Quantitative measurement of binary liquid distributions using multiple-tracer x-ray fluorescence and radiography

Benjamin R. Halls,1 Terrence R. Meyer,1,* and Alan L. Kastengren2

1Department of Mechanical Engineering, Iowa State University, Ames, Iowa 50011, USA
2X-Ray Science Division, Argonne National Laboratory, Argonne, Illinois 60439, USA

trm@iastate.edu

Abstract: The complex geometry and large index-of-refraction gradients that occur near the point of impingement of binary liquid jets present a challenging environment for optical interrogation. A simultaneous quadruple-tracer x-ray fluorescence and line-of-sight radiography technique is proposed as a means of distinguishing and quantifying individual liquid component distributions prior to, during, and after jet impact. Two different pairs of fluorescence tracers are seeded into each liquid stream to maximize their attenuation ratio for reabsorption correction and differentiation of the two fluids during mixing. This approach for instantaneous correction of x-ray fluorescence reabsorption is compared with a more time-intensive approach of using stereographic reconstruction of x-ray attenuation along multiple lines of sight. The proposed methodology addresses the need for a quantitative measurement technique capable of interrogating optically complex, near-field liquid distributions in many mixing systems of practical interest involving two or more liquid streams.

©2015 Optical Society of America

OCIS codes: (010.1030) Absorption; (110.7050) Turbid media; (110.7440) X-ray imaging; (260.2510) Fluorescence; (340.6720) Synchrotron radiation.

References and links

1. A. H. Lefebvre, Atomization and Sprays (Taylor and Francis, 1989).
2. T. R. Meyer, M. Brear, S. H. Jin, and J. R. Gord, “Formation and diagnostics of sprays in combustion,” in Handbook of Combustion (Wiley, 2010).
3. D. T. Campbell, S. D. Clapp, R. L. Proffit, and G. L. Cline, “Reactive stream separation photography,” AIAA J. 9(9), 1832–1836 (1971).
4. J. D. Dennis, S. F. Son, and T. L. Pourpoint, “Critical ignition criteria for monomethylhydrazine and red fuming nitric acid in an impinging jet apparatus,” AIAA Paper 2012–4325 in 48th AIAA/ASME/SAE/ASEE Joint Propulsion Conference and Exhibit, Atlanta, GA, 30 July–01 Aug. 2012.
5. M. Linne, “Imaging in the optically dense regions of a spray: a review of developing techniques,” Prog. Energ. Combust. 39(5), 401–440 (2013).
6. H. Purwar, S. Idlahcen, C. Rozé, D. Sedarsky, and J.-B. Blaisot, “Collinear, two-color optical Kerr effect shutter for ultrafast time-resolved imaging,” Opt. Express 22(13), 15778–15790 (2014).
7. R. Wellander, E. Berrocal, E. Kristensson, M. Richter, and M. Aldén, “Three-dimensional measurement of the local extinction coefficient in a dense spray,” Meas. Sci. Technol. 22(12), 125303 (2011).
8. G. Charalampous, Y. Hardalupas, and A. M. K. P. Taylor, “Novel technique for measurements of continuous liquid jet core in an atomizer,” AIAA J. 47(11), 2605–2615 (2009).
9. J. H. Rupe, “The liquid-phase mixing of a pair of impinging streams,” Jet Propulsion Laboratory Progress Report 20–195, 1953.
10. N. Ashgriz, W. Brocklehurst, and D. Talley, “Mixing mechanisms in a pair of impinging jets,” J. Propul. Power 17(3), 736–749 (2001).
11. V. McDonell, V. Phi, S. Samuelsen, A. Nejad, M. Shahnam, C. Guernsey, and R. Carlson, “Structure of sprays generated by unlike doublet injectors,” AIAA Paper 99–2464 in 35th AIAA/ASME/SAE/ASEE Joint Propulsion Conference and Exhibit, Los Angeles, CA, 20–23 June, 1999.
12. K. Jung, H. Koh, and Y. Yoon, “Assessment of planar liquid-laser-induced fluorescence measurements for spray mass distributions of like-doublet injectors,” Meas. Sci. Technol. 14(8), 1387–1395 (2003).

#223583 - $15.00 USD
Received 25 Sep 2014; revised 3 Dec 2014; accepted 2 Jan 2015; published 23 Jan 2015
© 2015 OSA
26 Jan 2015 | Vol. 23, No. 2 | DOI:10.1364/OE.23.001730 | OPTICS EXPRESS 1730
Based on these downstream measurements, three near-field interaction regimes have been postulated to occur in the early jet interaction process. In reflective mixing, the jets do not have sufficient momentum to coalesce, and their trajectories do not cross. This results in poor interaction and mixing. At higher momentum conditions, transmissive mixing occurs because of the dual effect of high impact velocity and the presence of jet instabilities that allow ligaments from each liquid to pass through the opposing liquid to the other side of the liquid sheet. This occurs through jet fragmentation within the impingement zone and can increase...
the contact area and mixing between the two fluids. At sufficiently high jet momenta, liquid transmission may occur without substantial mixing; however, an increase in post atomization turbulent dispersion typically leads to improved overall mixing. Residing between reflective and transmissive mixing is a thoroughly mixed condition [9,10]. Because these mixing phenomena have been inferred from downstream measurements, the local rate of liquid interaction at the impingement point and early onset of potential chemical reactions cannot be predicted. This ultimately limits fundamental understanding and detailed modeling of the mixing process for these and other types of jet mixing applications.

![Fig. 1. (a) Orthogonal backlit visible-light images of impinging jet spray and (b) methodology for measuring equivalent path length (EPL) from x-ray attenuation and fluorescence. Reabsorption path length (RPL) used to correct for fluorescence signal trapping. Dashed arrow in (a) indicates x-ray beam path.](image)

In the current work, x-ray radiography and fluorescence are used to address these challenges and to enable measurements of binary liquid distributions in the impingement region itself, thereby providing key information about the processes leading to the mixing of unstable liquid jets. Because the primary interaction of low-energy ($\leq 10$ keV) x-rays with sprays is through absorption and weak scattering, it is relatively straightforward to relate the attenuation [see Fig. 1(b)] to the path-length-integrated liquid density along the path of the x-ray beam. X-ray radiography, for example, has been demonstrated with high spatial resolution and signal-to-noise ratio in studies of liquid injection into gaseous environments using line-of-sight x-ray attenuation [14–16]. More specifically, studies of overall quantitative liquid distributions in impinging jets have recently been demonstrated [17].

Unfortunately, x-ray attenuation by itself cannot easily distinguish liquids because both liquids will appreciably absorb x-rays, leading to ambiguity between liquid path length and relative concentration of each liquid. The goal of this work is to investigate the feasibility of utilizing x-ray fluorescence as a means to study the interaction of impinging liquid jets, particularly in the complex near-field region. In x-ray fluorescence, the absorption of x-rays by atoms leads to the emissions of fluorescence photons, also in the x-ray regime, at unique photon energies characteristic of the atomic number of the element emitting the fluorescence. Like x-ray attenuation, x-ray fluorescence benefits from insensitivity to refraction and strong scattering, but it has the added advantage of allowing each fluid to be uniquely identified using tracer elements that fluoresce at particular, known x-ray energies. By seeding two or more liquid streams with different tracer elements (at the ppm level), the liquids that originate from each jet can be tracked simultaneously with high dynamic range. In this way, two liquids with similar x-ray attenuation cross-sections but different tracers can be excited with the same x-ray source and differentiated by the characteristic fluorescent emission of those tracers. This approach is commonly used for static elemental analysis [18], and researchers at the Advanced Photon Source (APS) at Argonne National Laboratory have also used fluorescence to study turbulent mixing in gases [19].
A significant challenge associated with utilizing this approach for studies of liquid distributions is that, just as the incident x-ray beam experiences absorption, the fluorescence signal is also subject to absorption, leading to potential bias errors due to substantial non-uniform reabsorption of the fluorescence x-rays generated at various points in the flow. As such, the fluorescence signal is not only a function of the local liquid mass fraction but also the total liquid path length from each measurement point to the fluorescence detector. This is illustrated as the reabsorption path length (RPL) in Fig. 1(b). Hence, in addition to demonstrating the use of x-ray fluorescence for studies of liquid mixing, this work evaluates different methods to correct for signal reabsorption and ensure quantitative accuracy. This includes a radiographic technique that relies on time-intensive attenuation measurements along multiple lines of sight (analogous to [7]) to determine RPL and correct for x-ray fluorescence reabsorption, as well as a multiple-tracer approach that allows for corrections to be performed on an instantaneous basis. The latter takes advantage of the unique path-length dependent absorption of fluorescence generated from different tracer species seeded into each fluid. The details of each approach, their implementation for reabsorption corrections, and the accuracy of the fluorescence measurements are addressed in the following discussion.

2. Experimental setup

Experiments of liquid mixing were conducted using an impinging jet injector with 510 µm diameter orifice passages directed at each other with an included angle of 60°, as shown in Fig. 1(a) [17]. The liquid jets exit into air at atmospheric pressure and collide, forming a liquid sheet with varying breakup characteristics. The orifice passages have chamfered entrances and length-to-diameter ratios of 47:1 to ensure fully developed flow. The liquid velocities are maintained at \( V = 18 \) m/s by a pair of rotameters that are accurate to within 2%. The liquid properties of the salt solutions where similar to water within the accuracy of the measurements of viscosity (~1%) and surface tension (~3%) using a falling ball rheometer and force tensiometer, respectively. The density of the solutions increased approximately 0.5%, resulting in jet Reynolds numbers of \( R_{\text{jet}} = \frac{VD}{\nu} \approx 9000 \), where \( D \) is the passage diameter and \( \nu \) is the bulk water kinematic viscosity. The consistency of the liquid solutions was held by mixing in bulk, and once the salts dissolved they would remain in solution.

Attenuation and fluorescence measurements were conducted at the APS 7-BM beamline, a synchrotron bending magnet beamline. The x-ray source creates a nearly collimated, initially polychromatic x-ray beam with high photon flux. The beamline consists of two enclosures, 7-BM-A and 7-BM-B. Enclosure A contains slits and a double multilayer monochromator to condition the beam size and to select a narrow range of x-ray energies, respectively. The monochromator can tune the photon energy from 5.1 to 12 keV (\( \Delta E/E = 1.4% \)). Enclosure B houses Kirkpatrick-Baez focusing mirrors [20], the injector affixed to a two-dimensional translation stage, and the x-ray detectors to measure attenuation and fluorescence. The total detected x-ray flux is \( 1.4 \times 10^{11} \) photons per second in the absence of the spray. A thorough description of the beamline can also be found in [21]. Point-wise raster scanning data were collected while the spray was traversed along the minor axis shown in Fig. 1(b) and across the incident beam at several locations downstream of the nozzle exit.

Experiments were conducted using either single or dual tracers in each water stream and employed photon energies of 10.4 or 10.6 keV, respectively. In each case the x-ray beam was focused to 5 µm (vertical) \times 6 µm (horizontal) full width at half maximum (FWHM). An unbiased 300 µm thick silicon PIN diode was used to detect the transmitted x-ray beam intensity for attenuation measurements. The PIN diode signal was normalized by an upstream beam intensity monitor to account for fluctuations in the incident beam. The relative x-ray attenuation along the path of the beam, and hence the amount of liquid along that path or the equivalent path length (EPL), was then measured using Beer’s Law. For a monochromatic x-ray beam, Beer’s Law can be written as:
where $I$ and $I_0$ are the x-ray intensities measured at the PIN diode with and without spray, respectively, $\mu$ is the attenuation coefficient, and $EPL$ is the equivalent path length of the x-ray beam through the fluid. In the case where the liquid is not contiguous along the path of x-ray beam, then $I$ is interpreted as an EPL of the liquid intersecting the x-ray beam. The attenuation coefficient, $\mu$, is the product of the attenuation cross-section and number density of the fluid, which can be considered constant in the case of identical water jets and for small fluorescence tracer species concentrations.

A silicon drift diode (SDD) photon counting detector capable of discriminating the spectral energy of each incoming x-ray photon with a resolution of 250 eV was used to measure the x-ray fluorescence generated by the interaction of the incident x-ray beam with the spray. The Kα fluorescence signal was integrated over a spectral region centered on the peak emission of each tracer. The SDD was placed at approximately 200 mm from the x-ray beam along the minor axis, as shown in Fig. 1(b). Elastic and Compton scattering were minimized by detecting at 90° to the excitation beam, and there was no appreciable x-ray fluorescence from water to interfere with tracer fluorescence. Note that the fluorescence signal captured by the SDD at a particular x-ray spectral energy is proportional to the density of the tracer element that fluoresces at that x-ray energy multiplied by the incident x-ray intensity. The fluorescence signal from each tracer element, therefore, can be directly related to EPL of the liquid originating from each jet integrated along the beam path. Calibration for absolute EPL was achieved by comparing the fluorescence signal in the pure liquid jets to the attenuation signal measured using the PIN diode, along with appropriate corrections discussed further below.

The main uncertainty using this fluorescence technique is the reabsorption of fluorescence photons en route to the SDD. Two methods for error correction are proposed here. The first method, termed the stereo method, uses one unique tracer in each fluid and employs stereographic reconstruction to estimate the 3-D shape of the spray, as shown for a single z-slice in Fig. 2(a), and therefore the amount of reabsorption that may occur. Fluorescence and attenuation measurements were made along the minor axis followed by attenuation measurements made along the major axis to produce orthogonal views of the spray. These two views were reconstructed using a back projection technique followed by an iterative arithmetic reconstruction technique to generate an estimate of the liquid mass fraction in the z-plane. This allowed for the determination of the signal reabsorption. In this case, it was not possible to perform an instantaneous radiographic measurement orthogonal to the path of the excitation beam. As such, this approach provides data for reabsorption corrections on a time-averaged basis and is susceptible to uncertainties due to instantaneous fluctuations in the nonlinear, path-length dependent absorption process [see Eq. (2)]. The fluorescent tracer elements for the stereo method were chosen to have absorption edges below the energy of the excitation beam (~10.4 keV), high fluorescence photon energy (> 8 keV) to minimize reabsorption, good fluorescence yield, and the ability to be easily dissolved in water. The two tracer elements initially included zinc (K-edge energy ~8.63 keV) and copper (K-edge energy ~8.04 keV) [22] seeded separately into each of the two jets. The second method for reabsorption correction, termed the four-salt method, uses two unique tracers in each fluid. The fluorescence photons from each pair of tracers follow the same path to the fluorescence detector but are at different x-ray energies; hence, they are absorbed at different rates for a given reabsorption path length (RPL) of liquid. The reabsorption ratio between the tracers can be used, therefore, to estimate the RPL and calculate the initial signal prior to reabsorption of the fluorescence photons. In the four-salt method, nickel (K-edge energy ~7.47 keV) was added to the zinc stream, and cobalt (K-edge energy ~6.92 keV) was added to the copper stream [22] to facilitate instantaneous corrections for reabsorption. As shown in Fig. 2(b),
each pairing (Ni-Zn and Co-Cu) maximized the energy ratio so that the fluorescence from one tracer of each pair would be more sensitive to reabsorption than the other. The concentration of each tracer element was 500 ppm by mass, which caused a negligible change in the overall attenuation coefficient of the liquids but allowed high fluorescence signal-to-noise ratio.

Several corrections are applied to the fluorescence data. The fluorescence signal of each liquid is based on the Kα lines associated with each tracer present in the spray, while signals from the Kβ lines are subtracted if they overlap with Kα lines. Matrix effects such as zinc Kα exciting copper are neglected due to the small concentrations of the tracer species. The effects of dead-time in the SDD are removed by multiplying the ratio of the input count rate by the output count rate [23]. The average dead-time was less than 10%, so the dead time corrections are considered minor. The effects of the attenuation of the incident beam as it passes through the spray were also corrected.

As noted earlier, an absolute correlation between the fluorescence data and the quantitative liquid distribution is obtained through comparison with the attenuation data. To obtain a quantitative fluorescence signal several possible sources of bias error are accounted for through linear corrections, including fluorescence signal reabsorption and mass conservation. Reabsorption of the fluorescence signal, $R_{	ext{salt}}$, occurs as the fluorescent photons pass through the spray, air, and the window of the SDD. Attenuation coefficients used in the analysis are tabulated in [24]. The correction is based on the RPL determined through stereographic reconstruction is shown in Eq. (2):

$$R_{\text{salt}}(x) = \frac{1}{N} \sum_{x} \frac{m(x,y)}{M} e^{-\mu_{\text{salt}}R_{\text{PL}}(x,y)}$$

where $R_{\text{salt}}$ is determined by Beer’s Law given the attenuation coefficient of the emitted x-ray energy and $R_{\text{PL}}$, weighted by the local mass, $m$, divided by the total mass, $M$, along the major axis, $y$, and averaged over $N$ pixels along the minor axis, $x$.

The two correction methods differ in how the value of RPL is determined. Figure 2(a) shows the results of the stereographic reconstruction technique used to estimate RPL from the cross-stream slice of the spray. From each slice the RPL can be weight-averaged along the major axis for each data point. A sample spectrum of fluorescence from the tracer elements for the four-salt method to calculate RPL is shown in Fig. 2(b). In the four-salt method, the ratio between the fluorescence signals of the two tracers in each stream is used to compute the amount of reabsorption, using the ratio in attenuation coefficient between the Kα photons of the two tracers. An example of the calculated signal ratio, $R$, is shown in Fig. 2(c), where zinc is divided by nickel and copper by cobalt. As $R$ is acquired at each instant, it facilitates efficient reabsorption corrections in post-processing.

![Fig. 2](image-url)
A calibration coefficient, $M_{\text{salt}}$, is applied to match the EPL (or proportionately the liquid mass) measured through attenuation. Equation (3) displays the equality of mass in each jet with the total attenuation mass in terms of spatially integrated EPL (assuming constant density). Each correction is applied at each location along the minor axis, except the mass conservation coefficient, which is a global value used at all locations along the minor and $z$ axes. The final fluorescence signal is a quantitative measurement of the EPL, which is proportional to the line-of-sight averaged mass for a given fluid density and area of interest.

$$M_{\text{Cu}} \int A(x) R_{\text{Cu}}(x) F_{\text{Cu}}(x) \, dx = M_{\text{Zn}} \int A(x) R_{\text{Zn}}(x) F_{\text{Zn}}(x) \, dx = 0.5 \int EPL(x) \, dx \quad (3)$$

Measurements utilizing the stereo and four-salt correction methods were conducted at separate times but at the same injector conditions.

The spatial resolution of the raster scans through the spray was limited by the selected point spacing of 25 µm. Noise in the attenuation and fluorescence signals was determined by the standard deviation in the flat field signal where no spray is present. The noise values equate to EPL values of 0.4 µm and 0.8 µm for the attenuation and fluorescence signals respectively. This gives rise to maximum signal-to-noise ratios (SNR) of 2300:1 and 800:1, respectively, for the attenuation signals (used for calibration from fluorescence signals to EPL) and fluorescence signals (used to measure the distribution of each liquid).

3. Results and discussion

The reliability of the stereo and four-salt correction methods can be tested by comparing the EPL of the free jets prior to impingement as measured using fluorescence with that measured directly through attenuation. The largest corrections are required in the copper-seeded free jet, as shown in Fig. 3, because the fluorescent photons must pass through a longer reabsorption path length. Single liquid jet profiles show significant errors resulting from reabsorption of fluorescence photons before corrections are applied. Figure 3(a) shows a slight over-correction of reabsorption, manifested as a right shifted asymmetric profile for the stereographic correction method. The four-salt method is able to capture the jet profile more accurately but with a slight undercorrection in the overall EPL. This undercorrection occurs because some signal from the zinc-seeded jet would appear on the side of the copper-seeded jet due to slight overlap of fluorescence signals on the SDD. Although a background subtraction method was used to reduce this interference, a small error would remain in some cases. As such, the EPL of the copper-seeded jet does not quite equal the EPL as measured by attenuation. After the corrections the error near the peak value in the copper-seeded jet are reduced from 60% to 2% and 45% to 3.5% for the stereo and four-salt methods, respectively.

![Fig. 3. Line plots of the equivalent path length (EPL) across a single jet –1.5 mm upstream of impingement comparing the (a) stereo and (b) four-salt correction methods. The Cu signal before corrections for reabsorption significantly underestimate the EPL measured via attenuation.](image-url)
An additional bias error in the attenuation and fluorescence signals appears in the data as intensity spikes (e.g., near −0.1 mm and −0.3 mm along the minor axes in Figs. 3(a) and 3(b), respectively). These are most likely because of droplets collecting on the injector face and falling through the x-ray beam. These transient errors are infrequent and do not appreciably affect the overall jet profiles or observations about the mixing process.

As might be expected, the jet liquid closest to the SDD experiences minimal reabsorption, and so another measure of accuracy is the symmetry in the corrected jet profiles. This is illustrated in Fig. 4, which shows excellent symmetry between the two jet profiles during the impingement process. While the results from the two correction methods are very similar, slight differences at downstream distance from the impingement point −0.5 mm are due to uncertainties in the location selected for interrogation.

As the two jets begin to merge at −0.5 mm, they largely maintain their round spatial profiles, as connoted by the EPL values being similar to the widths in the jet profiles. It is also possible to detect long tails in the EPL profiles in the jet propagation direction that give an early indication that some fraction of each fluid is beginning to penetrate across the opposing jet. At 0.5 mm, the liquid jets continue to merge, as indicated by the overlap in spatial profiles, and the sheet begins to spread along the major axis. This sheet formation leads to an increase in EPL to ~1200 µm with a corresponding compression of the jet profiles along the minor axis to a combined FWHM of only ~380 µm, reducing the reabsorption path. At 2 mm, the sheet distribution thickens as liquid breakup and dispersion continues.
From the downstream distance required to achieve overlap in the spatial profiles of the liquid jets and from the approximate downstream propagation velocity of the liquid jets, it is possible to estimate that the merging process is complete within a time frame of about 100 μs (or less if the jets accelerate at the point of impingement). Based on this time frame and the distance that each jet propagates during the merging process, it is also possible to estimate an average velocity along the minor axis that is about 25–30% that of the initial free jets. This is not consistent with rates of Fickian diffusion but is indicative of rapid deceleration along the minor axis, likely due to transmission of liquid fragments through the opposing fluid.

As the uncertainties in the correction methods may vary downstream, additional information about the accuracy of the measurements is listed in Table 1. The error is taken as the absolute difference between the attenuation signal and the sum of the fluorescence signals from the two fluids. All errors reported are averaged from data where the total EPL was greater than 100 μm and, therefore, include a wide range of EPL values. The error at the peak EPL is considerably lower than the values reported in Table 1. According to these data, the stereo and four-salt methods have typical errors between 1.5 and 5% and 4–8%, respectively, after corrections for reabsorption. The stereo method has the advantage of providing a direct measure of the RPL without spectral analysis. However, this requires additional radiographic data orthogonal to the x-ray beam path to apply the correction. The error with the stereo method may result from overcorrection of fluorescence reabsorption due to the SDD not being exactly perpendicular to the excitation beam. The main source of error with the four-salt method is separation of the fluorescence lines from the different elements, particularly subtraction of the $K\beta$ line from the $K\alpha$ line of the element of next higher atomic number. The four-salt method does not require any additional data collection and can be performed at each measurement instant, but care must be taken when choosing tracers and analyzing the data, particularly where spectral features overlap.

Table 1. Errors in fluorescence EPL measurements. Origin in z-location taken as approximate point of jet impingement.

| Z-Location [mm] | Stereo Method Error [µm] | Error [%] | Four-Salt Method Error [µm] | Error [%] |
|-----------------|--------------------------|----------|-----------------------------|----------|
| −1.5            | 10.5                     | 3.2      | 15.9                        | 4.3      |
| −0.5            | 16.1                     | 3.9      | 21.6                        | 4.5      |
| 0.5             | 24.7                     | 4.9      | 32.3                        | 4        |
| 2               | 4.1                      | 1.5      | 22.7                        | 5.3      |
| 9.5             | 6.4                      | 3        | 17.2                        | 8.1      |
| Average         | 12.4                     | 3.3      | 21.9                        | 5.2      |

4. Conclusions

X-ray fluorescence was utilized for measurements of liquid distributions in the near field of impinging jets at various stages of the merging process. Absolute calibration using x-ray attenuation measurements enabled detailed characterization of the effective path length (EPL) profiles of each liquid jet after corrections for beam attenuation and reabsorption of fluorescence photons. Two methods to account for the reabsorption of the fluorescence signal were discussed and compared, including a stereo method using multiple line-of-sight stereography and a four-salt method relying on differential absorption between a pair of tracers seeded into each liquid stream. Errors due to reabsorption are reduced by factors of 30 and 13 for the stereo and four-salt methods, respectively.
The ability to track individual liquid streams can provide key insight into the complex mixing phenomena that develops during the merging of two or more liquid jets. In the current work, data near the impingement region confirm the existence of transmissive mixing very early in the process. Further examination of various conditions using this approach can, therefore, deepen the understanding of impinging jet mixing and help to validate high-fidelity numerical simulations and/or detailed physicochemical models.

Acknowledgments

This work was funded, in part, by the Army Research Office (Dr. Ralph Anthenien, Program Manager). A portion of this research was performed at the 7-BM beamline of the Advanced Photon Source, Argonne National Laboratory. Use of the APS is supported by the U.S. Department of Energy under Contract No. DE-AC02-06CH11357. The authors express their gratitude to M. Johnson, C. Radke, A. Poniatsowski, J. Tiarks, D. Stoecklein, and W. Lubberden of Iowa State University for their technical assistance.