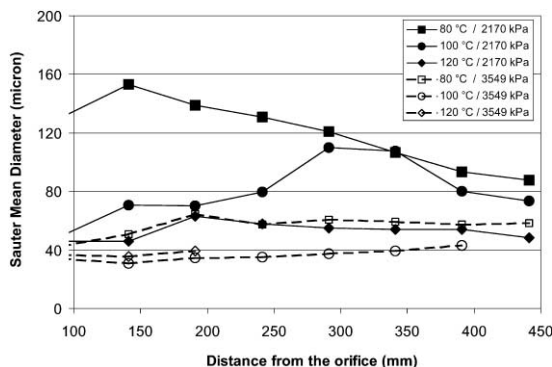


Fig. 12. Effect of liquid temperature on atomization of an alkylated aromatic HTF at 2,170 and 3,550 kPa; 0.38 mm orifice.

were >450 mm and generally larger than the distances for the 0.21 mm orifice. At the higher pressures, however, the aerosol distances were unexpectedly at the same distance (350 mm for 2,170 kPa at 80°C) or smaller distances (100 mm to 250 mm). At the pressure of 3,550 kPa, the measurements at 120°C were conducted up to only 190 mm from the orifice, because high obscuration, due to the multiple scattering from the dense aerosols, resulted in a bias to smaller SMD values. At 3,550 kPa and 100°C, the obscurations beyond 390 mm from the orifice were too large (greater than 70%) and the data accuracy was reduced, so the measurements were conducted only up to this distance.

4. Conclusions

This research demonstrates that for orifice sizes of 0.21 and 0.38 mm, significant amounts of aerosols are produced by HTF released into ambient air at high pressures and at temperatures below the HTF flash points. Several stages of stream breakup yield various amounts of aerosol droplets at various distances from the orifice depending on experimental conditions.

The aerosol droplet sizes at 2,170 kPa, and 3,550 kPa ranged from 30 to 70 micron with smaller droplet sizes measured at higher liquid temperatures, as expected, with lower viscosities and surface tensions. At higher pressures there are greater shearing forces and stream breakup results in the formation of smaller droplets. Aerosol droplet sizes are larger for the larger orifice sizes, and higher pressures and temperatures produce larger amounts of aerosol. Temperatures above the HTF flash point increase the hazard because of the potential for a vapor explosion.

As summarized in Table 1, the aerosol formation distances ranged from 150 to >450 mm (6" to >18 ") depending on conditions with the formation distances decreasing with increasing pressure and liquid temperature. The critical Weber number ranges for the aerosol formation distances (8–15) were consistent with but somewhat lower than the estimated values of critical Weber numbers (12–22) reported by Johnson and Woodward (1998). Aerosol formation distances can help guide the placement of guard surfaces or coalescing devices that could mitigate the formation of aerosols.

5. Future research

This research indicates conditions at which HTF aerosols can be formed with the aerosol drop-size distributions. Measurements for a variety of industrial HTFs are needed over the ranges of operating conditions for a variety of orifice sizes to develop experimentally extrapolated models of aerosol formation behavior.

Aerosol behavior models can be used to help predict the conditions, such as injection pressure, liquid temperature, density, viscosity, surface tension, orifice size, and distance from the orifice for formation of HTF aerosols. In addition, dispersions can be simulated in spaces with barriers to determine temporal and geometric variations of aerosol concentrations in air. This information is needed also for strategies to abate or prevent the formation of aerosols using internal additives or external guards. Finally, information about aerosol flammability as a function of drop-size distributions and HTF properties can be used to determine the upper and lower explosive limits of HTF aerosol/air mixtures.

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