Measuring flow profiles in heated miniature reactors with X-ray fluorescence spectroscopy

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ABSTRACT

Density profiles of flowing gases within an opaque, heated, silicon carbide (SiC) miniature flow tube were measured, for the first time, by X-ray fluorescence spectroscopy using krypton as the fluorescent species. The experiments were performed with the flow tube at various fixed exterior wall temperatures between 300 K and 1100 K and a highly focused X-ray beam ($5\text{ }\mu\text{m} \times 7\text{ }\mu\text{m}$; 15 keV). Spatially resolved raster scan maps of gas densities in the miniature reactor and the nearly supersonic jet eluting from the reactor were obtained. The intensity of the fluorescence signal was attenuated by the SiC, and a signal trapping model based on the tube morphology is required to convert the raw fluorescence signal into gas densities. This model was obtained, in part, from simultaneous measurements of the absorption of the incident X-rays by the SiC. Additional radiographic experiments with an unfocused beam were performed to image the SiC tube and complement the raster scans. Excellent correlation was found between the flow pattern revealed by the fluorescence measurements and the wall structure observed from the absorption measurements. Comments are made regarding refinements of the technique that will result in data of sufficient fidelity for validating computational fluid dynamic models of gas flows in heated miniature tubes.

1. INTRODUCTION

In situ, non-invasive measurements of the gas state inside a reactor can be challenging to make, but are necessary for accurately determining reaction conditions and extracting reliable chemical kinetic data where complex or poorly defined flowfields exist. For example, micro-reactors such as those based on Chen nozzles (resistively heated SiC tubes) [1–3] have recently been shown by numerical simulation to have highly non-linear axial temperature and pressure distributions that are strongly dependent on the bath gas (see Fig. 9 in Ref. [2]). Validations of these temperature and pressure distributions are necessary to accurately determine reaction conditions and extract reliable chemical kinetic data, however, there is little experimental data to verify these predictions.

To make *in situ* measurements in micro-reactors (e.g., Chen nozzles, channels in catalyst supports, etc.), two obstacles must be overcome. First, the reactors are typically opaque (e.g. metal, ceramic, silicon carbide (SiC)), precluding the use of conventional optical diagnostics. Second, the internal diameters are too small to insert a probe without severely disrupting the flow field. Generally, limited observations such as inlet and outlet pressures (P_{inlet} and P_{outlet}), flow rate, and exterior wall-temperature (T_{wall}) are reported. From these observations estimates of residence time and internal gas temperature are made assuming either laminar or plug flow, which may be incorrect, as demonstrated by Guan et al. [2]. In this paper we focus on the development of an X-ray fluorescence (XRF) method of probing gas density at high spatial resolution in heated opaque reactors. The XFR technique is specifically demonstrated for here for Chen type nozzles, but Kr-XRF has also been implemented to measure aerated sprays [4] and with other fluorescent media to examine impinging jet sprays [5] and pressure swirl atomizers [6].

Studies in Chen nozzles have elucidated mechanistic details for complex processes, such as dissociation of biofuel components (e.g. [7,8]) and formation of aromatics [9,10], as a function of T_{wall} . Recently there have been efforts to extend Chen nozzles to high temperature chemical kinetic measurements, [2] for which the gas temperature (T_{gas}), pressure, and residence time in the reactor must be known accurately. Typical estimates of the error in T_{gas} are ± 100 K and T_{gas} may never actually reach T_{wall} in a typical Chen nozzle [2]. Thus, methods of measuring gas properties (pressure, density and temperature) within the reactor are required to validate numerical models that can be used to extract kinetic data.

In the remainder of this paper, non-intrusive XRF measurements are used to probe flow fields within heated SiC tubes and directly measure one state property of the gases, the density. These experiments are an extension of prior XRF studies of gas mixing in jets [4-6, 11] and complement T_{gas} measurements in a miniature quartz reactor by Baraban et al. [12]. Unlike the previously cited studies of

reactions in SiC microreactors, which depend on inferring the gas properties within the tube from measurements of exterior properties (T_{wall} , inlet gas pressure and flow) and conceptual models of the interior flow, the current measurements provide a direct measurement of gas density. In the current work the gas density distribution within a SiC tube and in the nearly supersonic jet eluting from the reactor are measured with XRF. These measurements are complemented with simultaneous X-ray absorption measurements that reveal an intricate internal wall structure and provide data required to correct the raw XRF data. The current experiments provide relative measurements of gas density. Future improvements to the technique are discussed that will yield absolute measurements and higher fidelity data.

Fig. 1. Schematic of the SiC tube and the coordinate system. Translation in Z represents motion along the X-ray beam. After initial alignment the Z location was fixed.

2. EXPERIMENTAL SECTION

The SiC reactor has been described in detail elsewhere [1,2] and only brief details are given here. A schematic is shown in Fig. 1 along with the coordinate system. A SiC tube was suspended in a vacuum chamber, with one end of the tube connected to a gas supply line and the other open to the chamber. Krypton and helium were independently regulated by mass flow controllers and mixed far

upstream of the SiC tube, with a total flow of 187 sccm. The Kr concentration was selected to produce sufficient fluorescence signal and reasonable collection times (30 seconds/point). For measurements inside the tube this was 57% Kr by mass and 3% by volumetric flow (similar to the upper limit used in chemical studies in SiC tubes), although for imaging gases in the freely-expanding jet, where trapping of the photons by the SiC is no longer a concern, it was reduced to 6% by mass. P_{inlet} , Table 1, was monitored by a capacitance manometer and the pressure in the vacuum chamber by a MicroIon gauge (Granville Phillips). The vacuum chamber was evacuated by a turbo pump (550 L/s) to < 0.0001 Pa. Under gas load, chamber pressures were < 0.2 Pa.

Table 1. SiC tube dimensions and experimental ranges^a.

	i.d. / mm	l^b / mm	Δl^c / mm	T_{wall} / K	P_{inlet} /Pa	P_{outlet} /Pa
#1	0.6	30	18	300	3333	0.19
				1090	8666	0.17
#2	1.0	25	13	300	3070	0.19
				800	6400	0.18
				925	6930	0.17

^a Flow rate: 187 sccm. ^b tube length. ^c Electrode separation.

The SiC tube was resistively heated between two electrodes clamped on the exterior of the tube, see Table 1 for spacing. A thin layer of graphite pencil lead was applied to the tube surface to obtain the appropriate resistance to permit heating. Approximately 1 mm of tube extended below the bottom electrode and was therefore not heated resistively. However, this section of the tube was heated by conduction and by heat transfer from the flowing gases, thereby reducing T_{gas} near the exit. The outer wall temperature was measured 3 mm below the downstream edge of the upstream electrode by a type K thermocouple (0.25 mm bead diameter) which was held against the wall with 0.25 mm diameter tantalum wire. The connector wires for the thermocouple and electrical contacts were positioned to minimize obstruction of the incident and fluorescent X-rays. After approximately 24 hours of continuous heating, a hole developed in tube #1. The replacement, tube #2, Table 1, was shorter and the electrodes

thus closer together. Unfortunately, the thermocouple wires interfered excessively with raster scans of tube #2. Thus, the data presented from tube #2 are confined to the freely expanding jet downstream of the tube.

Fig. 2. Schematic of the experimental setup at 7-BM-APS.

2.1. X-ray diagnostics

The experiments were performed at the 7-BM beamline at the Advanced Photon Source (APS), Argonne National Laboratory [13]. A schematic of the setup is shown in Fig. 2. The beamline provided a nearly collimated incident X-ray beam of 15 keV mean photon energy, 1.2% spectral bandpass. Two sets of measurements were conducted using either an unfocused beam or a focused beam. With the unfocused monochromatic beam a series of absorption images (1.3 mm X by 1.75 mm Y) of tube #1 were taken, Fig. 3, using a LYSO:Ce scintillator 100 μm thick and a 5x long-distance optical microscope to image the transmitted X-ray beam.

The majority of the measurements used a focused beam to simultaneously measure absorption and fluorescence from the flowfield and tube. The beam was focused to a cross section of 5 μm by 7 μm full width at half maximum with a pair of 300 mm long Kirkpatrick-Baez focusing mirrors; the beam size remained effectively constant across the experimental apparatus. The incident flux was

approximately 1.6×10^{11} photons/s, which was monitored with a diamond photodiode (52 μm thick) prior to the reactor, Fig. 2. The amount of absorption was determined with a second detector (silicon PIN photodiode, 300 μm thick) after the reaction chamber. The absorption by the flowfield gas was negligible; absorption measurements were used only to better understand the shape of the tube and to provide corrections for the fluorescence data.

Fig. 3. Composite radiograph of 3.7mm of tube #1 (rotated 90 degrees). The upstream edge of the downstream electrode bounds the left edge. The image resolution is 1.29 $\mu\text{m}/\text{pixel}$, Image size 1930 by 2900 pixels. An image of more of the tube is shown in Fig. S1.

When a K-shell (1s) electron is ionized from krypton (14.326 keV K-edge energy [14]) due to absorption of an incident X-ray photon, the ion relaxes either through emission of an Auger electron or through decay of a 2p electron to the 1s level, resulting in the emission of an X-ray fluorescence photon (Kr $K\alpha_1 = 12.649$ keV [15]). The X-ray fluorescence emission is directly proportional to the incident X-ray flux and the path length-integrated density of krypton in the beam path. As such, the fluorescence signal can be used as a measure of the gas density. Since each element emits fluorescence photons at a unique energy, the krypton signal can be separated from the signal from other elements, Fig. 4.

Fig. 4. Example fluorescence signal from 15 keV X-rays penetrating a SiC tube and exciting Kr atoms within the tube.

While argon ($K\alpha_2 = 2.995$ keV [11]) XRF has previously been used as a flow tracer [11], this experiment required more energetic X-rays, both for the incident photons to penetrate the tube and to create fluorescence photons with sufficient energy to escape the SiC tube and reach the detector; fluorescence photons from argon would be completely absorbed by the SiC. In the current experiments, with a nominal tube wall thickness of approximately 500 μm , the transmission through one wall of the tube is 0.325 at 15 keV (absorption coefficient of $7.48 \text{ cm}^2/\text{g}$ [16]), but only 0.153 at the Kr $K\alpha_1$ energy (absorption coefficient of $12.45 \text{ cm}^2/\text{g}$ [16]). Thus, while the photons have enough energy to escape the tube in sufficient numbers to provide adequate signal, corrections are needed for the signal trapping of the fluorescence photons by the tube, as discussed later.

The X-rays enter and exit the vacuum chamber via thin (0.127 mm) polyimide windows, which absorb a small (but known) amount of the X-ray photons. The fluorescence photons were detected with a silicon drift diode, an energy-dispersive X-ray detector, positioned at 90° to the propagation direction of the incident beam in the horizontal plane to minimize the detection of scattered X-rays from the polarized incident beam. The SiC tube was positioned so that the centerline intersected the X-ray beam, and this position was defined as $X=0.0$ mm, Fig. 1. Raster scans were taken from -1.50 mm to $+1.50$ mm at 0.05 mm intervals in the X direction and at 1.0 mm vertical steps over the length of the SiC tube

by maneuvering the vacuum chamber on precision translation stages around the X-ray beam. The scan range in the X-direction extends beyond the walls of the tube into the vacuum chamber to give several measurements of background fluorescence and absorption by gases in the vacuum chamber and the tube walls as well as the gases within the tube. The step size in Y is coarser than required for high resolution but is adequate for the current experiments and is a reasonable compromise between resolution and time. Each complete raster scan took approximately eight hours. $Y=0.0$ mm was defined as the upstream edge of the downstream electrode, Fig. 1. An example signal from the fluorescence detector is shown in Fig. 4, where peaks due to Compton and elastic scattering are observed along with the Kr fluorescence. Additional peaks at lower energies are also seen, but do not interfere with the weak Kr signal. These are primarily assigned to either the stainless steel chamber walls, excited by scattered X-rays, or contaminants of the pencil lead that was used to improve electrical conduction.

There was insufficient range in the translation stages to scan the tube and the jet eluting from it in a single pass. Consequently, after the tube scans were completed the chamber was repositioned. The fluorescence signals are of most use in the jet scans because absorption of 15 keV X-rays by the low density gases in the vacuum chamber is negligible. However, the absorption signals allowed accurate location of the end of the SiC tube. The pressure differential at the tube exit is insufficient for a true supersonic jet to form, and the conditions lie somewhere between an effusive jet and a supersonic jet. To the best of our knowledge, these represent some of the first images of such a low pressure gas jet.

Fig. 5. Example absorption raster scans of SiC tube # 1. The white region at $Y = -12$ mm in (b) is probably the thermocouple, which may have moved in heating the tube. a) $T_{\text{wall}} = 300\text{K}$, $P_{\text{inlet}} = 3333$ Pa, $P_{\text{outlet}} = 0.19$ Pa, b) $T_{\text{wall}} = 1090\text{K}$, $P_{\text{inlet}} = 8666$ Pa, $P_{\text{outlet}} = 0.19$ Pa

Several calibration runs were attempted by operating the tube under incompressible flow conditions (i.e., a slow flow of the He/Kr mix through the tube at atmospheric pressure with helium filling the chamber). However, the buildup of Kr in the chamber gas made extraction of the calibration signal difficult, preventing conversion of the current data to absolute gas densities. For future studies, the SiC tube will be capped and loaded with different pressures of the He/Kr mixture to obtain a calibration curve.

Fig. 6. Krypton fluorescence images captured simultaneously with the absorption images in Fig. 5. The black bands indicate regions that were partially obscured by the thermocouple and tantalum wire. 57% Kr dilute in He by mass.

3. RESULTS AND DISCUSSION

The analysis of the absorption signal is straightforward. The beam intensity, I , at any point is normalized by the intensity when the tube is not present (either a bright field image for imaging with the unfocused beam or well outside of the tube for raster scans). The Beer-Lambert law is then applied. Since a monochromatic beam is used, no beam hardening effects are present. A raster scan image of the absorption signals (in terms of extinction lengths at 15 keV) from the unheated tube #1 ($T_{\text{wall}} = 300$ K,

$P_{\text{inlet}} = 3333 \text{ Pa}$) is shown in Fig. 5a and a corresponding image of the heated tube ($T_{\text{wall}} = 1090 \text{ K}$, $P_{\text{inlet}} = 8666 \text{ Pa}$) in Fig. 5b. In these images the inlet of the tube is at the most negative Y-values, Fig. 1. The images in Fig. 5 essentially show a vertical cross section of the SiC tube. The interior tube walls are far from smooth with dips and bumps that are on the order of $0.1 \times$ tube i.d. in magnitude. It is also clear that the tube walls are not of uniform thickness; unfortunately, in these experiments the thicker wall was pointing towards the silicon drift diode, which resulted in reduced fluorescence signal.

Extraction of the fluorescence signals shown in Fig. 6 is more complex. The fluorescence detector provides a spectrum of the collected photons for each measurement location. The Kr fluorescence peak, Fig. 4, is quite weak but distinct, despite its convolution with the low energy tail of the Compton scattering from the tube wall. This is accounted for by fitting the tail of the Compton scattering and subtracting it from the Kr signal. The resulting fluorescence signal is normalized by the incident flux and corrected for detector dead-time effects. Additionally, there is a low concentration of Kr in the vacuum chamber into which gases elute from the SiC tube. This weak fluorescence signal is measured from the points outside the tube and also subtracted. The absorption signal is then used to model the density of the tube, which is used for two purposes. First, it corrects for the attenuation of the incident beam in the tube wall before it reaches the internal flow. Second, it forms the basis of the error correction for the signal trapping model used to calculate the amount of fluorescent photons that escape the SiC tube. The signal trapping model assumes that the density of krypton is uniform across the tube at a given Y location and that the reactor is a cylindrical hollow tube. However, as Fig. 5 shows, the tube walls are not uniform and the bore is offset by $< 0.1 \text{ mm}$ in X. A 0.1 mm change in wall thickness between the incident X-ray beam and the fluorescence detector amounts to approximately a 30% change in the fluorescence signal. This offset of the tube bore in the X-direction is accounted for in the signal trapping model. The tube bore may also be offset in the Z-direction, although this cannot be determined in these experiments, and a change in wall thickness of in the Z-direction of 0.1 mm reduces the

fluorescence signal by approximately 15%, mainly due to the change in absorption of the incident beam. For this initial study the approximation of the tube shape is reasonable, but for quantitative measurements the signal trapping must be modeled more accurately. This could be achieved by taking tomographic images from which the 3-D shape and structure of the SiC tube at each point of the raster scan could be calculated. This would account for non-uniformities in the tube and remove any assumptions about its orientation. Such experiments were not possible during this study, but recent upgrades to the 7-BM facility will allow tomographic images to be obtained in future work, permitting a more complete signal trapping model to be developed and thus more accurate density maps.

Fig. 7. Plots of Kr fluorescence integrated across X at each Y measurement location. Conditions as in Fig. 6. Vertical scale in arbitrary units.

The fluorescence data from within the tube are shown in Fig. 6. The signal clearly decreases as the gas flows downstream. This is expected and consistent with Guan et al. [2], since the gas is strongly expanding as it passes through the SiC tube. While there is radial information contained in the images, caution should be exercised when interpreting points far from the centerline. The Kr fluorescence signals are quite weak and will decrease in intensity away from $X = 0$ as the effective path length of the incident X-ray beam through the wall of the SiC tube increases and more of the incident beam is absorbed. Furthermore, the tail of the Compton scattering interferes most with the weakest signals and thus introduces greater uncertainty into the data away from $X = 0$. Fig. 7 shows the axial trends in gas density integrated across each cross-section in X. This plot shows more quantitatively the decrease in gas

density as the gas flows downstream. The rate of decrease in gas density is considerably greater for the heated tube than the room temperature tube, indicating a stronger expansion of the gas.

A composite image of the fluorescence signal overlaid on the absorption signal is shown in Fig. 8. It is evident that the flowfield inside the SiC tube is strongly influenced by the wall shape. There appears to be a thin layer of low density gas very close to the wall, possibly a boundary layer, and where pockets appear in the wall there are also regions of apparently low density. These observations should be treated with caution and require confirmation with a more accurate density model of the tube.

Fig. 8. Composite of fluorescence and absorption data from Figs. 5a and 6a. The absorption signal is shown with a single color. The fluorescence signal is displayed as in Fig. 6a. The background has been reduced to a single shade to enhance contrast with the tube walls.

Fig. 9. Example raster scan image of krypton fluorescence in the expanding jet as gases elute from tube # 2. Dashed lines indicate the tube exit diameter. 3% Kr dilute in He by mass. $T_{\text{wall}} = 920\text{K}$, $P_{\text{inlet}} = 6930$ Pa, $P_{\text{outlet}} = 0.19$ Pa.

In Fig. 9 a fluorescence raster scan image is shown of the expanding jet from a heated SiC tube. Absorption measurements were used to locate the end of the tube and radial scans of the jet started 0.1 mm below the tube edge. A strong core of the jet is evident that is slightly shifted to positive X. This is likely due to the core of the tube not being concentric with the tube walls. The strong jet persists for approximately 2 mm before dissipating over several millimeters. Images of the expanding gas have been taken for a number of T_{wall} and additional images are available in Fig. S2. They all show similar structure and indicate that the optimum location of a skimmer for molecular beam sampling, e.g. for time-of-flight mass spectrometry analysis [8], is within 1-2 mm (2-4 tube diameters) of the tube exit, closer than expected for a supersonic jet.

4. CONCLUSIONS

XRF of krypton has been successfully used to obtain gas density maps inside heated, opaque micro-reactors as well as the freely expanding jet eluting from these reactors. These images yielded detailed information about the flowfields within the reactors and will be useful for testing numerical models of the flows. The simultaneous acquisition of absorption and fluorescence signals yielded images that revealed the influence of the irregular wall structure on the gas flow. Furthermore, the absorption measurements provided critical information for developing the density maps from the fluorescence signals. While these results are a promising proof of concept of the usefulness of this technique for non-invasively probing gas densities within opaque environments, these experiments have several deficits that hinder quantitative measurements of density. In particular, it is desirable to reduce the required Kr concentration, obtain information regarding the radial distribution of the gas density, and perform measurements calibrated to absolute density.

Clear paths exist to address these deficits. The largest sources of error are correcting the signal trapping, extracting the weak fluorescence signal from the spectral data, and obtaining absolute

calibrations. Density calibrations can be made, as noted earlier, by capping the tube and filling it with known pressures of gas. To better perform signal trapping, a tube with a smoother interior profile would be helpful, as would higher resolution data regarding the tube density; X-ray tomography would be well-suited to provide such data. A key enabler to reducing the required Kr concentration is to reduce the interference of Compton and elastic scattering in the X-ray spectra with the Kr K α signal. This interference can be reduced by using a Se- or Br-containing filter (termed a Z-1 filter in the X-ray literature) between the reactor and the detector, which preferentially absorbs scattering. The use of polycapillary X-ray optics to examine only a discrete region of the beam path would allow for more spatially resolved measurements while also greatly reducing the influence of scatter when measuring the gas in the tube interior. The use of chemically pure graphite on the outer wall of the tube would also result in a simpler spectrum, simplifying extraction of the Kr K α signal. Finally, use of higher energy X-rays (for example, using Xe as a tracer, with a much higher fluorescence photon energy, 29.5 keV) would result in far less absorption of both the incident beam and fluorescence in the tube wall, resulting in reduced acquisition times. Reducing the distance from the fluorescence detector to the sample will also increase the fluorescence signal, reducing the required integration time and Kr concentration. With these improvements, it seems reasonable to expect density measurements with $\pm 5\%$ precision or better in future measurements.

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Figure Captions:

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Fig. 3. Composite radiograph of 3.7mm of tube #1 (rotated 90 degrees). The upstream edge of the downstream electrode bounds the left edge. The image resolution is 1.29 $\mu\text{m}/\text{pixel}$, Image size 1930 by 2900 pixels. An image of more of the tube is shown in Fig. S1.

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Supporting Information

- S1. Radiograph of tube # 1 with an unfocused beam.
- S2. Raster scan images of tube #2 with T_{wall} of 300 and 925 K.
- S3. Raster scan image of freely expanding jet from tube #2 with T_{wall} of 800 K.