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Hard X-ray fluorescence spectroscopy of high pressure cavitating fluids in aluminum nozzles

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ABSTRACT

X-rays are frequently used to study the internal geometry of dense objects, and to measure the density of multiphase flows. However, quantitatively measuring fluid density inside a metallic object such as a high pressure spray nozzle is difficult. X-rays of sufficiently high energy to penetrate a metal object are not appreciably absorbed by the fluid inside. This requires the use of plastic or beryllium test sections, which are not suited to high pressure conditions. We present a high-energy X-ray fluorescence technique which can overcome this problem. The experiments were conducted at the 7-BM beamline of the Advanced Photon Source at Argonne National Laboratory. A hydrocarbon fluid was seeded with cerium nanoparticles. The fluid was pumped at high pressure through aluminum nozzles with inner diameters of 0.36–0.90 mm and wall thicknesses of 2–3 mm. A collimated, monochromatic 42.5 keV X-ray beam excited K-edge fluorescence from the cerium. The K_α emission lines at 34–35 keV were recorded by a cryogenic germanium detector. Changes in fluid density due to cavitation of the liquid inside the nozzle were measured by raster scanning the nozzle across the beam. A spatial resolution of 20 × 20 μm^2 was achieved with a slitted beam, which was improved to 5 × 10 μm^2 with X-ray focusing mirrors. The uncertainty in the path-integrated vapor fraction was 30–40 μm at 95% confidence. A limitation of this approach is that for low vapor pressure fluids, the nanoparticles increase the vapor pressure of the fluid and act as additional nucleation sites. These experiments demonstrate a path forward for measurements of multiphase flows inside metal components under conditions that are not feasible in optically accessible materials.

1. Introduction

The measurement of fluid flow inside nozzles, valves and other hydraulic components is critical to the study and design of many fluid systems. These include spray nozzles for fuel injection and spray coating, throttling valves and pump components for refrigeration systems and chemical processing, gas turbines and rockets, among many others. Where possible, optically accessible models for these devices can be constructed (Gavaises et al., 2014). However, compromises must be made in scale, working pressure, temperature, and geometry owing to the mechanical limitations of many materials.

As an example, diesel fuel injection nozzles with diameters of 100–200 μm can operate at pressures over 200 MPa and temperatures above 100 °C. Plastics (such as polycarbonates and acrylics) can be machined into complex shapes similar to real nozzles, but their yield stress is insufficient to be able to reach the design pressure at true scale, and this problem becomes worse with increasing temperature. As such, models are often scaled up and operated at room temperature (Gavaises et al., 2009). Improvements in the manufacture of real-size polycarbonate nozzles (He et al., 2016) has demonstrated operation up to 120 MPa, but many limitations remain. Optical distortion due to the difficulty of refractive-index matching while maintaining other fluid-mechanical properties must be considered (Budwig, 1994). Glasses (quartz, sapphire) (Falgout and Linne, 2016) can withstand extreme pressures and temperatures but are difficult to machine into complex shapes, requiring simplified geometries with flat windows, and limited optical access (Winklhofer et al., 2003). Furthermore, the working fluid is not always transparent. When a complex two-phase flow exists, the interfaces can scatter visible light to an extent that optical measurements become problematic (Powell, 2008). Similar problems occur in particle-laden and bubbly flows.

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X-rays provide an alternative route to measurement in such devices. X-ray imaging is commonly used to perform metrology in optically inaccessible materials (Lee et al., 2005). For example, high-speed X-ray imaging has been used to study the internal motion of steel fuel injection nozzle components (Moon et al., 2015; Matusik et al., 2016; Viera et al., 2016). The X-ray energies required to penetrate such objects are high (typically above 20 keV). As such, any low density or weakly absorbing materials inside the highly absorbing object (such as water or hydrocarbons inside a metal object) absorb a negligible amount of the X-ray beam. This contrast problem limits the usefulness of absorption techniques such as X-ray computed tomography (CT) (Halls et al., 2013; Shakya et al., 2014; Hu et al., 2014; Marchitto et al., 2015) where metal geometries are concerned. While phase contrast imaging can be used to observe gas-liquid interfaces inside metal objects (Swantek et al., 2014; Duke et al., 2014), quantitatively measuring the fluid density is not presently feasible with such techniques.

High flux synchrotron sources have been used for some time to make X-ray radiography and fluorescence measurements of optically dense external fluid flows such as fuel sprays (Kastengren et al., 2014; Xue et al., 2015), medical sprays (Mason-Smith et al., 2016), impinging jets (Halls et al., 2015; 2017) and bubbly flows (Heindel, 2011). These experiments are typically made at energies from 6 to 15 keV and have the advantage of providing quantitative, temporally and spatially resolved line of sight density measurements (Kastengren and Powell, 2014). In recent years, X-ray radiography and fluorescence techniques have been extended to internal flows in geometries made from thermoplastics (Duke et al., 2016) and low atomic number materials such as beryllium (Duke et al., 2017). However, these materials are limited to modest pressures and temperatures.

In order to measure fluid density in more absorptive metal components capable of withstanding higher temperatures and pressures, a means of obtaining sufficient contrast at high X-ray energies is required. The most effective means of achieving this goal is to use hard X-ray fluorescence spectroscopy with a heavy element tracer in the flow. In prior studies, tracers such as Ar, Ni, Zn, Co, Cu, Br and Kr have been used (Kastengren et al., 2011; Halls et al., 2015; Duke et al., 2015b; 2015a; Tranter et al., 2016). The K-edge fluorescence line energies for these elements are too low to transmit through a metal geometry of appreciable thickness. In a previous study of needle motion in a steel diesel injector tip, we found that cerium nanoparticles in the fluid (which were used as a contrast enhancing agent for X-ray radiography experiments of the spray outside the nozzle) absorbed enough of the high-energy X-rays as to be weakly visible on the imaging system (Duke et al., 2014). Although the absorption contrast from the cerium is weak due to its low concentration in the fluid, an incident beam with energy above the Ce K-edge (40.4 keV) generates spontaneous fluorescent emission at 34–39 keV, which is sufficiently energetic to transmit through several mm of aluminum (Bearden and Burr, 1967). Since the fluorescent emission occurs against a very low background, a modest concentration of nanoparticles is sufficient to obtain good signal-to-noise ratio (SNR).

In this paper, we demonstrate a novel high-energy X-ray fluorescence experiment to measure fluid density inside an aluminum nozzle with 2–3 mm wall thickness and 0.36–0.90 mm internal diameter. The flow was seeded with cerium nanoparticles, and the emitted X-ray fluorescence was measured using an energy-dispersive detector placed at a right angle to the incident beam. A spatial resolution of 20 × 20 μm^2 was achieved with a slitted beam, which was improved to 5 × 10 μm^2 with X-ray focusing mirrors. Measurements were made at internal pressures of up to 87 bar. A decrease in density due to cavitation of the liquid was clearly observed. The ability to perform quantitative density measurements in aluminium demonstrates a viable technique for making similar measurements under extreme conditions and in complex geometries which are not feasible with optically accessible materials.

Table 1
Bulk fluid properties for Viscor BR16 gasoline surrogate at 25 °C, with and without cerium nanoparticles.

	Without particles	With particles (4% by mass)
Fluid density	773 kg m^{-3}	817 kg m^{-3} (+6%)
Kinematic viscosity	1.13 ×10^{-6} m^{2}s^{-1}	1.23 ×10^{-6} m^{2}s^{-1} (+9%)
Surface tension	0.029 N m^{-1}	0.028 N m^{-1} (within error)
Vapor pressure (P_v)	360 Pa	500 Pa (+40%)
Critical cavitation number K_c	1.570	1.569 (within error)

Table 2
Cerium nanoparticle properties.

Median particle diameter	20 nm
Particle/fluid density ratio	8.3
Particle Knudsen number	0.06
Particle slip correction factor	1.08
Maximum particle Re	1.06 (at $U_{max} = 65$ ms^{-1})
Maximum particle Stokes number	0.53 (at $U_{max} = 65$ ms^{-1})
Particle Bond number	10^{-10}

2. Methods and materials

2.1. Nanoparticle and fluid properties

The test fluid used in the experiments was *Viscor 16BR*, a low-volatility gasoline surrogate. The fluid was doped with 4% by mass of cerium nanoparticles in the form of a hydrocarbon-soluble suspension (*Rhodia DPX9*, 10:1 v/v). Measurements of the fluid properties and the nanoparticles were made in order to determine the effects of the particles on the fluid and how faithfully the particles will follow the fluid. Tests of bulk fluid properties were made both with and without the nanoparticles (see Table 1). The fluid density and viscosity increased by 6% and 9%, respectively, and these changes were taken into account when determining the flow conditions.

In order to determine the size range of the nanoparticles, ultra-small-angle X-ray scattering measurements were taken of the fluid in plastic test sections. Further details can be found in Kastengren et al. (2017) and Ilavsky et al. (2009). The plastic nozzle was scanned at an energy of 21 keV, and the scattering contrast between the fluid with particles and the control was used to estimate a median particle diameter of approximately 20 nm. Particle properties based on these measurements are shown in Table 2. The particles are small enough to follow the flow faithfully, while being large enough to exhibit continuum behavior. The particles are not subject to significant diffusion with respect to the scale of the experiment. The Knudsen number is large enough however to invalidate the no-slip assumption when determining particle drag. Accounting for this, the maximum particle Stokes number under impulsive acceleration from zero to the maximum freestream velocity was determined to be 0.53, with particle Reynolds number on the order of unity. The particle lag distance is approximately 200 times smaller than the smallest length scale which can be probed by the X-ray beam in these experiments, and is thus a negligible contributor to the measurement error.

It is also necessary to determine whether the particles will remain in the liquid phase or can migrate into cavitation bubbles, given that the particles are used as a tracer for the liquid phase. Measurements of the fluid surface tension indicate that the particle Bond number is small enough that the particles will be unable to break the surface tension of any bubbles in the flow and will