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Understanding the Formation of Heat Transfer Fluid Aerosols in Air

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ABSTRACT

Mist or aerosol explosions present a serious hazard to process industries. Heat transfer fluids are widely used in the chemical process industry, are flammable above their flash points, and can cause explosions. Though the possibility of aerosol explosions has been widely documented, knowledge about the explosive potential of such aerosols is limited. Studying the formation of such aerosols by emulating leaks in process equipment will help define a source term for aerosol dispersions and aid in characterizing their explosion hazards.

Current research by the Mary Kay O'Connor Process Safety Center involves the non-intrusive measurement of such aerosol sprays using a Malvern Instrument Diffraction Particle Analyzer. Predictive models relating the aerosol formation distances, aerosol droplet sizes, and volume concentrations to bulk liquid pressures, temperatures, fluid properties, leak sizes and ambient conditions are developed. These models will be used to predict the conditions under which leaks will result in the formation of aerosols and ultimately help in estimating the explosion hazards of heat transfer fluid aerosols. Important information can be gleaned about the effects of various fluid properties on aerosol formation behavior. The goal is to provide information that will help improve process safety in industry.

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ABSTRACT

Heat transfer fluids are widely used in the chemical process industry, are flammable above their flash points, and can cause explosions. Though the occurrence of aerosol explosions has been widely documented, knowledge about the potential hazards of heat transfer fluid aerosols is limited. Studying the formation of these aerosols by emulating leaks in process equipment can yield a source term for aerosol dispersions and aid in characterizing their explosion hazards.

Current research by the Mary Kay O'Connor Process Safety Center involves the non-intrusive measurement of heat transfer fluid aerosol sprays using a Malvern Laser Diffraction Particle Analyzer. A predictive correlation relating aerosol droplet sizes to bulk liquid pressures, temperatures, fluid properties, leak sizes, and ambient conditions is described. This correlation will be used to predict the conditions under which leaks will result in the formation of aerosols and ultimately help in estimating the explosion hazards of heat transfer fluid aerosols. Important information can be derived from this correlation concerning the effects of various fluid properties on aerosol formation behavior. The goal is to provide information that will help improve process safety in industry.

Introduction

Accidents in the chemical industry almost always result in the loss of containment. Escaping fluids are released into the surroundings in the form of a liquid, a vapor, or both. Depending on the conditions, liquid releases may atomize to form an aerosol, which is a dispersion of liquid droplets in air. These droplets have the potential to disperse over a larger area than the bulk liquid. A potential problem arises when a combustible liquid is atomized. It is a common misconception that flammable liquids are safe below their flash points (Febo and Valiulis, 1995). Aerosols of flammable liquids at temperatures well below their flash points, however, can be as explosive as vapor-air mixtures.

Heat transfer fluids (HTFs) are high flash point synthetic liquids that are omnipresent in the chemical process industry. They are available in a wide range of properties and are used over a wide range of conditions. HTFs are generally considered benign below their flash points, but they are used at high pressures and upon leaking have the potential to form aerosols.

The main processes associated with the formation of aerosols from leaks are not well documented and there is a lack of knowledge about the droplet sizes that result from leakages under various conditions. This aerosol formation information is critically needed for the study of aerosol flammability and to develop measures to prevent aerosol formation with industrial uses of heat transfer fluids.

Experimental Methodology

An objective of this research is to study the atomization characteristics of heat transfer fluids. An experimental approach employing the Malvern Laser Diffraction Particle Analyzer (LDPA) for a non-intrusive analysis technique measures the formation distances, drop size distributions, and volume concentrations of aerosols created by plain orifice atomization to emulate leaks in a process system.

Figure 1 shows a schematic of the experimental apparatus, which consists of a pressurized fluid cell and delivery system, a spray collection and exhaust system, and the LDPA.

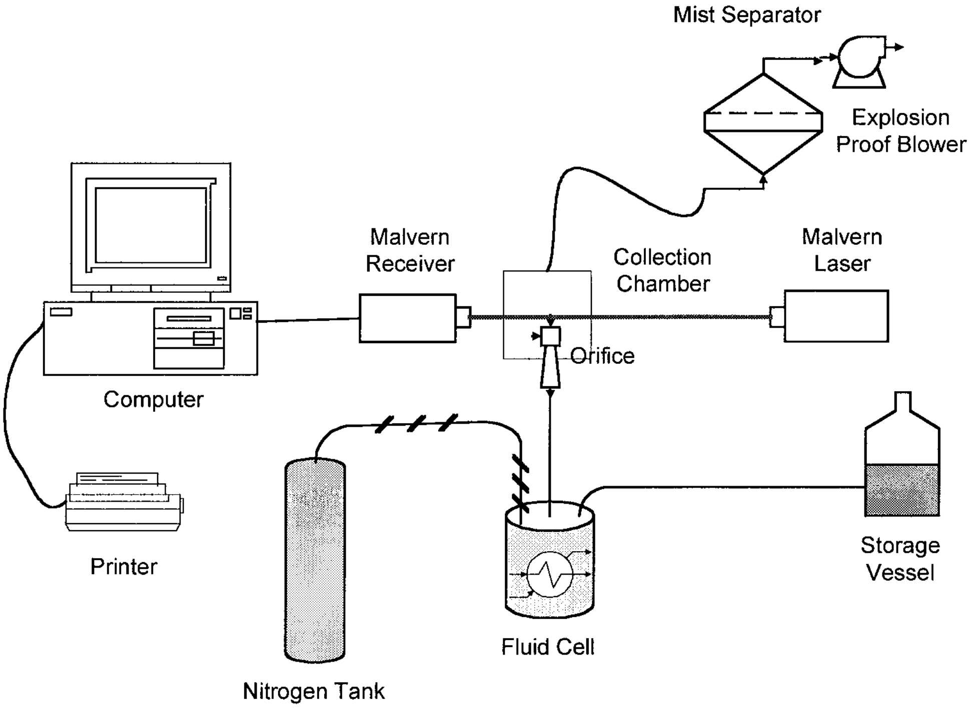

Figure 1. Experimental Aerosol Apparatus

The fluid cell is a 5.9 liter aluminum cylinder with an internal diameter of 17 cm and a height of 36 cm. To simulate leaks, orifices are drilled into brass plugs. The positioning system comprises of two precision rails for the X and Y directions. The X direction represents the direction along the centerline of the spray from the orifice, and the Y direction represents the radial direction perpendicular to the X direction. The exhaust system includes a collection chamber for the spray and a mist separator that removes aerosol droplets above 5 micron size and also includes an explosion proof blower to remove the vapor and residual aerosol phases.

The LDPA consists of a 2mW Helium-Neon laser tube and a ring diode detector. The laser beam is a collimated monochromatic beam of wavelength 780-662 nm and 1.8 mm in diameter. When the aerosol droplets pass through this beam, they diffract the light by amounts inversely proportional to the droplet size (Barth, 1984). This diffracted light falls on 31 concentric ring diodes with each ring detecting a specific size range of droplets. The light intensities on each of these diodes are converted into droplet size data. The LDPA measurement technique for aerosols has been widely studied in the area of automotive fuel spray combustion (Kihm et al., 1994).

The HTF in the fluid cell is heated to the required temperature using heating tape and is pressurized with nitrogen gas to propel it through the orifice. Measurements of the HTF temperature and pressure are made as close to the orifice as possible. Upon pressurization the heated HTF is sprayed through the laser beam and into the collection chamber. Each measurement of the drop size distribution is an averaged value of 500 diode sweeps. The uncertainty in the measurement of aerosol droplet sizes by the LDPA is estimated to be ± 5 micron. Photographs of the sprays at various conditions help to interpret the droplet size data in the aerosol formation or transition regions. The fragmenting liquid produces a variety of non-spherical shapes in these transition regions, and here the LDPA droplet sizes, which are calculated assuming spherical droplets, are not realistic.

Experimental Observations

The focus of this research has been to emulate the formation of an aerosol from a HTF leak in an industrial process and to study the effects of process operating conditions and the leak size on the fluid atomization behavior. The process conditions studied were the HTF temperature and pressure along with the leak size. A qualitative analysis was performed by Sukmarg et al. (2001), and the results of this research exhibit similar trends. At higher temperatures, liquids have lower densities, lower viscosities, and lower surface tensions and hence the effect of temperature on aerosol droplet size is indirect. Pressure has a direct influence on the atomization. Higher injection pressures increases the liquid velocity, which hastens the atomization to form smaller droplet sizes at shorter aerosol formation distances from the orifice.

Data Analysis

Atomization is the process by which aerosol is produced. Here, atomization is induced by forcing the liquid through a small orifice to emulate a leak in a process system. The liquid stream upon leaving the orifice is destabilized by the friction forces between the air and the liquid surface. These aerodynamic forces create surface disturbances, which can be large enough to overcome the stream surface energy and cause stream break-up. This break-up results in the formation of non-spherical segments known as ligaments, which break further until they are small enough to form stable spherical droplets (Lefebvre, 1989, Bayvel and Orzechowsky, 1993).

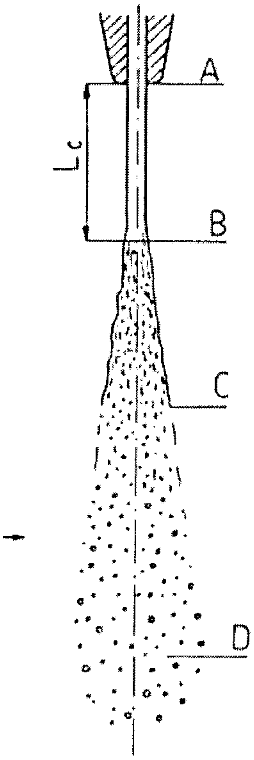

Figure 2. Disintegration of a liquid stream into aerosol

The atomization process can be partitioned into three zones as indicated in Figure 2: AB represents the compact stream, BC the disintegration zone, and CD the fully atomized zone. Stream atomization can be characterized by the dependence of droplet diameter on the droplet Weber number, We_D, which relates the shear forces that cause the stream breakup to the surface tension forces that hold the stream together. $We_D = \frac{\rho_g V^2 D}{\sigma}$, where ρ_g is the air density, V is the jet velocity, D is the droplet diameter, and σ is the fluid surface tension (Ohnesorge, 1936). 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 (CD). The critical Weber number range between the stable steam or droplet region and the atomization region was estimated by Johnson & Woodward (1998) to be 12 to 22 which is similar to the range determined from the data as shown in Figure 3.

Figure 3 shows three regions **I**, **II**, and **III**. Region **I** represents streams that do not atomize because of a low injection pressure. For streams that do break up, region **II** represents the spray before complete atomization (similar to BC in Figure 2), while region **III** represents complete atomization, characterized by low Weber numbers representing the small drop sizes (similar to CD in Figure 2).

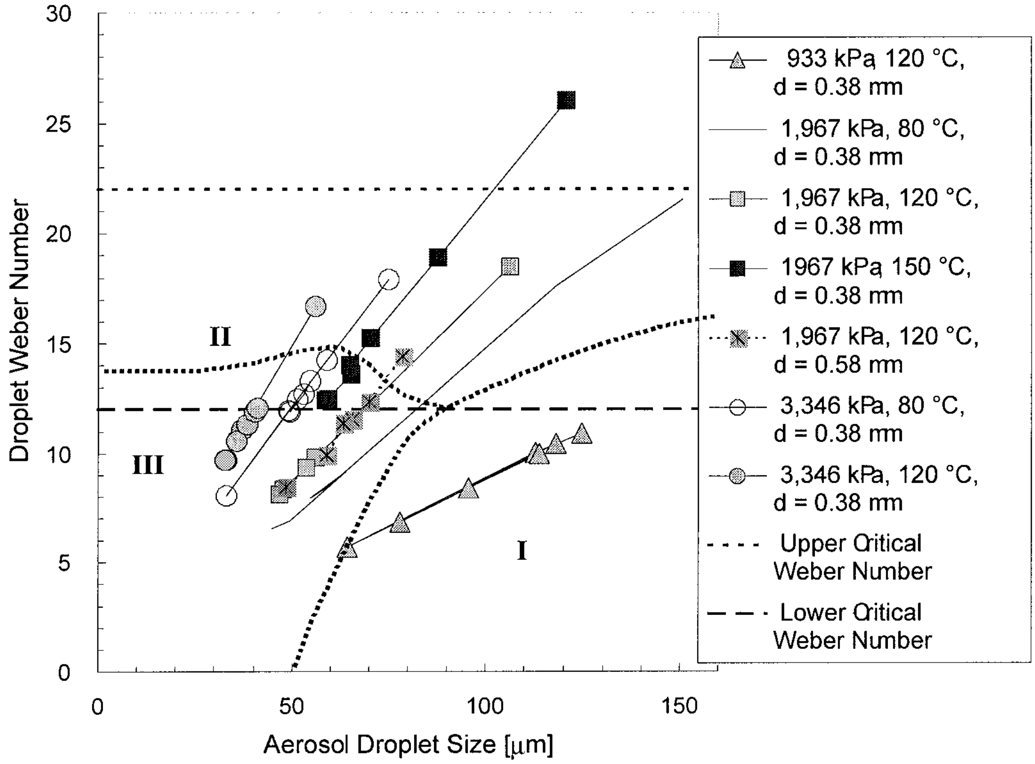

Figure 3. Weber Numbers for an alkylated aromatic HTF

With aerosol the data following established trends, the next step involves developing predictive correlations that relate the aerosol droplet sizes to the injection conditions and fluid properties. Modeling of the atomization process is very important in fuel combustion, where the fuel is generally sprayed before it is ignited to increase the combustion efficiency. A vast amount of research in this regard has confirmed that droplet size is the most important parameter of combustion efficiency. To circumvent the fact that no theory could completely and accurately describe the atomization process, various methods were adopted to describe the atomization process quantitatively.

Dimensional analysis is a popular quantitative method, because the use of dimensionless groups decreases the number of experiments required to obtain an empirical expression. The resulting expression is based on actual experimental data and hence is readily applicable to the system. Elkotb (1982), Park et al. (1996), Bayvel and Orzechowski (1993), and Kihm and Chigier (1991) have provided analyses of the important parameters required to characterize the atomization process.

The operating conditions, temperature and pressure, and the orifice diameter are the basic parameters that must be related to the aerosol droplet size. The temperature mainly affects the physical properties of the fluid, which in turn affect the atomization process. Pressure however has a more direct influence on the atomization process. Higher pressures translate into higher spray velocities, increasing shear at the liquid-air interface, which magnifies the instabilities on the liquid stream, causing a faster and more effective atomization.

The basic parameters that are important to the atomization process are:

D	Aerosol mean droplet size, D
L	The characteristic dimension of the orifice, e.g., the orifice diameter, d_0
X	Axial distance from the orifice
V	Initial velocity of the exiting liquid stream
σ	Liquid surface tension
ρ_L	Liquid density
ρ_G	Gas (air) density
μ_L	Dynamic liquid viscosity
μ_G	Dynamic gas (air) viscosity

The basis of dimensional analysis is the selection of the dimensionless parameters by combining the above parameters.

$$D,\ d_0,\ X,\ V,\ \sigma,\ \rho_L,\ \rho_G,\ \mu_L,\ \mu_G \tag{1}$$

Using the Buckingham Pi principle, the following relationship is developed (Bayvel and Orzechowsky, 1993).

$$\frac{D}{d_0} = f\left(\frac{\rho_G V^2 d_0}{\sigma}, \frac{\rho_L \sigma d_0}{\mu_L^2}, \frac{\rho_L}{\rho_G}, \frac{\mu_L}{\mu_G}, \frac{X}{d_0}\right) \tag{2}$$

or

$$\frac{D}{d_0} = f\left(We, Lp, M, N, \frac{X}{d_0}\right) \tag{3}$$

As discussed above, the Weber number (We) represents the effect of external forces on the stream break up and droplet development and is the ratio of the dynamic forces contributed by the ambient air to the surface tension. A higher Weber number indicates a dominance of the dynamic forces indicating break up of the stream.

$$We = \frac{\rho_G V^2 d_0}{\sigma} \tag{4}$$

Laplace number (Lp), the contribution of the liquid properties to the atomization process, is the ratio of the surface tension forces to the viscous forces within the liquid.

$$Lp = \frac{\rho_L \sigma d_0}{\mu_L^2} \tag{5}$$

The density ratio (M) denotes the ratio of the liquid density to the air density.

$$M = \frac{\rho_L}{\rho_G} \tag{6}$$

The viscosity ratio (N) denotes the ratio of the liquid viscosity to the air viscosity.

$$N = \frac{\mu_L}{\mu_G} \tag{7}$$

Also, implicit within this array of parameters is the Reynolds number (Re), which represents the ratio of the liquid inertial forces to the viscous forces.

$$\mathrm{Re} = \frac{d_0 V \rho_L}{\mu_L} \tag{8}$$

The following relationship holds for the Reynolds number

$$\mathrm{Re} = \sqrt{We\, Lp\, M} \tag{9}$$

Therefore, we may also use Re as a term in the dimensionless equation instead of Lp or We.

Correlations and Predictions

The correlations for all fluids exhibited a good agreement with the data. The coefficient of multiple regression is above 74% for the droplet sizes, and most of the values predicted from the correlation are within the ± 5 micron droplet size range, as shown in Figure 4.

Correlation Validation

The correlation validation is performed on the basic parameters that are described by the dimensionless groups. All the obtained equations are expanded in terms of the basic parameters and the exponent on each parameter is examined for its physical significance. For the droplet diameter, *D*, the expression is

$$\frac{D}{d_0} = 2.6\,(\mathrm{Re})^{-0.34}(We)^{-0.30}\left(\frac{\rho_L}{\rho_G}\right)^{-0.66}\left(\frac{\mu_L}{\mu_G}\right)^{-0.32}\left(\frac{X}{d_0}\right)^{-0.166} \tag{10}$$

$$D = 2.6(\rho_G)^{0.36}(\mu_G)^{0.32}(\rho_L)^{-0.32}(\mu_L)^{0.016}(\sigma)^{0.3}(V)^{-0.955}(X)^{-0.166}(d_0)^{0.52} \tag{11}$$

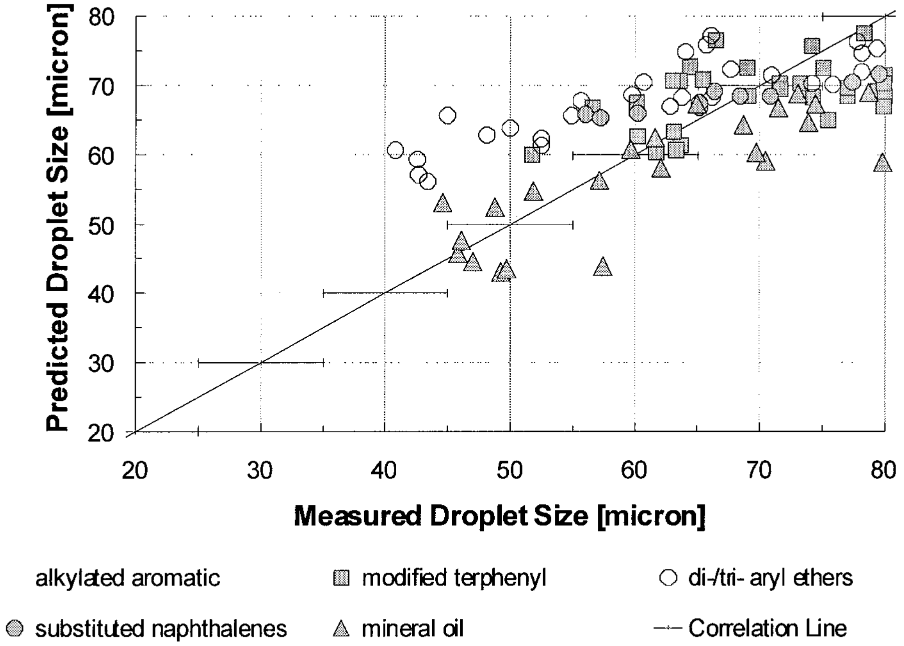

Figure 4. Predicted droplet sizes for six heat transfer fluids

In the expression for droplet sizes, the exponent for liquid viscosity is positive, so that higher viscosities result in larger aerosol droplet sizes and also larger aerosol formation distances. Therefore, while a higher viscosity hinders atomization, under conditions where atomization does occur, the droplet sizes will be larger.

The exponent for surface tension is positive, which indicates that, while higher surface tension produces larger droplets, it also hinders aerosol formation (Tabata et al., 1985).

The exponent of liquid density is negative, which means denser fluids produce smaller droplets with atomization occurring much closer to the orifice. This can be rationalized by the fact that denser liquids have a higher kinetic energy and consequently smaller droplets develop.

Liquid velocity always has a negative exponent. Higher velocities are caused by higher injection pressures and result in smaller droplets as well as shorter aerosol formation distances (Tabata et al., 1985).

The smaller orifice size tortures the liquid stream to a greater extent, resulting in smaller droplets and hence confirms the positive exponent. This correlation has a good agreement with experiment and its implications can be rationalized by theory as is important for the validity of any correlation.

Conclusions

This research has demonstrated that there is a dependence of aerosol droplet size distributions on the operating conditions and the leak size. The most important conclusion for process safety is that significant quantities of aerosol are formed from

HTFs at conditions well below their flash points. However, there are threshold conditions below which significant amounts of aerosols were not formed in the tested ranges. Operating conditions and leak size also had a significant effect on the droplet size, atomization distance, and the amount of aerosol generated.

The correlation helps in defining source terms for leaking fluids forming aerosols. During the design process the engineer must consider the following in addition to the design criteria:

- The HTF with the higher density will form smaller droplets on leaking.
- The HTF with the higher viscosity is less likely to form aerosol.
- The HTF with the higher surface tension will form larger droplets on leaking.
- Higher operating pressures will produce aerosols closer to the leak and smaller droplet sizes.

HTF selection can be based on which liquids are less likely to form aerosols. Design criteria also can incorporate the aerosol formation information to arrive at operating conditions that are less likely to produce aerosols.

Using the correlation described here to define source terms for leaks forming aerosols will help optimize existing dispersion models by relating the dispersion to the process operating conditions. Finally, the study of aerosol combustion as a function of drop-size distributions, concentrations, and fluid properties will be help estimate the upper and lower explosive limits of HTF aerosol/air mixtures. This information will help industry understand the phenomenon of aerosol explosions and thereby improve process safety.

Acknowledgements

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References

Barth, H.G. Particle and Droplet Sizing Using Fraunhofer Diffraction, 'Modern Methods of Particle Size Analysis', Wiley & Sons, Chapter 5, 135-172. (1984).

Bayvel, L., and Orzechowski, Z. 'Liquid Atomization', Taylor&Francis, Washington DC. (1993).

Elkotb, M.M., "Fuel Atomization for Spray Modeling," Prog. Energy Combust Sci. 8, 61-91 (1982).

Febo, H.L., & Valiulis, J.V. ‘Heat Transfer Fluid Mist Explosion Potential an Important Consideration for Users’, AIChE Loss Prevention Symposium. Norwood/Main: Factory Mutual Research Corporation. (1995).

Johnson, D.W., and Woodward, J.L. “Release: A model with data to predict aerosol rainout in accidental releases”, Center for Chemical Process Safety, New York: AIChE (1998).

Kihm, K.D., and N. Chigier, “Effect of Shock Waves on Liquid Atomization of a Two-dimensional Airblast Atomizer,” Atomization and Sprays, 1, 113-136 (1991).

Kihm, K.D., Terracina, D.P., Payne, S.E., and Caton, J.A. ‘Synchronized Droplet Size Measurements for Coal-Water Slurry Sprays Generated from a High-Pressure Deisel Injection System’, Journal of the Institute of Energy, 67, 2-9 (1994).

Lefebvre, A.H., ‘Atomization and Sprays’, Taylor&Francis, PA (1989).

Ohnesorge, W. ‘Formation of Drops by Nozzles and the Breakup of Liquid’ Jets. Z. Angew. Math. Mech. 16, 355-358. (1936).

Park, B.K., J.S. Lee, and K.D. Kihm, “Comparative Study of Twin-fluid Atomization Using Sonic or Supersonic Gas Jets,” Atomization and Sprays, 6, 285-304 (1996).

Sukmarg, P., Krishna, K., Rogers, W.J., Kihm, K.D., and Mannan, M.S., ‘Non-Intrusive Characterization of Heat Transfer Fluid Aerosol Sprays Released from an Orifice’, Journal of Loss Prevention in Process Industries, in press (2001).

Tabata, M., M. Arai, and H. Hiroyasu, “Effect of Fuel Viscosity and Surface Tension on Diesel Spray Drops,” Proceedings of the ICLASS 85, London, England, July 8-10, IIB/1/1 (1985).