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Flammability of heat transfer fluid aerosols produced by electrospray measured by laser diffraction analysis

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ABSTRACT

Accidental release of pressurized high flash point heat transfer fluids can result in fire and explosion hazard scenarios in the process industry. An experimental investigation on ignition of aerosols of a heat transfer fluid is carried out, and characterization of aerosol and its ignition process by non-intrusive laser diffraction technique is reported. Propagation speed of the aerosol combustion flame front as analyzed from the laser diffraction measurement agrees with high-speed visual camera observation. Flammability of the aerosol, which is based on the chances of the global flame appearance in the aerosol, is mainly controlled by aerosol droplet size and the droplet volume concentration.

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1. Introduction

In the process industry, the release of a hydrocarbon at high pressure can develop clouds of small droplets suspended in the air, and it can create an explosion hazard in presence of an ignition source (Bowen & Shirvill, 1994; Durkee, 2003), which can cause fatalities and huge economic losses to the industry.

Before happening of aerosol explosion, three main events take place consecutively: aerosol formation, dispersion and ignition. Most fluids tend to form aerosols upon pressurized release. Characteristics of the aerosols depend on the properties of the liquid, including density, viscosity, surface tension, and physical parameters including fluid temperature, operating pressure, heat transfer coefficients, release geometry. Sukmarg & Krishna (2002) studied the aerosol formation by leakage of hydrocarbons through small orifices from a pressurized container. Krishna (2001); Krishna (2003); Krishna, Rogers, and Mannan (2004) introduced a methodology to select heat transfer fluids based on the study of the their tendency to form aerosols. It was suggested that liquids of higher viscosity or surface tension tend to form aerosols with higher droplet size, and liquids under higher operating pressures tend to develop smaller droplet size.

There is a general misconception that fluids are safe below their flash points. But aerosol can be ignited below the fluid's flash point, because the evaporation rates are drastically increased for liquids dispersed in the form of fine aerosol droplets with large droplet surface area. Ignition of aerosols can occur with ignition sources, such as an electric spark, static electricity, naked flames, hot surfaces, and impact friction (Ballal & Lefebvre, 1975, 1981; Danis, 1987; Danis, Cernansky, & Namer, 1985; Singh, 1986).

Eichhorn (1955) made the distinction between flammability of gas mixtures and flammability of aerosols, and proposed a conceptual diagram which indicated both the vapor and aerosol flammability regions with upper and lower boundaries. The dew point line separates the vapor and aerosol flammability regions. However, compared with well-established and widely tested methods for identifying flammability region of vapors, there is a lack of research data for characterization of the aerosol flammability region.

Different from ignition of gas mixtures, there exist no distinctive lines separating the flammable and non-flammable regions for aerosols. Instead a transitional region was found where the aerosol becomes more flammable with increase of its ignition frequency, which is defined as the ratio of the number of successful ignitions over the number of trials to ignite the aerosol (Aggarwal and Cha, 1987; Aggarwal and Sirignano, 1984). From the numerical analysis of aerosol ignition process by Aggarwal and Sirignano (1984), the ignition of aerosol is thought to be a statistical process rather than a deterministic process. To study the flammability of aerosols, it is

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critical to consider several factors, such as liquid/gas phase temperature, fuel concentration, droplet velocity, droplet concentration, turbulence for mixing of aerosol droplets with air, and most importantly, aerosol droplet size. The average aerosol droplet size can be measured by introducing the Sauter Mean Diameter (SDM), which is the surface area moment mean diameter expressed as:

$$D_{[3][2]} = \sqrt{\frac{\sum_{i-1}^{m} D_i^3 \Delta n_i}{\sum_{i-1}^{m} D_i^2 \Delta n_i}}$$

where D_i and n_i are droplet diameter and the number of the droplets with diameter D_i.

Flame propagation velocity is another important factor which can influence the ignitability of aerosols, and is closely related to the aerosol droplet size which controls the liquid fuel vaporization process. Polymeropoulos (1984) established a model for prediction of the laminar flame propagation velocity in a quiescent combustible mixture containing liquid fuel droplets, fuel vapor and air. There is a good agreement between predicted values and experimental data by Ballal and Lefebvre (1975, 1981) for larger size (60–100 μm) droplets in iso-octane aerosols. However, the difference between predicted and experimental values of flame propagation velocity for lower range of droplet size reveals the difficulty to accurately measure the flame front propagation speed and relevant properties of aerosols (droplet size and droplet concentrations) right in front of the flame front.

There is still a lack of data for aerosol flammability and combustion behaviors, particularly for those from high flash point hydrocarbons, which are widely used in the process industry. And it is hard to understand the potential explosion hazard from these aerosols in the process industry. To systematically study the relationship between aerosol flammability and its properties, it is necessary to simplify the problem by producing aerosol droplets of uniform size. One method as applied in the current work is the electrospray, which allows the atomization of liquids with minimal power consumption at low pressures. In electrospray, conductive liquid is injected through capillary nozzles which are charged with electric potential. An electric field is created by placing a ground electrode at a short distance from the nozzle. The meniscus of the liquid takes a conical shape upon outlet of each nozzle, which is called the cone-jet mode. Uniform small droplets are created from a fine jet issued from the cone and dispersed due to the coulombic forces among the droplets. The aerosol droplet size can be easily changed by adjusting the voltage value on the nozzles or the liquid flow rate.

In this paper, a new method to study the combustion behavior of heat transfer fluid aerosols fabricated by electrospray is introduced. The method is based on the detection of the aerosol combustion flame using Laser Diffraction Analysis (LDA). Characteristic changing patterns in the droplet-size measurement data by LDA along time were identified, which correspond to the moments of flame appearance in the aerosol and its propagation through the laser beam measurement area. Analysis of each pattern yielded propagation velocity of the flame front, which was further compared with the velocity visually observed by a fast camera. Good agreement between the two methods elucidates the possibility for further study of aerosol combustion behavior with the new method. Meanwhile frequency of the flame pattern appearances can also be related to flammability of the aerosol. By changing properties of aerosols a trend is revealed in frequency of the flame appearance, as will be discussed below.

2. Materials and methods

2.1. Aerosol generation by electrospray

A commercial heat transfer fluid, PNF, is applied in the current work for production of monodisperse aerosols. Main properties of PNF are summarized in Table 1.

Electrospray is characterized by the capability of fine control on the aerosol droplet-size distribution (Mejia, He, & Cheng, 2009. Fig. 1a shows a schematic of the electrospray setup for the production of uniform aerosol droplets of heat transfer fluid PNF. The electrospray system applies electrostatic field for dispersing liquid into minute droplets (Deng & Gomez, 2006, 2007). The nozzles are maintained at several kilovolts relative to a ground electrode, which is positioned at a certain distance down from the nozzle tips (Fig. 1b). The liquid meniscus at the outlet of each capillary takes a conical shape (cone-jet) under the influence of the electric field between the nozzles and the ground electrode, as shown in Fig. 1c. The jet further breaks up downstream into a spray of fine, charged droplets. And aerosol droplet sizes are mainly controlled by liquid flow rate from the nozzles, the applied voltage on the nozzles, and the electric field configuration.

The electrospray setup consists of a function generator (Stanford Research System, Ds-345) to generate the voltage signal and a high voltage amplifier (Trek Inc. 610E) to raise the signal to the desired high voltage level. An electric wire connects the high voltage amplifier output to the top of the nozzles. PNF fluid is pumped from an infusion syringe pump (KDS 220). The size of the syringe is 2.5 ml each. The nozzles used in the experiment are stainless steel capillary tubes of 0.01″ i.d. and 0.02″ o.d.

2.2. Aerosol characterization

A laser diffraction particle analyzer (SprayTec, Malvern Inc.), which consists of a 2 mW Helium–Neon laser tube and a ring diode detector, was used to characterize the aerosols formed from the heat transfer fluid. The aerosol droplets that pass through the laser beam will scatter light at angles directly related to their sizes. As the droplet size decreases, the observed scattering angle increases logarithmically. The laser beam was a collimated monochromatic beam of wavelength 632.8 nm and 10 mm in diameter. Intensities of the diffracted light on the diodes in the detector were converted into droplet-size data online by a computer.

Parameters calculated by the laser diffraction particle analyzer include the percentile diameters $D_{[x]}$, which are the sizes in μm that specifies x% percent of the aerosol droplets whose sizes are below the value. In this paper the $D_{[10]}$, $D_{[50]}$ and $D_{[90]}$ values are used to indicate the aerosol droplet-size distribution. The Surface Area Moment Mean Diameter $D_{[3][2]}$, also known as the Sauter Mean Diameter (SMD), is calculated based on the droplet-size distribution at every sampling moment.

Liquid volume concentration of the aerosol droplets obtained by Malvern laser is calculated from the Beer–Lambert law, and can be calculated by:

Table 1
Main characteristics of the studied heat transfer fluid PNF.

Properties	Heat transfer fluid: PNF
Appearance	Transparent colorless
Composition	Hydrotreated mineral oil
Average molecular weight	350 (g/mol)
Flash point	174 °C (345 F)
Fire point	196 °C (385 F)
Density	7.25 lb/gal (24 °C)
Viscosity	11.0 cSt at 40 °C

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