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The Development and Application of Laboratory X-ray Particle Velocimetry and
Thermodynamic Measurement Techniques

by

Jason T. Parker

A dissertation submitted in partial satisfaction of the

requirements for the degree of

Doctor of Philosophy

in

Engineering – Mechanical Engineering

in the

Graduate Division

of the

University of California, Berkeley

Committee in charge:

Assistant Professor Simo Mäkiharju, Chair

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Professor Evan Variano

Summer 2023

The Development and Application of Laboratory X-ray Particle Velocimetry and
Thermodynamic Measurement Techniques

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Jason T. Parker

Abstract

The Development and Application of Laboratory X-ray Particle Velocimetry and Thermodynamic Measurement Techniques

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Flow visualization methods are as old as the study of fluid dynamics, yet there remain major limitations to the types of flows these laboratory methods can measure. In particular, flows with numerous moving refractive interfaces and flows with opaque fluids or environments are all difficult or impossible to measure with conventional optical methods. A wide range of important fluid flows have one or more of these characteristics, including suspensions, bubbly flows, and scalar mixing flows. X-ray image-based particle velocimetry can overcome many of the limitations of optical methods, but has remained difficult to implement in a laboratory setting. This dissertation presents work on the development of X-ray particle velocimetry in the laboratory, with the hope of vastly increasing the accessibility and applicability of these techniques. Additionally, this dissertation presents a study on vitrification thermodynamics using X-ray image-based measurement techniques to demonstrate the broad applicability of said techniques.

New developments in laboratory X-ray imaging have the power to reveal interesting physics in a variety of fields by enabling researchers to see what they once could not. This dissertation hopes to demonstrate the utility of these techniques and inspire new ones.

I would like to dedicate this dissertation to my family, especially to my father, a fellow Golden Bear. Their love and support throughout my doctoral studies has been invaluable.

Contents

Contents	ii
List of Figures	iv
List of Tables	x
1 Introduction to Flow Visualization	1
1.1 A Brief History of Flow Visualization	1
1.2 What Makes A Good Flow Tracer?	3
1.3 Introduction to PIV and PTV	7
2 Theoretical Background For X-ray Imaging	12
2.1 X-ray Spectrum	12
2.2 Absorption and Emission Processes	12
2.3 Scattering, Diffraction, and Refraction	18
2.4 X-ray Imaging Processes	25
2.5 Laboratory X-ray Sources	33
2.6 Applications of X-ray Imaging for Flow Visualization	36
2.7 Chapter Summary	37
3 Simulating X-ray Images for Flow Visualization	38
3.1 Background	38
3.2 Experimental Setup	39
3.3 Simulation Methodologies	42
3.4 Results and Discussion	50
3.5 Conclusions	58
4 Developing General-Use XPV Tracer Particles	65
4.1 Background	65
4.2 Experimental Setup	66
4.3 Tracer Particle Properties	71
4.4 Results and Discussion	78
4.5 Conclusions	85

5	<i>O</i>(1 kHz) Imaging and Tomographic Laboratory X-ray Particle Tracking Velocimetry	88
5.1	Background	88
5.2	Experiment Design	90
5.3	Results and Discussion	101
5.4	Conclusions	105
6	Direct Comparison of Isobaric and Isochoric Vitrification of Two Aqueous Solutions	110
6.1	Background	110
6.2	Methods	111
6.3	Results and Discussion	115
6.4	Conclusions	121
7	Conclusions and Future Steps	123
7.1	X-ray Particle Velocimetry	123
7.2	Thermodynamic State Measurements and Cryopreservation	127
7.3	Final Thoughts	129
	Bibliography	130

List of Figures

1.1	A spherical particle in a unidirectional, uniform flow field of constant velocity after an impulse acceleration at $t = 0$	6
1.2	The basic algorithm for PIV.	8
1.3	PIV and PTV setups are similar except for the particle seeding density. A laser is turned into a sheet, which illuminates a slice of particles. The camera captures the motion of the illuminated particles. From <i>Particle Image Velocimetry – A Practical Guide</i> [3].	9
1.4	A simple PTV algorithm. Many modifications can be made to this algorithm to improve the accuracy of particle tracking.	10
2.1	The electromagnetic radiation spectrum (adapted from [14]), of which X-rays are just a small part. X-ray wavelengths range from a few hundred nanometers to a few picometers.	13
2.2	Fluorescent emission and photoionization are depicted together in this simplified diagram of an atom. They do not necessarily occur together, but may in many situations discussed in the chapter.	14
2.3	Bremsstrahlung occurs when an electron is accelerated by the presence of an atomic nucleus. As we will learn later, an accelerating electron emits electromagnetic radiation.	15
2.4	The Excillum Metaljet E1+ spectrum [16] and a SpekCalc [17]–[19] simulation of the YXLON FXE225.99 source [20]. The Metaljet spectrum shows spikes at the characteristic emission energies. The solid tungsten target FXE225.99 source only shows bremsstrahlung emission because the fluorescent emission of tungsten is not excited at 55 kV.	15
2.5	The mass attenuation coefficient for the element iodine, a common X-ray contrast agent. Note the sharp increase at approximately 33 keV. This is the K fluorescence edge of iodine.	17
2.6	Scattering, diffraction, and refraction are all form of redirected radiation due to the presence of accelerating electrons (either free or bound to atoms).	18

2.7	A diagram of reflection and refraction at an interface. During reflection both rays are in the same medium, so $k_i = k_{reflect}$, implying that $\phi_i = \phi_{reflect}$ as a direct result of the first boundary condition. During refraction, the medium changes, so $k_i \neq k_r$. However, according to the first boundary condition again, we know the component parallel to the interface must be equal. We then arrive at equation 2.29.	24
2.8	Roentgen captured this picture of his wife Bertha's hand on November 8, 1895, signaling the discovery of X-rays.	26
2.9		28
2.10	A diagram of a typical PCD sensor [13]. A voltage is applied across a high-Z semiconductor material; charge bumps collect the emitted photoionization electrons, generating a pulse in the detector circuitry.	29
2.11	A diagram (not to scale) showing how an electron beam focused onto a tungsten target generates X-rays. The size and shape of the tungsten target may vary, but the operating principle is the same.	34
2.12	The heel effect is apparent in this plot of the mean columnar photon count in a flat field image taken with a conical target source.	35
2.13	The X-ray source spectrum from the Excillum Metaljet E1+ liquid metal jet X-ray source [16] and a SpekCalc [17]–[19] simulation of the YXLON FXE225.99 solid tungsten target source.	36
3.1	A diagram of the X-ray imaging system. The SOD and SDD are 1 cm and 47.6 cm, respectively.	40
3.2	(a) A slice of the nano-CT scan captured by project collaborator Till Dreier at Lund University. (b) The measured and fit PDF of the particle radius of the AGSF-33 tracer particles.	42
3.3	The AGSF-33 settling speed magnitude distribution. Glycerine slows the particles due to its higher viscosity. Some particles sink, but the magnitude of the settling/rising speed is shown here to accommodate the log-scale axis.	43
3.4	SpekCalc source spectrum of a 22.5° half-angle conical tungsten target at 55 kV with a 1 mm thick aluminum window.	45
3.5	A point source generates rays along which the Beer-Lambert law is evaluated at the detector plane. The detector itself is not in the geometry. Here, an AGSF-33 particle is placed on the front surface (surface towards source) of a glass slide. Figure not to scale.	46
3.6	The MCNP geometry is similar to the BL simulation setup of figure 3.5, but includes a CdTe-detector crystal of finite thickness (omitting housing and electronics for simplicity) and accounts for the finite X-ray source spot. Figure not to scale.	48
3.7	An incoming photon can enter one pixel, deposit sufficient energy to be counted, then scatter to another pixel where it is counted once more.	49

3.8	A single photon in an MCNP simulation is detected in two separate pixels. Multi-detections such as this one lower the image SNR.	49
3.9	A sample image from the experiment images collected at 55 kV, 500 μ A for a 100 ms exposure time. The filtered image shows higher particle contrast, which would improve the PIV correlation peak or the PTV centroid localization. . . .	51
3.10	A normalized BL simulated Poisson-noisy image. The simulated source flux was matched to the experimentally calculated value. The average pixel intensity is 4,661 photons.	52
3.11	Gaussian blur degrades the SNR of the BL simulated image. In the future, a finite source focal spot may be simulated by adding Gaussian blur. The average pixel intensity for all images is 4,661 photons.	53
3.12	The MCNP simulation image of an AGSF-33 particle on a glass slide. The MCNP image shows blurring from the finite focal spot. The average pixel intensity is 5,074 photons.	54
3.13	The MCNP, BL, and experiment image photon detection distributions. A small peak to the left in the MCNP distribution is due to the depressed photon counts in the MCNP edge pixels. MCNP is more accurate than BL, but still does not match experiment exactly.	54
3.14	The image SNR from experiments and BL simulations as a function of the source voltage. The MCNP SNR for the 55 kV image is shown for comparison.	56
3.15	The image contrast and noise intensity are plotted as a function of the source voltage. BL underestimates the noise intensity, likely due to the simplistic scattering model.	57
3.16	An MCNP-simulated image of multiple particles on a glass slide. The imaging settings are identical to the single-particle image.	57
3.17	The BL-simulated image of multiple particles on a glass slide. The settings are identical to the single-particle simulations.	58
3.18	The error between the hand-calculated polychromatic spectrum and the MATLAB-simulated spectrum using our BL code is less than one millionth of a percent. (At low photon energies, the number of photons at that energy approaches zero, hence the singularity.)	60
3.19	The scenario with the maximum lost area given a continuous, non-convex shape with dimensions greater than or equal to a pixel side-length. A sub-pixel is assumed to be uniformly covered or uncovered based on the state of its center. .	61
3.20	The maximum lost area scenario for a hole with a diameter greater than or equal to the pixel side length.	62
3.21	As the number of sub-pixels increases, the maximum area lost decreases rapidly. The homogeneity assumption criteria is met for $N \geq 121$ (11×11).	63
3.22	Example particle history tracks from an MCNP PTRAC file.	64
4.1	The experimental setup (not to scale). A syringe pump pushes glycerol and tracer particles through a vertically mounted pipe.	68

4.2	Particle A will be magnified less than particle B because it is farther from the X-ray source. Particle A will also appear to move slower than particle B.	69
4.3	The depth-wise direction is towards the detector. The analytical velocity profile is depth-averaged.	71
4.4	The probability distribution of the terminal speed magnitude for sieved, uniformly distributed, 45-53 μ m AGSF-33 tracer particles in the Stokes drag regime in water and pure glycerol at 23.7°C. The distribution in water is effectively symmetric about zero; only the velocity magnitude is shown here to accommodate a log-axis plot to highlight the water-glycerol distribution contrast.	73
4.5	The CW particle diameter distribution – measured using a non-aqueous based dispersion (ISO 13320) with a Saturn DigiSizer – provided by Particle Testing Authority. While the particles are polydisperse from the factory, sieving and future manufacturing improvements can reduce the polydispersity.	74
4.6	An SEM image of the CW tracer particles used in these experiments prior to sieving. Note the significant polydispersity prior to sieving.	75
4.7	In this SEM image of a broken particle the coating thickness can be measured. However, we find that the coating thickness varies significantly due to the new manufacturing process not yet being fully refined. The particle shown is also non-spherical, another challenge in manufacturing new tracer particles.	75
4.8	The Stokes terminal speed probability distribution of the CW tracer particles in water and pure glycerol.	76
4.9	a) CW tracer particles and b) AGSF tracer particles imaged with a 15 ms exposure time, 500 mm SDD, 38 mm SOD, a 55 kV acceleration voltage, and a 500 μ A target current. The CW tracer particles demonstrate higher contrast than the AGSF-33 tracer particles for the same exposure time. A cluster of AGSF-33 particles is circled as an example. Un-clustered AGSF-33 particles should nominally be the same size as the CW particles.	79
4.10	The measured SNR for the AGSF-33 and CW tracer particles. The CW SNR is shifted higher as a result of the higher contrast compared to the AGSF-33. Note that due to clustering of most of the AGSF-33 particles the true SNR of individual AGSF-33 particles is likely lower than observed.	80
4.11	The DAVP as measured by the CW and AGSF-33 tracer particles compared against the analytical DAVP. The shaded regions are the measurement variation between the 15.9 and 84.1 velocity percentiles.	81
4.12	The measured x -component velocity, $v(x, y)$, for the CW and AGSF-33 tracer particles. The CW tracer particles, being more localizable and less clumped, exhibit smaller measurement variance compared to the AGSF-33. The shaded regions are the measurement variation between the 15.9 and 84.1 velocity percentiles.	82

4.13	The velocity vectors overlaid on the hundredth frame of the (a) CW and (b) AGSF-33 footage. The greater variability of the AGSF-33 x -component velocity due to variability of the apparent particle centroid location is apparent, whereas the CW particles offer better contrast and are more robustly localized. The color scale is inverted for PTV processing in DaVis.	83
5.1	The X-ray imaging setup. The flow setup is placed in front of the X-ray source aperture. A LAB Motion RT100S air bearing rotation stage controls rotation of the pipe.	91
5.2	(a) Schematic of the flow setup where the test section is connected to supply lines via a swivel joint that rotates freely. (b) The inside of the flow setup shows a brass pipe mounted concentrically within the plastic pipe. The Taylor bubble and KI jet are injected via this brass pipe.	92
5.3	(a) Multiple batches, such as this one, of CW particles are examined with a nanofocus X-ray source (NanoTube N3, Excillum AB). (b) The prototype nature of these particles is evident from the various coating thicknesses, broken particles, and polydispersity.	95
5.4	The measured and fit particle size distribution. We fit a log-normal distribution to the particles, which exhibit clear polydispersity.	96
5.5	The settling speed distribution of the CW tracer particles in water and glycerine. Using glycerine dramatically reduces the spread in settling speeds compared to water.	97
5.6	The X-ray flux from the MetalJet E1+ LMJ source used [16] versus a traditional solid anode microfocus X-ray source [17]–[19]. A LMJ is able to withstand greater focal spot power density. As a result the spectrum is $20\times$ brighter for the same focal spot size and source acceleration voltage.	98
5.7	A $5 \times 5 \times 5$ median-filtered vertical slice through the central plane from a reconstructed Taylor bubble experiment. Only the pipe inner diameter is shown here for clarity. The Taylor bubble exhibits some motion blur manifested as a roughly $200\text{ }\mu\text{m}$ thick “halo” around the bubble, while in reality the interface is sharp. Fast moving particles may similarly warp, but many particles do not. There is a sufficient number of unwarped particles to successfully track them.	101
5.8	The depth-averaged velocity profile of a pipe flow with a 45 mm/s center line speed ($Re = 0.40$) agrees well with the expected profile. The gray region depicts the variation of the velocity measurement between the 16^{th} and 84^{th} percentiles to approximate the 2σ error bounds for a non-Gaussian-distributed variable. The data are normalized by the theoretical center line speed.	102
5.9	The TXPTV radial flow profile from a single scan shows good agreement with the theoretical Poiseuille profile. The 16^{th} and 84^{th} percentiles are shown to approximate 2σ error bounds for a non-Gaussian-distributed variable. Unlike 2D-projected XPTV, TXPTV can measure the true radial flow profile. The data are normalized by the theoretical center line speed, $U = 0.08\text{ mm/s}$	103

5.10	The particle pathlines flowing around the Taylor bubble moving through the field of view. The bubble-glycerine interface is shown in blue. A black circle on the bottom indicates the theoretical pipe interface.	104
5.11	The evolution of the jet, but with the intensity values converted to KI-water solution concentration. The pipe wall is masked out for clarity. The reconstructed slices are Gaussian filtered with an $11 \times 11 \times 11$ kernel and a 7 pixel standard deviation.	106
6.1	Vitrification chambers enabling control of boundary conditions. A two-piece assembly with metal-on-metal sealing surfaces generates the isochoric (rigid) confinement. Replacing the rigid plug with a movable piston produces isobaric conditions while ensuring equivalent thermal and surface conditions as the isochoric system. This configuration was employed in the trials with CVS1. Critical cooling rates of $\leq 5^\circ\text{C}/\text{min}$ are easily achievable with 50 wt% DMSO, and the solution is left uncovered in order to allow the free interface to deform. In this configuration, the chamber is capped with Kapton tape to prevent intrusion of liquid nitrogen during cooling.	113
6.2	The experimental setup consisting of a cone beam X-ray CT source and a photon counting detector. A vitrified chamber is placed into the beaker that sits atop a stack of three linear stages and one rotation stage. The beaker is partially filled with liquid nitrogen below the sample to ensure the chamber does not warm over the duration of the CT scan.	114
6.3	CT scan voxel intensity histograms for entire reconstructed volume of CVS1 and 50 wt% DMSO vitrified under isochoric and isobaric conditions, respectively. These examples show the contrast between the aluminum walls, the vitrified solution, and the vapor. The histograms are aligned based on the aluminum peaks.	116
6.4	CT scan voxel intensity histogram for a single unfiltered reconstructed slice of the CSV1 solution vitrified under isochoric confinement. Peaks are visible for the three present phases: aluminum, vitrified CVS1, and ice. Inset: the unfiltered reconstructed slice showing globular ice formation.	117
6.5	Reconstructed CT scans of 50 wt% DMSO in isobaric (left) and isochoric (right) systems showing vapor cavity formation. Vitrified solution and vapor phases are segmented and shown in purple and blue, respectively.	118
6.6	Reconstructed CT scans of CVS1 in isobaric (left) and isochoric (right) systems showing partial ice formation. Vitrified solution and ice phases are segmented and shown in purple and blue, respectively.	119
6.7	Segmented ice phases from reconstructed CT scans of vitrified CVS1 in isochoric (top) and isobaric (bottom). Globular ice formation in the isochoric system is reminiscent of bubbles formed during vapor cavitation.	120

List of Tables

3.1	BL, MCNP, and experiment image photon detection distribution moments. . . .	52
4.1	The Stokes number and terminal velocity ratio for the AGSF-33 and CW tracer particles for flow $Re = 0.0018$. Note that all dimensionless numbers are much less than unity in glycerol, so the particles can be expected to accurately trace the flow. AGSF-33 particles would also work as flow tracers in water at these speeds, but the prototype CW particles considered here would need to be used in flows faster than $2000 \mu m/s$ to neglect buoyancy effects in water.	78
5.1	The experiment parameters for each of the three experiments that are conducted. *At the nozzle exit; we measure roughly 19 nozzle diameters downstream.	93
5.2	The mean settling speed to center line speed (U) ratio and Stokes number of the CW tracer particles that we use in these experiments. For the speed ratio, we compare to the slowest flow speed we measured ($U = 0.08 \text{ mm/s}$); for the Stokes number we use the highest flow speed we measured ($U = 45 \text{ mm/s}$). This is to ensure our Stokes number and buoyancy measurement bias are low for all of our flow conditions.	96
5.3	The expected maximum particle blur due to pipe rotation and particle motion in units of nominal particle diameter, d_p ($50 \mu m$). Maximum expected flow velocities and radial distances are used to calculate these values. Values greater than one indicate that some blurring is possible.	100
6.1	Average cooling rates ($^{\circ}C/min$) during plunge cooling in liquid nitrogen of CVS1 and 50 wt% DMSO under isochoric and isobaric conditions.	114
6.2	Ice volume fractions (%) of ice for the CVS1 and vapor volume fractions (%) for 50 wt% DMSO under isochoric and isobaric conditions.	121

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Chapter 1

Introduction to Flow Visualization

It is important to first put this dissertation in its proper context. This work is the latest contribution in a long lineage of flow visualization advancements. In this chapter, I will provide an overview of the history of flow visualization and the current scientific environment in which this work was conducted. Then, I will introduce what makes a good flow tracer. Lastly, I will discuss the basics of particle image velocimetry (PIV) and particle tracking velocimetry (PTV) algorithms.

1.1 A Brief History of Flow Visualization

Sight is perhaps the primary human sense. As such, visualization is invaluable to scientific experimentation. The itch of curiosity is best scratched when one can *see* what happens. The itch only grows stronger when the subject is invisible to the human eye. The motion of a uniform fluid is one such subject.

The human eye requires contrast to detect motion. A uniform fluid offers no contrast by which the human eye can track its motion, or even whether motion exists. Flow visualization is the art of deliberately adding contrast to a fluid to observe its motion. These contrasting elements, commonly referred to as tracers, take many forms; among them are dyes, particles, smoke, droplets, and bubbles. There are other means of introducing contrast, such as in schlieren photography, which uses density differences that create a contrast in refractive index. The inventive ways in which fluid dynamics experimentalists have introduced contrast for flow visualization is astounding; they are not comprehensively covered in this work. For a comprehensive treatment of the subject, the reader is referred to *The Handbook of Experimental Fluid Mechanics* by Tropea et. al. [1]. This thesis will focus on image-based measurements that make use of tracer particles to introduce contrast.

Although rudimentary flow visualization such as drawing and naked eye observation existed prior to the advent of photography (Leonardo Da Vinci, in particular, conducted fascinating flow visualization experiments of the vascular system [2]), the beginning of modern flow visualization starts with photography. Photographs provide the ability to:

1. store and reproduce results,
2. capture a dynamic process at an instant in time, and
3. rigorously measure distances and sizes captured at that instant in time.

In these three capabilities we find the seeds of quantitative flow visualization. At the advent of the photograph, exposure times – the time that the sensor is exposed to collect an image – were too long to capture fast-moving processes without being blurred beyond comprehension. The first photographs, taken by Joseph Nicéphore Niépce in the first half of the 19th century, required exposure times of hours. Taking pictures of a person sitting still would have been impossible, let alone taking pictures of moving fluids.

It was not until the end of the 19th century and the beginning of the 20th that photography could start to capture fluid motion. Advances in film chemistry, shutter mechanisms, and lighting all combined to enable pioneers such as Prandtl, Mach, Marey, and Ahlborn to conduct some of the first flow visualization experiments with photography [3]. Ahlborn even captured sub-surface cross section footage by using slits of light [4]. Originally, these experiments used doubly-exposed films to capture the motion of tracers between two instants in time. Then, the images were optically correlated to ascertain the fluid motion. These early experiments, however, could only yield qualitative results such as the shape of streamlines and the location of stagnation points. The techniques to extract quantitative data, i.e., fluid velocity, did not yet exist. That would have to wait until the advent of lasers and modern computers in the latter half of the 20th century.

The invention of the laser is another turning point in the development of quantitative flow visualization. Lasers opened up the possibility of new fluid flow measurement techniques such as laser Doppler velocimetry (LDV). Although LDV and other laser-based techniques can measure tracer velocity, they do not use images as their raw data, instead relying on the properties of coherent laser light, the Doppler effect, or both. They are not image-based flow visualization techniques. This dissertation focuses on image-based measurements, so the reader is again referred to *The Handbook of Experimental Fluid Mechanics* [1] for more information on other laser-based fluid measurement techniques. For image-based measurements, the laser has been used to illuminate the tracer particles in precisely timed increments and in thin, two-dimensional sheets to isolate planar velocity components. Short, bright illumination times and thin illumination sheets improve the accuracy of the flow visualization measurements. Short illumination times reduce blurring and velocity accuracy, while thin illumination sheets better isolate the planar velocity components.

Modern computers brought with them the speed and precision necessary to execute digital image correlation algorithms that could extract quantitative measurements. With the advent of digital computers, single-exposure images became more commonly used for their suitability for determining direction of flow in digital image correlations. In 1983, Sutton et. al. [5] published the first displacement measurement of a deformed body using digital image correlation. They calculated the displacement of random speckles spray painted on

a plexiglass beam. It is not a large leap from random speckles on a beam to randomly distributed tracers in a fluid. The principle of digitally correlating two images between which a displacement occurred is the same in both cases. Other algorithms can track the motion of individual tracers. Section 1.3 will discuss the digital image correlation algorithm and particle tracking algorithm in greater detail.

The paradigm of flow visualization remains remarkably unchanged since the beginning of the 1980s. Computers are more powerful, lasers and tracer particles are better, and cameras have more pixels and higher frame rates. The foundation upon which these advancements have been built, though, has not significantly changed. Despite all of the advancements over the last two centuries, flow visualization still suffers from a number of limitations. Chief among them is what is dubbed “optical accessibility.” In other words, for the laser light to illuminate the tracers and for the camera to capture their motion, the system must be transparent to visible wavelength light. While this is not a challenge for transparent fluids in transparent containers, such as water in acrylic, there are many systems that do not meet this criterion. Blood, for example, is opaque. Another challenge for visible wavelength light is that refraction distorts it. If, for example, the fluid system contains two or more fluids, one of which is dispersed throughout the other, then the laser sheet will refract across the multiple moving boundaries between the two fluids. Refraction will distort the laser sheet too much, making it unusable for illuminating the tracers. One example of this refractive opacity is the air-water mixture coming out of a sink faucet appearing white. Refractive opacity is commonly observed in multiphase systems.

The next paradigm of flow visualization must address the limitations of the current paradigm. X-ray light is both able to penetrate opaque objects and experiences negligible refraction in many cases. X-ray imaging is an ideal candidate for the next frontier of image-based flow visualization. X-rays, and to some extent gamma rays, have been used for flow visualization for a few decades [6]–[11]. Most of these systems are used to observe dynamic structures of multiphase flows or conduct void fraction measurements, the latter of which will be discussed in detail in chapter 2. However, X-ray imaging has not commonly been used for particle-based velocimetry. There are a number of reasons why, including: 1) low spatiotemporal resolution imaging, 2) lack of quantitative general-use tracer particles, and 3) the limitations of 2D-projected imaging. Overcoming these challenges to provide a path towards wide applicability is the subject of this dissertation.

Chapter 2 will introduce X-ray physics and X-ray in the context of X-ray image-based flow visualization. The remainder of this chapter will introduce the basics of image-based flow visualization techniques that use tracer particles.

1.2 What Makes A Good Flow Tracer?

There are many different types of flow tracers, each with their own criteria for being “good”. In this section we will focus on tracer particles. These could be droplets, bubbles, or solid particles that are manufactured or formed in a controlled manner and seeded into the flow.

I will refer to all of these as particles. This precludes natural particulate matter that can be used in ultrasonic velocimetry, for example. Broadly speaking, in order to be good flow tracers, particles must be:

1. Density-matched with the fluid,
2. Small compared to the flow features,
3. Spherical (unless also capturing vorticity measurements),
4. Able to generate contrast with the fluid.

The reason for item one is straightforward: a particle that floats or sinks is experiencing an acceleration that the fluid is not, which adds error to the velocity measurement. A particle that is not density-matched with the fluid will also have inertia under acceleration that the fluid does not. Item four is necessary because a measurement is not possible with an invisible particle. Items two and three are less straightforward.

Item two refers to the spatial resolution of the measurement taken with a particle. If a particle has a diameter d_p , then the highest spatial resolution possible is $d_p/2$. No other particles can fit within the radius of another particle, so it would be impossible to take a measurement with another particle in that space. Another way to consider this limitation is if a particle is up against a wall. The particle is unable to measure the velocity within $d_p/2$ of the wall. A smaller particle maximizes the spatial resolution.

Furthermore, a smaller particle will be able measure larger velocity gradients in the flow. If a large particle experiences different velocities across its diameter, which velocity is it tracing? As we will see later, good tracer particles are assumed to experience a uniform translational acceleration in a uniform velocity field. In other words, they are smaller than the smallest length scale in the flow. A smaller particle, however, may be more difficult to detect or may not be neutrally buoyant because it is more dense. Particle size should be balanced against the other requirements for a good flow tracer.

Item three essentially means that the particle behavior should behave isotropically (independent of direction). A spherical particle will respond the same way regardless of the direction of the flow. A non-spherical particle will experience a different acceleration based on its orientation and the flow direction. Non-spherical particles may also exhibit an oscillating or random contrast in imaging.

Stokes Number

A commonly used metric for determining how well a particle traces the flow is the Stokes number. The idea underlying the Stokes number is simple: if the response time of the particle to a change in acceleration is smaller than the shortest characteristic time scale of the flow, then the particle can be treated as a responsive flow tracer. While this idea does not explicitly address sphericity, it is implicit. This idea must hold true regardless of flow direction, necessitating a spherical particle. The ratio of the response time to the

characteristic time scale of the flow is the Stokes number, shown in equation 1.1 where τ_p and τ_f are the particle response time and flow time scale, respectively.

$$St = \frac{\tau_p}{\tau_f} \quad (1.1)$$

To get an estimate of the particle response time, we will make some simplifying assumptions:

- The particle occupies a uniform velocity field. This implicitly assumes the particle is much smaller than any velocity gradients.
- The particle experiences an impulse acceleration at time $t = 0$.
- The flow is unidirectional. Further, the same must hold true in all directions, necessitating a spherical particle.
- The particle is neutrally buoyant.
- The relative velocity between the particle and the surrounding flow is small enough that the particle experiences Stokes drag.

The final item refers to a purely viscous flow that occurs at low Reynolds numbers ($Re \ll 1$). Stokes drag is given by

$$F_D = 3\pi\mu d_p(U_f - U_p), \quad (1.2)$$

where μ is the dynamic viscosity of the fluid, U_f is the velocity of the fluid, and U_p is the velocity of the particle. A diagram of the particle in the flow field can be seen in figure 1.1.

Newton's second law applied to the particle results in the following equation of motion:

$$F_D = 3\pi\mu d_p(U_f - U_p) = \frac{\pi}{6}\rho d_p^3 \frac{dU_p}{dt} \quad (1.3)$$

with the initial condition $U_p(t = 0) = 0$ without loss of generality. Here, the density $\rho = \rho_p = \rho_f$ because the particle is neutrally buoyant. Next, we define a slip velocity $U_s = U_f - U_p$, so the equation of motion becomes

$$\frac{dU_s}{dt} + \frac{18\mu}{\rho d_p^2} U_s = 0 \quad (1.4)$$

$$U_s(t = 0) = U_f \quad (1.5)$$

Solving the above simple ODE returns an exponential rise or decay function depending on whether the step change in fluid velocity is positive or negative. Regardless, the particle response time can be taken as the exponential time constant

$$\tau_p = \frac{\rho d_p^2}{18\mu}. \quad (1.6)$$

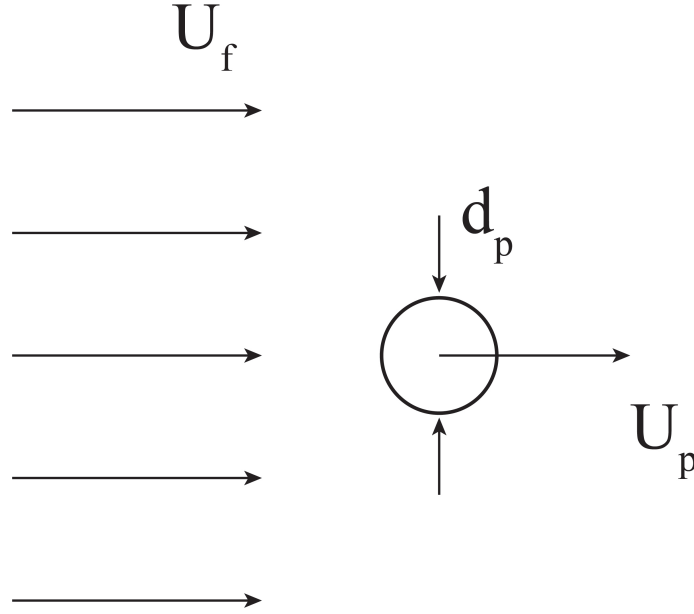


Figure 1.1: A spherical particle in a unidirectional, uniform flow field of constant velocity after an impulse acceleration at $t = 0$.

The Stokes number is then

$$St = \frac{\rho d_p^2}{18\mu\tau_f}. \quad (1.7)$$

To accommodate consideration of particle-fluid density differences, the Stokes number is often written as

$$St = \frac{|\rho_f - \rho_p| d_p^2}{18\mu\tau_f}. \quad (1.8)$$

In order for a particle to be considered a faithful flow tracer, equation 1.8 must be much less than unity. Note that this only holds under the assumptions listed above, which pose constraints on the particle design in order for it to be an accurate flow tracer.

Equation 1.8 has some interesting features. A smaller density difference will reduce the Stokes number linearly, but decreasing the particle diameter will reduce it quadratically. That means, under certain circumstances, if the diameter is sufficiently small, density-mismatched particles can still function as accurate flow tracers. In this case, any bias errors due to buoyancy effects must be carefully characterized and should be much lower than the flow speed. Nevertheless, this feature is useful since many particles cannot be perfectly matched to the fluid density. They can still remain useful tracers, though!

1.3 Introduction to PIV and PTV

Particle Image Velocimetry (PIV) and Particle Tracking Velocimetry (PTV) are two of the most common forms of quantitative flow visualization. Each has numerous spin-offs and sub-genres (stereo-, tomo-, micro-, etc.) that we will not touch on here. This section will instead give an overview of how these algorithms work conceptually and some good rules of thumb for each.

Particle Image Velocimetry

PIV has its origins in digital image correlation, as mentioned previously. Conceptually, the idea is to find how much a small group of particles has moved between two frames. The process for doing so is:

1. Two frames are captured in quick succession with a known time between them, Δt .
2. These frames are then split up into smaller pieces called windows. The window locations and sizes are the same for both frames.
3. Then, the corresponding windows in each frame are cross-correlated. This finds the point of maximum overlap of the particles in each window.
4. The point of maximum overlap has likely shifted from the center of the window. That shift is how many pixels the group of particles has moved, and in what direction.
5. With a known pixels-to-distance conversion (calibration) and the time difference Δt the shift in pixels can be converted to a velocity.

This process is repeated for each window to get the velocity of the particles in each of the windows. What we are left with is a grid of velocities – a powerful tool for comparing to numerical results.

Figure 1.2 depicts the PIV algorithm. Notice how the algorithm above only applies to 2D images. That is by design. PIV is traditionally conducted in two spatial dimensions and captures two in-plane velocity components (2D2C). While there are many versions of PIV that enable three-dimensional, three-component measurements, we will focus on 2D2C measurements here for simplicity.

If we take a 2D image, though, we will see particles along the entire depth-wise direction of the image. How do we make sure that particles in one plane are not correlated with particles in another at a different depth? That is where lasers and imaging optics come in. A schematic showing a typical PIV setup is shown in figure 1.3. A laser is formed into a planar sheet using laser optics. When the camera captures an exposure, the laser fires, illuminating only the particles that are in the plane of the laser sheet. The imaging optics of the camera are set such that only the illuminated plane is in focus.

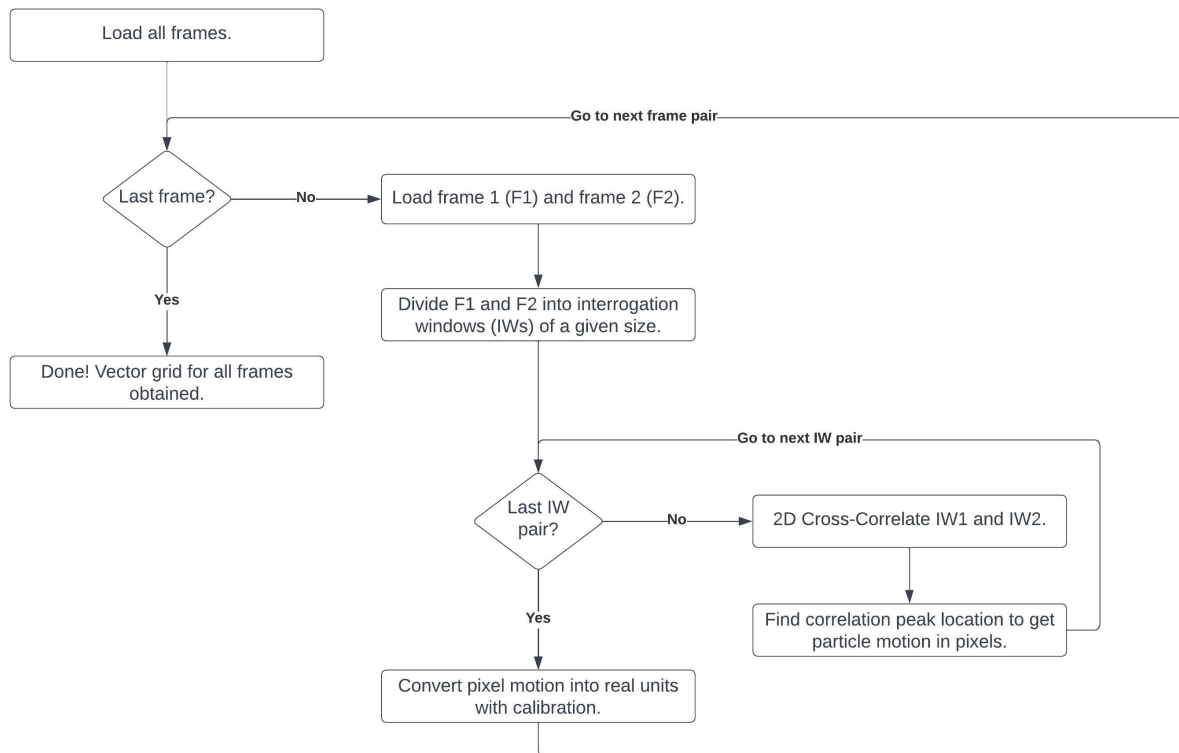


Figure 1.2: The basic algorithm for PIV.

Since PIV uses cross-correlations, the particle seeding density (the number of particles per unit area) should be high, but not so high that the particles become indistinguishable. Generally, a good rule of thumb is to have about 10 particles per window. If there are too few, the uncertainty of the velocity in the window may be too high. If there are too many, particles may get confused for one another.

One drawback of PIV is that it may struggle to deal with flow regions with large velocity gradients. Since PIV requires that the images be broken into windows for cross-correlating, and each window corresponds to one velocity measurement, the spatial resolution of the measurement is much lower than the spatial resolution of the image. Near-wall measurements and large gradient flows are difficult to measure with PIV. There are means to overcome this, such as reducing the field of view to increase the spatial resolution of the velocity measurement, but ultimately PIV is limited in this way.

Particle Tracking Velocimetry

PTV is an alternative to PIV that addresses some of the limitations of PIV. It uses a nearly identical hardware setup to 2D2C PIV. Instead of discerning the motion of small groups of

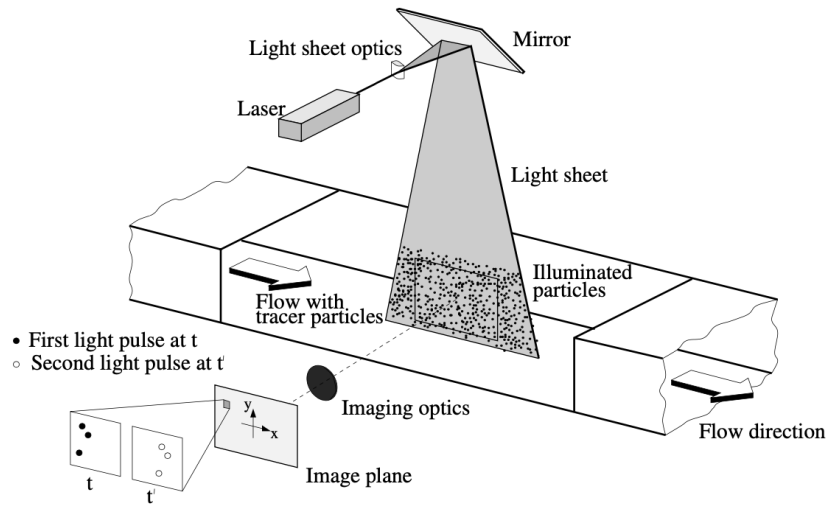


Figure 1.3: PIV and PTV setups are similar except for the particle seeding density. A laser is turned into a sheet, which illuminates a slice of particles. The camera captures the motion of the illuminated particles. From *Particle Image Velocimetry – A Practical Guide* [3].

particles, though, PTV tracks the motion of individual particles. This greatly increases the spatial resolution of the velocity measurement to half the size of the particle, as discussed in section 1.2. The drawback, however, is unpredictable measurement locations. Measurements are only captured along the pathlines of the particles. PIV, on the other hand, captures a grid of velocities with regular spacing across the entire image.

One of the simplest algorithms for PTV is the Nearest-Neighbor algorithm (NNA), depicted in figure 1.4. The NNA is as follows:

1. Capture two sequential frames with known Δt .
2. Identify the centroids of all of the particles in each frame.
3. For each centroid in the first frame, find the nearest centroid in the second frame and label them as the same particle.
4. With a known pixel-to-distance (calibration), convert the distance between the particle centroid in the first and second frame to a displacement. Calculate the velocity with the known Δt .

There are numerous ways to improve the algorithm above, but in its simplest form, that is the NNA. One easy way to improve the algorithm above is to conduct PIV first to estimate the local velocity. Then, shift the centroids from the first frame by the PIV velocity times Δt and search a neighborhood around that shifted centroid location. This estimates where the particles in that window should be in the second frame, making it easier to find the particle

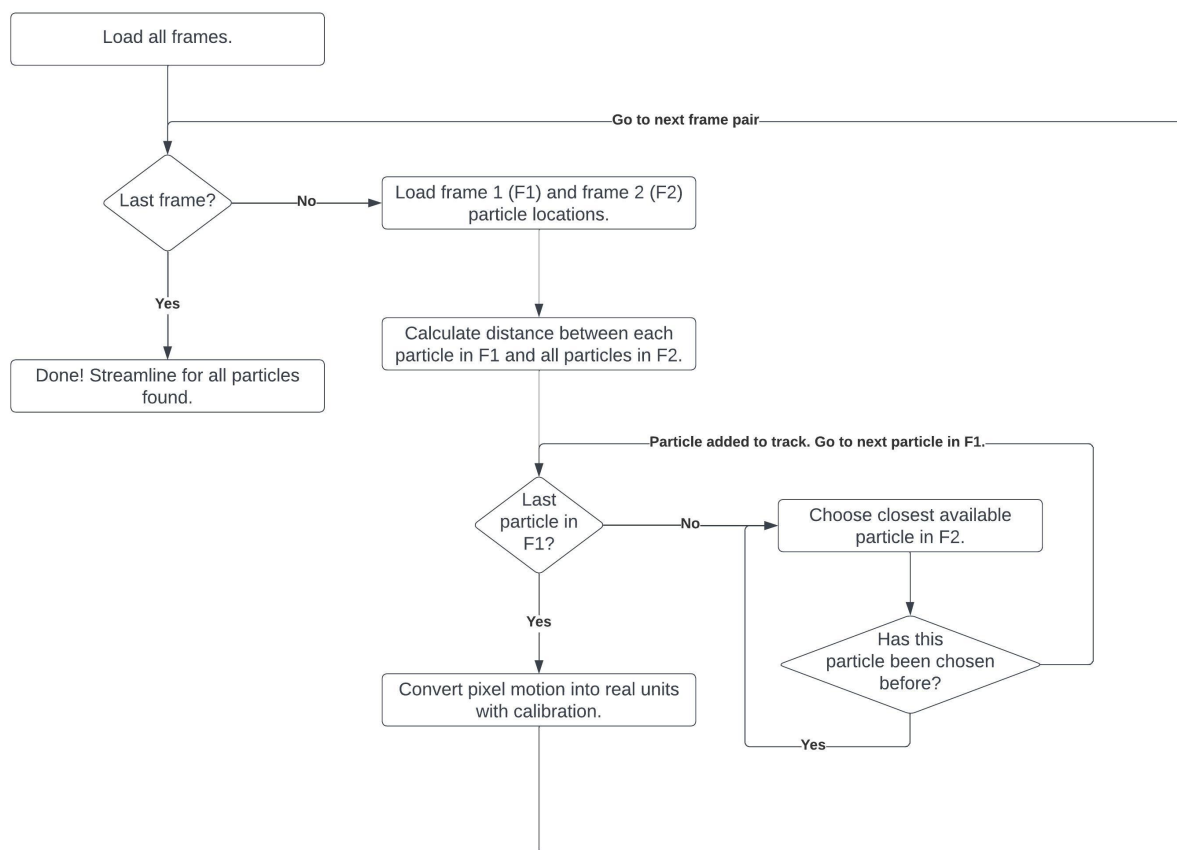


Figure 1.4: A simple PTV algorithm. Many modifications can be made to this algorithm to improve the accuracy of particle tracking.

in the next frame. Another way to improve the algorithm is to add more factors besides shortest distance to a cost function. This could include particle size, particle acceleration, sphericity, or other characteristics. The centroid with lowest cost function is then chosen as the “nearest neighbor”.

Since PTV tracks individual particles, if the seeding density is too high the particles will get confused for one another. Typically, PTV has a smaller seeding density than PIV. PTV is best used for flows with large velocity gradients or where the spatial resolution of the velocity measurement must be high. Converting the pathlines into an organized grid is difficult, though, often requiring long duration measurements to reach statistical certainty and to span the physical domain. In such cases, PTV is temporally limited.

What about X-ray PIV and PTV?

X-ray PIV and PTV (XPIV and XPTV, respectively) are techniques contained within a class of measurement called X-ray Particle Velocimetry (XPV). XPIV and XPTV are both based on the same principles described in this chapter. There are new constraints due to the challenges of X-ray imaging, though. For example, both PIV and PTV rely on a laser to briefly illuminate particles in a plane of interest. X-ray lasers and X-ray optics are not commonly available in laboratories, though, so illuminating tracer particles in a thin sheet of X-ray light is infeasible. Furthermore, X-ray image contrast is typically generated with absorption of transmitted light, not scattered light as with visible light. Capturing the X-ray light scattered from tracer particles in a thin X-ray sheet would be a dark image indeed. As a result, if one only has a single X-ray source and detector (typically due to cost limitations) and no means to capture multiple angles of the flow, the only option is to take depth-averaged measurements. This is known as 2D-projected XPIV/XPTV measurements. In almost all cases, this is problematic since the flow velocity changes over the course of the image depth. Tomographic and stereographic XPIV and XPTV are necessary to address the shortcomings of X-ray imaging for flow visualization. These will be discussed in detail in chapter 5.

Additionally, tracer particle design becomes more challenging because they have to simultaneously attenuate X-rays and be accurate flow tracers. Chapters 3 and 4 will touch on the challenges associated with tracer particles.

The next few chapters of this dissertation seek to address the challenges associated with XPV with the ultimate goal of introducing these as usable laboratory measurement techniques.

Chapter 2

Theoretical Background For X-ray Imaging

This chapter will provide a crash-course introduction to the physics of X-rays and how it pertains to X-ray flow visualization and imaging technology. A lot of this material is adapted from Prof. David Attwood's *X-rays and Extreme Ultraviolet Radiation* course at UC Berkeley. For a detailed treatment of these topics, I refer the reader to *X-Rays and Extreme Ultraviolet Radiation: Principles and Applications* by Dr. Anne Sakdinawat and Prof. David Attwood [12]. That book provides thorough explanations of basic X-ray physics as well as applications in radiation producing machinery and synchrotrons. Later portions of this chapter are drawn from *Handbook of X-ray Imaging Physics and Technology* by Russo et al. [13].

2.1 X-ray Spectrum

First, what are X-rays? X-rays are a range of wavelengths on the spectrum of electromagnetic radiation – light, shown in figure 2.1. The softest X-rays are generally considered to start at wavelengths of a few nanometers; hard X-rays extend to picometers. This part of the electromagnetic spectrum exhibits unique properties that can be useful for experiments, the most famous of which being that X-rays can pass through materials that are opaque to the visible wavelengths. To explore these properties, we first need to understand photon absorption and emission processes.

2.2 Absorption and Emission Processes

Emission Processes

A photon is a particle of light containing a packet of energy. The amount of energy that a photon carries is determined by its wavelength according to equation 2.1. The energy of a

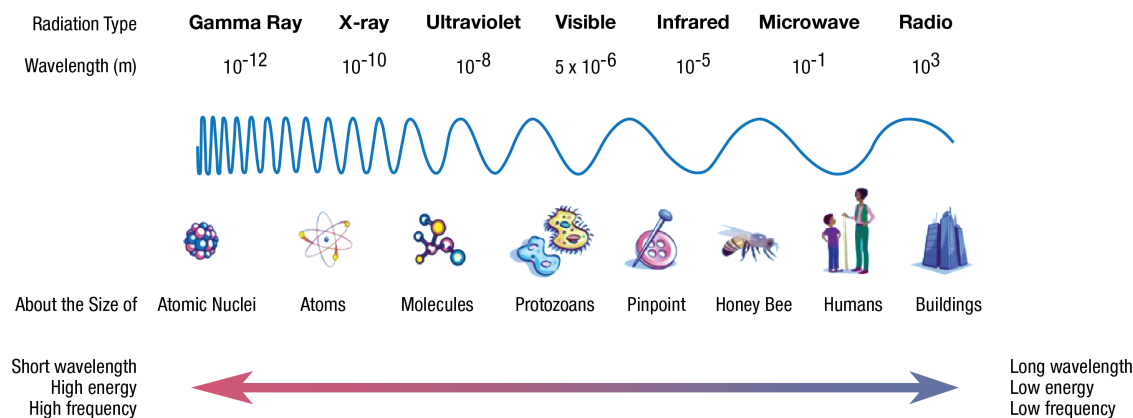


Figure 2.1: The electromagnetic radiation spectrum (adapted from [14]), of which X-rays are just a small part. X-ray wavelengths range from a few hundred nanometers to a few picometers.

photon is given by $\hbar\omega$, where $\hbar$ is Planck's constant, h , divided by 2π , and ω is the light wave frequency in radians per second. Equation 2.1 encapsulates the idea that the color of light is equivalent to the energy of the photon transmitting that light. That is why a source that emits photons of many different energies is called a polychromatic source.

$$\hbar\omega \cdot \lambda = hc \quad (2.1)$$

There are two ways that a photon can interact with or be produced by an atom of a material that it encounters in its path: photoionization and fluorescent emission. In photoionization, an incoming photon excites an electron that is orbiting an atomic nucleus to the point that the electron escapes the pull of the nucleus. The photon is considered absorbed in this scenario, depicted in figure 2.2. There is also an electron hole where the emitted electron used to be. Electrons from higher energy levels can drop into this lower energy hole, producing a photon of energy equal to the difference between the higher and lower electron energies. This process, shown in figure 2.2, is known as fluorescent emission. When a photon is emitted by fluorescent emission, it is considered “characteristic radiation”. The photon energy is unique to the atom that produces it because it is equal to the difference between the higher and lower electron energies, which themselves are unique to the structure of the atom. Quantum mechanics is necessary to determine the allowed energy transitions for different atoms. We avoid that discussion here; the allowed energy transitions can be found in tables provided by the National Institute of Standards and Technology (NIST) [15]. For details on the quantum mechanical origins of characteristic emissions, see [12]. These characteristic emission energies can be used to identify materials, an oft-used technique in chemistry, material science, and biology.

Another way that photons may be emitted is known as bremsstrahlung, German for “braking radiation.” To understand bremsstrahlung (and the scattering processes discussed

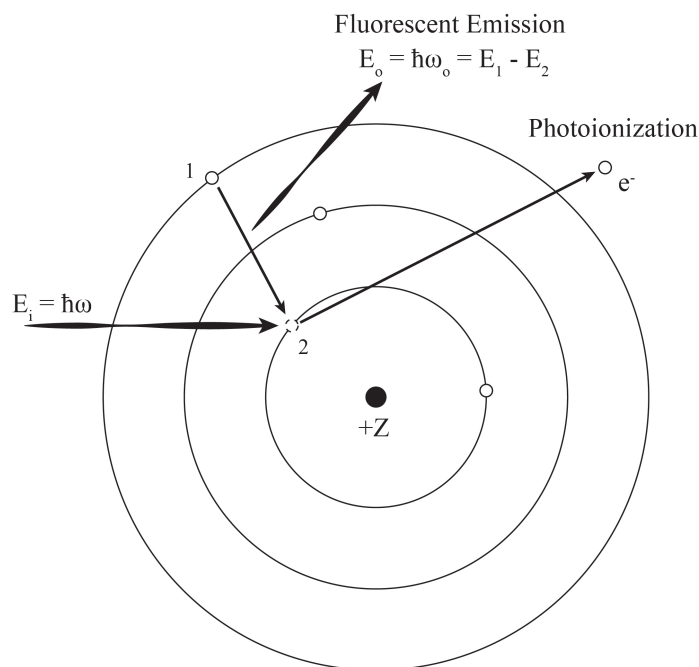


Figure 2.2: Fluorescent emission and photoionization are depicted together in this simplified diagram of an atom. They do not necessarily occur together, but may in many situations discussed in the chapter.

in later sections) one should first understand that because electrons are negatively charged particles, and light is just oscillations in the electric field, light can accelerate electrons via electromagnetic force. Crucially, this is also true in reverse. An accelerated electron can produce photons, since it is simply perturbing the electric field. This reverse process is how bremsstrahlung works. A decelerated electron will emit a photon of energy equal to that which it has lost, hence the term “braking radiation.” Bremsstrahlung is one of the most common ways that X-rays are generated in radiation producing machinery, from synchrotrons to laboratory tube sources. In tungsten target X-ray sources, for example, dense, proton-laden tungsten atoms are effective at bending electron paths, making them a good choice for generating X-rays in a small package that fits in the lab. A diagram depicting bremsstrahlung can be seen in figure 2.3.

Bremsstrahlung can occur alongside other emission processes like fluorescent emission. While fluorescent emission is a quantized process that occurs at discrete energy increments determined by atomic structure, bremsstrahlung is a continuous process. The acceleration experienced by an incoming electron is determined by the distance of its closest approach to the atomic nucleus, otherwise known as the impact parameter, b . The impact parameter can change continuously, so bremsstrahlung can emit a continuous spectrum of photon energies. Fluorescent emission and bremsstrahlung are often seen together in a source spectrum. For

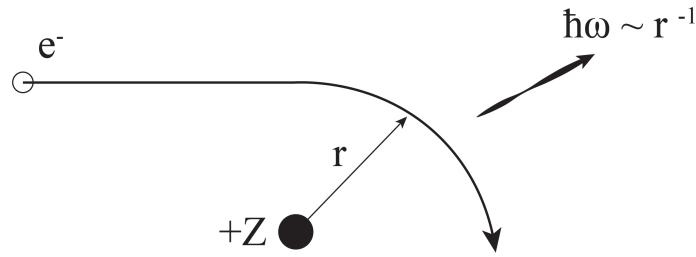


Figure 2.3: Bremsstrahlung occurs when an electron is accelerated by the presence of an atomic nucleus. As we will learn later, an accelerating electron emits electromagnetic radiation.

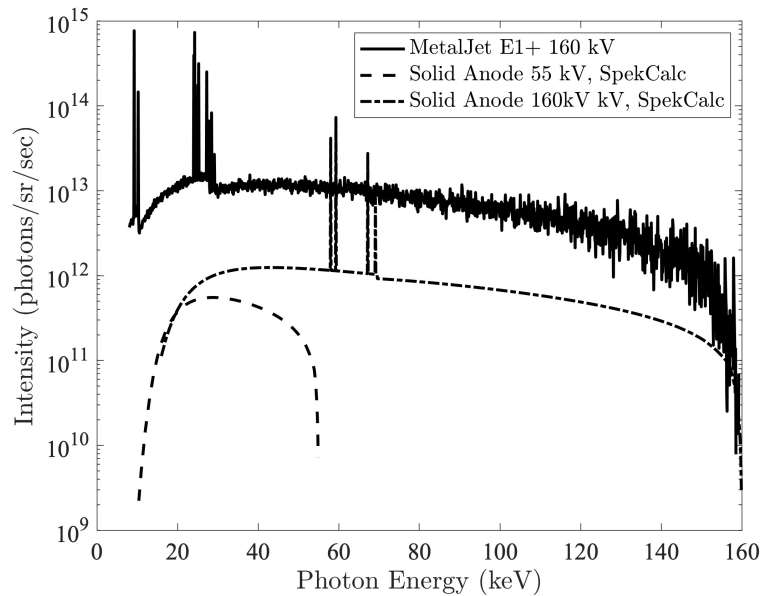


Figure 2.4: The Excillum Metaljet E1+ spectrum [16] and a SpekCalc [17]–[19] simulation of the YXLON FXE225.99 source [20]. The Metaljet spectrum shows spikes at the characteristic emission energies. The solid tungsten target FXE225.99 source only shows bremsstrahlung emission because the fluorescent emission of tungsten is not excited at 55 kV.

example, the Metaljet E1+ source spectrum shown in section 2.5 has spikes at the characteristic emission energies, but a continuous curve of energies elsewhere. The graphic is reproduced in figure 5.6 for convenience.

Absorption Processes

Previously, it was mentioned that photoionization is one way that photons are absorbed, that is, when they impart all of their energy. In fact, there are a number of processes that can lead to a photon being absorbed, which we will look at in more detail in following sections. For now, however, let us take a macroscopic perspective. Numerous experiments have demonstrated the Beer-Lambert law, one of the most useful relationships in X-rays. The Beer-Lambert law, shown in equation 2.2, relates the number of photons exiting a material, I , to the number of photons entering a material I_0 , the thickness of the material x , and the material properties: density ρ and mass attenuation coefficient $\mu(\epsilon, Z)$, where ϵ is the photon energy and Z is the atomic number of the material.

$$I(\epsilon) = I_0(\epsilon)e^{-\rho\mu(\epsilon, Z)x} \quad (2.2)$$

One reason equation 2.2 is so useful is that it is linear. Multiple materials along the same beamline will all absorb photons, attenuating the beam. Each attenuation follows the Beer-Lambert law, so equation 2.2 can more usefully be written as

$$I(\epsilon) = I_s(\epsilon)\exp\left(-\sum_i \rho_i\mu_i(\epsilon, Z)x_i\right) \quad (2.3)$$

where I_s is the source intensity before attenuation, and subscript i indicates the material i . Furthermore, the mass attenuation coefficient is also linear, so multiple coefficients can be combined to calculate the coefficient of a compound or mixture. The mass attenuation coefficients for all elements are well known, and can be found in tables provided by NIST [15]. An example mass attenuation coefficient curve for the element iodine can be seen in figure 2.5. The sharp increase is termed the K-edge for iodine. It is the energy of the first fluorescence of iodine. Larger elements will have multiple such edges.

The Beer-Lambert law is incredibly useful for obtaining first-order estimates of photon intensities (the number of photons), and can even be used to simulate X-ray images. Chapter 3 discusses this use case. The primary limitation of the Beer-Lambert law is that it is a macroscopic relationship that only describes the transmission of photons. It does not describe emission processes such as the ones described above, or scattering, diffraction, and refraction, which are discussed in the following section. These processes are buried in the linear mass attenuation coefficient μ .

Nevertheless, the Beer-Lambert law can also be useful for fluid dynamics experiments. In chapter 1, reference was made to using X-rays for volume fraction measurements in multiphase flows. Let us assume for simplicity that we have a monochromatic X-ray source (i.e., that the source output X-ray photons at one energy) capturing images of a two-phase system: gas and liquid. The Beer-Lambert law tells us the intensity of photons detected along a certain ray path of length L . Suppose we want to find the fraction α of length L that is in gas in the multiphase system. α is the so-called void fraction.

$$I_g = I_0e^{-\rho_g\mu_gL} \quad (2.4a)$$

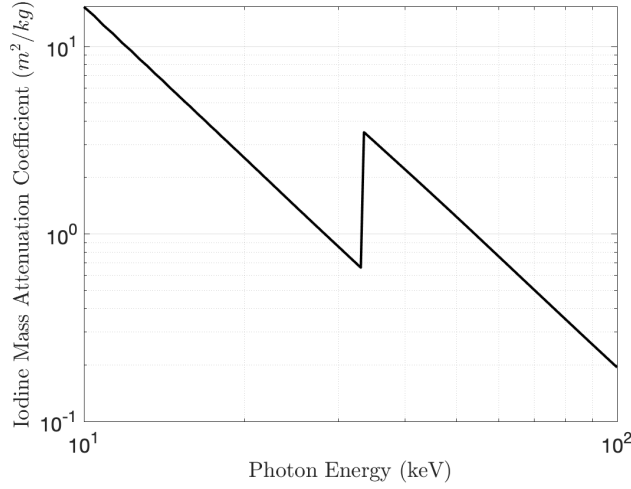


Figure 2.5: The mass attenuation coefficient for the element iodine, a common X-ray contrast agent. Note the sharp increase at approximately 33 keV. This is the K fluorescence edge of iodine.

$$I_l = I_0 e^{-\rho_l \mu_l L} \quad (2.4b)$$

$$I_m = I_0 e^{-\rho_g \mu_g s_g - \rho_l \mu_l s_l} \quad (2.4c)$$

Here, I_0 is the source intensity, ρ is the material density, μ is the material mass attenuation coefficient, and s is the material length along the ray path. Equations 2.4a, 2.4b, and 2.4c are the detected intensity for pure gas, pure liquid, and mixed gas-liquid systems, respectively. Equations 2.4 can be rearrange to solve for the void fraction, shown in equation 2.5.

$$\alpha = \frac{l_a}{L} = \frac{\ln(I_m/I_l)}{\ln(I_g/I_l)} \quad (2.5)$$

Equation 2.5 is the basis of an experimental technique in which one captures images of the experiment filled with pure gas, then with pure liquid, and lastly with an unknown mixture of gas and liquid. Using equation 2.5 one can then calculate the void fraction at each point in the image by using the images of pure gas and pure liquid. It is important to note that we assumed a monochromatic source, which most laboratory X-ray sources are not. With a typical energy-integrating detector (which will be defined later in this chapter) this introduces error. With an energy-resolving photon counting detector, however, one can calculate the void fraction for each energy bin, thereby reducing the polychromatic source error. Nevertheless, equation 2.5 can provide a decent estimation of the void fraction of gas in a two-phase system and is a good example of the utility of the Beer-Lambert law.

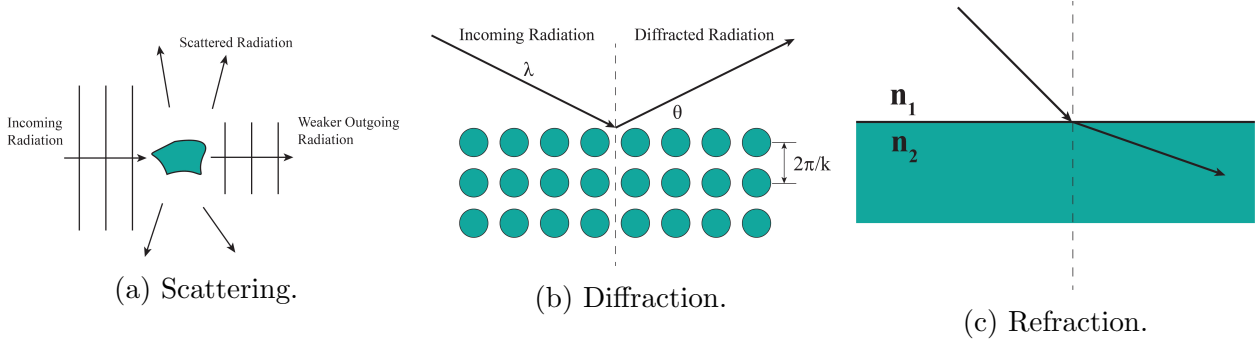


Figure 2.6: Scattering, diffraction, and refraction are all form of redirected radiation due to the presence of accelerating electrons (either free or bound to atoms).

2.3 Scattering, Diffraction, and Refraction

The Beer-Lambert law calculates the number of *transmitted* photons, that is, the photons that are not deflected from their original path. A photon that is scattered, diffracted, or refracted is not considered a transmitted photon and, under the Beer-Lambert law, is treated as attenuated. A more complete description of photon transport processes, however, must include scattering, diffraction, and refraction. To start, let us define each of these processes.

A scattered photon is a photon that, through interactions with one or more electrons, is redirected in a new direction. The hallmark feature of photon scatter is that it is a random process. Diffraction and refraction are more deterministic. In X-ray imaging, scattered photons play an important role because they add to image noise without contributing to the image signal. Diffraction is when a regular pattern of atoms, e.g. a crystal, create a pattern of constructive and destructive interference that results in the incoming light being redirected in a new direction. Refraction is the bending of light that occurs when it passes from one medium to another due to differences in the speed of light in each medium. Figure 2.6 depicts each of these processes. In order to understand scattering, diffraction, and refraction, a brief overview of classical models for light-atom interactions is necessary.

The Electromagnetic Wave Equation and Accelerating Electrons

Ultimately, light is the propagation of waves in the electromagnetic field. The wave equation is given in equation 2.6, where c is the speed of light in a vacuum, $\mathbf{E}(\mathbf{r}, t)$ is the electric field, ϵ_0 is the dielectric constant in a vacuum, $\mathbf{J}(\mathbf{r}, t)$ is the charge current density, and $\rho(\mathbf{r}, t)$ is the charge density. This equation can be derived from Maxwell's equations, which can be found in [12].

$$\left(\frac{\partial^2}{\partial t^2} - c^2 \nabla^2 \right) \mathbf{E}(\mathbf{r}, t) = \frac{1}{\epsilon_0} \left[\frac{\partial \mathbf{J}(\mathbf{r}, t)}{\partial t} + c^2 \nabla \rho(\mathbf{r}, t) \right] \quad (2.6)$$

Another important relation is the conservation of charge, given in equation 2.7.

$$\nabla \cdot \mathbf{J} + \frac{\partial \rho}{\partial t} = 0 \quad (2.7)$$

Scatter, refraction, and diffraction are all solutions to equations 2.6 and 2.7. The first assumption we will make is that electrons behave as point sources. Electrons have a negative charge, so they interact with incoming waves in the electric field. As an electron accelerates due to this interaction, it too creates waves in the electric field, thus acting like a source. Importantly, we assume that electrons themselves are much smaller than the wavelengths of incoming radiation, allowing us to treat electrons as a point source. As a result, we can write the current density as

$$\mathbf{J}(\mathbf{r}, t) = -e\delta(\mathbf{r})\mathbf{v}(\mathbf{r}, t) \quad (2.8)$$

where e is the electron charge, $\delta(\mathbf{r})$ is the Direct delta function, and $\mathbf{v}(\mathbf{r}, t)$ is the electron velocity. Armed with these assumptions, one can search for Green's function solutions to the wave equation by taking a Fourier transform to algebraize the wave equation, as seen in **attwood**.

Single Free Electron Scatter

One important canonical solution is that of an oscillating free electron, provided in equation 2.9, where $\mathbf{a}_T(\mathbf{r}, t)$ is the transverse component of the electron acceleration.

$$\mathbf{E}(\mathbf{r}, t) = \frac{e\mathbf{a}_T(t - r/c)}{4\pi\epsilon_0 c^2 r} \quad (2.9)$$

This is the first half of describing scattering by a free electron. We must describe the acceleration that the electron experiences due to incoming radiation. For that, we turn to Newton's second law. For non-relativistic oscillation frequencies (i.e., those that produce electron velocities much smaller than the speed of light), we can assume that Lorentz forces from the incoming magnetic field are negligible compared to those of the electric field. So, we can write the electron acceleration as

$$\mathbf{a}(\mathbf{r}, t) = -\frac{e}{m}\mathbf{E}_i(\mathbf{r}, t), \quad (2.10)$$

where m is the mass of an electron and $\mathbf{E}_i(\mathbf{r}, t)$ is the incoming (also called incident) electric field radiation. Thus, the radiation emitted by a free electron accelerated by an incoming electric field is

$$\mathbf{E}(\mathbf{r}, t) = \frac{e^2 E_i \sin \Theta}{4\pi\epsilon_0 m c^2 r} e^{-i\omega(t-r/c)}, \quad (2.11)$$

where Θ is the angle relative to the transmission direction, and ω is the oscillation frequency of the incoming electric field wave. Equation 2.11 describes the most simple form of scattering. It provides a useful illustration of the physical phenomena that give rise to scattering.

Single and Multiple Bound Electron Scatter

Procedures similar to the one presented above are used to find the scattered radiation due to single bound electrons and multiple bound electrons such as those found in atoms. In the case of bound atoms, Newton's second law takes the form of harmonic oscillation in which an electron oscillates at a resonant frequency that corresponds to the strength of its attraction to the atomic nucleus. Equation 2.12 gives the solution for a single bound electron accelerating in an incoming electric field.

$$\mathbf{a} = \frac{-\omega^2}{\omega^2 - \omega_p^2 + i\gamma\omega} \frac{e\mathbf{E}_i}{m} \quad (2.12)$$

Here, γ is a small dissipative coefficient; ω_p is the resonant frequency of the bound electron; ω is the frequency of the incoming radiation.

When there are multiple bound electrons, as in all atoms except hydrogen, the acceleration of each individual electron must be taken into account. Furthermore, radiation emitted from excited electrons becomes incoming radiation for other electrons in the atom, coupling the excitation of electrons in an atom. However, if we assume that the coupling effects are negligible compared to the strength of the incoming radiation, there is a natural extension to the bound electron solution.

If we first define a coordinate system where r is the vector location of the atom nucleus (assumed to be a point at the origin) to the observer and r_p is the vector distance of the electron to the observer, then the scattered radiation is the sum of the emitted radiation from each electron,

$$\mathbf{E}(\mathbf{r}, t) = \frac{e}{4\pi\epsilon_0 c^2} \sum_{p=1}^Z \frac{\mathbf{a}_{\mathbf{T},p}(t - r_p/c)}{r_p}. \quad (2.13)$$

Once more, we can use Newton's second law to calculate the transverse component of the electron acceleration in order to find the scattered radiation as a function of the incoming radiation. Since we have assumed that the scattered radiation from one electron does not affect the others, each electron independently accelerates as though excited by the incoming radiation, i.e., the solution for an individual accelerating bound electron (equation 2.12 applies here as well with an additional wrinkle). We do not assume that the incoming wavelength is much larger than the atom, so phase of the incoming radiation is not the same for each electron. Nor is the phase of the emitted radiation seen by the observer the same for all electrons. The incoming electric field can then be written as

$$\mathbf{E}_i(\mathbf{r}, t) = \mathbf{E}_i e^{-i(\omega t - \mathbf{k}_i \cdot (\mathbf{r} - \mathbf{r}_p))}, \quad (2.14)$$

where $\mathbf{k}_i$ is the vector wave number and E_i is the corresponding vector amplitude of the incoming radiation. Inserting equation 2.14 into equations 2.12 and 2.13, we find the scattered radiation from a multi-electron atom due to incoming radiation, shown in equation 2.15.

$$\mathbf{E}(\mathbf{r}, t) = -\frac{r_e}{r} \sum_{p=1}^Z \frac{-\omega^2 E_i \sin \Theta}{\omega^2 - \omega_p^2 + i\gamma\omega} \exp \{-i[\omega(t - r/c) + \Delta \mathbf{k} \cdot \Delta \mathbf{r}_p]\} \quad (2.15)$$

Here, we have defined three new variables:

$$r_e = \frac{e^2}{4\pi\epsilon_0 mc^2} \quad (2.16)$$

$$\Delta \mathbf{k} = \mathbf{k} - \mathbf{k}_i \quad (2.17)$$

$$\Delta \mathbf{r}_p = \mathbf{r} - \mathbf{r}_p \quad (2.18)$$

where r_e is called the classical electron radius. Equation 2.15 was simplified slightly by assuming $|\mathbf{r}| \gg |\mathbf{r}_p|$, that is, the observer is much farther away from the atomic nucleus than the electrons.

Equation 2.15 mathematically describes scatter, diffraction and refraction. Fundamentally, all three phenomena are just different forms of electrons interacting with incoming radiation.

Scattering

Incoming radiation is said to be scattered when it is redirected across a wide range of directions by interacting with electrons in a material. There is no pattern to the constructive and destructive interference with radiation from nearby accelerated electrons. This causes the scattered direction of photons to appear random, although some directions are more likely than others. With large numbers of atoms and photons, scattering is best describes by a random process. Energy is deposited in the material when a photon is scattered due to the dissipation term in equation 2.12. Scattering as a random process is discussed in greater detail in the context of Monte Carlo simulations in chapter 3.

Diffraction

Unlike scattering, diffraction is caused by constructive and destructive interference in a periodic electron distribution combining to effectively redirect light in a single direction. This implies that the phase of the light emitted from the electrons is the key element. If we examine the $\Delta \mathbf{k}$ term in equation 2.15, and note that the dispersion relation for light in a vacuum implies that $|\mathbf{k}| = |\mathbf{k}_i| = \omega/c$, we arrive at one form of the Bragg equation:

$$|\Delta \mathbf{k}| = 2 |\mathbf{k}_i| \sin \theta, \quad (2.19)$$

where θ is the half angle between the transmission wave vector, $\mathbf{k}$, and the incident wave vector, $\mathbf{k}_i$. By definition, $k_i = 2\pi/\lambda$ and $|\Delta \mathbf{k}| = 2\pi/d$ where d is the electron distribution period. Thus, we get the typical Bragg equation,

$$\lambda = 2d \sin \theta. \quad (2.20)$$

The Bragg dictates that for a given electron distribution period, e.g. the length of a crystal unit, a wavelength of light λ will be deflected by angle θ .

Refraction

The Complex Atomic Scattering Factor

Refraction occurs at the boundary between two different media. The speed of light changes across the boundary, causing the light to bend accordingly. To identify how the refraction of light occurs, we first need to define the complex atomic scattering factor, $f(\Delta\mathbf{k}, \omega)$. The complex atomic scattering factor is directly connected to the index of refraction. We can reorganize equation 2.15 such that the emitted radiation is some scale factor multiplied by the incident radiation:

$$E(\mathbf{r}, t) = -\frac{r_e}{r} \sum_{p=1}^Z \frac{\omega^2 e^{-i\Delta\mathbf{k} \cdot \Delta\mathbf{r}}}{\omega^2 - \omega_p^2 + i\gamma\omega} E_i \sin \Theta e^{-i\omega(t-r/c)} \quad (2.21)$$

Now, we can define the complex atomic scattering factor as

$$f(\Delta\mathbf{k}, \omega) \equiv \sum_{p=1}^Z \frac{\omega^2 e^{-i\Delta\mathbf{k} \cdot \Delta\mathbf{r}}}{\omega^2 - \omega_p^2 + i\gamma\omega}. \quad (2.22)$$

The $\Delta\mathbf{k} \cdot \Delta\mathbf{r}$ term in equation 2.22 is difficult to work with. When $\Delta\mathbf{k} \cdot \Delta\mathbf{r} \rightarrow 0$, the exponential approaches unity. This occurs in either of two circumstances: the long wavelength limit, and the forward scattering limit. Expanding the dot product and substituting $k_i = 2\pi/\lambda$, we get

$$|\Delta\mathbf{k} \cdot \Delta\mathbf{r}| \leq \frac{4\pi a_0}{\lambda} \sin \theta, \quad (2.23)$$

where a_0 is the Bohr radius, the radius in which the charge of an atom is largely contained. In other words, it poses a lower limit on the wavelength in this regime. Equation 2.23 approaches zero when either $a_0/\lambda \ll 1$ or $\theta \ll 1$. These are referred to as the long wavelength and forward scattering limits, respectively. In general, for all but the softest X-rays, the long wavelength limit does not hold. To investigate refraction further, we will assume we are in the forward scattering limit, which is independent of wavelength. In either of these limits, we denote the complex atomic scattering factor with a superscript, $f^0(\omega)$; it is now only depending on the frequency of the incoming light.

In general, each electron in an atom has a different oscillation strength, which will add an oscillation strength coefficient g_p in front of the complex scattering factor numerator. Here, though, we will ignore these oscillation strength factors in order to focus instead on the conceptual behavior of light in the X-ray spectrum. For more information, see [12].

Since $f^0(\omega)$ is complex, we can break it into its real and imaginary components:

$$f^0(\omega) = f_1^0(\omega) - i f_2^0(\omega) \quad (2.24)$$

where both $f_1^0(\omega)$ and $f_2^0(\omega)$ are real-valued functions. Similar to mechanical vibration, the real term corresponds to phase shift, while the imaginary term corresponds to amplitude decay. Later, these two components will be linked to the index of refraction in the X-ray spectrum.

The Index of Refraction

By deriving the electromagnetic wave equation in a medium, where the incoming radiation propagation is altered by the existence of forward scatter, one arrives at a modified form of the wave equation where the refraction index $n(\omega)$ alters the speed of light. This refraction index is related to the forward complex scattering factor by

$$n(\omega) = 1 - \frac{n_a r_e \lambda^2}{2\pi} [f_1^0(\omega) - i f_2^0(\omega)], \quad (2.25)$$

where n_a is the atomic number density and λ is the wavelength of the light in a vacuum. It should be noted here that in order to arrive at equation 2.25, we assume that the frequency $\omega^2 \gg \frac{n_a r_e \lambda^2}{2\pi}$. It is common when discussing the X-ray portion of the spectrum to rewrite the index of refraction as a perturbation from unity. That is because X-rays are of such high frequency that ω is much larger than most resonance frequencies, making $n(\omega)$ approach unity. We can write

$$n(\omega) = 1 - \delta + i\beta, \quad (2.26)$$

where

$$\delta = \frac{n_a r_e \lambda^2}{2\pi} f_1^0(\omega), \quad (2.27a)$$

$$\beta = \frac{n_a r_e \lambda^2}{2\pi} f_2^0(\omega). \quad (2.27b)$$

That X-rays have an index of refraction close to unity is a major benefit for their application in fluid flow visualization. In multiphase fluid flows, refraction over multiple warped, moving interfaces renders flows effectively opaque to visible light. X-rays, on the other hand, would pass through with often negligible refraction, making it possible to explore these flows. Alternatively, one could use the small changes in the phase due to refraction across interfaces to generate phase contrast. This is different from typical X-ray imaging, which uses absorption to generate contrast. While phase contrast focuses on the δ term, typical absorption contrast relies on the β term. In fact, β is related to the mass attenuation coefficient by

$$\mu(\omega) = \frac{2r_e \lambda}{A m_u} f_2^0(\omega), \quad (2.28)$$

where m_u is the atomic mass unit and A is the number of atomic mass units.

Snell's Law

Finally, we are prepared to determine the law of refraction - Snell's law. Snell's law is a consequence of the boundary conditions for Maxwell's equations and the dispersion relation for light. First, the boundary conditions for Maxwell's equations:

1. Field components of $\mathbf{E}$ parallel to the interface must be continuous.

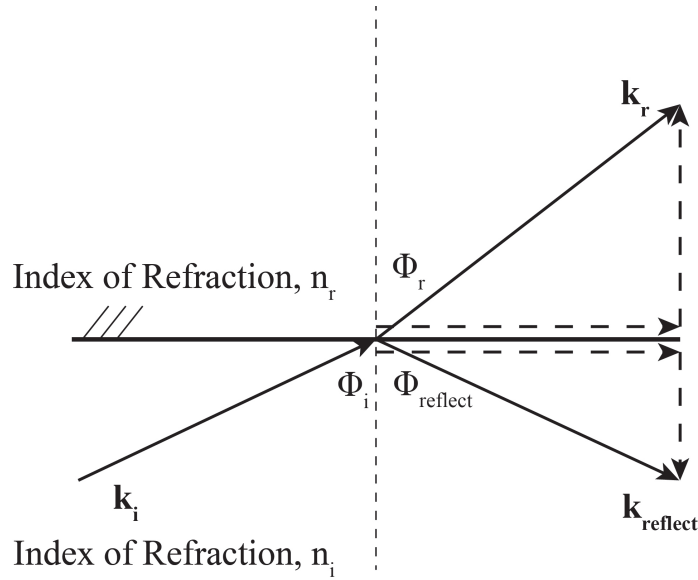


Figure 2.7: A diagram of reflection and refraction at an interface. During reflection both rays are in the same medium, so $k_i = k_{reflect}$, implying that $\phi_i = \phi_{reflect}$ as a direct result of the first boundary condition. During refraction, the medium changes, so $k_i \neq k_r$. However, according to the first boundary condition again, we know the component parallel to the interface must be equal. We then arrive at equation 2.29.

2. Field components of $\mathbf{B}$ perpendicular to the interface must be continuous. $\mathbf{B}$ is the magnetic field.

The first of these boundary conditions is relevant to Snell's law. It implies that the angle of departure at an interface, that is, the angle of light reflect away from the interface, is equal to the angle of incidence. This further implies that the parallel component on the other side of the interface is equal to the parallel component of departure, as shown in figure 2.7. Thus,

$$k_i \sin \phi_i = k_r \sin \phi_r, \quad (2.29)$$

where k_r and ϕ_r are the wave number and angle of the refraction light to the perpendicular, respectively. We can combine the dispersion relation, $\omega = kc/n$, and that both waves must oscillate at the same frequency to arrive at

$$\frac{k_i c}{n_i} = \frac{k_r c}{n_r}. \quad (2.30)$$

Combining equations 2.29 and 2.30, we obtain Snell's law:

$$n_i \sin \phi_i = n_r \sin \phi_r. \quad (2.31)$$

Section Summary

This section provides a crash-course in the X-ray physics that are relevant to this dissertation. It highlights important models in scattering, diffraction, and refraction, and what their origin is. Ultimately, each is a different behavior of the same solution for emitted radiation from electrons excited by incident radiation. Diffraction and refraction are special types of scattering, while scattering colloquially refers to the undirected redirection of radiation by electrons. Diffraction is defined by Bragg's law; refraction by Snell's law.

We also examined why X-rays have an index of refraction close to unity, and what implications that has for X-ray fluid dynamics and X-ray imaging. When $n(\omega) \approx 1$, high volume fraction multiphase flows are no longer opaque from refraction over multiple interfaces, as they are with visible light. Additionally, we broke down two different ways to generate contrast in X-ray images and how those connect with the index of refraction. Most commonly, X-ray imaging involves generating contrast with absorption, i.e., making use of the β amplitude decay term. One can, with sufficiently coherent light, also generate contrast with by measuring the phase different generated by the δ term. In the next section, we will use the knowledge in this section to discuss the details of X-ray imaging processes.

2.4 X-ray Imaging Processes

Now that we know how light travels and interacts with different media, we can begin exploring how light is converted into electrical signals that become images. In other words, what processes govern X-ray imaging. There are a number of excellent references for this subject material, including *The Handbook of X-ray Imaging: Physics and Technology* by Russo et al. [13]. Much of the material in this section is from that book and my own experience in the course of my doctorate. Similar to the previous section, I will provide a high-level overview of X-ray imaging technology to better help the reader understand conceptually what occurs when an X-ray image is captured, and what to consider to improve their technique.

Scintillating Detectors

X-ray imaging dates to the discovery of X-rays themselves. Roentgen famously captured a picture of his wife, Bertha's, hand with a ring on it on November 8, 1895, seen in figure 2.8. Roentgen discovered X-rays by noticing the fluorescence of barium platinocyanide on a nearby workbench while working with a cathode ray tube. As with Roentgen's discovery, many X-ray detectors today use X-ray fluorescence to capture images. These are known as scintillating detectors. Scintillating detectors are made with materials that, when excited by incoming X-ray photons, fluoresce in the visible light spectrum.



Figure 2.8: Roentgen captured this picture of his wife Bertha's hand on November 8, 1895, signaling the discovery of X-rays.

Operating Principle

In this context, a scintillator is a material that will scintillate (flash or otherwise emit light, typically in the visible light range) when it is hit with X-rays. Basically, a scintillator will convert incoming X-rays into visible light. The benefit of this is that one can use visible light optics and sensors to capture X-ray images, both of which are relatively cheap and readily available. Currently, X-ray optics are difficult to obtain, highly-specialized, and often rely on coherent, monochromatic light. Recently, though, there have been advances in X-ray optics that may allow for limited laboratory use in the future [21].

Scintillation occurs by the processes we touched on in section 2.2. Incoming X-ray will excite electrons, knocking them loose of the nucleus in what is known as photoionization. Higher energy electrons, or loose electrons themselves will then be pulled into the hole left by the departing electron. This releases lower energy photons at energies characteristic to the scintillating material. By choosing the appropriate materials, these lower energy photons will be in the visible light range. Typical scintillator materials in X-ray detectors include cesium iodide (CsI), cesium fluoride (CsF), sodium iodide (NaI), and many others [13].

When a scintillator converts X-rays into visible light, the energy information about the original X-ray energy is lost and is instead translated into brightness. As a result, scintillating detectors do not have any energy resolution. Many laboratory X-ray sources are polychromatic, so valuable information about the incoming photons is lost by using scintillating detectors. Nevertheless, in applications where spatiotemporal resolution is more important than energy resolution, or when energy resolution is not important, scintillating detectors are useful.

Blurring Effects

In addition to losing energy resolution, scintillating detectors also experience blurring effects. When X-rays impact the scintillator, they are often scattered through the scintillator and cause multiple atoms to fluoresce. This causes blurring in the resulting X-ray image because the location of the original incoming X-ray is lost. Thinner scintillating materials mitigate this effect, but also have a lower scintillation efficiency – the ratio of scintillation events to the number of incoming X-rays.

Afterglow Effects

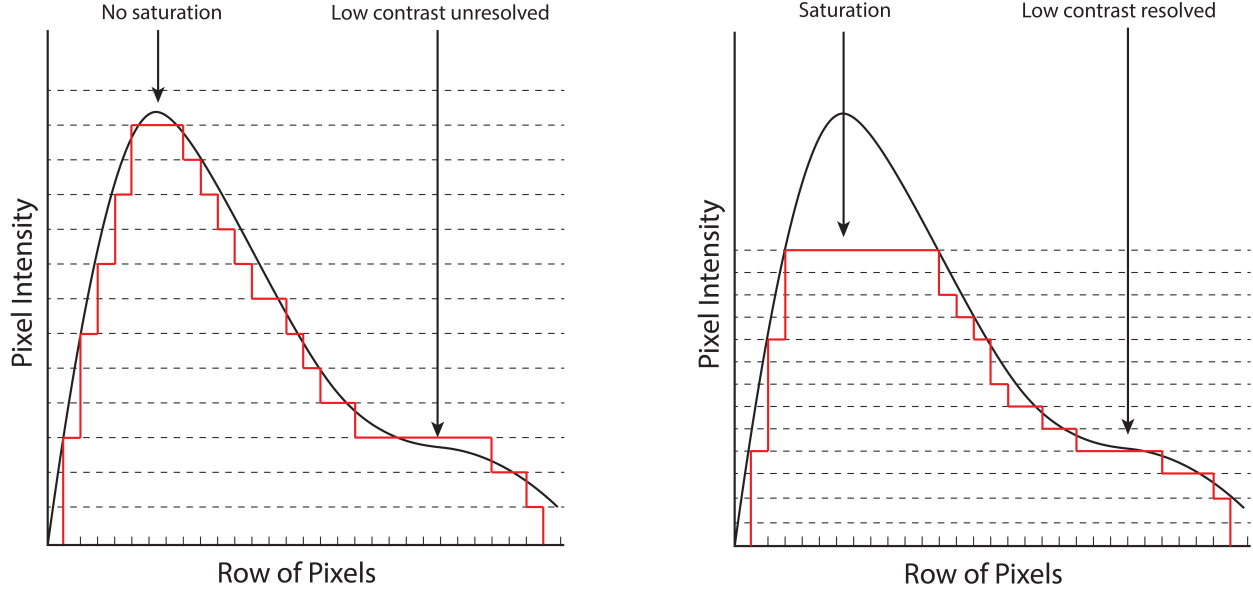
Another issue that scintillating detectors often experience is afterglow. The scintillator will glow for a short period of time after an X-ray is converted to visible light. When photons arrive faster than the afterglow dissipates it will build up, leaving glow artifacts in images that are not real. This problem is particularly pernicious when the incoming X-rays are relatively high energy because they cause a larger initial scintillation.

Pixel Saturation

Scintillating detectors may have tunable dynamic ranges. That is, the maximum and minimum intensity values can be adjusted to avoid saturation in imaging situations where part of the image is bright but other parts are dark. Since the imaging sensor has a constant bit depth, intensity resolution suffers when the dynamic range is too large. Figure 2.9 gives an example of pixel saturation and dynamic range for a detector with 13 thresholds. By adjusting the range over which the thresholds bin intensities, one can tune the intensity resolution and pixel saturation. Areas of slight contrast will appear uniform with a large dynamic range, but fewer pixels will saturate. The user should be mindful of finding an appropriate balance between intensity resolution and dynamic range, much as they would for visible light photography. This can sometimes be difficult when there are areas of the FOV with vastly different amounts of contrast, brightness, or both. In those instances, beam hardening can be useful. Beam hardening will be discussed in greater detail in the following section on PCDs.

Image Noise

Scintillating detector noise originates in two places: the detector itself and the incoming photons. These two sources of noise superpose with one another to create the noise that the detector outputs in the image. The detector noise is typically modeled as Gaussian white noise and independent of the incoming image. It is the result of thermal and electronic fluctuations in the visible light sensors (typically CCD or CMOS). The incoming photons are a Poisson noisy signal as a result of the emission statistics of the source. We will discuss the signal noise in the following section on PCDs. Equation 2.32 describes the image noise from a scintillating detector, where N is the random variable of photons generated from



(a) A line profile captured with a 13-threshold detector. No pixel saturation occurs, but regions of low contrast will appear uniform.

(b) A line profile captured with a 13-threshold detector. Saturation occurs in regions of high intensity, but low contrast regions are resolvable.

Figure 2.9

noise, μ is the expected thero-electronic noise intensity, σ is the standard deviation, and λ is the rate parameter of the incoming photons.

$$N(t) \sim \text{Gauss}(\mu, \sigma) + \text{Poiss}(\lambda t) \quad (2.32)$$

The combination of Gaussian and Poisson noise poses issues for filtration since it is difficult to ascertain the relative strength of each. Different filters are designed to address different forms of noise. Furthermore, the existence of Gaussian white noise independent of the image noise increases the noise floor above which a signal is usable. Longer exposure times are necessary to overcome the additional noise and achieve a sufficient SNR.

Photon-Counting Detectors

Most scintillating detectors are energy-integrating detectors (EIDs). Energy-integrating refers to the fact that scintillating detectors do not resolve the energy of the incoming photons, but instead add up the energy contribution from deposited X-ray photons. A high energy photon is brighter than a low energy photon, but both are added together indiscriminately. The output pixel intensity is proportional to the total deposited energy. A more recent type of detector can resolve the incoming X-ray photon energy. These are known as photon counting detectors (PCDs).

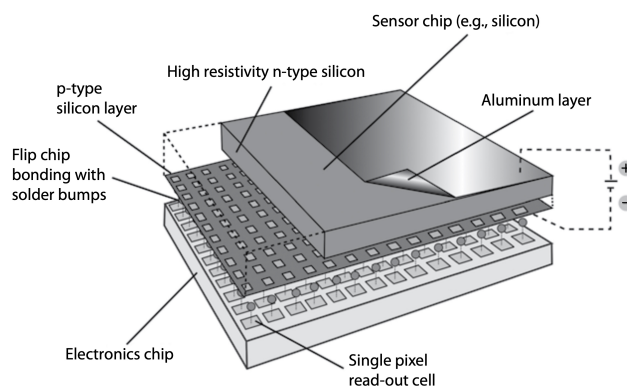


Figure 2.10: A diagram of a typical PCD sensor [13]. A voltage is applied across a high-Z semiconductor material; charge bumps collect the emitted photoionization electrons, generating a pulse in the detector circuitry.

PCDs are unique in that instead of converting X-ray photons into visible light photons, they count the individual electrical pulses that X-rays generate as they are absorbed into a material with a large electrical potential across it.

Operating Principle

PCDs directly detect photons, which has a number of benefits compared to scintillating detectors. A diagram of a typical PCD can be seen in figure 2.10. A large voltage is applied across a uniform semiconductor crystal with small charge bumps on the back side. The semiconductor crystal can be made from a number of different materials such as cadmium telluride (CdTe), gallium arsenide (GaAs), and high purity germanium (HPGe), among others. All of these materials are high-Z, that is, they have large atomic numbers. High-Z materials are able to efficiently absorb X-ray photons, meaning they have a higher detection efficiency (DE) - the ratio of photons that enter the crystal and the number of photons that are absorbed by the crystal. These materials are also resistive so a large voltage can be applied across them without shorting.

When an X-ray photon enters the crystal, it is most likely going to be absorbed and deposit energy in the crystal. These materials are selected such that photoionization, discussed in chapter 2, is the most common emission phenomenon. The large potential applied across the crystal drives these newly emitted electrons towards the charge bumps, which collect them as a current. When a photon is absorbed and emits electrons, this is registered as a pulse in the electrical circuitry of the detector. These pulses are what is counted in a PCD.

Interestingly, there are no delineated pixel boundaries, and no electronics between pixels. The entire crystal is active area, unlike visible light sensors. This means the detector is able to more efficiently detect photons. The downside, however, is that charge sharing is

possible. Charge sharing occurs when the charge cloud generated by an absorbed photon is spread between more than one charge bump, which would count two lower energy photons instead of a single higher energy photon. Charge sharing is one of the primary impediments to smaller PCD pixels.

Another difficulty with single-crystal detectors is more practical in nature. It is difficult to manufacture large high-Z semiconductor crystals without defects. Defects in the material can create non-uniformity in the detection quantum efficiency (DQE), a measure combining the DE and the charge collection efficiency (CCE) of the material. Since PCDs do not use optics, they are reliant on large detection areas to increase the FOV and spatial resolution (via geometric magnification). Being unable to manufacture large crystals requires the use of tiling, which introduces gaps in the detector sensor that have to be artificially corrected for. Hopefully, in the future, improvements in manufacturing these materials makes PCDs cheaper and larger.

Photon Pile-Up

A unique feature of PCDs is photon pile-up (PPU). PPU occurs when the rate of incoming X-rays creates charge pulses that are too close to each other to distinguish. The electronics have a dead time between pulses; if photons arrive in that dead time they are not counted. In what are called “paralyzable” PCDs, photons arriving during the dead time can eventually freeze the detector as it tries to catch up. “Non-paralyzable” detectors use re-triggering to mitigate detector paralyzation. Nevertheless, non-paralyzable detectors will still undercount the number of incoming photons.

PCD manufacturers often report a photon count rate limit, typically reported in photons per pixel per second. Beyond that limit, the detection rate of the electronics is insufficient for counting the incoming photons. For a given dead time, one can model the event detection rate as a function of the true event rate for both a paralyzable and non-paralyzable PCD (wieloposki, gardner, yu fessler, kno11), where equation 2.33a is the paralyzable model and equation 2.33b is the non-paralyzable model.

$$D_p = I e^{-I\tau} \quad (2.33a)$$

$$D_{np} = \frac{I}{I\tau + 1} \quad (2.33b)$$

Here, I is the incoming photon rate, τ is the dead time, and D is the detected photon rate.

PPU has adverse effects on the image quality. When photon detections are lost to PPU, there is a diminishing return in image quality for a brighter X-ray source. In the case of the paralyzable PCD, the image quality may actually degrade after a certain incoming photon rate. Make sure that the incoming photon rate does not exceed the photon count rate limitations set by the manufacturer. Be aware, PPU is a function of the incoming photon rate, so increasing or decreasing exposure time will not resolve PPU.

Pixel Saturation

Unlike scintillating detectors, PCDs do not have adjustable saturation intensities. The saturation intensity of a PCD is simply a function of the bit depth of the pixels ($2^b - 1$). A smaller bit depth requires either a shorter exposure time or a dimmer source to prevent saturation.

A small bit depth also limits the dynamic range of a single exposure, the difference between the highest intensity and lowest intensity pixel value. One can harden the beam to make the dynamic range smaller. Hardening the beam means placing a material (e.g. aluminum, copper) in front of the X-ray source to attenuate lower energy photons. This has the effect of shifting the average photon energy from a polychromatic source up and reduces the source photon energy bandwidth. If, for example, in one part of the field-of-view (FOV) there is strong attenuation from a metallic object, but in another part of the FOV there is only attenuation from air, the dynamic range of the image will be quite high. By hardening the beam, photon counts from low energy photons in the air pixels will be reduced without significantly lowering the photon counts from the metallic objects pixels, since the low energy photons would be attenuated by that object regardless of hardening the beam.

Alternatively, one can take multiple short, unsaturated exposures and sum them to create a single image with a total exposure time that would otherwise saturate the detector. This is uniquely possible with PCDs because they are “noiseless,” that is, they measure exactly zero in the absence of signal. The next section will discuss PCD image noise. EIDs have a finite noise intensity even in the absence of X-ray light, so adding multiple exposures introduces additional noise that would not be there in a single, longer exposure. Ostensibly, this “noiseless” feature of PCDs gives them “infinite” dynamic range since one could take an arbitrary number of unsaturated exposures to get larger dynamic range.

Image Noise

Image noise in a PCD is different than a scintillating detector. While scintillating detectors experience Gaussian white noise from electrical and thermal signals, PCDs do not. The noise contribution to PCD images – setting aside PPU for now – is determined entirely by the Poisson process of photon emission from the source and photon detection in the pixels. As a result, PCDs have lower noise intensities, and thus a higher signal-to-noise ratio (SNR) compared to scintillating detectors for the same number of photons. That PCD pixel intensity is, to the first order, a Poisson random variable makes it usefully intuitive thanks to the nature of Poisson processes.

A Poisson process is one in which the random variable is integer valued, positive, and monotonically increasing. The Poisson distribution is a single-parameter distribution determined by the rate λ . The probability mass function of the Poisson distribution is given by equation 2.34, where $n \in \mathbb{N}$, t is time, and N is a Poisson random variable. A Poisson process is considered homogeneous if λ is a constant. We denote a Poisson random variable

as $X \sim \text{Poiss}(\lambda)$.

$$P(N = n) = e^{-\lambda t} \frac{(\lambda t)^n}{n!} \quad (2.34)$$

Photon emission is well-modeled as a homogeneous Poisson process. We can choose N to be the number of photons emitted from the X-ray source over the course of time t . A unique feature of Poisson processes is that λt is the expected, or mean, value *and* the variance. This can be written

$$\mathbb{E}[N] = \bar{N} = \mathbb{V}[N] = \lambda t \quad (2.35)$$

This property will be useful in chapter 3 where we make use of it to estimate the image noise and the SNR of a tracer particle. First, though, we need to understand how a Poisson emission process becomes a Poisson detection process.

Another great property of Poisson processes is the *thinning* property. Thinning is best described by example. Consider photons emitted by a Poisson process that encounter an attenuating material. There is a probability p that they are attenuated, and a probability $1 - p$ that they are not. In other words, each photon experiences a binary process where only one of two things happens, either of which are independent of the Poisson emission process. When many photons experience the same binary process independent of one another, the total random process is called a Bernoulli process. Let us put n photons through a Bernoulli process with survival rate p to find what the probability is that m photons survive and k photons are attenuated. Using the law of total probability, we find

$$P(M = m, K = k) = \sum_{n \geq 0} P(M = m, K = k \mid N = n) P(N = n). \quad (2.36)$$

We know that out of a total if m are survive and k are attenuated, then with 100% probability $n = k + m$ photons are attenuated. Thus, we can rewrite the above as

$$P(M = m, K = k) = P(M = m, K = k \mid N = m + k) P(N = m + k). \quad (2.37)$$

The first probability is given by the probability mass function of a binomial random variable with success rate p ; the latter is given by the probability mass function of a Poisson random variable. We then find that

$$P(M = m, K = k) = \binom{m+k}{m} p^m (1-p)^k e^{-\lambda t} \frac{(\lambda t)^{m+k}}{(m+k)!} \quad (2.38)$$

$$= \left(e^{-p\lambda t} \frac{(p\lambda t)^m}{m!} \right) \left(e^{-(1-p)\lambda t} \frac{[(1-p)\lambda t]^k}{k!} \right) \quad (2.39)$$

It follows, then, that $P(M = m)$ and $P(K = k)$ are independent Poisson processes with rates $p\lambda$ and $(1-p)\lambda$, respectively. Focusing just on those photons that survive, we find that a Poisson process that passes through a Bernoulli process is a thinned a Poisson process. This holds true regardless of how many Bernoulli processes (i.e., attenuating materials) the

photons pass through. In fact, if we take the survival probability through a material to be $p = e^{-\rho\mu x}$ and calculate the expected value, we recover the Beer-Lambert law:

$$\mathbb{E}[M] = p\lambda t = \bar{N}e^{-\rho\mu x}. \quad (2.40)$$

Similarly, the DQE of the detector acts as a success probability in Bernoulli process. The nominal PCD image noise is thus Poisson distributed.

If PPU occurs, however, then the image noise may no longer be Poisson [22]. The Poisson noise model is remarkably robust, though. For example, even if pixels are binned by summing intensities to improve contrast, the noise remains Poisson with a deterministic expected value. This is because two Poisson processes added together yields yet another Poisson process by equation 2.41.

$$Poiss(\lambda_N + \lambda_M) = Poiss(\lambda_N) + Poiss(\lambda_M) \quad (2.41)$$

2.5 Laboratory X-ray Sources

Tungsten Target Sources

One of the most common means of generating X-rays in the laboratory is with tungsten target sources (TTSs). TTSs are simple to operate and maintain, and, for many laboratory experiments, TTSs provide ample flux to acquire a good image. Like all laboratory sources, though, TTSs are orders of magnitude dimmer than synchrotrons.

A diagram depicting the internal operation of a TTS can be seen in figure 2.11. TTSs operate by first emitting electrons from a cathode. The emitted electrons are then accelerated by a strong electric potential called the source acceleration voltage. The electrons are also focused by magnetic fields into a narrow beam that impacts the tungsten target at the focal spot. As the electrons impact the tungsten target, X-rays are generated by bremsstrahlung. As discussed previously in this section, bremsstrahlung produces a continuum of photon energies, so the X-rays emitted from the tungsten target are polychromatic, meaning many wavelengths of light are emitted. In some cases, a polychromatic source is a drawback, but for rudimentary imaging purposes, a polychromatic source helps achieve enough flux to get good image quality.

Some TTSs have conical targets, emitting X-rays at an angle to the electron beam. Others have disc targets where X-rays are emitted in the same direction as the electron beam. Conical targets allow for a higher overall flux, but introduce a heel effect in which the emission is not uniform. A notable heel effect can be seen in figure 2.12. The mean columnar photon count increases across the detector. Besides the heel effect, conical targets tend to have larger focal spots as well. For example, a circular conical beam will make an elliptical focal spot on a conical target, increasing the spot size. A larger focal spot means that conical targets have stronger blurring artifacts and less spatial resolution than disc targets. Generally speaking, disc targets are used for lower flux imaging where image sharpness is

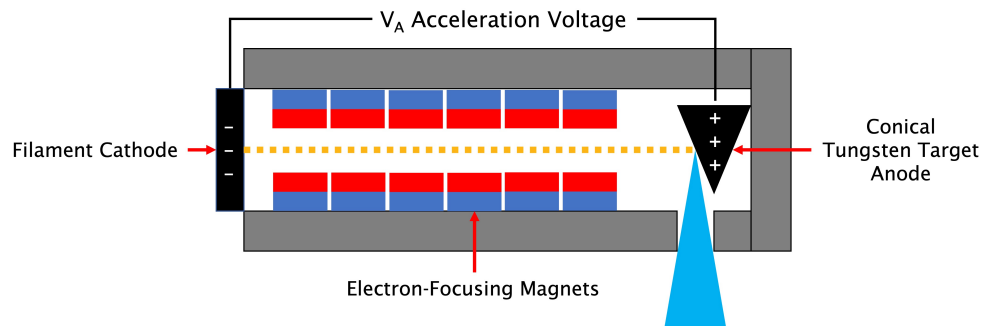


Figure 2.11: A diagram (not to scale) showing how an electron beam focused onto a tungsten target generates X-rays. The size and shape of the tungsten target may vary, but the operating principle is the same.

important, while larger conical targets are used when higher flux is needed to capture a good image.

TTSs are often limited by the heat generated from electrons slamming into the target. Active cooling can be used to increase this limit, but it is nevertheless easily reached. Eventually, to obtain a higher photon flux, the electron beam needs to be widened to disperse the heat throughout the target more so that it does not melt. Enlarging the electron beam leads to a larger focal spot, however, reducing image sharpness.

For high speed imaging with low exposure times, TTSs can be a limitation. Unless the subject being imaged offers high contrast, TTSs may not be bright enough to capture images with a good SNR.

Liquid Metal Jet Sources

Liquid metal jet (LMJ) sources, on the other hand, can overcome the heat generation limitation of TTSs. Instead of using a solid tungsten target, LMJ sources hit a high speed jet of liquid metal with electrons to generate X-rays. The liquid metal is recirculated, so the electron beam never heats a single metallic element for too long, meaning that higher electron fluxes are possible. In this way, LMJs are able to produce much greater X-ray flux than TTSs. Additionally, the electron beam does not need to de-focus for heat management purposes, so the focal spot size is constant in LMJs over a broad range of electron beam power.

LMJs make use of low-melting temperature metals like gallium and indium. Unlike tungsten, both of these metals have relatively low characteristic emission peaks; the emitted X-ray spectra tend to be softer. That is, for the same acceleration voltage, LMJs have more low energy photons compared to a tungsten target. Figure 2.13 shows the X-ray spectrum of the Excillum Metaljet E1+ source at 160kV acceleration voltage. The characteristic emission energies are at lower, softer energies. A softer source spectrum means higher contrast, but

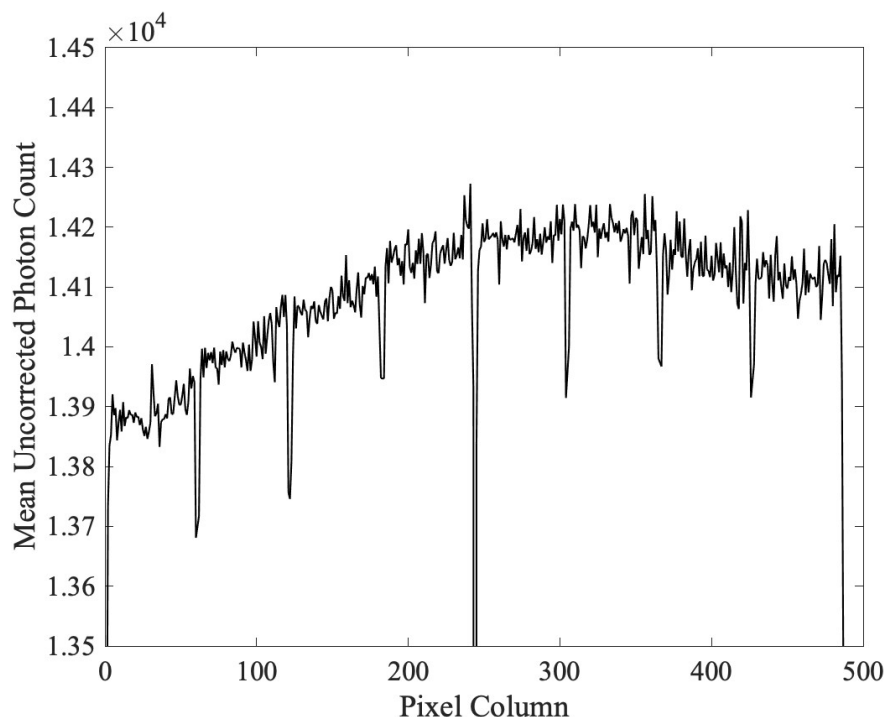


Figure 2.12: The heel effect is apparent in this plot of the mean columnar photon count in a flat field image taken with a conical target source.

also more attenuation. For thick metallic objects, LMJs may struggle to produce enough high energy photons, making the greater X-ray flux moot. In general, though, the relatively soft spectrum does not limit the usefulness of LMJs.

As of this writing, LMJs are still a relatively new technology. Companies such as Excillum are developing and selling LMJ sources, but production volumes are small and prices are prohibitively high due to research and development costs. Over the next few years, however, the cost of these sources should continue to fall as the performance continues to improve.

LMJ sources will propel X-ray imaging to higher speeds, which has benefits for flow visualization. These benefits will be discussed in chapters 3 and 5. Furthermore, since LMJ sources have such high flux, monochromators and other X-ray optics may become feasible for use in the laboratory in the future. These devices make possible a wide array of experiment techniques such as phase contrast imaging, diffraction pattern imaging, and dual energy imaging that were previously only available at synchrotrons.

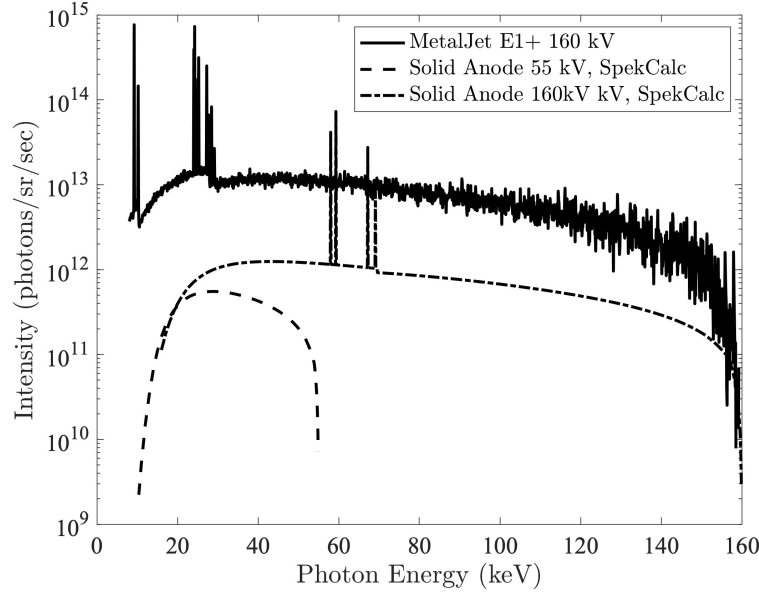


Figure 2.13: The X-ray source spectrum from the Excillum Metaljet E1+ liquid metal jet X-ray source [16] and a SpekCalc [17]–[19] simulation of the YXLON FXE225.99 solid tungsten target source.

2.6 Applications of X-ray Imaging for Flow Visualization

Armed with an understanding of X-ray physics, we can understand what makes X-ray imaging useful for flow visualization. X-rays have two important qualities that makes them useful for highly refractive and opaque fluid flows: 1) the index of refraction of X-rays is near unity; 2) many materials are transparent to X-rays. Together, these qualities mean that X-ray light will not be substantially warped by refraction in, for example, a multiphase fluid flow. Similarly, opaque flows such as blood flow could be studied.

X-rays have a number of other properties that can make them useful. In many multiphase flows material identification is important. X-ray attenuation and fluorescence properties are a direct function of the chemical composition of a material. K-edges were introduced in section 2.2; they are often used to identify materials with dual-spectrum or dual-threshold imaging. PCDs can, for example, place the threshold immediately above and immediately below the K-edge, dramatically darkening the objects comprised of that material in the flow. Fluorescent emission can similarly be used to uniquely identify materials because the emission energy is unique to the electron structure of the elements that comprise the material.

Lastly, X-rays can be used to measure the concentration of solutes in a flow. Combined with material identification techniques, this could be extended to the concentration of multiple solutes in a flow. Chapter 5 discusses how this can be done. There are many other

benefits to X-rays that make them useful (crystallography, small-angle scattering, etc.) that are too numerous to mention. The unique capabilities of X-ray light recommend it for use in fluid dynamics experimentation.

Some examples of fluid flows that would greatly benefit from examination with X-ray flow visualization techniques are:

1. Flows with transient high curvature refractive interfaces
2. High volume fraction multiphase flows
3. Porous media flows
4. Internal biological fluid flow
5. Scalar mixing and particle transport flows
6. Phase change and chemical reaction flows

among many others.

2.7 Chapter Summary

In this chapter we laid the foundation for a basic understanding of X-ray physics and imaging technology. The material in this chapter is assumed to be understood in the following chapters. We covered what X-rays are, what unique properties they have, and the ways that they interact with materials. Then, we applied these concepts to better understand X-ray imaging and source technologies. The reader is encouraged to look at *X-rays and Extreme Ultraviolet Radiation* by Sakdinawat and Attwood and *Handbook of X-ray Imaging Physics and Technology* by Russo et al. for a more detailed treatment of this material. Hopefully this chapter suffices for now, though!

Chapter 3

Simulating X-ray Images for Flow Visualization

Armed with the basics of X-ray physics, imaging, and sources, we can begin to explore applications of this knowledge. In this chapter we will delve into using the Beer-Lambert law discussed in chapter 2 to simulate X-ray images quickly. We will compare the images obtained from the Beer-Lambert simulations with images from Monte Carlo N-Particle (MCNP) simulations – the gold standard simulation method – and experiments. The content for this chapter is pulled from [23] with some modifications to reflect work that has occurred since.

3.1 Background

X-ray flow visualization equipment is expensive. An X-ray source and detector are expensive enough on their own before adding in the cost of the experiment and tracer particles. It can also be time-intensive to identify the best detector and source settings for a given experiment. The best settings change with the geometry, materials, fluid, and tracer particles that are used. Given all of this, a fluid dynamicist would like a quick, easy-to-use tool to identify the best experiment design, tracer particles, source settings, and detector settings. Additionally, if one would like to develop new tracer particles that are specifically designed for X-ray particle flow visualization, then a simulation tool would be handy for testing new designs prior to manufacturing prototypes.

X-ray particle velocimetry (XPV), which encompasses X-ray particle tracking and particle image velocimetry (XPTV and XPIV, respectively), is in its relative infancy. Lee and Kim [24] ran the first XPV experiment by conducting 2D-projected XPIV on flow in a pipe. They measured the depth-averaged velocity profile (DAVP). Since then, a lot of the work done to develop XPV further has been conducted at synchrotrons [25]–[30]. Synchrotrons are extraordinarily bright – much brighter than laboratory sources – making it easier to capture high signal-to-noise ratio (SNR) images at short exposure times.

Synchrotrons, however, pose their own problems. It is notoriously difficult to get time

at synchrotrons, making XPV an inaccessible technique. Synchrotrons also have beam sizes that are only a few millimeters across and impose tight geometric constraints on experiments. In fluid dynamics, having the ability to scale experiments to the appropriate size is vital because the ratio of forces changes with physical scale. Being able to conduct XPV in the laboratory is critical for democratizing the technique, and making it more useful. Capturing high SNR images of micro-sized particles is difficult in the laboratory since not only are the particles small, but ideally they are also density-matched with the surrounding fluid. Small, density-matched particles do not generate a lot of X-ray attenuation contrast.

Many previous laboratory experiments [31]–[34] used tracer particles that are $O(1\text{ mm})$ in diameter or were much denser than the fluid in order to obtain a sufficient SNR to conduct XPV. While these particles may have been suitable for the experiments for which they were used, smaller, neutrally buoyant tracer particles are necessary to study a wider range of flows.

Besides a brighter source and more attenuative tracer particles, another way to improve SNR is to decrease the noise level. This can be achieved with a photon counting detector (PCD). As discussed in chapter 2, PCDs remove the Gaussian noise associated with thermal and electronic signal noise. PCDs also reduce blurring as well as post-scintillation visible light scattering and refraction effects.

In this chapter, we explore simulating X-ray images from photon counting detectors (PCDs) using Beer-Lambert ray tracing (BL) and MCNP so that laboratory XPV users can design their experiments appropriately, conjure up new tracer particles designs, or both. The work this chapter is based on, [23], was the first to demonstrate that silver-coated hollow glass tracer particles could be used as neutrally buoyant XPV tracer particles. We capture images of a particle on a glass slide to validate our simulations.

3.2 Experimental Setup

X-ray Imaging Setup

A schematic of the experimental setup is shown in figure 3.1. The X-ray source used for these experiments is an YXLON FXE225.99 TwinHead. TwinHead refers to the ability to swap the source heads between a transmission and directional head. This source is capable of achieving up to 225 kV with either head, with a maximum tube power of 64 W with the transmission head and 320 W for the directional head. The directional head is brighter, owing to its higher wattage, but sacrifices sharpness due to a larger, non-circular focal spot. For these experiments, the focal spot size was approximately $6\text{ }\mu\text{m}$ with the directional head. The directional source head has a water-cooled tungsten target and a millimeter-thick aluminum window. For our experiments, the directional head is used for higher photon flux.

The detector used is a Dectris Pilatus3 X 100K–M cadmium telluride (CdTe) detector with a 1 mm thick CdTe panel, 487×195 ($172\text{ }\mu\text{m}$)² pixels, and a single photon-energy threshold. For this study the energy threshold is set to 20 keV. The Pilatus3 detector is

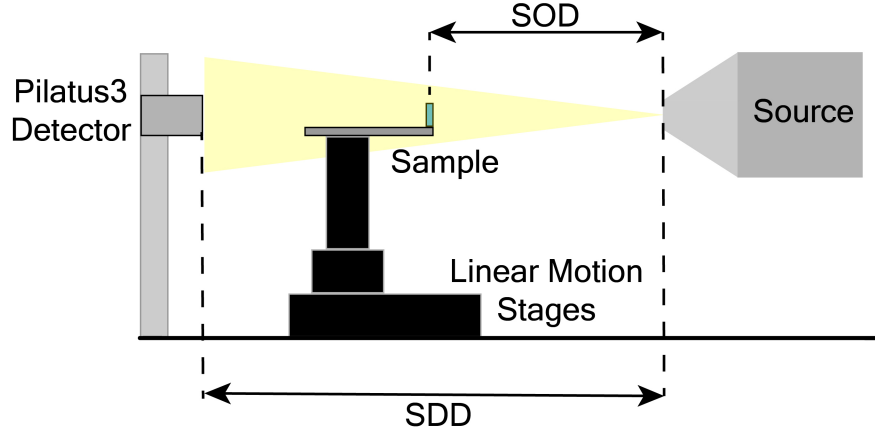


Figure 3.1: A diagram of the X-ray imaging system. The SOD and SDD are 1 cm and 47.6 cm, respectively.

capable of counting approximately 10^6 photons per pixel per second without correction, which introduces a limit for image quality. The detector can record images at a rate of up to 500 frames per second, but for these experiments it is operated at fps Hz with an exposure time of 100 ms. The detector temperature was maintained with a chiller at $23.0 \pm 0.1^\circ\text{C}$ and a continuous dry nitrogen gas purge was used to prevent the buildup of humidity within the detector. The raw data is recorded by the Pilatus3 detector. Prior to recording the images of the particles, a series of flat field images are recorded to generate a correction map. This correction map eliminates detector artifacts and weak pixels when applied to a data image.

Three ThorLabs NRT150 linear stages control the placement of the object to be imaged. Each stage has a 150 mm range of motion, $0.1 \mu\text{m}$ minimum incremental motion, and a calibrated on-axis accuracy of $2 \mu\text{m}$. Samples are held in place with 3D printed mounts that are bolted to the linear stages. When imaging the particles on a glass slide, as shown in figure 3.1, the source-to-object distance (SOD) is 1 cm to maximize the geometric magnification effect. The source-to-detector distance (SDD) is 47.6 cm, so the geometric magnification $M = 47.6\times$.

Tracer Particle

For quantitative XPV, the tracer particle must be neutrally buoyant and have a Stokes number much less than unity [3]. The AGSF-33 particles from Potters Industries LLC are selected for this study as a potential candidate for XPV because they are nominally neutrally buoyant in water and their silver coating enhances X-ray attenuation contrast. With a nominal diameter of $50 \mu\text{m}$, AGSF-33 particles will have a Stokes number less than unity for a wide range of flows of interest and can provide reasonable spatial resolution in XPV.

The particle geometry must be known to accurately simulate a particle. While the actual shape of a particle may deviate from a perfect sphere, we assume spherical symmetry so multi-layer particles of arbitrary composition can be easily modeled. In the case of AGSF-33, the particles are known to be hollow Pyrex spheres that have been coated with silver that accounts for approximately 33% ($\gamma = 0.33$) of the particle mass. The glass wall and silver coating thickness can be calculated by assuming neutral buoyancy in water and using the manufacturer's nominal outer diameter, 50 μm . Let R_0 be the inner radius and R_1 be the outer radius of the Pyrex wall, and R_2 the outer radius of the silver coating. In this case, $R_2 = D/2 = 25 \mu\text{m}$. Then, given the densities of air, Pyrex, silver, and known value of γ , R_1 and R_0 are readily calculated by equations 3.1 and 3.2, respectively.

$$R_1^3 = \frac{\rho_{silver} - \gamma\rho_{average}}{\rho_{silver}} R_2^3 \quad (3.1)$$

$$R_0^3 = \frac{(1 - \gamma)\rho_{average}R_2^3 - \rho_{pyrex}R_1^3}{\rho_{air} - \rho_{pyrex}}, \quad (3.2)$$

where ρ_i is the density of material i . For AGSF-33 we find $R_0 = 21.9 \mu\text{m}$ and $R_1 = 24.7 \mu\text{m}$. These radii are used in the BL and MCNP simulations.

Particle Settling

While the AGSF-33 particles are nominally matched with the density of water they, like most tracer particles, may have a non-uniform density within a batch. Density mismatches with the fluid induce relative motion due to buoyancy, which can bias velocity measurements. While for visual light particle velocimetry the buoyancy induced velocity may often be neglected, the first flows that will be measured with XPV tend to be low speed or creeping flows due to the present technological limitations. Particle settling speeds and buoyancy must be considered at these flow speeds. When the relative velocity is low enough for Stokes drag to be appropriate ($Re_{relative} \ll 1$), the speed with which particles will rise or settle in a fluid can be calculated from equation 4.4.

$$U_{St} = d_p^2 \frac{\rho_{fluid} - \rho_{particle}}{18\mu} g \quad (3.3)$$

Here, d_p is the particle diameter, ρ_i is total density of i , μ is the dynamic viscosity of fluid, and g is the acceleration due to gravity. We took nano-CT scans of a sample of the AGSF-33 particles to ascertain to the size distribution. Based on the segmented CT scans, a slice of which can be seen in figure 3.2a, we take the particle diameter to be log-normal distributed. The AGSF-33 size distribution is shown in figure 3.2b along with the probability density function (PDF) we use for the Monte Carlo simulations. We assume, for simplicity, that the density is normally distributed and statistically independent of the particle diameter. Potters Industries reports the particle density to be between 0.9 and 1.1 g/cm^3 . We truncate the density distribution at these values. This truncation does not affect the Monte Carlo

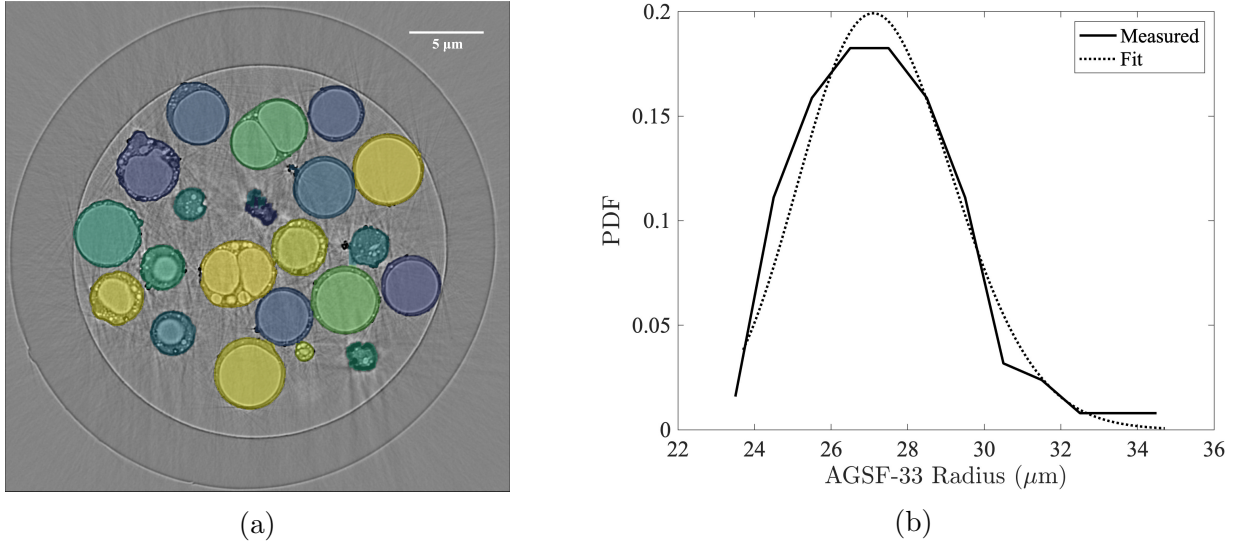


Figure 3.2: (a) A slice of the nano-CT scan captured by project collaborator Till Dreier at Lund University. (b) The measured and fit PDF of the particle radius of the AGSF-33 tracer particles.

calculation results significantly, but we sought to make our calculations as representative of reality as possible. We then simulate equation 4.4 10^8 times to ensure that the Monte Carlo calculations converge. The results of the calculation can be seen in figure 3.3. On average, AGSF-33 particles rise in water at roughly $0.01 \mu\text{m/s}$ and rise in glycerine at $0.42 \mu\text{m/s}$. The standard deviations for water and glycerine are $59.1 \mu\text{m/s}$ and $0.1 \mu\text{m/s}$, respectively. The large standard deviation for water can pose problems for measurements. Monodispersity in the particle buoyancy is necessary to avoid errors in the velocity measurement. In glycerine, however, the settling speed becomes more monodisperse due to the higher viscosity. For all but the slowest flows, AGSF-33 tracer particles in glycerine should be suitable with regards to buoyancy induced velocity error. For faster flows $O(0.5 \text{ mm/s})$ or more, the AGSF-33 particles will still be suitable in water.

3.3 Simulation Methodologies

Beer-Lambert Simulation

Equation 3.4 is a version of the Beer-Lambert law discussed in chapter 2. It which describes the attenuation of photons at energies ϵ as they pass through J media of thickness x_j that have mass attenuation coefficient $\mu_j(\epsilon)$ and density ρ_j . The attenuation coefficient for each material is obtained from the NIST X-ray attenuation database [15]. In this form, we assume the incoming photon intensity is a polychromatic spectrum. To emulate how a PCD works,

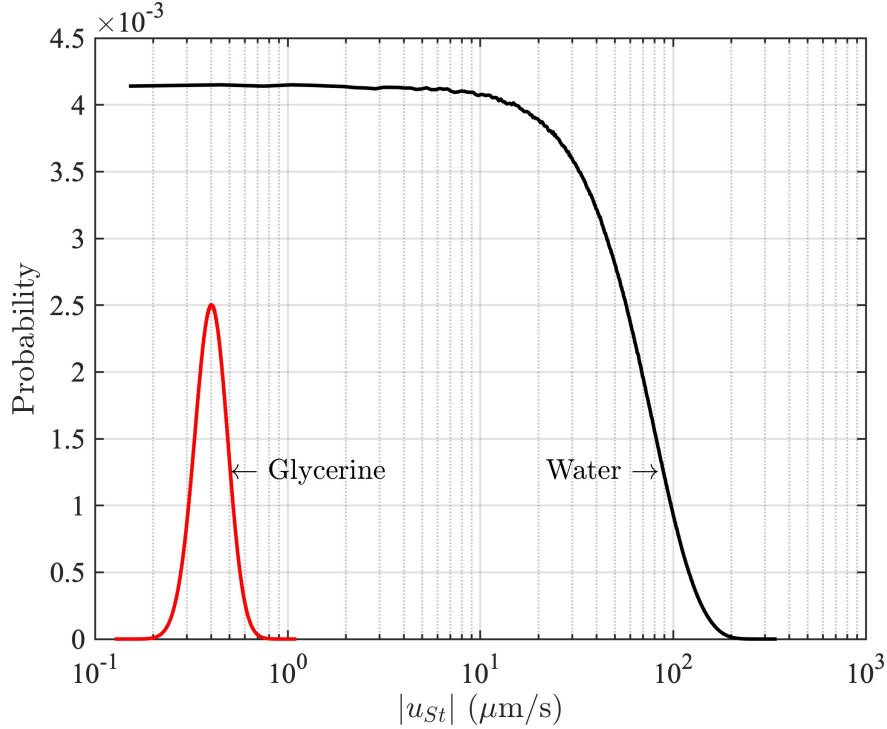


Figure 3.3: The AGSF-33 settling speed magnitude distribution. Glycerine slows the particles due to its higher viscosity. Some particles sink, but the magnitude of the settling/rising speed is shown here to accommodate the log-scale axis.

we integrate over all energies above the detector energy threshold because any detected energy pulse is added to the count indiscriminately.

$$I = \int_{E_{thresh}}^E Q(\epsilon) I_0(\epsilon) \Delta t e^{-\sum_{j=1}^J \rho_j \mu_j(\epsilon) x_j} d\epsilon \quad (3.4)$$

Here, E_{thresh} is the detector threshold energy, E is the maximum photon energy, Q is the efficiency of the detector, Δt is the exposure time, I is the detected photon intensity, and I_0 is the source photon intensity, the spectrum for which can be seen in figure 3.4.

Equation 2.2 is a steady state form of the Beer-Lambert equation. Assuming the subject being imaged does not move or change in composition over the course of a single exposure (XPV exposures are ideally shorter than the characteristic time of a flow) the steady state form can predict a detector image. A diagram of the BL simulation geometry as implemented in the present study can be seen in figure 3.5. The SDD is 47.6 cm as measured from the center of the detector. An AGSF-33 particle is set on a glass slide and the SOD as measured from the center of the particle to the source is 1 cm. The AGSF-33 particle geometry is discussed in Section 3.2.

A ray-tracing MATLAB code was developed to calculate the number of photons that are detected by each pixel. Critically, each pixel is divided into sub-pixels to account for geometry projection variations across a single pixel. The error associated with sub-pixel variation was found to be negligible (below 10% in the worst case scenario) when 121 (11×11) sub-pixels are used. The Dectris Pilatus3 crystal surface components are to the first order transparent to the incoming radiation, so the pixels have a 100% fill factor. We therefore consider each sub-pixel to detect photons equally. The convergence of the pixel photon count with increasing sub-pixels is discussed in chapter supplementary material section A at the end of this chapter. The photon intensity for each sub-pixel is calculated using equation 3.4. For each object along the ray, the thickness x_j is calculated by taking the Eulerian distance between the two intersection points (entry and exit) of the ray with the object. The detected photon intensity for all sub-pixels within a pixel are summed to calculate the number of detected photons for that pixel. In this way, the number of detected photons are calculated for each pixel in the detector, thus initially simulating a noiseless X-ray image taken with a PCD.

While PCDs exhibit no Gaussian thermal noise, the emission process of the X-ray source and the binomial nature of photon attenuation introduce Poisson noise. To simulate a more realistic image, Poisson noise is added to the image calculated with equation 2.2. To add Poisson noise, we take each noiseless calculated pixel value to be the expected value of a Poisson distribution for that pixel. A random value is selected from said distribution to be the noisy pixel value. It is important to note that adding pure Poisson noise assumes that the effects of photon pileup are negligible [22].

The source intensity spectrum $I_0(\epsilon)$, shown in figure 3.4, is calculated using SpekCalc [17]–[19]. The actual source intensity spectrum and its spatial distribution are not known precisely. In order to calculate the number of photons to simulate for a given exposure time, experimental flat images are used to calculate the average number of photons emitted from the source that are detected. The solid angle of the detector is known; the source has a 15° cone angle, so the total number of source photons is calculable, assuming uniform emission. Attenuation through 47.6 cm of air was accounted for by taking the average air attenuation ratio across all photon energies, then calculating an estimate for $\sum_{\epsilon} I_0(\epsilon)$ based on equation 3.4. In reality, the source emission is not spatially uniform due to the heel effect, and the attenuation of X-rays through air is strongly energy-dependent. Improved source characterization could eliminate errors originating from spectrum and flux approximations, improving simulation accuracy. In the absence of an X-ray source and detector to measure the source flux, SpekCalc has been found to adequately estimate the source flux in addition to the intensity spectrum. In this study, we approximately measure flux as described above, but rely on SpekCalc for the intensity spectrum.

While BL simulations are computationally inexpensive, the approach entails several simplifying assumptions. Most consequential of these is that BL simulations assume all scattered photons are attenuated and not detected. In general, this is not true. Many scattered photons may experience a shallow scattering angle and will be detected even if they are scattered from their original ray. Photons may also experience scatter within the detector,

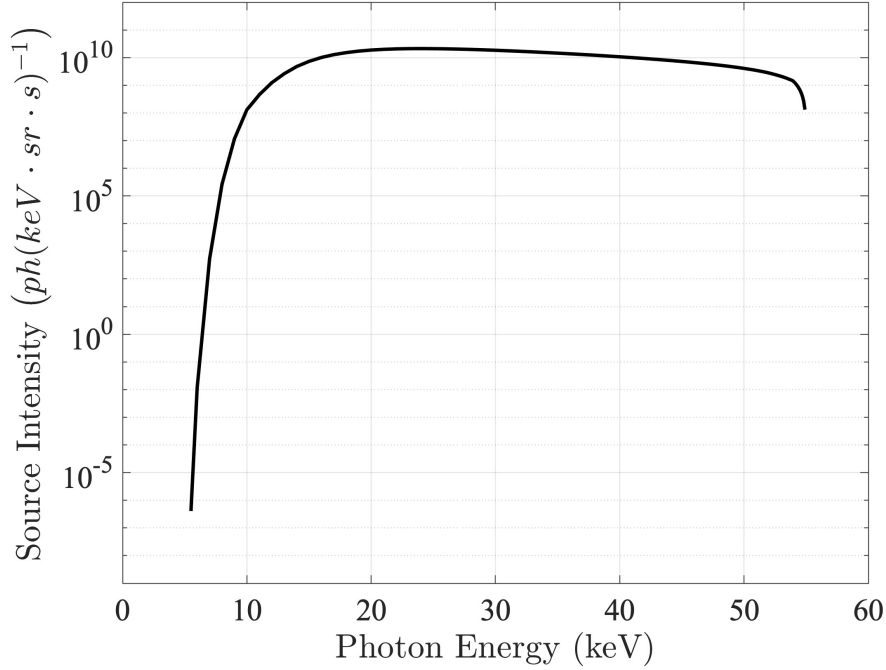


Figure 3.4: SpekCalc source spectrum of a 22.5° half-angle conical tungsten target at 55 kV with a 1 mm thick aluminum window.

a process discussed in greater detail in section 3.3. As a result of the scattering attenuation assumption, BL will underestimate the number of detected photons and noise intensity.

For computational efficiency, source photons are emitted uniformly from a single, infinitesimal point. We assume uniform emission for simplicity, but in reality the source emission is not uniform due to the heel effect. An infinitesimal source neglects the blurring effects from a finite focal spot. Blurring from a finite focal spot reduces image contrast. One could account for a finite focal spot in BL simulations with distributed emission locations, spatial non-uniformity of emissions, and randomized emission flux. However, the computational advantage of BL simulations as a design tool would diminish if we consider thousands of emission sources and hundreds of sub-pixels.

Although we make simplifying assumptions to reduce the computational cost of the BL simulations, the MATLAB code developed for this study is not optimized for computational efficiency. The calculations are done on a CPU and are not parallelized. Parallelization on a GPU is low-hanging fruit for improvement. There is a wealth of literature on ray-tracing methods [35]–[38] that the reader can refer to for pathways to speed up the BL simulation method. For more recent advances in ray-tracing, we refer the reader to [39], [40]. For the purposes of this study, we simply consider the accuracy of the BL simulations compared to real X-ray images as the benchmark of simulation performance. This work is simply meant to show that BL simulations can be used as a computationally inexpensive potential design

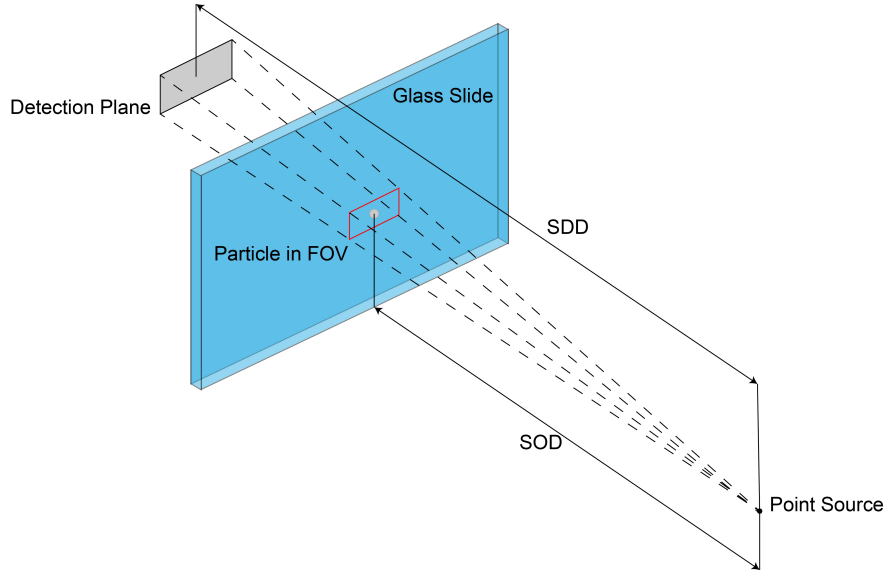


Figure 3.5: A point source generates rays along which the Beer-Lambert law is evaluated at the detector plane. The detector itself is not in the geometry. Here, an AGSF-33 particle is placed on the front surface (surface towards source) of a glass slide. Figure not to scale.

tool.

Lastly, X-ray fluorescence [12] is not considered in the BL simulations. We ran identical simulations using MCNP to observe the effects of scattering, fluorescence, and a finite focal spot. MCNP is discussed in section 3.3.

Monte Carlo N-Particle Simulations

Monte Carlo N-Particle (MCNP) is a particle transport simulation code developed by Los Alamos National Lab [41]. In MCNP, each photon is assigned an energy, departure direction vector, and initial position in the focal spot based on known or assumed distributions. Using databases compiled from experiment data over the past five decades, MCNP calculates the probability of different photon interactions with a medium as well as the probability of the outcome of said interactions. For example, as a photon passes through water, there are probabilities for that photon being scattered, absorbed, or passing through unperturbed. By simulating as many photons as the experiment source emits for a given exposure time, one can reconstruct the detector image. Although MCNP is more computationally expensive than the BL simulations discussed in 3.3, it incorporates scattering effects both in the domain and the detector, a finite source size, and fluorescence.

MCNP is a highly optimized program that has been used – particularly in the nuclear engineering and radiation protection communities – for nearly five decades. The user provides the MCNP program with an input file that dictates the domain, geometry, source

characteristics, data to be exported, etc., without modifying the MCNP code itself.

MCNP Simulation Setup

The MCNP domain geometry defined for this study can be seen in figure 3.6. A 6 μm radius finite source emits a 15° conical beam with uniform photon flux [20]. The source photon energy probability density function is calculated from the same SpekCalc spectrum used for the BL simulation. Unlike the BL simulations, which do not have a detector in the domain, the detector in MCNP is modeled as a uniform CdTe panel in the domain. In order to reduce unnecessary photon tracking, the domain is 60 cm by 20 cm by 20 cm with non-reflective boundary conditions. In other words, it is assumed that any photons that leave this domain will not scatter back to the detector.

MCNP simulates a path for each source photon with the Monte Carlo method based on the probabilities of an interaction between the photon and the domain object(s). If a photon enters the CdTe panel, the location and energy of that photon are outputted as the photon scatters or is absorbed and deposits energy within the CdTe panel. Then, in post-processing, we count how many photons entered each pixel and deposited enough energy to be detected, i.e., deposited energy greater than or equal to our experiment energy threshold. In a PCD such as the one we use in this study, the simulated image pixel intensity is simply the number of photons that are counted for each pixel. The post-processing to generate the image is done in MATLAB.

Notably, MCNP does not simulate the number of source photons emitted per unit time, but rather simulates a fixed number of photons that in the physical world corresponds to an actual emission time with some probability. Using the same procedure as was described in 3.3, MCNP simulated approximately the same number of source photons as the BL simulations and experiment images, thus approximating an image with an exposure time equal to the experiment images.

MCNP Processing Methodology

The output method of choice for our purposes is the particle track (PTRAC) file. A PTRAC file contains every interaction that every photon has with the environment. To simulate the image formed at the detector, two filters are applied to the PTRAC file in order to restrict the output to relevant interactions. The first filter reduces the reported interactions to only those that occur within the detector. The second filter is applied to only include interactions that would generate a signal within the photon-counting detector. A snippet of a PTRAC file can be seen in figure 3.22 in chapter supplementary material B along with a detailed explanation. A PTRAC file output gives us the ability to change pixel size and detector energy thresholds a posteriori. Since MCNP simulations take so long to run, it is beneficial to be able to reuse one set of output data. As mentioned previously, the PTRAC file(s) are inputted into MATLAB for post-processing to generate the simulated image.

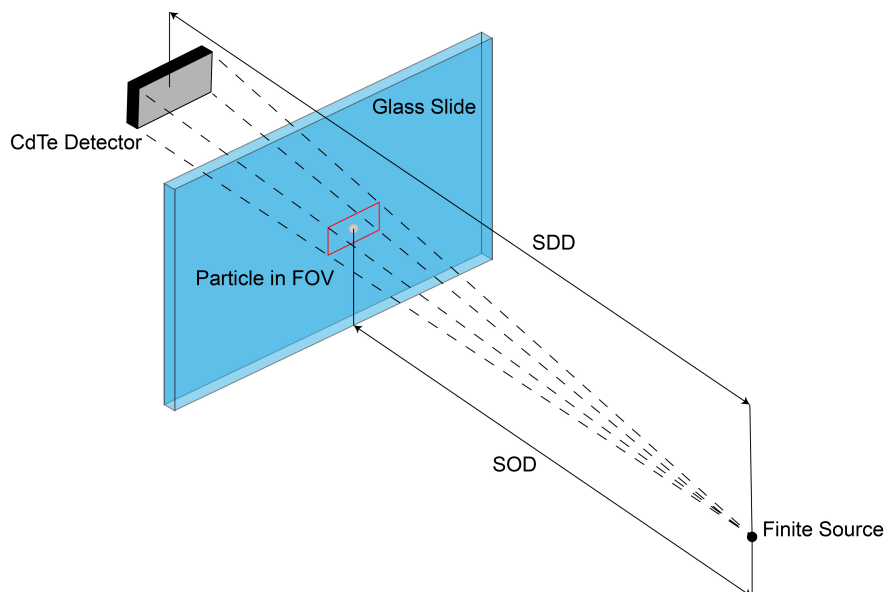


Figure 3.6: The MCNP geometry is similar to the BL simulation setup of figure 3.5, but includes a CdTe-detector crystal of finite thickness (omitting housing and electronics for simplicity) and accounts for the finite X-ray source spot. Figure not to scale.

For the present study, a major benefit of using a PTRAC file output in combination with a CdTe panel in the domain is the ability to simulate multi-detection photons. Multi-detection photons are photons that deposit sufficient energy to be detected in one pixel before scattering to an adjacent pixel and being detected again, a process we refer to as internal scattering. A single photon can then be detected two or more times. Figure 3.7 depicts how multi-detection could occur. Figure 3.8 shows a multi-detection event from an MCNP simulation.

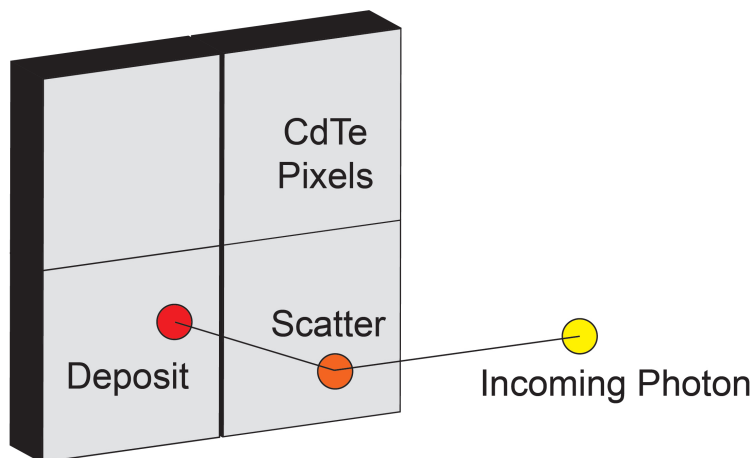


Figure 3.7: An incoming photon can enter one pixel, deposit sufficient energy to be counted, then scatter to another pixel where it is counted once more.

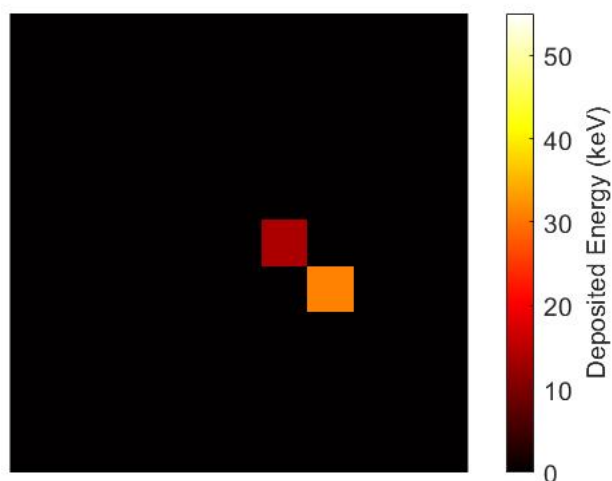


Figure 3.8: A single photon in an MCNP simulation is detected in two separate pixels. Multi-detections such as this one lower the image SNR.

In order to ascertain the impact of internal scattering, one set of MCNP simulation data was processed in two ways. First, the simulation data was processed such that a photon was detected once in the pixel it first enters. Processing in this way will not count multi-detections. Next, the simulation data was processed to include internal scattering effects. The difference between the former and latter processing is approximately 8% of the average

multi-detection pixel count. Internal scattering clearly should be included when simulating a PCD image. The ability to do so in MCNP is an advantage over BL.

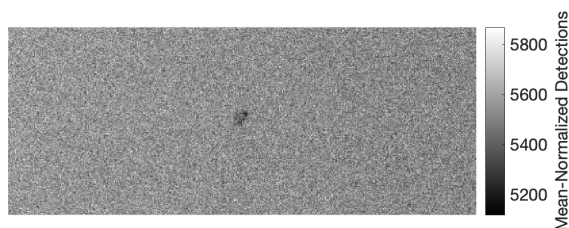
3.4 Results and Discussion

Experiment and Simulation Image Comparison

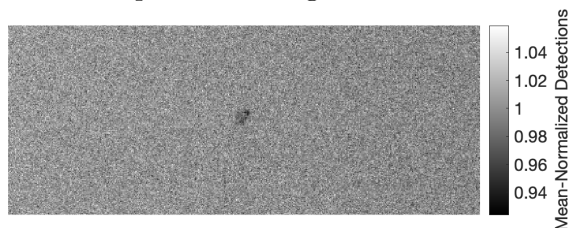
Figure 3.9 shows a flat field corrected image of a particle taken at a source voltage of 55 kV, target current of 500 μA , and a 100 ms exposure time. The nominal detector threshold is 20 keV. The SOD is 1 cm; the SID is 47.6 cm. In figure 3.9 we see a single AGSF-33 particle taped to a 1 mm thick glass slide. Although the particle is clearly detectable, the ability to localize the particle for PIV can be enhanced with simple de-noising techniques specific to Poisson-noisy images. Figure 3.9c shows the image in figure 3.9 after applying a PURE-LET deconvolution [42], a CLAHE filter [43], and finally a median filter. After filtration, the particle is even more visible. PIV image correlation could be performed on an image containing many of these particles. For the purposes of this paper, however, we will not attempt to optimize settings for localizing a single particle in the shortest possible exposure. Instead, we will focus our discussion on the simulation accuracy and design utility. Figure 3.10 shows the BL image with Poisson noise added. The simulation settings are the same as the experimental settings for figures 3.9 and 3.9c, with the source flux matched. The BL simulation assumes a point source, which produces an image without the realistic blurring due to a finite focal spot. In the BL image, the silver coating is clearly resolved, whereas in the experiment image the coating is blurred with the rest of the particle. Simple blurring can be added to the BL image without the computational cost of simulating multiple source locations if we add Gaussian blur, as seen in figure 3.11. Clearly, blurring degrades SNR; figure 3.11c qualitatively resembles the experiment data. As previously noted in section 3.3, more could be done to quantitatively address image blurring in BL simulations while balancing computational expense.

The most significant limitation of the BL simulation, however, is that it cannot simulate internal or external scattering. As noted in the caption of the normalized images, the average intensity of the BL image is notably lower than the experiment data. The depressed photon detection count can be attributed to the attenuation assumption for scattered photons. Scattered photons in BL are not detected, which in general is not true. Many scattered photons may experience shallow scattering angles, and so could still be detected. Furthermore, photons may be detected multiple times due to scattering within the detector panel.

The MCNP image, seen in figure 3.12, matches the experiment image better than the BL image seen in figure 3.10. Qualitatively, the first thing we notice is that the particle is slightly blurred, primarily due to MCNP modeling an idealized circular finite focal spot. If all blurring were attributable to focal spot blurring, the blur kernel should be approximately 1.6 pixels across. The MCNP image qualitatively matches figure 3.11a, which has a kernel standard deviation of $\sigma = 1$ pixel. However, the experiment image qualitatively agrees best



(a) Flat field corrected X-ray image of an AGSF-33 particle on a glass slide.



(b) The mean-normalized X-ray image of data shown in (a). The average pixel intensity is 5,540 photons.



(c) A PURE-LET, CLAHE, and median filtered version of (a).

Figure 3.9: A sample image from the experiment images collected at 55 kV, 500 μ A for a 100 ms exposure time. The filtered image shows higher particle contrast, which would improve the PIV correlation peak or the PTV centroid localization.

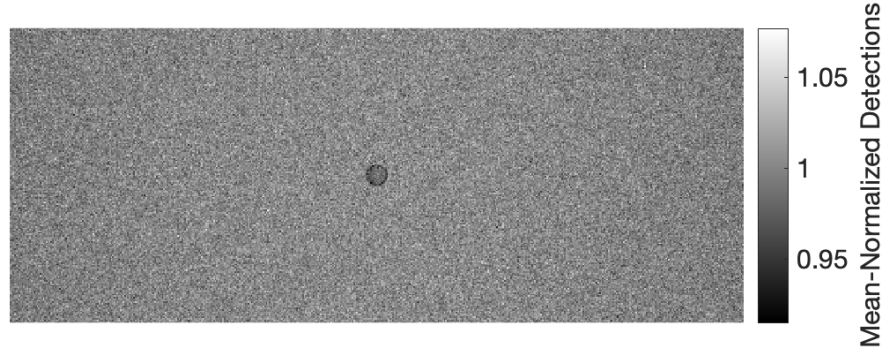


Figure 3.10: A normalized BL simulated Poisson-noisy image. The simulated source flux was matched to the experimentally calculated value. The average pixel intensity is 4,661 photons.

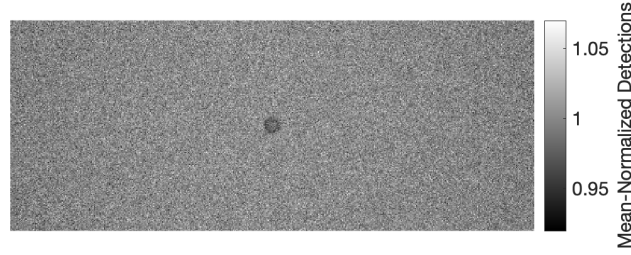
	Mean	Variance
Experiment	5,540	5,622
MCNP (with edges)	5,074	7,344
MCNP (without edge pixels)	5,079	5,604
BL	4,661	4,803

Table 3.1: BL, MCNP, and experiment image photon detection distribution moments.

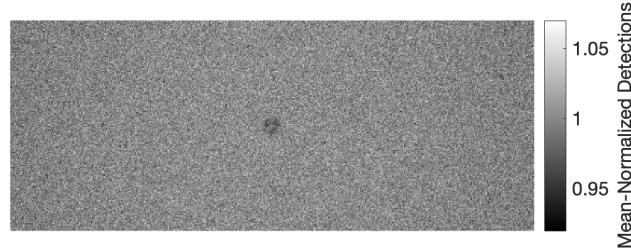
with figure 3.11c, where the kernel standard deviation is $\sigma = 3$ pixels. This suggests that additional blur is attributable to a non-circular focal spot, the resulting heel effect, detector imperfections, or some combination thereof.

Quantitatively, the average photon detections per pixel in the MCNP image approaches that of the experiment image, suggesting that scatter attenuation is the main reason for depressed photon detections in BL. Figure 3.13 compares the distribution of photon detections per pixel from the experiment, BL, and MCNP. The distribution moment data are shown in table 3.1. While MCNP more closely predicts the experimental statistics, it still does not align exactly with what was measured. This is to be expected, since MCNP cannot account for detector crystal imperfections, anisotropy in the detector, or non-Poisson noise behavior due to photon pile-up in the detector. MCNP did not simulate photons scattering to the detector from outside the domain or the detector housing and electronics, as these external materials were not included in the simulation geometry. Furthermore, neither MCNP nor BL can account for inaccuracies in the simulated source spectrum or calculated source flux. Improving the source characterization is one way to improve the simulation accuracy in the future.

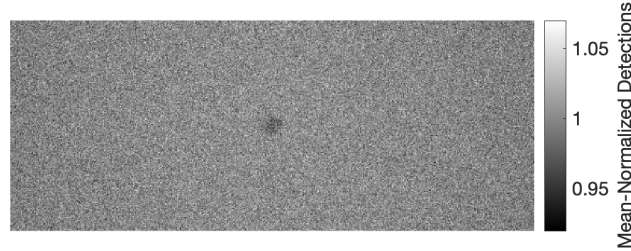
In figure 3.13 there is a slight bump in the MCNP histogram at about 4,800 photon detections. This bump is an artifact of the MCNP model geometry. Our model geometry



(a) 55 kV, 500 μ A BL simulation with Gaussian blur; $\sigma = 1$, blur span = 5 pixels square.



(b) 55 kV, 500 μ A BL simulation with Gaussian blur; $\sigma = 2$, blur span = 9 pixels square.



(c) 55 kV, 500 μ A BL simulation with Gaussian blur; $\sigma = 3$, blur span = 13 pixels square.

Figure 3.11: Gaussian blur degrades the SNR of the BL simulated image. In the future, a finite source focal spot may be simulated by adding Gaussian blur. The average pixel intensity for all images is 4,661 photons.

does not have any material surrounding the CdTe panel, so the edge pixels have less adjacent material from which photons may scatter back into the detector pixels. Statistics in table 3.1 are provided for the MCNP image with and without the edge pixels.

Despite the greater computational expense, it may be best to use MCNP for imaging conditions in which scatter, fluorescence, and focal spot blurring are more substantial. In the following section, however, we demonstrate that in some cases, BL can offer effectively the same accuracy as MCNP at a fraction of the computational cost.

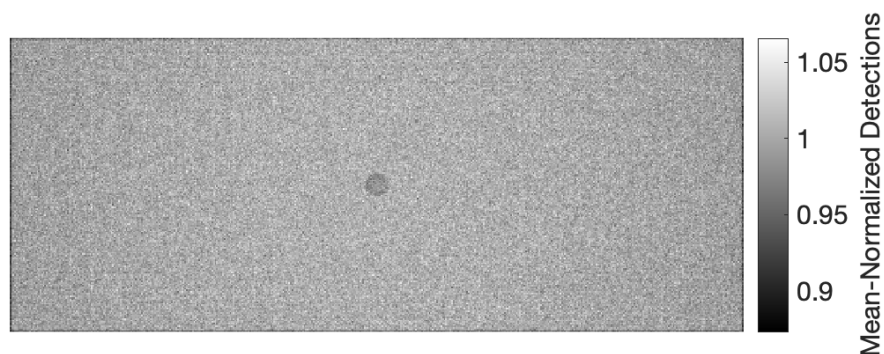


Figure 3.12: The MCNP simulation image of an AGSF-33 particle on a glass slide. The MCNP image shows blurring from the finite focal spot. The average pixel intensity is 5,074 photons.

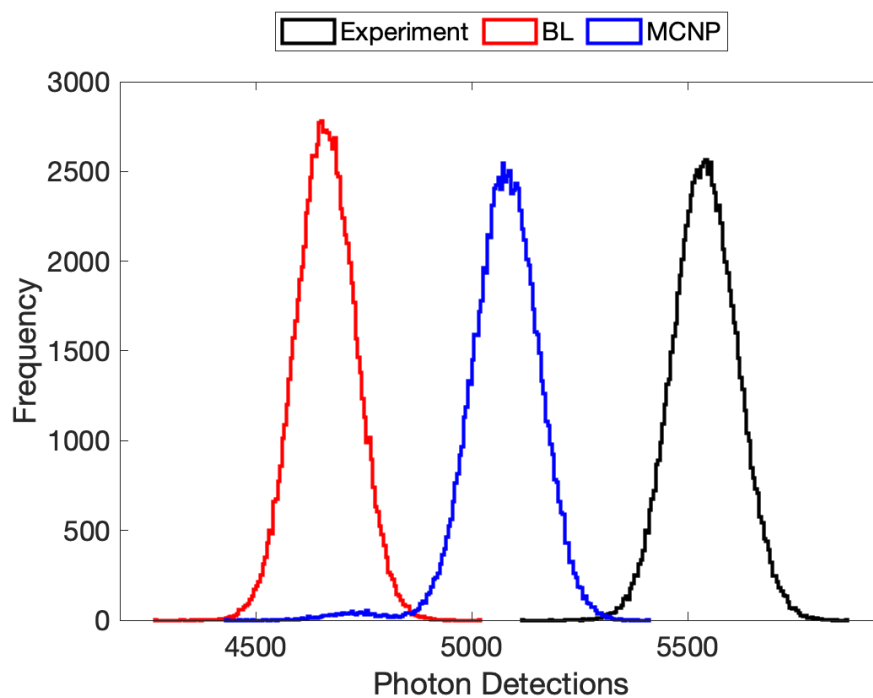


Figure 3.13: The MCNP, BL, and experiment image photon detection distributions. A small peak to the left in the MCNP distribution is due to the depressed photon counts in the MCNP edge pixels. MCNP is more accurate than BL, but still does not match experiment exactly.

Predicting Image SNR For XPV System Design

MCNP and BL simulations can be used as tools to help XPV experiment designers methodically improve their system. BL simulations prove useful in design applications such as predicting image SNR behavior. SNR can be used as a metric to gauge the detectability of tracer particles in an image. XPV system performance improves as the particle detectability improves. Figure 3.14 shows the SNR as a function of source voltage for the experimental, BL, and MCNP images. As expected, the BL simulations overestimate the SNR; MCNP is more accurate, but only slightly. Practically speaking, BL simulations are the preferred predictive tool by virtue of being computationally inexpensive. As figure 3.14 demonstrates, BL simulations are able to explore a wide parameter space relatively quickly, unlike MCNP.

The SNR is calculated as the ratio of the particle-background contrast and the background noise intensity. The particle contrast was calculated by taking the difference in average pixel value for the particle and for all pixels within one particle diameter of the particle edge (i.e., the background pixels). The contrast was then divided by the standard deviation of the background pixels. The standard deviation approximates the background noise intensity. For the BL SNR, the standard deviation was calculated assuming a perfect Poisson distribution, so the standard deviation is taken to be the square root of the average background pixel value.

The BL simulations predict the SNR behavior best in the low source voltage regime where scattering is less likely and the Poisson noise regime holds. However, the predicted and measured curves diverge at higher source voltages as BL underestimates the noise intensity and overestimates the contrast, as seen in figure 3.15. The contrast is higher in the BL images because there are no blurring effects, as discussed in figure 3.11. MCNP, on the other hand, incorporates blurring effects due to scatter and a finite focal spot with ease, and as a result predicts contrast slightly more accurately. Both MCNP and BL underestimate the noise intensity. This makes sense, considering both BL and MCNP are limited to Poisson noise and idealize the detector. Experiment images must contend with detector artifacts and anisotropy in the CdTe crystal. The noise intensity is also a function of the number of source photons, so improving source flux modeling is crucial for improving SNR prediction.

It is important to note that in PCDs in particular the image noise is not perfectly Poisson due to pileup effects [22]. In some situations it may be advantageous to consider energy integrating or scintillator-based X-ray imagers. When the photon flux rate is small compared to the maximum photon count rate of the imager, the effects of pileup can be neglected and the noise can be taken to be Poisson. The BL and MCNP simulations neglect pileup effects. As the source voltage increases, modeling image noise as Poisson becomes less tenable, which could partially explain the noise intensity divergence seen in figure 3.15, in addition to the effects mentioned previously.

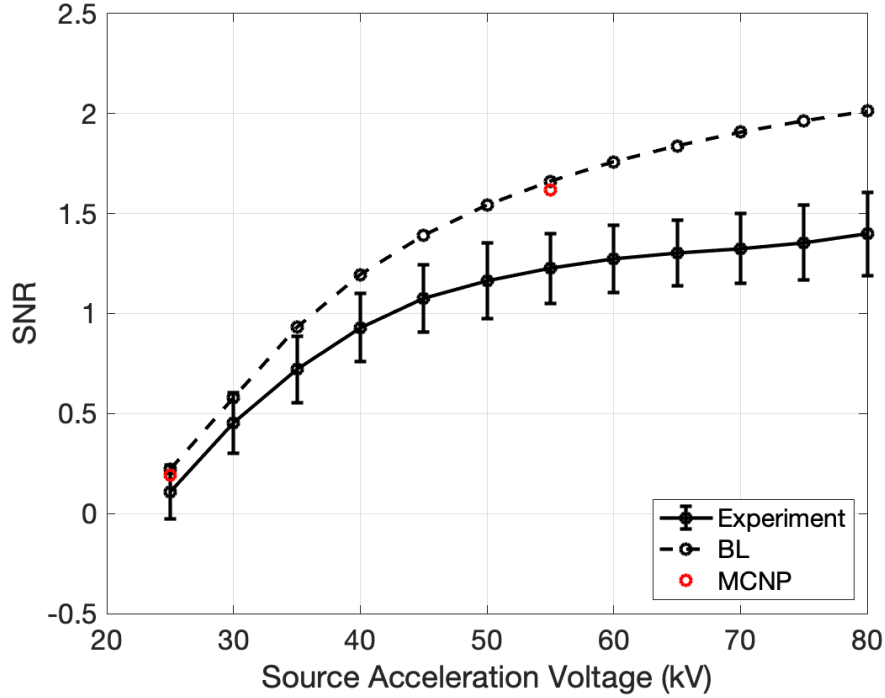


Figure 3.14: The image SNR from experiments and BL simulations as a function of the source voltage. The MCNP SNR for the 55 kV image is shown for comparison.

Multiple-Particle Simulations

In this study, we focus on single-particle images. They are simpler to simulate and provide an easily replicable geometry for measuring SNR in an experiment. However, in realistic XPV experiment geometries, particles will be seeded at much higher densities, which would increase the image noise intensity due to scattering from the particles to the detector. We examine this effect by simulating multiple particles on a glass slide. Here, we did not investigate the effects of overlap because the likelihood of photon scattering from one particle to another, then having enough energy to be detectable, is extremely low. To confirm this, we ran a simple MCNP simulation with a 30keV pencil beam pointed through the center of two tracer particles in a vacuum. The two particles are separated by $1 \mu\text{m}$. Photon scatter between particles and to the detector should be much more prevalent under these circumstances than normal XPV seeding conditions. We choose a 30keV beam because that is the average photon energy of our polychromatic 55kV source spectrum. After simulating 1.2 million photons, we found the photon counts from MCNP to be only 4.4% lower than BL (we neglected multi-detection for these simulations). For overlapping particles, direct transmission attenuation is the primary phenomenon for simulating overlapped particle images. Both BL and MCNP simulate this phenomenon accurately. Furthermore, we did not consider overlapping particles in order to isolate the effect that multiple particles' scatter to

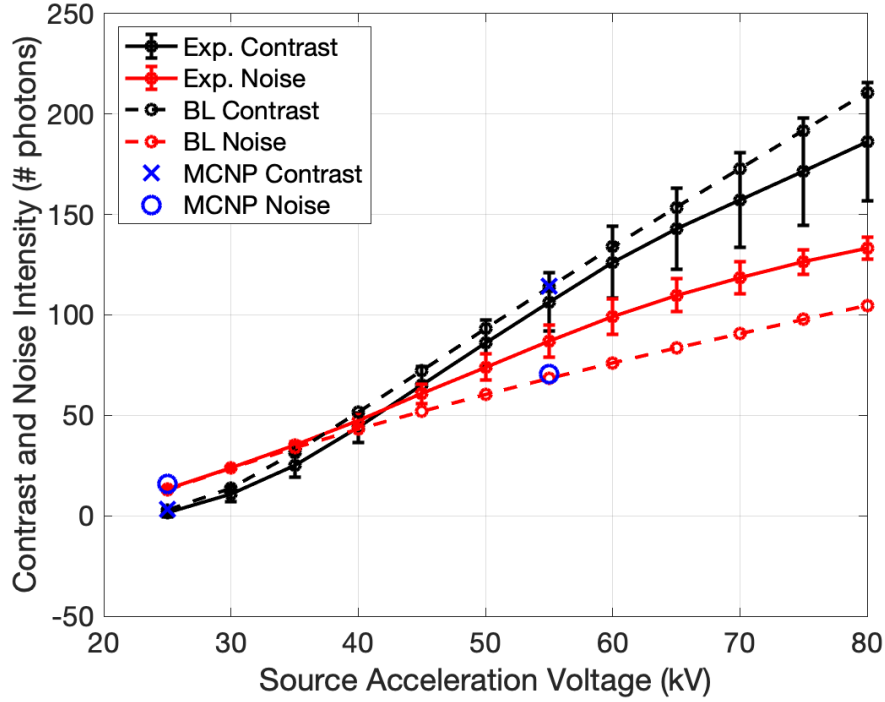


Figure 3.15: The image contrast and noise intensity are plotted as a function of the source voltage. BL underestimates the noise intensity, likely due to the simplistic scattering model.

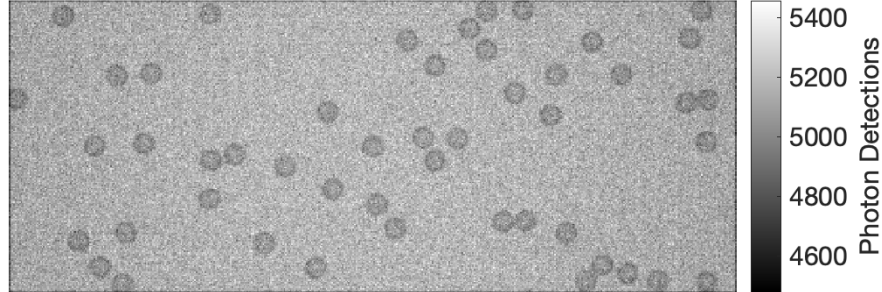


Figure 3.16: An MCNP-simulated image of multiple particles on a glass slide. The imaging settings are identical to the single-particle image.

the detector will have on image noise intensity.

To demonstrate that the effect of scatter from the particles to the detector is small, and that our simulation tools can simulate multiple-particle images, we provide MCNP and BL images of multiple particles on a glass slide, seen in figures 3.16 and 3.17. The particle images cover approximately 10% of the detector imaging area, a typical percentage for low density seeding applications that may be more likely to be used in early XPV experiments.

To calculate the SNR, we use a smaller background area compared to the single-particle

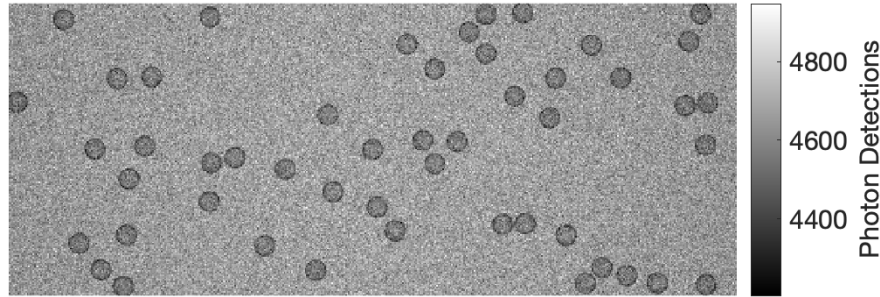


Figure 3.17: The BL-simulated image of multiple particles on a glass slide. The settings are identical to the single-particle simulations.

simulations in order to avoid including pixels with other particle images. Using identical background areas and particles, the background noise intensity in the multi-particle MCNP simulation increases approximately 4% compared to the single-particle MCNP simulations; the contrast decreases by roughly 1%. The SNR decreases by 5%. In the multi-particle BL simulation, the SNR changes by less than 1%. The background noise intensity is constant at 68 detections. The contrast increases from 106 to 107 detections from the single-particle simulation to the multi-particle simulation. As expected, the BL simulations are insensitive to multiple particles in the domain because it cannot simulate scatter.

In realistic imaging scenarios, scatter is dominated by the surrounding liquid, containers, and mounting equipment. Hence, the observed impact of multiple particles on noise intensity and contrast are likely exaggerated in the multi-particle simulations, where the tracer particles comprise a larger percentage of the geometry than they likely would in early in-lab XPV experiments. These results imply that a single-particle simulation is an adequate simplification for estimating SNR for practical design of experiments purposes. In particular, single particle simulations are adequate for comparing tracer particle designs.

3.5 Conclusions

BL and MCNP simulations are both able to simulate approximate X-ray images relatively accurately. BL simulations are substantially faster than MCNP simulations, and offer greater flexibility and opportunity for improvement in terms of both speed and accuracy. The 55 kV, 500 μ A, 100 ms MCNP simulation calculates all of the photon track data in 6.9 hrs. Then, we completed processing the MCNP output data in MATLAB in 10.5 hrs for a total MCNP computation time of 17.4 hrs. The 55 kV, 500 μ A, 100 ms non-optimized, MATLAB-based BL simulation ran in 0.56 hrs on the same workstation that the MCNP data was processed on. In spite of the higher computational cost, MCNP may still be helpful. For geometries where phenomena such as focal spot blurring, scattering, and X-ray fluorescence are more dominant effects, using MCNP simulations may still be prudent despite the higher computational cost.

While MCNP currently offers an advantage over BL simulations by modeling the physics more accurately, BL simulations in the future will be able to rapidly simulate complex image behavior with empirical modeling. For example, BL simulations could take focal blurring into account by adding Gaussian blur, improve noise intensity approximations by changing the noise probability distribution, and more accurately emulate photon detection with detector point spread functions in the spatial and energy domains. These are all computationally inexpensive alternatives to the computationally expensive MCNP simulations, and will further degrade its accuracy advantage over BL.

MCNP simulations may be difficult to improve upon besides using better source spectrum and flux approximations specific to the experiment. MCNP is also agnostic to time, so modelling photon counting dead time and pileup must be done after the MCNP simulation by assigning event times based on the statistics. Without such an addition, MCNP is incapable of predicting non-Poisson image noise. BL simulations, on the other hand, have ample room for improvement with regard to noise modeling, source spectrum and flux modeling, and quantitative blurring. Furthermore, the BL code for this study is far from optimized. An optimized and parallelized GPU code will only add to the computational expense advantage that BL has over the already-optimized MCNP code. Efforts to improve BL may be more fruitful than trying to use MCNP as an XPV design tool. In all but the most extreme cases it is hard to justify the need to use MCNP for the design of XPV experiments.

The techniques developed in this study can be used to harness improvements in X-ray imaging hardware and flow tracers designed for X-ray imaging. As laboratories adopt XPTV as a measurement tool, they can apply BL or MCNP simulation methods to quantify the expected performance of their source, tracer particle, and imager systems for a variety of flows.

Chapter Supplementary Material A: BL Simulation Validation

Depth and Attenuation Validation

The BL equation is evaluated along rays that extend from the source to the center of a sub-pixel. It is implicitly assumed that the photon flux across a sub-pixel is homogeneous.

The first validation test was conducted by calculating the true depth and attenuation ratio by hand. A hand calculation for an individual pixel was compared to the simulation results. Then, the monochromatic photon attenuation ratio and the polychromatic intensity spectrum were calculated by hand to compare to simulation. Both the depth and attenuation errors can be attributed to computational round-off.

An 86 mm $\times$ 86 mm pane of glass with a depth of 172 μ m is placed 1 cm from the point source such that the upper left corner aligns with the origin. The ray from the source to the pixel (270,122) passes through the front and back faces of the glass pane. The path length

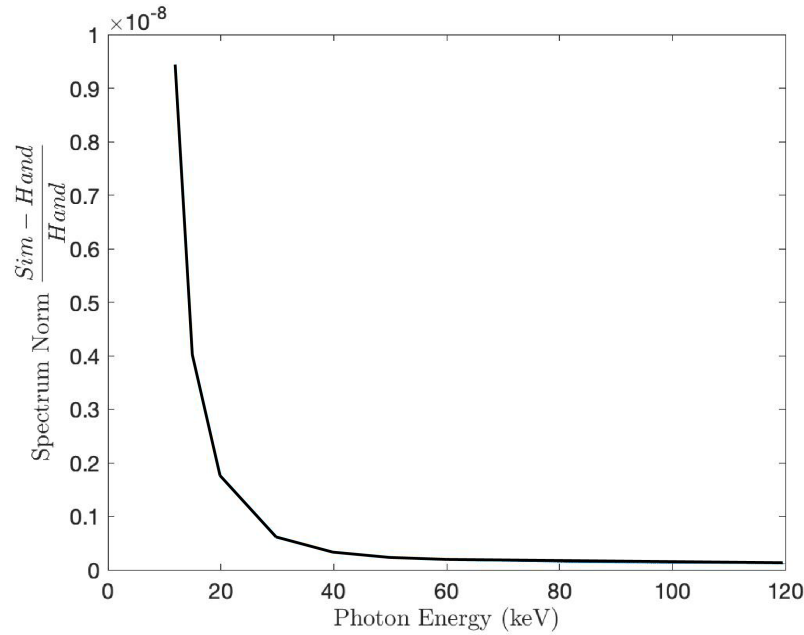


Figure 3.18: The error between the hand-calculated polychromatic spectrum and the MATLAB-simulated spectrum using our BL code is less than one millionth of a percent. (At low photon energies, the number of photons at that energy approaches zero, hence the singularity.)

of the ray passing through the glass can be calculated by

$$l = \frac{t}{\cos \theta \cos \phi} \quad (3.5)$$

where t is the glass pane thickness, θ is the angle between the ray and the axis pointed in the thickness direction, and ϕ is the angle between the ray and the direction perpendicular to the thickness direction. The difference between the hand-calculated depth and the simulation-calculated depth for the pixel (270,122) is -3.429×10^{-12} m.

For the same (270,122) pixel, the error between the hand-calculated (using path length calculated above) and simulated attenuation ratio $\frac{I}{I_0}$ is -6.711×10^{-11} . From figure 3.18 it is clear that the error between the polychromatic spectrum calculated by hand and with the simulation is order of round off error.

Sub-pixel Homogeneity

In order to validate the homogeneity assumption (i.e. that a sufficient number of sub-pixels was considered), an upper bound on the maximum “lost area” is established. Lost area is the area of a pixel that is erroneously attributed as covered (or not covered). For

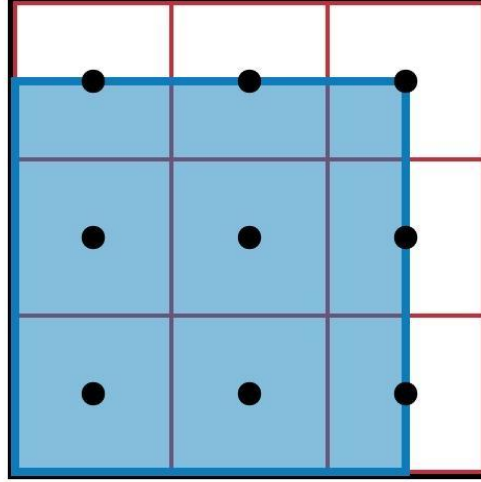


Figure 3.19: The scenario with the maximum lost area given a continuous, non-convex shape with dimensions greater than or equal to a pixel side-length. A sub-pixel is assumed to be uniformly covered or uncovered based on the state of its center.

example, in Figure 3.19, the lost area is the white region. The sub-pixels detect the blue material projection, but the projection does not physically cover the entire pixel. Assuming a continuous, non-convex, projected shape whose dimensions are larger than a pixel side length, the maximum lost area can be readily calculated. The scenario depicted in Figure 3.19 is the maximum possible lost area that meets the stated assumptions. The area lost as a fraction of the pixel area is given as a function of the number of sub-pixels, N , by

$$\frac{A_{lost}}{A_{pixel}} = \frac{1}{\sqrt{N}} - \frac{1}{4N}. \quad (3.6)$$

A pixel is sufficiently homogeneous when the maximum lost area is no more than 10% of the pixel area. For $N = 121$ sub pixels (11×11), the maximum lost area is less than 9%.

If the non-convex assumption is relaxed to allow for holes, the maximum lost area bound increases. Figure 3.20 shows the maximum possible lost area after relaxing the non-convex assumption. The lost area was calculated in MATLAB; the results are plotted in figure 3.21. It is clear that the criteria for the homogeneity assumption, discussed above, is again met for $N \geq 121$ (11×11).

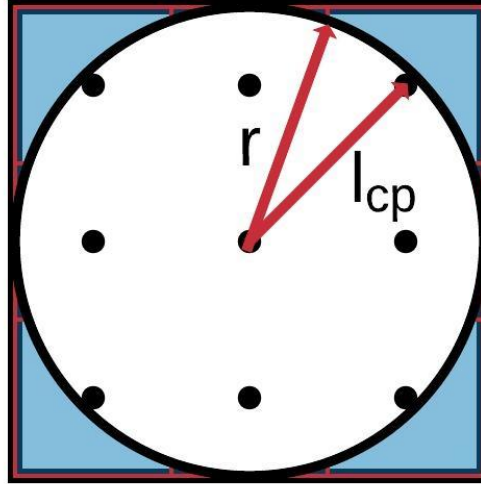


Figure 3.20: The maximum lost area scenario for a hole with a diameter greater than or equal to the pixel side length.

Chapter Supplementary Material B: MCNP PTRAC File Explanation

MCNP [41] with PTRAC files is a powerful simulation tool, but care must be taken when analyzing the data as the PTRAC file format is not intuitive. We will briefly make a few comments on this so that anyone interested on building on this study may better navigate some of the challenges.

The PTRAC file denotes different types of interactions – henceforth referred to as events – and the associated event data: location, direction, and post-event photon energy. The physically relevant event types are: surface, collision, and termination events (sur, col, and ter, respectively). Each event has an associated event ID, which is a number reported in the PTRAC file. A surface event flags a photon entering the CdTe detector panel; its event ID is 3000. With the filters discussed prior, a surface event flags a photon entering the detector panel. A collision event flags a photon imparting some energy to the detector; its event ID is 4000. Collision events can refer to either a scattering event or an absorption event. Termination events flag the end of a photon’s recorded track history; its event ID is 5000. When a photon is absorbed, identical collision and termination events are reported in the PTRAC file. In addition to the physical event types, there is a fourth event type: history termination events. Their event IDs are 9000. History termination events denote the end of recording a photon track. This may occur when a photon is absorbed or when it leaves the

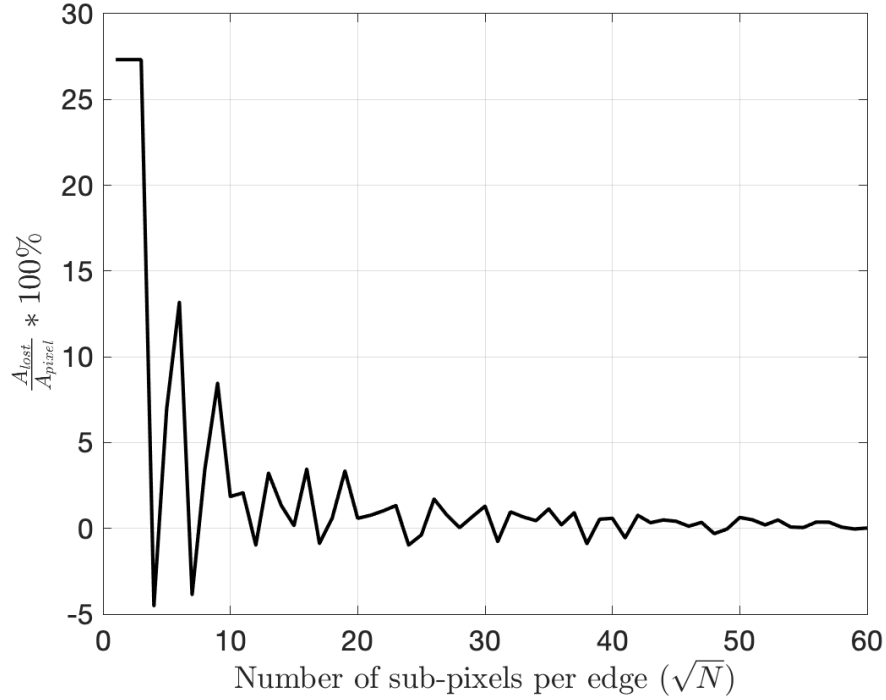


Figure 3.21: As the number of sub-pixels increases, the maximum area lost decreases rapidly. The homogeneity assumption criteria is met for $N \geq 121$ (11×11).

domain of interest (in this case, the detector panel). When a photon is absorbed, a history termination event identical to the collision and termination events is recorded.

Figure 3.22 shows a snippet of the PTRAC file from one of our trial MCNP simulations. In this example, the first particle track is for the fifth source particle. As with every particle track in this PTRAC file, the history begins with a surface event. The “5 3000” line at the very beginning states that the initial event for the track history of source particle five is a surface entry event. The data associated with this event can be seen on the third line. The first three numbers on line three are the x-, y-, and z-coordinates in centimeters; the next three numbers are the direction of travel component vectors after the event occurs. The last three numbers are photon energy in MeV, particle weighting, and track time in shakes.

In the case of this first particle track, the photon enters the detector at (50, -0.40716, 1.4224) centimeters with 0.31738 MeV of energy. The photon then collides (event ID 4000 on line two) at (50.002, -0.40718, 1.4245) cm and departs with 0.23153 MeV of energy. Note that the particle direction of travel is reversed in the x-direction due to this collision; the particle leaves the detector (event ID 9000 on line 4) before being absorbed, i.e., terminated (event ID 5000). In this track history, the photon deposits 0.08585 MeV of energy in the detector.

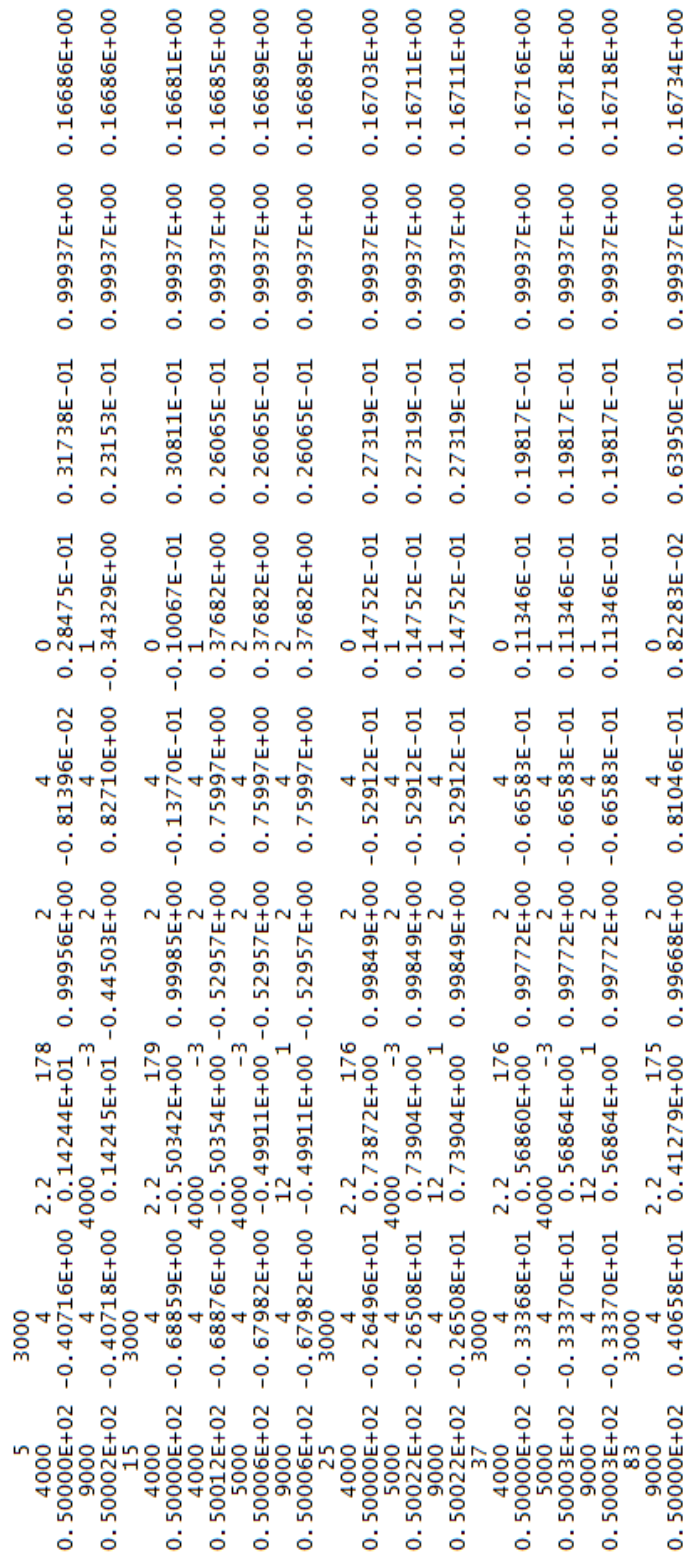


Figure 3.22: Example particle history tracks from an MCNP PTRAC file.

Chapter 4

Developing General-Use XPV Tracer Particles

4.1 Background

As discussed in the previous chapter, tracer particles do not produce high image contrast. They weakly attenuate X-rays by virtue of being small. Tracer particles designed specifically for XPV are necessary to widen the range of flows that can be studied. Higher contrast tracer particles are easier to locate and therefore reduce the exposure time necessary to achieve a usable image SNR. Shorter exposure times are necessary to study faster flows.

In order to achieve decent particle image contrast, most previous in-lab XPV experiments have relied on particles that are either near order of magnitude density mismatched with the fluid, are on the order of a millimeter in diameter, or both [33], [44], [45]. However, most particles that are not neutrally buoyant bias the velocity measurements and become unevenly distributed. Large particles, even if they are nominally density matched, limit the spatial resolution and may not trace the flow, particularly in large velocity gradients or if flow features are on the order of magnitude of the particle size. Recently, Mäkiharju et al. [46] investigated a creeping flow with $60\text{ }\mu\text{m}$ tracer particles. In that study, it was noted that significant limitations were imposed by the slow scan times that were necessary to achieve particle image contrast. As part of that study, improving the tracer particles was identified to be of paramount importance to laboratory XPV.

XPV imposes unique requirements on tracer particles that conflict with one another. XPV tracer particles must attenuate X-rays significantly more (or less) than the ambient fluid to generate contrast, which favors large, density-mismatched particles. At the same time, tracer particles must be neutrally buoyant and have sufficiently low inertia to trace the flow accurately, which favors small, density-matched particles. In addition, small tracer particles maximize the spatial resolution of the measurement. Thanks to modern manufacturing processes it is now feasible to develop $O(10\text{ }\mu\text{m})$ prototype custom tracer particles for XPV applications.

In this chapter we introduce new hollow carbon (C) tungsten-coated (W) microsphere XPV tracer particles (CW) that were designed using the simulation tools in [23], which were discussed in the previous chapter. We compare the performance of the CW tracer particles to the performance of silver-coated hollow glass tracer particles (AGSF-33), a previously available visible light PTV tracer particle. Tracer particle performance is evaluated by measuring Poiseuille pipe flow with 2D-projected XPTV. The methods used in this study provide a useful template for comparing the performance of new tracer particles.

Additionally, images of the tracer particles are captured with a variable single-energy-threshold photon counting detector (PCD), demonstrating that PCDs can be used for XPTV in the laboratory. PCDs open up the possibility of future experiments tracing multiple particle species, scalar fields, or both with K-edge material detection. We will touch on the benefits and challenges of PCDs in X-ray flow visualization.

4.2 Experimental Setup

The experimental setup can be seen in figure 4.1. The source to detector distance (SDD) and source to object distance (SOD) are 500 mm and 38 mm, respectively. The SOD is measured from the X-ray emitting tungsten target surface to the center of the pipe. The source emits a conical beam, which geometrically magnifies objects in the field-of-view (FOV) by a factor of approximately $M = 13.1$. The detector has a panel size of 83.8 mm $\times$ 33.5 mm. Due to geometric magnification, the FOV in the object plane is 6.37 mm $\times$ 2.55 mm at the central plane of the pipe.

The available quantity of the new custom particles made a closed loop impractical. Tracer particles are seeded directly into the 6.35 mm (0.25 in.) inner diameter pipe by pouring 100 mg of AGSF-33 particles or 150 mg of CW particles mixed with glycerol into the top of the pipe at an angle, leaving room for the air in the pipe to exit. Assuming a uniform particle distribution in the mixture, this corresponds to roughly 6,200 AGSF-33 particles or 8,200 CW particles in the FOV. However, some particles may not be observable due to broken coatings or particle overlap. In particular, we found that the CW particles required a larger mass of particles since the number density at 100 mg was lower than the AGSF-33 particles. CW particles with broken coatings will contribute to the mass, but will not show up in an X-ray image because they have low attenuation. The coatings themselves will sink out of the viewing section. Lastly, uneven particle distribution in the pipe and sinking or rising out of the FOV both reduce the number of observed particles, especially for the prototype CW particles. The LaVision PTV algorithm we use detects 100-200 particles in the FOV. The number of observed particles is larger, though, because the algorithm does not detect every particle.

The particle-glycerol mixture is at equilibrium at 23.7 °C as measured by an Ambient Weather WH31E temperature and humidity sensor adjacent to the X-ray source. A Harvard Apparatus 11 Plus syringe pump pushes the glycerol at a constant flow rate $Q = 0.215$ mL/min. The 11 Plus syringe pump has a 0.9° step rotation for high accu-

racy flow rates. The Reynolds number of the flow, $Re = Ud/\nu = 0.0018$, is of comparable order of magnitude as Lee and Kim’s experiment at a synchrotron, which, based on data they provide, is calculated to be $Re_{LK} = 0.0052$. Here, U is the centerline velocity and d is the pipe inner diameter. The laminar development length for the current experiment is $l = 0.06ReD = 0.69 \mu\text{m}$. Given that the pipe length upstream of the measurement FOV is significantly longer than the entrance length, the flow can be considered fully developed.

The pipe is mounted on a 3-axis linear motion system comprised of three Thor Labs NRT150 stages. The NRT150 stages have an on-axis accuracy of $2 \mu\text{m}$ and a minimum incremental motion of $0.1 \mu\text{m}$. A Starrett No.98 machinist level with a sensitivity of 0.024° is used to ensure that the pipe is mounted vertically.

The Dectris Pilatus3 PCD and YXLON FXE225.99 X-ray source are the same as were used in our prior study [23], where a fuller description of the imaging system is available. We note here, though, that given the $6 \mu\text{m}$ source focal spot size, the SDD, and the SOD, the penumbral blurring would span 0.42 pixels. Thus, in this configuration, penumbral blurring does not impact our measurements. Under different circumstances, however, the penumbral blurring can be significant and should be evaluated for any XPTV configuration. The detector has a 1 mm thick CdTe panel to capture photons in pixels of size $172 \times 172 \mu\text{m}$. It is important to note here that for this pixel size and our configuration’s $13.1\times$ geometric magnification, a $50 \mu\text{m}$ particle would appear to be $(50 \mu\text{m} \times 13.1/172 \mu\text{m} =)$ 3-4 pixels across. Sixteen tiles comprise the detector panel, leading to a 487×195 pixel detection area. The Dectris Pilatus3 can achieve frame rates up to 500 Hz. Due to the relatively dim X-ray source use, we chose a low flow speed such that images captured with 15 ms exposure times would not exhibit motion blur, but would have sufficient photon counts to achieve a usable SNR. Frame rates up to about 70 Hz could be used based on this exposure time. For PTV, however, it is preferable to have a notable particle shift between each frame, so we captured images at 2.5 Hz to allow sufficient particle motion between frames. The detector energy threshold is set to 20 keV to reduce photon scattering noise. The X-ray source is operated at an acceleration voltage of 55 kV with a target current of $500 \mu\text{A}$.

Image Processing Methods

Filtering and PIV Software

Raw particle images are corrected with a flat field correction map, inverted, and then imported to LaVision DaVis 8.4 for filtering and PTV processing. We use standard DaVis algorithms without modification to demonstrate that XPTV data can be processed with existing algorithms. Details on the filters and particle tracking settings can be found in the chapter supplementary material.

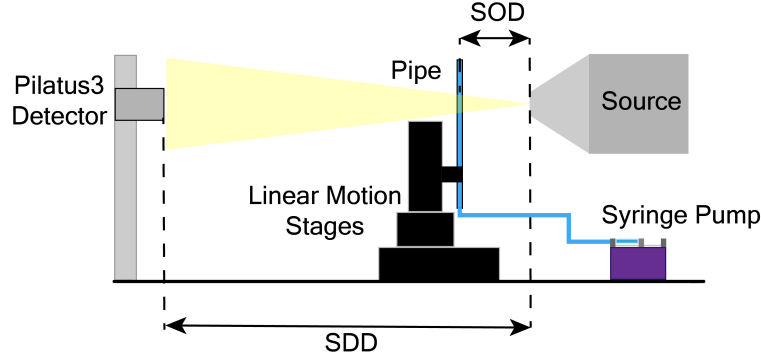


Figure 4.1: The experimental setup (not to scale). A syringe pump pushes glycerol and tracer particles through a vertically mounted pipe.

Scaling and Uncertainty

Image pixel-to-mm scaling is done by identifying the pipe inner diameter in the image. The pipe inner diameter is known to be 6.35 mm, and we count the number of pixels required to traverse the inner diameter of the pipe. The time step Δt is the time between the beginning of each exposure. For all of the images collected in this study, $\Delta t = 400$ ms to ensure that the particles travel sufficiently far between exposures for particle displacement to be significantly larger than the uncertainty in localizing the particle centroid. Here, localizing refers to determining the location of a particle centroid. Given the 15 ms exposure time, the acquisition duty cycle for these experiments is 3.75%.

As noted in section 4.1 and [46], the density mismatch between fluid and particles can be a significant source of uncertainty for XPTV and XPV. Laboratory systems with dim X-ray sources are currently limited to slow flows where the density mismatch has a more pronounced effect. Discussion of the effects of buoyancy and flow tracing accuracy is reserved for section 4.3.

There are additional uncertainty contributions unique to XPV and XPTV, though. Most laboratory X-ray sources have conical beams, leading to geometric magnification, as mentioned previously. The geometric magnification factor M is a function of the distance from the source, so it changes along the beam direction. Accordingly, the FOV will change over the depth of the experiment geometry. In our experiment, the FOV changes from approximately $5.84 \text{ mm} \times 2.33 \text{ mm}$ at the front of the pipe to $6.90 \text{ mm} \times 2.76 \text{ mm}$ at the back. In all planes along the beam direction, the entire pipe is in the FOV. Similarly, the motion of a particle image is a function of the magnification. Images of particles closer to the source will appear larger and will move faster than images of particles farther from the source, even when the particles are the same size and have same exact velocity.

Consider, for example, two identical particles A and B in a Poiseuille pipe flow, each at the same radius, but particle B is closer to the source than particle A, as shown in figure 4.2. Being at the same radius, both particles will travel at the same velocity. Since particle A is

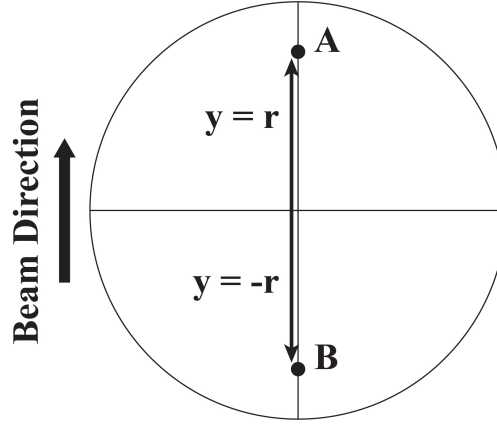


Figure 4.2: Particle A will be magnified less than particle B because it is farther from the X-ray source. Particle A will also appear to move slower than particle B.

farther from the X-ray source, it will be magnified less than particle B, and will appear to move less between frames than particle B even though it has traveled the same real distance. Given that there is no information on particle depth in a 2D-projected measurement, the pixel-to-mm scaling assumes that all particles are located in the central plane at $y = 0$. Equation 4.1 gives the measured displacement, after calibration, Δz of a particle that has moved a real distance ΔZ , accounting only for the effects of magnification. Recall that SOD is measured to the center of the pipe.

$$\Delta z = \frac{M(y)}{M(y=0)} \Delta Z = \frac{SOD \cdot \Delta Z}{SOD + y} \quad (4.1)$$

Considering only the effects of magnification, the relative error, ϵ , in the measured displacement versus the real displacement will increase with particle distance y from the central plane and is given by

$$\epsilon = \frac{|\Delta z - \Delta Z|}{\Delta Z} = 1 - \left(1 + \frac{|y|}{SOD}\right)^{-1}. \quad (4.2)$$

The velocity relative error is the same as equation 5.3. Note that the relative error depends on SOD and domain size, but not directly on SDD. We denote the ratio of the experiment depth $\delta = 2y_{max}$ to the SOD as the magnification aspect ratio. A good rule of thumb to avoid magnification error is to keep the magnification aspect ratio below 0.2. The maximum relative magnification error would then be 10%. In practice, SDD could affect the error by modifying how many pixels a particle image occupies on detector, contributing to the resolution error of localizing a particle centroid.

For this experiment, where the SOD is 38 mm and the maximum $y = 3.2$ mm, the relative magnification error never exceeds 9% of the flow speed. The relative magnification error is at its maximum at the pipe wall, where the velocity approaches zero. At the center of the pipe,

where the calibration is defined, the relative magnification error is zero. Experimenters with large depths of field should be cautious about the error introduced by magnification effects. Using fully tomographic XPTV, such as in [46], avoids issues with geometric magnification altogether. Alternatively, if the tracer particles are sufficiently monodisperse, one could determine the depth location based on the particle image size, correct for the magnification error, or both. We discuss this in greater detail in section 4.4.

Depth-Averaged Velocity Profile

In this study, 2D-projected images of flow in a pipe are captured. Particles all along the X-ray beam direction (depth-wise direction y in figure 4.2) of the pipe are imaged at the same x -location. As a result, for particle tracking velocimetry, one is not sampling a single value but rather the distribution of velocities along the depth-wise direction of the pipe. Lee and Kim [24] compared the measured average velocity of the depth-wise distribution to the analytical depth-averaged velocity profile (DAVP). Figure 4.3 depicts the depth-wise velocity variation in the pipe. Lee and Kim [24] assumed a uniform distribution of ideal tracer particles throughout the pipe, and that all particles contribute to the distribution equally. Under these conditions, the DAVP for a parallel-beam projection is simply the depth integrated average velocity of the fluid, given by

$$\langle u(x, y) \rangle_y = \frac{4Q}{3\pi R^2} \left[1 - \left(\frac{x}{R} \right)^2 \right] = \frac{2}{3} u(r = x) \quad (4.3)$$

where $\langle u(x, y) \rangle_y$ is the DAVP, $u(r)$ is the analytical solution for Poiseuille pipe flow, R is the pipe radius, and Q is the volumetric flow rate. Hidden in equation 5.5 is a cosine of the angle between the radius and the x -direction on the detector. Since this cosine is unity for this axisymmetric system with vertical alignment, it is omitted from equation 5.5 for clarity. For consistency throughout the paper, the DAVP will be written as a function of x , while the cross-sectional velocity profile will be written as a function of radius r . The varying magnification due to a cone-beam instead of a parallel beam is also not considered here since the magnification error is relatively small. In section 4.2 the magnification error is calculated to be more than an order of magnitude smaller than the flow speed.

It is worth noting that Lee and Kim compared their PIV measurement to the DAVP, whereas we are comparing a PTV measurement to the DAVP. Fouras et al. [26] points out that PIV calculates a modal average velocity, not the mean average that Lee and Kim compared against. By comparing a PTV DAVP measurement to the analytical DAVP, we omit any error associated with the discrepancy between the distribution mode and mean.

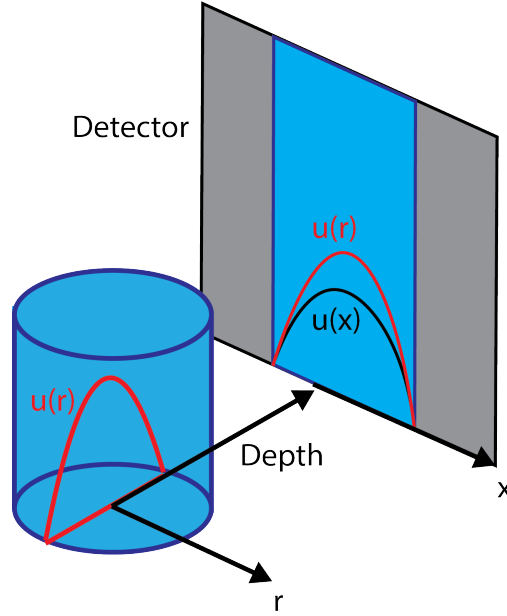


Figure 4.3: The depth-wise direction is towards the detector. The analytical velocity profile is depth-averaged.

4.3 Tracer Particle Properties

Silver-Coated Hollow Glass Spheres (AGSF-33)

Silver-coated hollow glass spheres are commonly used in visible wavelength PTV applications, so they are readily available for use in XPTV. The silver coating attenuates X-rays better than uncoated plastic or glass microspheres owing to the relatively high mass attenuation of silver. Higher mass attenuation elements such as gold, silver, and tungsten achieve better image contrast provided that the surrounding materials are comprised of lower mass attenuation elements such as hydrogen, oxygen, and carbon. The opposite is true for dense, heavy element fluids like liquid metals. In these experiments, we use AGSF-33 silver-coated hollow glass spheres, a previously available off-the-shelf tracer particle. AGSF-33 particles were previously shown to be suitable XPTV tracer particles [23], [46]. 100 mg of AGSF-33 tracer particles are added to 20 mL of glycerol for the experiments. Assuming uniformly 50 μm AGSF-33 tracers and an average density of 1 g/cm^3 , the concentration of AGSF-33 tracers is roughly 76,000 particles/mL.

For these experiments, the AGSF-33 particles are sieved to between 45 and 53 μm to improve monodispersity. We assume a uniform distribution across this size range. Although the AGSF-33 particles are on average matched with the density of water, they, like most tracer particles, have a non-uniform density within a batch. According to the manufacturer, Potters Beads, the particles have density between 0.9 and 1.1 g/cm^3 . Density mismatches

with the fluid induce relative motion due to buoyancy, which can bias velocity measurements, particularly in low speed flows if the velocity due to buoyancy is the same order of magnitude as the flow itself. Although buoyancy-induced velocity is often neglected in visible light PTV, early XPTV experiments will study low speed flows where buoyancy-induced velocity should be considered. When the relative velocity is low enough that $Re(u_{rel}) \ll 1$, and Stokes drag is appropriate, the terminal speed with which particles will rise or settle in a fluid can be calculated from equation 4.4.

$$u_{St} = d_p^2 \frac{\rho_{fluid} - \rho_{particle}}{18\mu} g \quad (4.4)$$

Here, d_p is the particle diameter, ρ_i is density of i , μ is the dynamic viscosity of the fluid, and g is the acceleration due to gravity. The Stokes settling speed can be thought of as a bias error in the velocity measurement. The Stokes settling speed is proportional to the particle diameter squared, which incentivizes the use of smaller particles. Smaller particles also have a lower Stokes number, which is discussed in section 4.3. The trade off of using a smaller particle, however, is less X-ray attenuation, which, according to the Beer-Lambert law, is exponentially dependent on the particle size. These opposing design criteria are an example of the challenges and opportunities of the XPTV tracer design space. Exploring smaller tracer particle designs that incorporate composite- and single-element hollow shells of high mass attenuation elements while remaining neutrally buoyant is crucial for improving XPTV.

For a coated composite particle such as AGSF-33 the particle density $\rho_{particle}$ is given based on the coating mass fraction γ as

$$\rho_{particle} = \frac{\rho_{pyrex}(r_{pyrex}^3 - r_{air}^3) + \rho_{air}r_{air}^3}{(1 - \gamma)r_{silver}^3} \quad (4.5)$$

where r_i is the outer radius of material i . To obtain an approximation of the effect of a particle-fluid density mismatch, we run Monte Carlo simulations of the Stokes settling speed. We follow the guidelines set out in the ISO Guide on the propagation of distributions via the Monte Carlo method [47]. For simplicity, we assume that the diameter and density are independent random variables. We take the density to be normally distributed with a 0.033 g/cm³ standard deviation, truncated at 0.9 g/cm³ and 1.1 g/cm³. We assume that Potters Beads' quality control has removed particles outside of this range. We use a truncated distribution to best reflect the manufacturer specifications. However, with an untruncated distribution, the average settling speed and standard deviation in glycerol do not change. The average settling speed in water does not change either; the standard deviation in water increases by 1 μ m/s. These calculations show that truncating the distribution is not a strong assumption. Randomly taking values from these probability distributions, we simulate equation 4.4 10^7 times for particles in water and glycerol each at 23.7 °C. The resulting Stokes velocity distribution is shown in figure 4.4. In water, the distribution mean is $\leq O(\pm 0.01 \mu\text{m/s})$; the standard deviation 47 $\mu\text{m/s}$. Using pure glycerol increases the

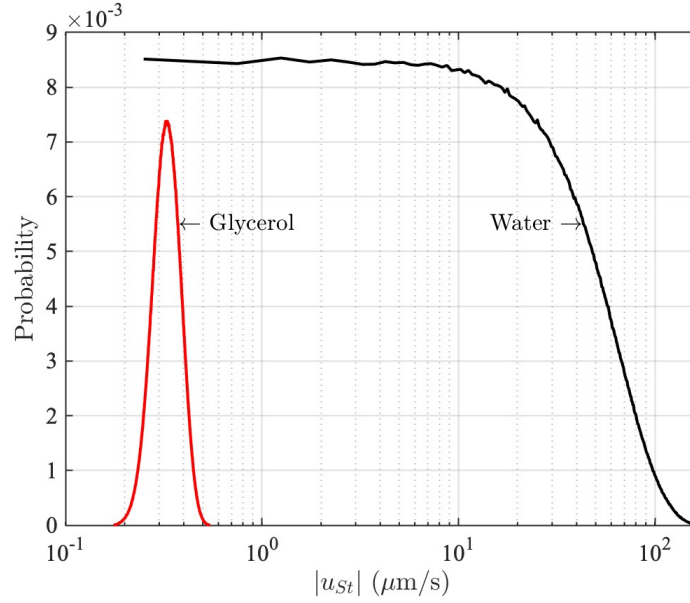


Figure 4.4: The probability distribution of the terminal speed magnitude for sieved, uniformly distributed, 45-53 μm AGSF-33 tracer particles in the Stokes drag regime in water and pure glycerol at 23.7°C. The distribution in water is effectively symmetric about zero; only the velocity magnitude is shown here to accommodate a log-axis plot to highlight the water-glycerol distribution contrast.

fluid viscosity by an order of magnitude, dramatically reducing the particle Stokes velocity spread, as seen in figure 4.4. In glycerol at 23.7°C, the AGSF-33 particles rise at an average speed of 0.34 $\mu\text{m/s}$, with a standard deviation of 0.05 $\mu\text{m/s}$. It is worth noting that a tighter density distribution would be possible using sorting techniques such as floating and sinking separation. However, for simplicity, the authors used the particles as-is to best emulate an off-the-shelf product usage.

Tungsten-Coated Hollow Carbon Spheres (CW)

Compared to the AGSF-33 particles, the new CW particles conceived and initially designed in the FLOW Lab and manufactured by Ultramet offer higher X-ray image contrast. A tungsten coating has a higher mass attenuation coefficient than silver, thereby generating greater contrast. In order to maintain neutral buoyancy and maximize the coating thickness, the tungsten is coated onto hollow carbon microspheres. Simulations using the Beer-Lambert method in [23] show that the CW particles can achieve a $3.4\times$ improvement in the SNR compared to the AGSF-33 particles on a glass slide. The SNR improvement implies that CW particles will exhibit higher contrast in a fluid as well. The diameter distribution of the tungsten-coated hollow carbon spheres as-manufactured can be seen in figure 4.5. The

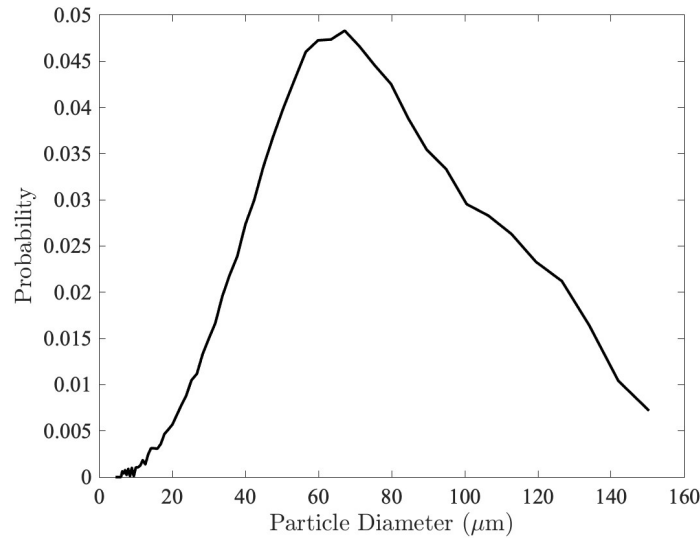


Figure 4.5: The CW particle diameter distribution – measured using a non-aqueous based dispersion (ISO 13320) with a Saturn DigiSizer – provided by Particle Testing Authority. While the particles are polydisperse from the factory, sieving and future manufacturing improvements can reduce the polydispersity.

polydispersity shown in figure 4.5 is apparent in the scanning electron microscope (SEM) image shown in figure 4.6. Figure 4.7 shows a nonspherical particle with a broken coating, demonstrating the prototype nature of these early CW particles. We take advantage of the broken coating to measure the coating thickness. Sieving reduces the polydispersity, but in general monodispersity in all properties will improve if there is wide demand and manufacturers can fine-tune their scalable manufacturing processes for making these new types of particles. In this study, the particles are sieved to the same diameter range as the AGSF-33 particles to ensure a fair contrast comparison. 150 mg of CW tracer particles are added to 20 mL of glycerol for the experiments. Assuming uniformly $50\text{ }\mu\text{m}$ diameter particles with average particle density of 1.13 g/cm^3 , the concentration of CW tracers is roughly 100,000 particles/mL. Given the polydispersity that the CW particles exhibit and that some of these prototype particles had broken coatings, their concentration cannot be accurately estimated based on the mass of particles added. Broken particle coatings contribute to the added mass, but deposit themselves below the viewing section and are not visible. Additionally, the particle that the coating belonged to may no longer produce sufficient contrast to be seen in an X-ray image. Owing to these difficulties of directly controlling concentration, we controlled the mass of particles added for both the CW and AGSF-33 tracer particles such that we achieve a similar observed number density.

We run the same settling speed Monte Carlo simulations for the CW particles as for the AGSF-33 particles in section 4.3. Based on the observed settling behavior and SEM images, the tungsten coating thickness is approximately $0.25\text{ }\mu\text{m}$. However, in SEM images, some

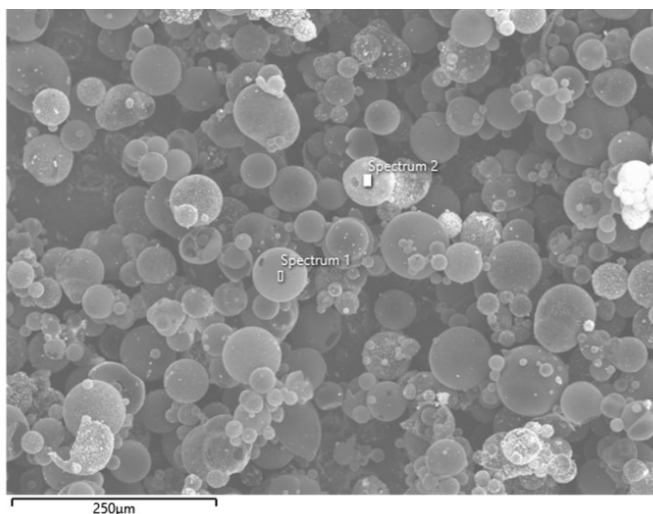


Figure 4.6: An SEM image of the CW tracer particles used in these experiments prior to sieving. Note the significant polydispersity prior to sieving.

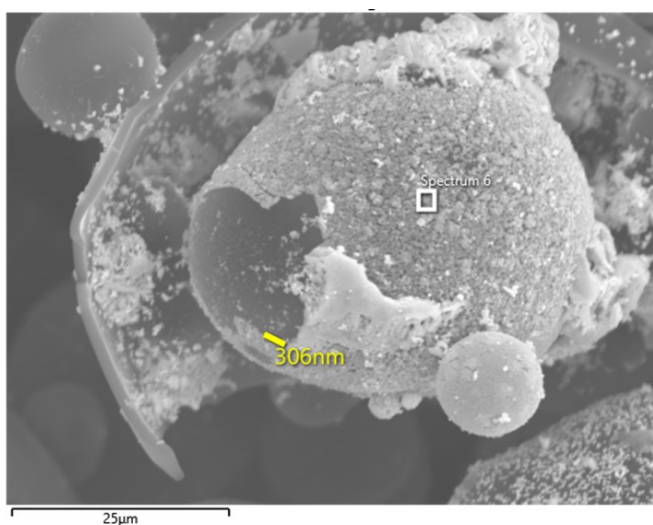


Figure 4.7: In this SEM image of a broken particle the coating thickness can be measured. However, we find that the coating thickness varies significantly due to the new manufacturing process not yet being fully refined. The particle shown is also non-spherical, another challenge in manufacturing new tracer particles.

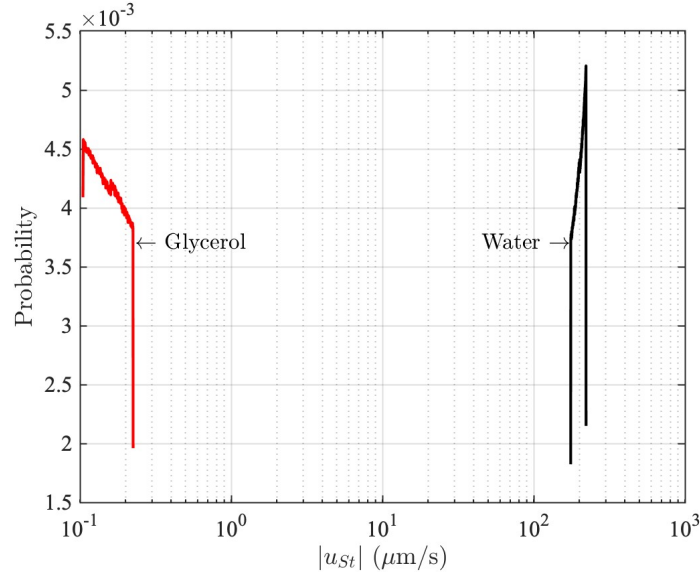


Figure 4.8: The Stokes terminal speed probability distribution of the CW tracer particles in water and pure glycerol.

particles are observed to have a $0.5 \mu\text{m}$ thick coating or more, demonstrating the manufacturing variability of the CW particles in these first batches. The average hollow carbon microsphere density is $\langle \rho_{\text{carbon}} \rangle = 0.5767 \text{ g/cm}^3$. Using these data alongside the diameter distribution in figure 4.5, we can estimate the CW particles' Stokes velocity distribution shown in figure 4.8. In water at 23.7°C , the CW particles rise an average speed of $200 \mu\text{m/s}$ with a standard deviation of $13.3 \mu\text{m/s}$. While the mean CW particle settling speed in water is much greater than the AGSF-33 particles, in glycerol at 23.7°C , the average Stokes terminal speed of the CW particles is $0.16 \mu\text{m/s}$, with a standard deviation of $0.035 \mu\text{m/s}$ – a much tighter and slower distribution than in water. While these first CW particles are outperformed by AGSF-33 particles in traditional tracer particle metrics like neutral buoyancy in water and polydispersity, as more monodisperse carbon microspheres are sourced and the tungsten coating thickness is better controlled, the traditional tracer particle metrics of CW will become competitive with AGSF-33 in water as well.

CW Particle Manufacturing

The CW particles are manufactured by Ultramet using a chemical vapor deposition (CVD) process to coat carbon microspheres with tungsten. The carbon spheres are hollow, averaging a diameter of $61.74 \pm 0.03 \mu\text{m}$ and a density of $0.5767 \pm 0.0009 \text{ g/cm}^3$. Diameter and density are measured using a non-aqueous based dispersion (ISO 13320) using a Saturn DigiSizer and nitrogen pycnometry (ISO 12154), respectively, provided by Particle Testing Authority. The tungsten coating is applied to the microspheres with a powder bed CVD reactor. The

CVD process uses a tungsten gaseous precursor that reacts with and deposits onto the carbon sphere surface. The fluidized bed is used to fluidize the particles so that the tungsten precursor can permeate the powder and react evenly with the particles. This process is done under a vacuum so any byproducts can be removed and neutralized. The coating thickness is varied by modification of the CVD process conditions. The coated spheres are sorted based on their diameter using various wire mesh sieves. These groups, where d_p represents a particles diameter, are: $d_p \in (0, 38]$, $(38, 45]$, $(45, 53]$, $(53, 63]$, and $(63, 250]$ μm . As mentioned in previous sections, CW particles with $d_p \in (45, 53]$ μm are used in this study to provide the best comparison to the equivalently sized AGSF-33 particles.

Tracer Particle Contribution to Uncertainty

A summary of the settling speed Monte Carlo simulations for CW and AGSF-33 particles can be seen in table 4.1. Ideally, the Stokes terminal velocity is at least an order of magnitude less than the characteristic velocity of the flow, i.e., $u_{St} \ll U$. When $u_{St} \ll U$, the buoyancy induced velocity can be neglected. Table 4.1 shows that this condition is satisfied for both particles in glycerol.

Another source of error is the particle's dynamic response to the flow. The particle response time – the Stokes time – is an indicator of the tracer particle's ability to follow a flow accurately. Assuming the relative velocity between the spherical particle and the surrounding fluid is such that the $Re(u_{rel}) \ll 1$, and the fluid acceleration is constant, the characteristic response time of the particle is given by equation 4.6 [3].

$$\tau_p = d_p^2 \frac{\rho_p}{18\mu}. \quad (4.6)$$

The Stokes number, shown in equation 4.7, is the ratio of the Stokes time to the characteristic time of the flow, τ_f . For pipe flow, $\tau_f = D/U$, where D is the pipe diameter.

$$St \equiv \frac{\tau_p}{\tau_f} = \frac{\rho_p d_p^2}{\rho_f D^2} \frac{Re}{18} \quad (4.7)$$

A particle is said to follow the flow accurately when $St \ll 1$. Equation 4.7 shows that smaller diameter particles are needed to measure higher Reynolds number flows, again motivating the need to explore new tracer particle designs to expand the applicability of XPTV to more flows of interest. Table 4.1 presents the Stokes number of AGSF-33 and CW particles in both pure glycerol and water at 23.7°C. Both particles have sufficiently small Stokes numbers for the pipe flow investigated in this study.

	AGSF-33	CW
u_{St}/U (Glycerol)	0.0013	0.00036
u_{St}/U (Water)	0.017	382
$St \times 10^9$ (Glycerol)	5.2	6.1
$St \times 10^9$ (Water)	6.6	7.7

Table 4.1: The Stokes number and terminal velocity ratio for the AGSF-33 and CW tracer particles for flow $Re = 0.0018$. Note that all dimensionless numbers are much less than unity in glycerol, so the particles can be expected to accurately trace the flow. AGSF-33 particles would also work as flow tracers in water at these speeds, but the prototype CW particles considered here would need to be used in flows faster than $2000 \mu m/s$ to neglect buoyancy effects in water.

4.4 Results and Discussion

Tracer Particle Measurement Comparison

The benefits of the CW tracer particles stem from their marked contrast improvement. Figure 4.9 demonstrates the higher contrast of the CW tracer particles compared to the AGSF-33. Figure 4.10 shows the SNR improvement of the CW tracer particles over the AGSF-33, with SNR approximated as

$$SNR \approx \frac{|\langle I \rangle_{r \leq r_p} - \langle I \rangle_{r_p < r \leq 3r_p}|}{\sqrt{\langle I \rangle_{r_p < r \leq 3r_p}}} \quad (4.8)$$

where I is the pixel intensity, r is defined as the radius from the particle centroid, r_p is the particle radius, and $\langle \cdot \rangle$ indicates mean. The average CW particle SNR is 47, while the average AGSF-33 particle SNR is 25, for a CW to AGSF-33 SNR ratio of 1.8. Equation 4.8 implies that if other particles are within a particle diameter of the particle edge (the definition we use for local), then the SNR calculation could be affected. Figure 4.10 shows the general behavior of the AGSF-33 and CW tracer particle SNR, but should not be taken as a high accuracy measurement of individual particles' SNR due to the effects of dense particle seeding and 2D-projection imaging. A higher SNR improves particle localization, which yields higher precision measurements. Figure 4.9 also shows the AGSF-33 particles clustering, which we did not observe for the CW particles. Since the AGSF-33 particles cluster, the actual SNR improvement by CW may in fact be higher than we observed.

Figure 4.11 shows the DAVP measured with the AGSF-33 and CW tracer particles. We use the PTV algorithms in LaVision DaVis 8.4, and the details of the processing steps and settings can be found in the chapter supplementary material. As mentioned previously, the glycerol-particle mixture is poured into the pipe at an angle, leaving room for air to escape the pipe. However, this resulted in particles being pushed to one side by shear

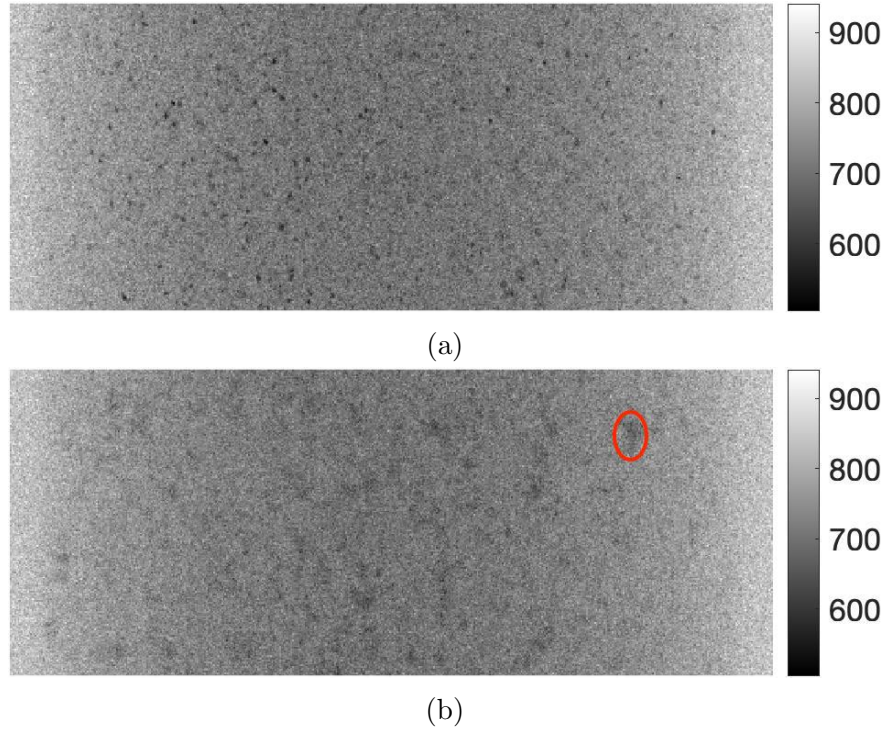


Figure 4.9: a) CW tracer particles and b) AGSF tracer particles imaged with a 15 ms exposure time, 500 mm SDD, 38 mm SOD, a 55 kV acceleration voltage, and a 500 μA target current. The CW tracer particles demonstrate higher contrast than the AGSF-33 tracer particles for the same exposure time. A cluster of AGSF-33 particles is circled as an example. Un-clustered AGSF-33 particles should nominally be the same size as the CW particles.

lift. Chipped or broken particles, which are smaller, are more susceptible to these shear lift forces and are preferentially pushed to one side. These broken particles are also more buoyant than the Monte Carlo simulations in section 4.3 suggest. Indeed, an artifact in the velocity profile at random locations could sometimes be observed. We did not observe the same seeding distribution behavior with the AGSF-33 particles, as they are mass produced via a more refined process and broken particles have been more effectively removed. For a clear quantitative comparison less sensitive to particle distribution bias we take the average and standard deviation of both halves of the pipe combined to calculate a projection of the radial velocity profile. Both the CW and AGSF-33 particles recover the DAVP, as figure 4.11 shows. Close to the wall, the data do not go to zero because particles have a finite diameter and do not stick to the wall.

Figure 4.11 also highlights how measurement variance is exacerbated by 2D-projected XPTV. Tracer particles at the front and back walls of the pipe appear at the same x -location as particles at the center, so there is an unavoidably wide spread in the measured

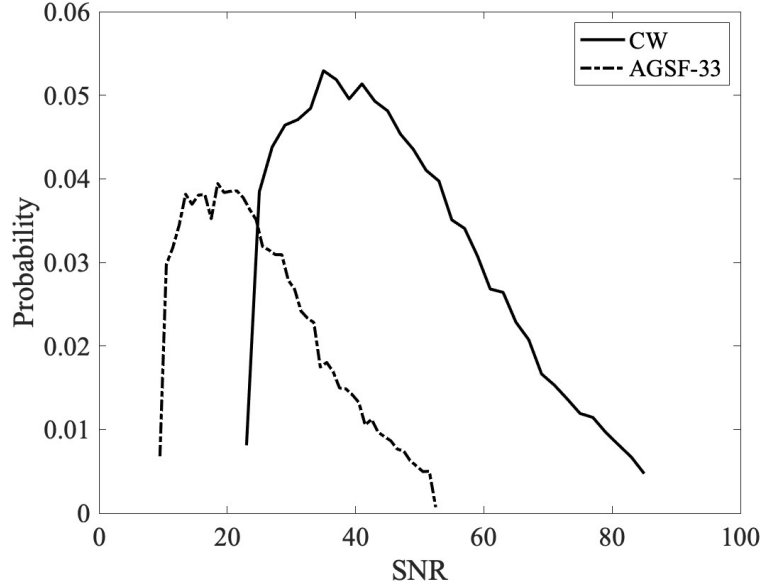


Figure 4.10: The measured SNR for the AGSF-33 and CW tracer particles. The CW SNR is shifted higher as a result of the higher contrast compared to the AGSF-33. Note that due to clustering of most of the AGSF-33 particles the true SNR of individual AGSF-33 particles is likely lower than observed.

velocities. Stereo or tomographic XPTV methods are necessary to alleviate the issues with 2D-projection and are topics of ongoing research. Light sheets like those used in PTV are not possible for XPTV. Most XPTV systems, including the one used in this study, are based on transmitted light instead of scattered light, so even if optics were available for in-lab XPTV, creating an X-ray light sheet would not be helpful. [31], [32], [48] use stereo X-ray imaging techniques, wherein two source-detector pairs are used to triangulate tracer particle locations from 2D-projection data. If multiple sources are not available, and the flow is slow enough that particle motion during object or source-imager rotation is below the desired velocity resolution, tomographic velocity data can also be obtained with a single source-detector pair. Recently, [46] and subsequently [49] used tomographic XPTV in which the object or source-detector pair is rotated in order to triangulate particle locations in 3D.

There is, however, an alternative to multi-source-detector pair or rotating systems for capturing 3D data. Provided that sufficiently monodisperse tracer particles are used, one can recover 3D velocity information by tracking the particle image size, which changes as a function of depth due to geometric magnification. Such a system would only require one source-detector pair. To briefly illuminate how such a system could be configured, equation 4.9 shows the minimum resolvable displacement in the depth-wise direction for any given particle location(s), pixel width, SDD, and particle diameter. The minimum resolvable displacement is determined by how far a particle must move in the depth-wise direction to

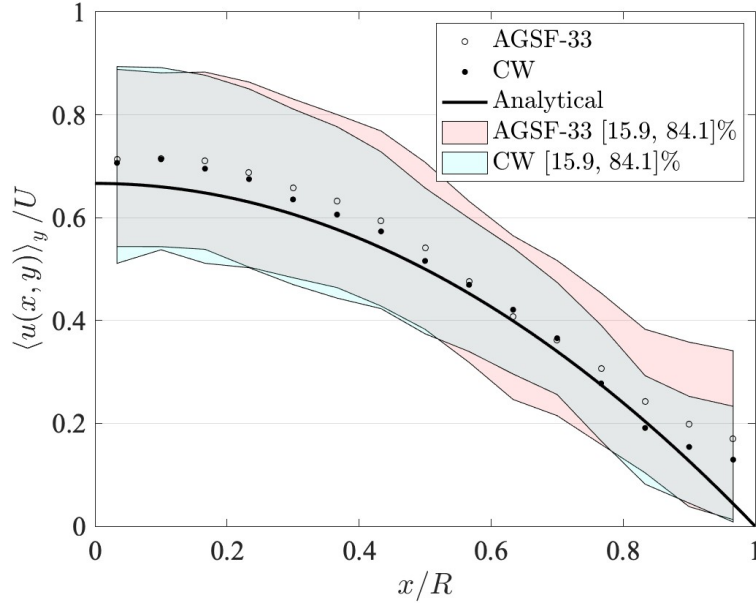


Figure 4.11: The DAVP as measured by the CW and AGSF-33 tracer particles compared against the analytical DAVP. The shaded regions are the measurement variation between the 15.9 and 84.1 velocity percentiles.

change its image diameter by one pixel.

$$(\Delta y)_{min} = (y_2 - y_1)_{min} = y_1 y_2 \left(\frac{w_{px}}{d_p SDD} \right) \quad (4.9)$$

Here, y_1 and y_2 are the depth-wise location from the source at two different times; w_{px} is the pixel width. Numerous trade-offs must be balanced to find a configuration that works. The depth-wise displacement resolution changes as a function of depth-wise location, so experiment depth is an important consideration. Another consideration is that a smaller tracer particle means a lower depth-wise displacement resolution. Lastly, a larger SDD increases the depth-wise displacement resolution, but makes the FOV smaller. Optimal values for all of these parameters will change for each experiment configuration.

One could also filter for particle images of a certain size, effectively creating an artificial 2D imaging plane akin to a laser sheet. XPTV tracer particles with a diameter tolerance that is tight enough for these techniques are, to the best knowledge of the authors, not presently available. Such tight tolerances for the particle diameter may not be an insurmountable requirement, though.

Figures 4.11 and 4.12 also show that CW tracer particles are more localizable due to their higher contrast and they did not clump like the AGSF-33 tracer particles. That is, the centroid of the CW tracer particles is able to be identified with greater precision than the AGSF-33 tracer particles. As a result, both the axial and x -component velocity mea-

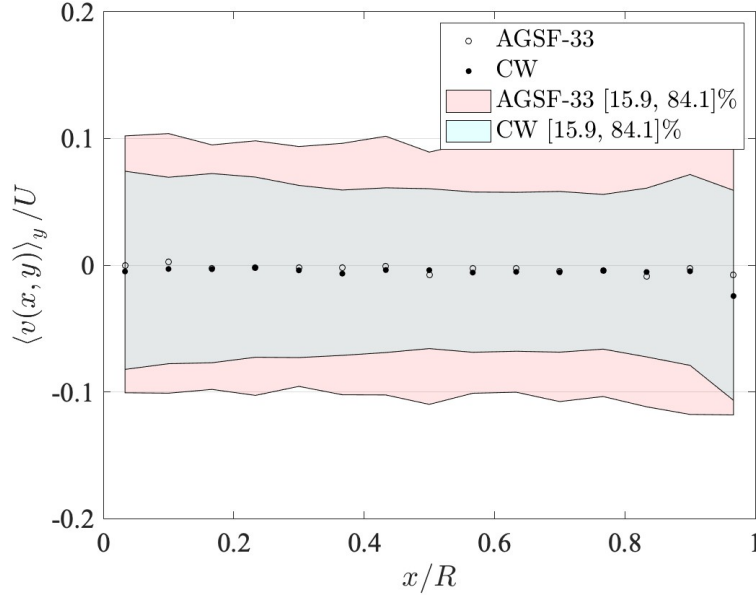


Figure 4.12: The measured x -component velocity, $v(x, y)$, for the CW and AGSF-33 tracer particles. The CW tracer particles, being more localizable and less clumped, exhibit smaller measurement variance compared to the AGSF-33. The shaded regions are the measurement variation between the 15.9 and 84.1 velocity percentiles.

measurements with the CW tracer particles demonstrate less spread than the AGSF-33 tracer particles. Better localizability is crucial because it reduces measurement error in velocity and improves spatial resolution.

Figure 4.13 shows the velocity vectors superimposed on an image. We note from inspection of many such images that: 1) the PTV algorithm does not produce spurious vectors a human observer would not create and 2) some particles that a human would trace are not traced by the algorithm. These data could likely be improved by optimizing the processing parameters or by using more advanced PTV algorithms. Improving image quality by using higher contrast particles, brighter sources, and better detectors will also help. These measurements demonstrate that time-resolved, $O(1 \text{ cm})$ domain XPTV at nearly 70 Hz is possible in the laboratory with both AGSF-33 and CW tracer particles. We achieve this result in spite of a dim laboratory X-ray source by using a single-threshold PCD and high-contrast tracer particles. Exploring the performance limits of CW tracer particles is the subject of future work. Brighter laboratory X-ray sources combined with advanced tracer particles will enable even faster XPTV. Particle contrast is proportional to photon flux, implying that with new X-ray sources that are 100 times brighter [50], frame rates on the order of 1 kHz are possible.

Despite the need for future improvements in monodispersity and tungsten coating thickness control, CW tracer particles outperform AGSF-33 tracer particles in terms of image

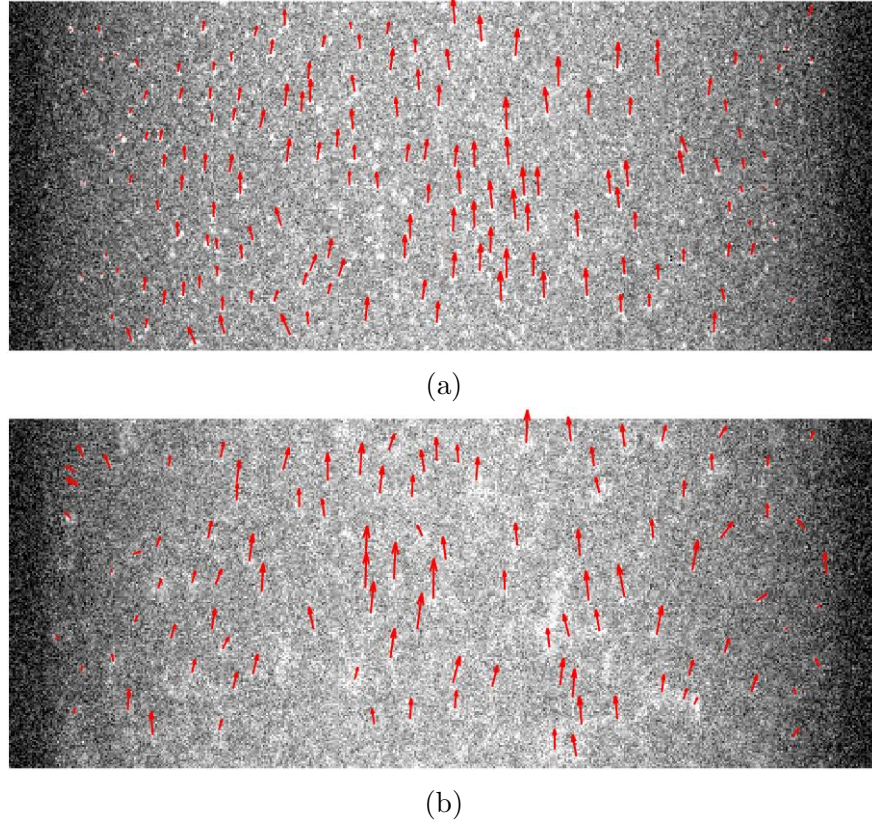


Figure 4.13: The velocity vectors overlaid on the hundredth frame of the (a) CW and (b) AGSF-33 footage. The greater variability of the AGSF-33 x -component velocity due to variability of the apparent particle centroid location is apparent, whereas the CW particles offer better contrast and are more robustly localized. The color scale is inverted for PTV processing in DaVis.

contrast and localizability while still measuring a comparable DAVP with satisfactory agreement to the analytical solution. As the tracer manufacturing process improves, CW tracer particles will enable the measurement of higher speed flows, especially when combined with brighter laboratory X-ray sources. The success and future improvement of the CW tracer particles should motivate the community and seeding particle manufactures to explore the XPTV tracer particle design space and push the boundaries of flows that can be studied with this technique.

Benefits and Challenges of PCDs

For the same source flux, PCDs can achieve a higher SNR and less blurring than scintillating detectors [13]. PCDs directly detect photons, dispensing with visible wavelength scatter and

glow artifacts in scintillators, which both contribute to blurring. Directly detecting photons also avoids the thermal and electronic noise associated with CCD and CMOS sensors. Despite these advantages, at this stage PCDs have a number of drawbacks that require consideration.

Generally speaking, SNR increases monotonically with increasing source brightness, enabling higher frame rates. For PCDs, however, the SNR can rapidly degrade due to photon count pile-up. That is, as the number of photons arriving at a pixel per unit time exceeds the circuit's ability to distinguish individual events, there ceases to be an increasing benefit to a brighter source. Current PCD nominal count rate limits are $O(10^6 - 10^7)$ photons/pixel/second, often with some form of retriggering, software correction, or both to mitigate the effects of pile-up. For the Dectris Pilatus3 detector used here, the nominal count rate limit is 10^7 photons/pixel/second when using retriggering and a count rate correction. We turn retriggering and the correction off for these experiments, however, because Dectris does not recommend using them with a polychromatic X-ray source. They are calibrated at a synchrotron for use with a monochromatic source. Unfortunately, most laboratory X-ray sources are polychromatic, including the one used here. Without retriggering and the count rate correction, the Dectris Pilatus3 can handle up to about 10^6 photons/pixel/second. Once pile-up begins to dominate, SNR degradation is observed and image artifacts such as detector tile boundaries become uncorrectable. Although we do not observe pile-up effects in these experiments, the count rate limit of PCDs is an important limitation on the source brightness that bounds the achievable SNR. Furthermore, pile-up effects can diminish the returns of a brighter source much earlier than the point of SNR degradation [13], making it an important consideration when designing an X-ray imaging system with a PCD.

A scintillating detector would not face the same limitations. X-rays are turned into visible wavelength photons regardless of their arrival rate, and the saturation limit for a CCD or CMOS sensor can often be adjusted. In practice, then, a scintillating detector could achieve a higher SNR than a PCD detector by enabling imaging with a brighter source than the PCD could handle. Indeed, scintillating detectors are widely used for imaging at synchrotrons, while PCDs are not placed directly in a synchrotron beamline. Currently, scintillating detectors can achieve faster frame rates or higher image quality than PCDs when a high flux source such as a synchrotron is used.

In addition to the temporal resolution advantage, scintillating detectors also have a spatial resolution advantage. Pixel sizes on currently available PCDs are relatively large – $O(100 \mu\text{m})$. Scintillating detectors offer much smaller pixel sizes, achieving higher spatial resolution.

Why, then, use a PCD for X-ray flow visualization? First, although PCDs currently suffer from count rate limitations, due to improving circuitry, detector materials and reduced pixel sizes, future PCDs can be expected to accommodate increasing count rates. When pile-up is not the limiting factor, the theoretical SNR advantage of PCDs is apparent. Furthermore, despite the current drawbacks, there are benefits to PCDs even now that in some cases may outweigh the limitations. Chiefly, PCDs can directly measure the deposited photon energy, in effect capturing “color” X-ray images. PCDs that are now coming to market are able to resolve the incoming photon energy with two or more thresholds, enabling K-edge material

detection.

K-edge material detection makes use of the fluorescence absorption edge(s) in the mass attenuation coefficient of a material. Many materials have similar mass attenuation coefficient curves, making them difficult to distinguish by image contrast alone, especially with a dim source producing low SNR images. Copper and iron, for example, have similar mass attenuation coefficients across a wide range of photon energies. Fluorescence absorption edges, however, are unique to each element because they are a direct consequence of an atom's stable electron orbit energies. Copper has an absorption edge at 8.9 keV, while iron has one at 7.1 keV. With an energy-resolving PCD, one could identify these absorption edges and so distinguish materials in the flow. It would be possible to track tracers made of different materials, a process analogous to fluorescent tracer particles with different colors in visible light flow visualization. Additionally, one could observe the scalar mixing of two or more phases while simultaneously measuring the fluid velocity field with tracer particles, a difficult experiment with visible light techniques, and an all but impossible one in opaque media or containers. For certain experiments, this unique capability of PCDs may outweigh the drawbacks.

4.5 Conclusions

This study demonstrates quantitative laboratory XPTV by measuring the 2D-projected velocity profile of Poiseuille flow in a round pipe with two different tracer particles. By capturing images of tracer particles in 15 ms exposure times, we show the potential to speed up laboratory XPTV to nearly 70 Hz, a limit that can easily be extended with brighter sources and improved detectors. We show the value of high-contrast tungsten-coated, hollow carbon microsphere tracer particles designed specifically for quantitative XPTV. The tungsten-coated CW tracer particles offer a contrast enhancement over the previously available AGSF-33 tracer particles, making them easier to localize and, in principle, allowing the same SNR with a shorter exposure time. As the manufacturing process for the CW and other custom particles improves, their utility as XPTV tracer particles can be expected to surpass AGSF-33. Continuing exploration of the XPTV tracer particle design space is important for developing application- and technique-specific tracers. For example, to develop smaller particles or optimize the attenuation contrast for application-specific fluids. The metrics used in this study, and methods previously discussed in [23], can be used for testing future tracer particle prototypes.

In addition to demonstrating the value of tracer particles developed specifically for X-ray flow visualization, we explored the feasibility of using PCDs for X-ray flow visualization. By showing that PCDs can acquire laboratory XPTV images at speeds comparable to scintillating detectors, we show that energy-thresholding material detection techniques are feasible for future laboratory XPTV experiments. With PCDs improving rapidly, material detection and other energy-thresholding techniques can improve tracer particle detection beyond what is capable with scintillating detectors, opening up new opportunities to use X-ray light to

the experimentalist's advantage. In fact, Simultaneous multi-species and scalar field flow tracing experiments are possible with energy-resolving PCDs, for example. As laboratory X-ray sources get brighter, and PCDs get faster, these techniques will be of even greater utility. The flexibility of the system that we propose lends itself to growing alongside X-ray source and detector technology.

XPTV, XPV, and related X-ray flow visualization techniques make possible the study of flows that opacity and refraction make difficult or impossible to observe with visible light. By demonstrating the potential of XPTV with PCDs and advanced tracer particles in the laboratory, this promising technique can proliferate in laboratories across the fluid dynamics community.

Chapter Supplementary Material

Image Pre-Processing

Flat-field correction is applied to all of the images. As part of this correction, dead pixels are replaced with the median value of the surrounding pixels. A flicker correction is also applied as the X-ray source intensity exhibits an $O(0.1\%)$ variation at approximately 0.25 Hz and integer-multiple frequency harmonics, which is presumed to be the controller update frequency of the anode current feedback loop. The flicker correction involves determining the 1st and 99th percentile pixel values for each image. Those pixel values are then established as the saturation limits for their respective image. The images are then inverted so that the particles are bright on a dark background, which the Lavisision DaVis processing algorithms expects. After inversion, the images are converted into 16-bit images. Then, the images are exported to undergo the following filtration and other operations in DaVis:

1. Mask the area outside of the region of interest.
2. Subtract the average intensity of all frames.
3. Spatial bandpass filter to features that are 3-6 pixels in length.
4. Subtract 3,700 counts to isolate the particle images as positive integer values in preparation for the next step. This value may change as a function of particle SNR.
5. Set any negative pixel value to 0.

PIV Processing

PIV is used as a pre-processing step to the PTV in order to establish an approximate particle velocity and a search radius for the particle tracking algorithm. The PIV settings chosen to provide a low spatial resolution estimation of the fluid velocity are as follows:

1. First pass: 64×64 pixel window, 50% overlap, single pass.

2. Second pass: 32×32 pixel window, 50% overlap, double pass.
3. Remove groups with < 5 vectors.
4. Allowable axial vector range: 3 to 19 pixels, encompassing range physically possible given range of particle densities.
5. Allowable x-component vector range: -4 to 4 pixels. Nominally zero, but due to shifts in the detected centroid, the x -component velocity can be measured as non-zero. This also allows for the x -axis of the images to potentially be misaligned with tube axis.

PTV Processing

After the PIV processing step, DaVis' particle tracking algorithm is called. For each frame and tracked particle the centroid location, and x - and axial-velocity components are calculated. This data is imported to MATLAB to bin the velocities by x -location and calculate the average axial velocity component. The PTV settings are as follows:

1. Particle size range: 3 to 12 pixels.
2. Allowed vector range relative to reference (i.e., velocity from PIV): 10 pixels, while maximum displacement was 8 pixels.

Chapter 5

$O(1\text{ kHz})$ Imaging and Tomographic Laboratory X-ray Particle Tracking Velocimetry

5.1 Background

We combine X-ray-specific tracer particles, a photon counting detector, and a liquid metal anode X-ray source to achieve $O(1\text{ kHz})$ X-ray imaging speeds in the laboratory, $15\times$ faster than previous comparable studies. To demonstrate the capabilities of these imaging speeds, we conduct three experiments: 2D and 3D X-ray particle tracking velocimetry (XPTV) of Poiseuille pipe flow, 3D XPTV of flow around a Taylor bubble, and 3D scalar mixing with a laminar jet. These experiments demonstrate the performance improvement of combining the aforementioned elements, the applicability to multiphase flows and deforming systems, and the potential to capture scalar and vector quantities simultaneously. Most importantly, these experiments are conducted in the laboratory, showing that in-lab X-ray particle velocimetry techniques are now usable for a wider range of flows of interest.

Particle tracking velocimetry and particle image velocimetry are commonly used in contemporary fluid dynamics experiments. One of the major limitations of these techniques, though, is their inability to measure flows that are opaque to visible wavelengths of light. The opacity could be due to either the surrounding media, the fluid itself, or multiple refractive interfaces. X-rays, on the other hand, have an index of refraction near unity, and many materials are transparent to X-rays. Recently, researchers have started to combine X-ray imaging and particle-based velocimetry to study multiphase flows [7]–[9], [31], [45], [51], [52], porous media flows [32], [46], [49], [53], biological flows [25], [28], [29], [54], and many others that are difficult to observe with visible light image-based measurement techniques. So far, however, the temporal resolution of X-ray velocimetry and other X-ray imaging-based techniques is insufficient to resolve many flows of interest. Wider availability of X-ray particle tracking velocimetry (XPTV) and particle image velocimetry (XPIV) could offer previously

unobtainable insight into many of these flows.

Unlike their visible light counterparts, 2D and tomographic XPTV and XPIV – all of which comprise a class of measurement we call X-ray particle velocimetry (XPV) – are not currently capable of reaching frame rates $O(1\text{-}100\text{ kHz})$ needed for many in-lab applications (i.e., outside of synchrotrons). Although continuously improving, non-scintillator based X-ray imaging detectors are orders of magnitude slower than visible light high-speed cameras. Even if X-ray imaging detectors were fast enough, the relatively modest polychromatic photon flux generated by most in-lab X-ray sources imposes another limitation. A brighter X-ray source enables shorter exposure times while retaining usable image quality. As a result, most prior work in XPV has been done at synchrotrons, which can achieve photon fluxes order of $O(10^{17})\text{ ph/m}^2/\text{sec}$ [55]. Laboratory X-ray sources are orders of magnitude dimmer than a synchrotron.

[24] captured the first XPIV measurement at the Pohang Light Source. They generated contrast by attenuation, with edge enhancement due to refraction. Their experiment measured the 2D-projection of Poiseuille flow. Im et al. [56] used phase contrast imaging at the Advanced Photon Source. Their study also used tomography to reconstruct the velocity field of pipe flow in 3D. Measurements of simple, low speed flows eventually gave way to more complex, high speed flows such as the cavitating sheet experiment recently performed by Ge et al. [30] at the Advanced Photon Source synchrotron. Synchrotrons, while incredibly bright, impose practical limitations on the types of experiments one can run. Typically, synchrotrons beam widths are on the order of a few millimeters, making large experiment geometries impossible. Multiphase flows in particular are notoriously challenging to scale because complete similarity is rarely achieved in scaled experiments. Furthermore, synchrotron beam time is in finite supply and high demand. For most users, using a synchrotron requires costly, inconvenient travel. When one does get time on synchrotrons, it is often for a few hours to a few days, making long term or complicated experiments impractical to conduct.

The limitations posed by synchrotrons motivate research to make XPV techniques practical for laboratory use. Prior work has demonstrated that XPV is feasible in a laboratory setting with currently available equipment [23], [33], [46], [57]. However, most of these experiments use exposure times $O(10\text{-}100\text{ ms})$ – too slow for most flows of interest. Many of these systems also use large, high Stokes number particles [33], [45], [58]. While they may be appropriate for the selected application, large particles are not suitable for every flow. For energetic or low-speed flows they may not be accurate flow tracers due to inertial and buoyancy effects, respectively.

The three primary limitations for in-lab XPV currently are detector performance, tracer contrast, and source brightness [46]. The means to address these shortfalls work best in combination: “noiseless” photon counting detectors (PCDs) [13] with new, high contrast, neutrally buoyant flow tracers [57], and brighter in-lab sources such as liquid metal jet (LMJ) anode sources [50]. Previous work has demonstrated the effectiveness of the former two approaches [57]. This study combines for the first time a LMJ X-ray source with X-ray imaging-specific tracer particles and a PCD.

Laboratory-scale LMJ X-ray sources can be orders of magnitude brighter than typical

laboratory solid target sources for a given focal spot size $O(10\text{ }\mu\text{m})$. A jet of liquid metal acts as the anode, overcoming the thermal limitations of stationary or rotating anode sources. That is, LMJ sources can achieve orders of magnitude higher output power density, achieving greater brightness without defocusing the focal spot.

For this study, we use an Excillum MetalJet E1+ X-ray source to capture up to 1 kHz X-ray footage of *i*) laminar pipe flow, *ii*) Taylor bubble flow, and *iii*) laminar jet scalar mixing. We demonstrate that LMJ sources, coupled with a PCD and contrast-optimized nominally neutrally $O(50\text{ }\mu\text{m})$ tracer particles, enable particle tracing in $O(10\text{ mm})$ domains with exposure times roughly $15\times$ lower than previously achieved. 1 kHz frame rates enable domain-resolving full 360 degree computed tomography (CT) up to 3 Hz.

These advancements make resolving flow with speeds $O(100\text{ mm/s})$ possible with an $O(50\text{ }\mu\text{m})$ tracer particle. Such flow speeds are well within the range of human blood flow [59], for example. Many porous media flows fall into this category as well [46], [49]. In effect, this study demonstrates that laboratory XPV is currently usable for studying previously inaccessible flows of interest.

This paper is organized as follows: section 5.2 details the flow experiment designs, X-ray source, tracer particles, and detector; section 5.3 discusses the experiment results; finally, section 5.4 provides the conclusions and future outlook for laboratory XPV. Additional details on image processing and particle tracking algorithms are provided in the appendices.

5.2 Experiment Design

Flow Setup

Three experiments are conducted with one setup to explore what is achievable with a brighter source in combination with improved flow tracers and a PCD. As discussed in section 4.1, many of the early synchrotron XPV experiments that focused on technique development measured developed pipe flow. It is straightforward to compare to theory; we replicate that experiment in the laboratory as the first of the XPV experiments for this study. This also enables clear comparison to [57], which used an identical pipe, fluid, and nearly identical tracer particles. This comparison usefully elucidates the performance improvement due to the brighter source. We study developed pipe flow with both 2D-projected and tomographic XPTV (TXPTV). The second experiment considers flow around a Taylor bubble, the results from which can be compared to [46], which used a similar setup to study a Taylor bubble with a traditional solid-target X-ray source and silver coated hollow tracers particles developed for visible light PIV. Lastly, we study the 3D mixing of a potassium iodide (KI)-water solution jet being injected into glycerine with high-speed CT.

Figure 5.1 depicts the X-ray imaging setup. For these experiments, the source-to-object distance (SOD) is 56.5 mm; the source-to-detector distance (SDD) is 521.5 mm. We measure the former from the focal spot to the center of the pipe; the latter from the focal spot to the

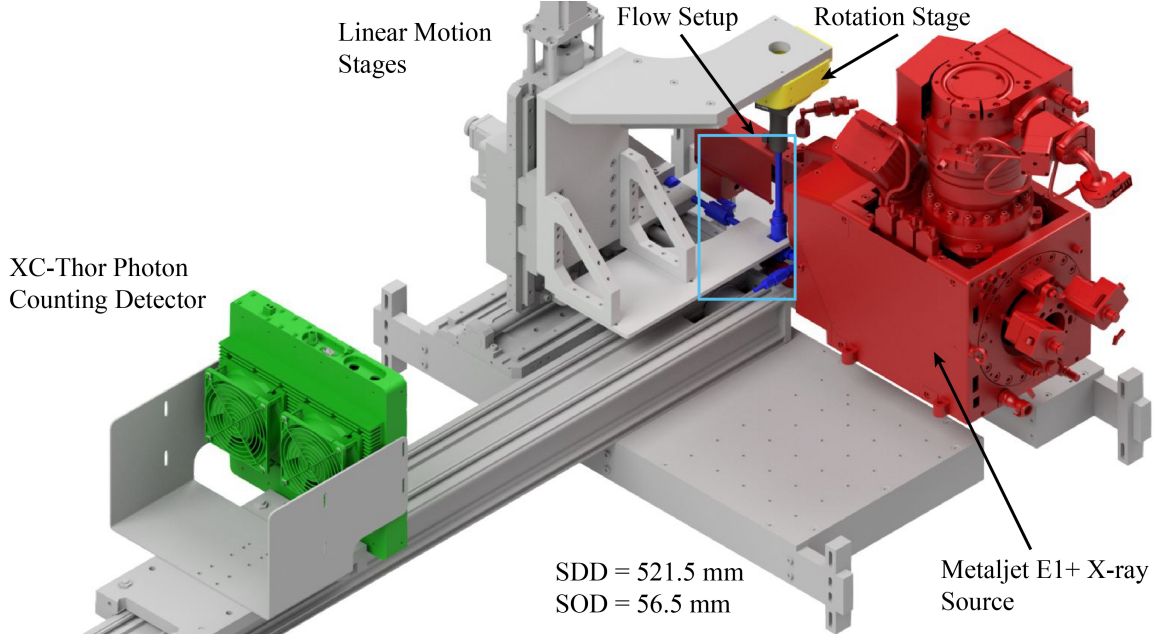


Figure 5.1: The X-ray imaging setup. The flow setup is placed in front of the X-ray source aperture. A LAB Motion RT100S air bearing rotation stage controls rotation of the pipe.

face of the detector pixel plane. The field of view is $22.2 \text{ mm} \times 2.8 \text{ mm}$ due to geometric magnification.

Figure 5.2 shows the flow experiment setup. A plastic 6.35 mm (0.25 in.) inner diameter, 9.53 mm (0.375 in.) outer diameter polycarbonate pipe is held at the top to linear motion stages (Optics Focus MOX-06-150, MOX-06-200, and MOX-06-100-B) and a LAB Motion Systems RT100S air bearing rotation stage. The RT100S air bearing stage has a maximum rotation speed of 3 Hz, which constrains maximum the CT scan rate in these experiments. The pipe is connected to a T-joint that enables particle and fluid seeding, as well as pumping with a Harvard Apparatus Pump 33 Dual Drive System (HAP33DDS). The valve to the syringe pump is closed during seeding while the valve to the seeding syringe is open, and vice versa during experiment runs. The HAP33DDS pump is able to drive two syringes independently and simultaneously. The first syringe contains pumping fluid (in this case glycerine); the other contains air for the Taylor bubble or KI-water solution for the jet. The HAP33DDS is accurate to 0.25% of the flow rate and can achieve flow rates ranging from (1.02 pL/min to 106 mL/min) depending on the syringe size. Our experiments use glycerine and range from $Re = 8.7 \times 10^{-4}$ for the pipe flow TXPTV experiments to $Re = 486$ for the KI jet. A 3.18 mm (0.125 in.) diameter brass pipe is inserted concentrically and terminates below the plastic pipe in the T-joint as seen in figure 5.2b. For the Taylor bubble experiments, this pipe is connected to the second syringe, which is full of air. The Taylor bubble is released from this pipe into the plastic pipe test section. For the KI jet experiments,

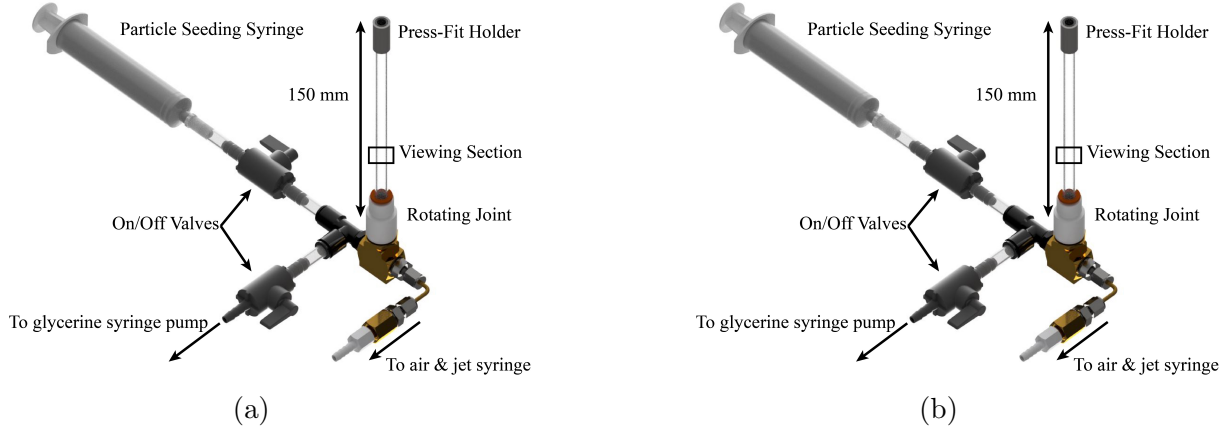


Figure 5.2: (a) Schematic of the flow setup where the test section is connected to supply lines via a swivel joint that rotates freely. (b) The inside of the flow setup shows a brass pipe mounted concentrically within the plastic pipe. The Taylor bubble and KI jet are injected via this brass pipe.

this pipe is connected to a syringe full of KI-water solution, which is then ejected from the brass pipe into the test section.

For these experiments, we use pure glycerine as the working fluid. The glycerine temperature is nominally constant at 27.6°C , the density is 1260 kg/m^3 , and the dynamic viscosity is $0.73 \text{ Pa}\cdot\text{s}$. Glycerine has an entrance length two orders of magnitude shorter than water for the same velocity. This allowed us to test higher velocities while retaining a fully-developed pipe flow. Glycerine also retarded the rise of the Taylor bubble, making it possible to reconstruct its morphology with the 3 Hz full angle CT acquisition rate. The glycerine is seeded with hollow carbon tungsten-coated microsphere (CW) tracer particles.

Experiment Parameters

For the 2D-projected pipe flow experiments the glycerine-tracer mixture is pumped at 42.75 mL/min , yielding a center line speed of 45 mm/s ($Re = 0.40$). Images are captured at 1 kHz .

A much slower flow is used for the TXPTV experiments to accommodate the lower temporal resolution. For the pipe flow experiments, the pipe is rotated at 720 degrees per second (dps) for a full 360° CT temporal resolution of 2 Hz . The fluid is pumped at 0.0760 mL/min , resulting in a 0.08 mm/s center line flow speed ($Re = 8.7 \times 10^{-4}$). At 720 dps , the centripetal acceleration is $0.05g$ at the pipe wall, where g is the acceleration due to gravity on earth. At 720 dps , then, we can safely neglect the effect of centripetal acceleration on the flow. Before image acquisition begins the pipe is allowed to rotate for at least 5 seconds so that liquid will be experiencing solid body rotation when the data

Experiment	Rotation Speed (dps)	Reynolds Number	Max. Flow Speed (mm/s)
2D Pipe Flow	0	8.7×10^{-4}	45
3D Pipe Flow	720	0.40	0.08
3D Taylor Bubble	1080	0.014	0.5
3D KI Jet	720	486	78.1*

Table 5.1: The experiment parameters for each of the three experiments that are conducted.

*At the nozzle exit; we measure roughly 19 nozzle diameters downstream.

is captured. The characteristic time scale for flow inside a rotating cylinder is given by $\tau = R^2\nu^{-1}$, where ν is the kinematic viscosity of glycerine and R is the pipe radius. At the temperature we operate at, $\tau = 0.02$ s – shorter than the duration we wait before capturing data. At low Re , the swirling flow from entering the pipe should not have a significant effect [60]. Our TXPTV pipe flow measurements do not detect azimuthal velocity components.

In the Taylor bubble experiments 0.5 mL of air is injected through the ejection pipe within the T-joint. The CW-laden glycerine is already seeded into the viewing section. There is no pump-induced co-flow; the only flow is due to the Taylor bubble motion. The bubble rises at approximately 1.9 mm/s ($Re = 0.014$). Images are captured at a 1 kHz frame rate; the pipe is rotated at 1080 dps (3 Hz). Although for the Taylor bubble $\tau = 0.65$ s (assuming air fills the pipe), we do note a slight azimuthal component in our results. At this rotation speed the centripetal acceleration in the pipe at the wall is $0.11g$. This could begin to distort the Taylor bubble shape. As shown later, though, this centripetal acceleration is not a major concern for the tracers since they are nominally density matched with the liquid. To increase the CT acquisition rate without concern for centripetal acceleration effects, one could rotate the source-detector pair as opposed to the flow loop, or utilize multiple source-detector pairs.

In the KI jet experiments, the KI-water solution is nearly saturated at 1.4 g/mL of KI. We inject the solution at 37.21 mL/min. $Re = 486$ based on a nozzle ejection speed of 78.1 mm/s, the density of the KI-water solution (1650 kg/m^3), the nozzle diameter, and the dynamic viscosity of water at 27.6°C . The jet is observed roughly 19 nozzle diameters downstream. Images are captured at 500 Hz and the pipe is rotated at 720 dps (2 Hz). For finer temporal interrogation of the jet evolution, we reconstruct the CT scans by overlapping the projections used for each reconstruction by 90%. While this does not increase the true temporal resolution (which would mitigate motion artifacts) it leverages data contained in the 500 projections per scan to show the jet evolution in finer temporal detail. Velocimetry data is not captured in this experiment. Instead, we measure the evolution of the KI-water solution concentration in 3D. The parameters for all of the experiments can be found in table 5.1.

Tracer Particles

Tracer particle properties determine how quantitative a particle-based velocimetry measurement can be. Ideally, tracer particles should be neutrally buoyant, small, and have low inertia. Neutral buoyancy ensures that there are no bias errors in the vertical velocity component measurement or due to centripetal acceleration. Small particles maximize the spatial resolution of the measurement. Smaller particles can also perform better in presence of steep velocity gradients [61]. Lastly, low inertia (for which being a small particle helps) means that the particle will more readily accelerate with the surrounding fluid. The parameter used to determine how well particles trace the flow is the Stokes number, St , defined by equation 5.1. When $St \ll 1$, the particles can be said to be accurate flow tracers.

$$St \equiv \frac{\tau_p}{\tau_f} \quad (5.1)$$

In equation 5.1, τ_p is the characteristic response time of the particle to an impulse uniform acceleration and τ_f is a characteristic time of the flow. Typically τ_f is taken to be a length scale divided by a characteristic velocity. For a particle that approaches neutral buoyancy, the response time can be calculated by

$$\tau_p = d_p^2 \frac{\Delta\rho}{18\mu}. \quad (5.2)$$

where d_p is the particle diameter, $\Delta\rho$ is the difference between the fluid and particle densities, and μ is the dynamic viscosity of the fluid. From equations 5.1 and 5.2 it is clear that a small, neutrally buoyant tracer particle is necessary to take an accurate measurement.

CW Tracers

Nominally neutrally buoyant 45–53 μm tungsten-coated hollow carbon microsphere (CW) tracers are used in this study. The CW tracer particles, designed with tools developed in [23], were shown in [57] to have higher contrast and better localizability than similarly sized silver-coated hollow glass tracer particles, so they are used for the high-speed measurements in these experiments. The CW particles are manufactured by Ultramet in California.

We use a Monte Carlo calculation to approximate the settling speed of the tracer particles. Inherent to our calculations are the assumptions that 1) the tracer particles are spherical and 2) the relative velocity between the settling or rising particles is low enough that the particles are in the Stokes flow regime. These particles are still in the prototype stage, however, so they exhibit strong polydispersity and uneven coating thicknesses. We assume a 200 nm mean tungsten coating thickness that is Gaussian-distributed with a standard deviation of 20 nm. Based on nanofocus X-ray images of the particles, we measure the particle diameter distribution to use in the Monte Carlo calculations. An example nanofocus image can be seen in figure 5.3; the particle size distribution based on all such batches is shown in figure 5.4. We find the particles have an average diameter of 50 μm , as designed. The standard

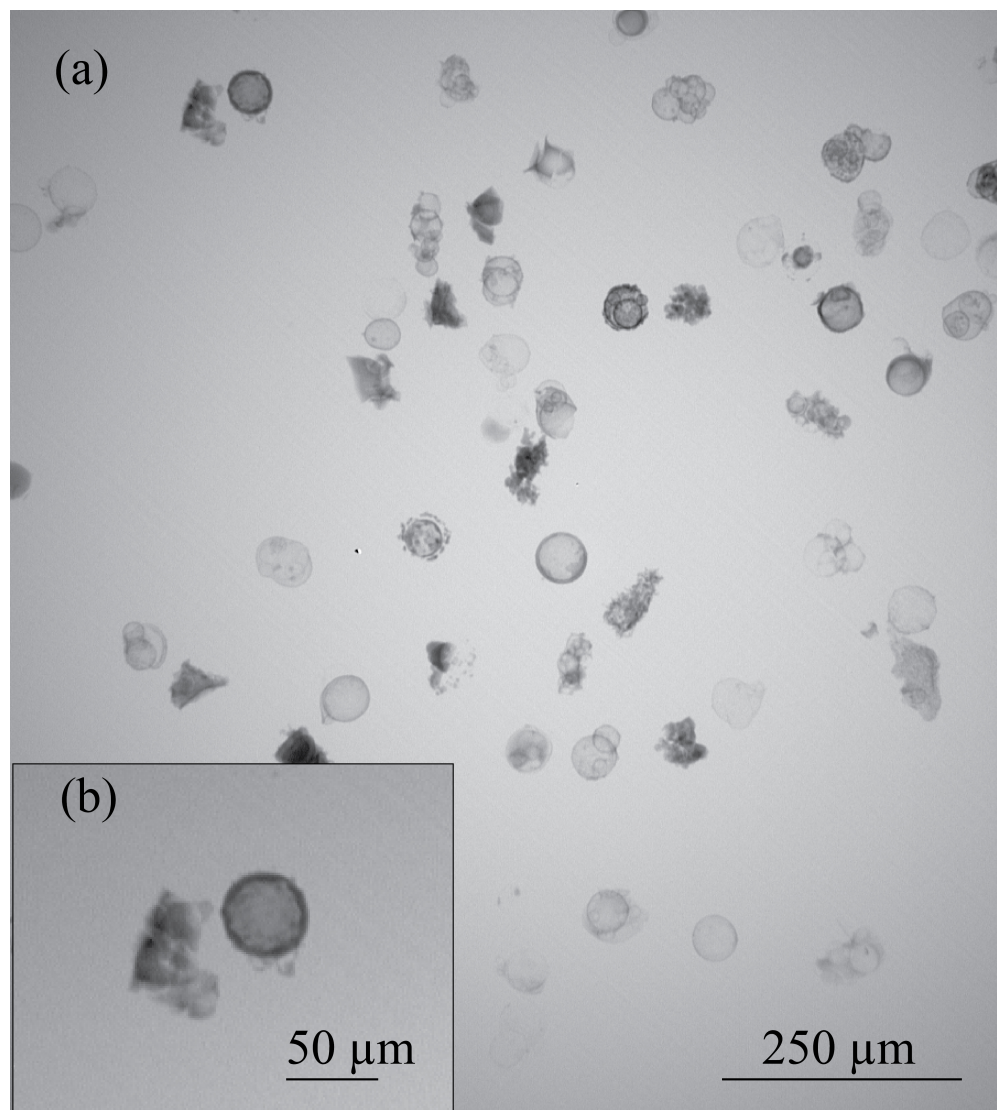


Figure 5.3: (a) Multiple batches, such as this one, of CW particles are examined with a nanofocus X-ray source (NanoTube N3, Excillum AB). (b) The prototype nature of these particles is evident from the various coating thicknesses, broken particles, and polydispersity.

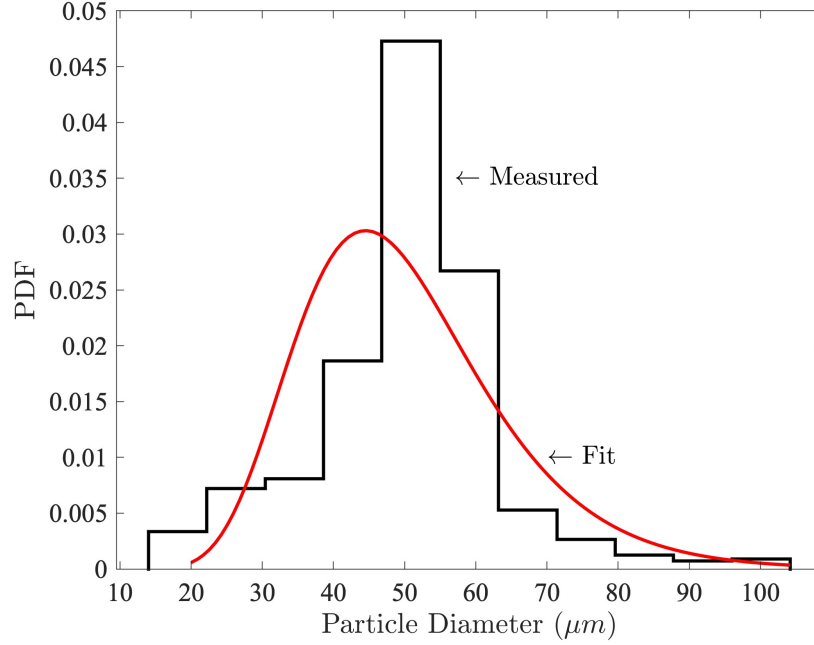


Figure 5.4: The measured and fit particle size distribution. We fit a log-normal distribution to the particles, which exhibit clear polydispersity.

Fluid	$ \langle u \rangle_{st} /U$	$St \times 10^6$
Glycerine	6.8×10^{-4}	2.8
Water	0.019	2300

Table 5.2: The mean settling speed to center line speed (U) ratio and Stokes number of the CW tracer particles that we use in these experiments. For the speed ratio, we compare to the slowest flow speed we measured ($U = 0.08 \text{ mm/s}$); for the Stokes number we use the highest flow speed we measured ($U = 45 \text{ mm/s}$). This is to ensure our Stokes number and buoyancy measurement bias are low for all of our flow conditions.

deviation is large, as expected given the polydispersity, at $13 \mu\text{m}$. In figure 5.5 we show the settling velocity distribution of the CW particles in glycerine and water based on Monte Carlo calculations with 10^7 trials. Table 5.2 lists the settling speed as a ratio of the characteristic flow speed and the Stokes number of CW particles in both water and glycerine. In glycerine, each of these values is much less than unity, so we can take the CW particles to be ideal flow tracers.

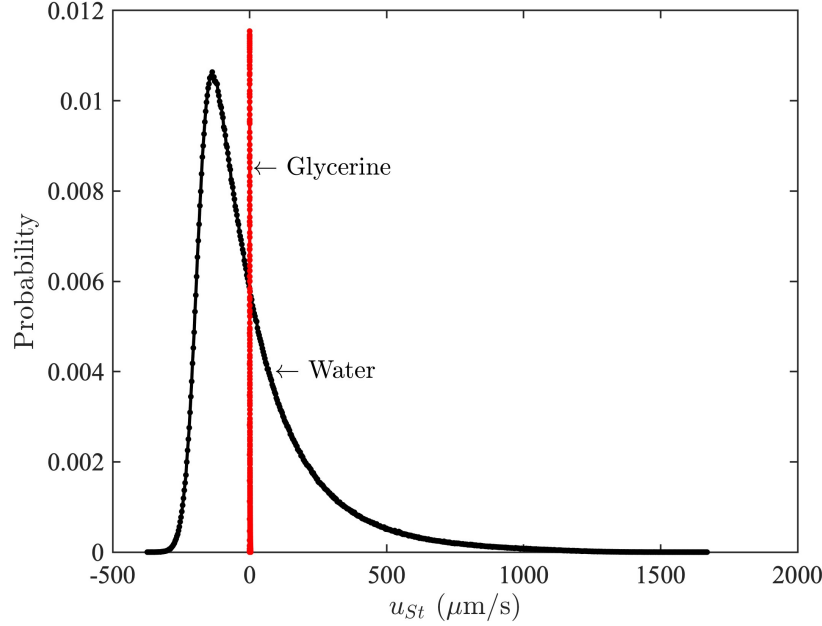


Figure 5.5: The settling speed distribution of the CW tracer particles in water and glycerine. Using glycerine dramatically reduces the spread in settling speeds compared to water.

The MetalJet X-ray Source

The critical element that enables $O(1 \text{ kHz})$ frame rates is the MetalJet E1+ X-ray source from [16]. This source is an order of magnitude brighter than a traditional solid anode microfocus X-ray source. For example, the MetalJet E1+ model used is roughly $20\times$ brighter for the same focal spot size and source acceleration voltage than the solid anode source used in [57]. The apparent X-ray focal spot size is approximately $30 \text{ }\mu\text{m}$ at the settings used in this study. We operate the MetalJet E1+ LMJ at a 160 kV source acceleration voltage and 4.375 mA for an output power of 700 W. The spectra for the MetalJet E1+ and a traditional solid anode microfocus tube used in [23] and [57] can be seen in figures 5.6 for a rough comparison of a LMJ source to a typical solid anode microfocus source. Here, brightness is defined as the total number of photons emitted per steradian per second. While convenient, this definition does not account for different photon energy spectra between sources. Different spectra may affect the image quality improvement depending on the materials in the field of view.

LMJ X-ray sources use a liquid metal jet target – in this case, a gallium-indium mixture – instead of a solid tungsten target. The benefit of using a liquid target is that the heat generated from electrons hitting the target is rapidly advected away and the anode regenerates such that pitting and material vaporization are not a concern like they would be for a solid anode target. By removing heat from the focal spot rapidly, the target can withstand greater power from the electron beam before vaporization is a concern. In these experiments, we run the LMJ source at 160 kV, 4.375 mA (700 W) compared to 55 kV, 0.5 mA (25.5 W)

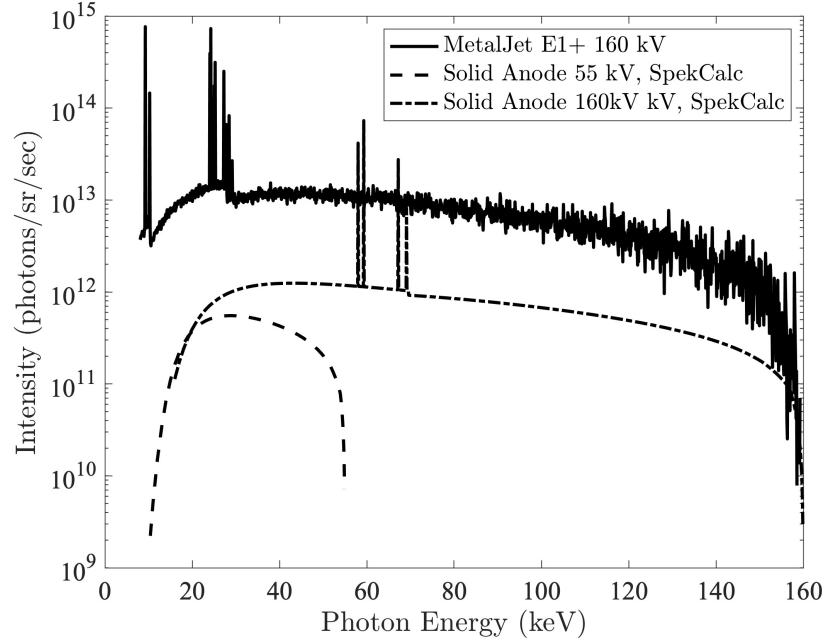


Figure 5.6: The X-ray flux from the MetalJet E1+ LMJ source used [16] versus a traditional solid anode microfocus X-ray source [17]–[19]. A LMJ is able to withstand greater focal spot power density. As a result the spectrum is $20\times$ brighter for the same focal spot size and source acceleration voltage.

for the source in [57], resulting in a nominally $83\times$ brighter source compared to that study. When matching the focal spot size and source acceleration voltage, the MetalJet E1+ source is $20\times$ brighter than the YXLON source in [57]. Such a bright source can improve the image signal-to-noise ratio (SNR), decrease exposure time, or both. How much a brighter polychromatic source helps is dependent on the materials in the experiment geometry, and what photon energies they most attenuate.

Image Processing

We collect data images with an XC-Thor FX20.1.256 PCD. For this study the flat field corrections are applied by the detector software prior to any custom processing of data. The flat field correction map is generated by the detector software with images taken after placing 0, 2, 7, and 21 mm thick plates of aluminum in front of the source one-by-one and collecting flat images at the source settings to be used.

For 2D-projected XPTV processing, the flat field corrected images are inverted, then imported to LaVision DaVis 8.4. We use standard DaVis algorithms. With TXPTV, we reconstruct the field of view using the ASTRA Toolbox [62], [63]. Then, we segment the particles from the fluid in MATLAB to extract the particle centroid locations. A particle

tracking code in MATLAB was developed to trace the particle movement through multiple reconstructions. Our workflows for the pipe flow, Taylor bubble flow, and KI jet flow are detailed in appendices.

Calibration and Uncertainty for 2D XPTV

We define the pixel-to-mm calibration by counting the pixels required to span the known pipe outer diameter – 9.5 mm (0.375 in.). As discussed in [57], the actual pixel-to-mm scaling changes as a function of particle distance from the source due to geometric magnification. However, because we collect 2D-projected images, this depth information is lost, so a single calibration value is used for the entire experiment depth. A good rule of thumb is to keep the relative error associated with geometric magnification, termed the relative magnification error (RME), below 10%. [57] derives the equation for the RME, shown to be

$$\epsilon(y) = \frac{|\Delta z - \Delta Z|}{\Delta Z} = 1 - \left(1 + \frac{|y|}{SOD}\right)^{-1}, \quad (5.3)$$

where Δz and ΔZ are the measured tracer particle motion and the true particle motion, respectively; y is the depth-wise location of the tracer particle measured from the center of the experiment. In order to keep the maximum RME below 10%, the magnification aspect ratio (MAR) should be kept below 0.2. The MAR is defined in equation 5.4, where δ is the experiment depth in the beam direction.

$$MAR = \frac{\delta}{SOD} \quad (5.4)$$

Equation 5.4 assumes symmetry in the depth-wise direction ($\delta = 2 y_{max}$). For all $y < y_{max}$, the RME will then be less than 10%. For these experiments, the $MAR = 0.11$.

Another source of measurement variance that is unique to 2D-projected data is the depth-averaged velocity profile (DAVP). In 2D-projected data, particles at the back wall, center, and front wall can all appear at the same horizontal location across the pipe. As a result, when collecting particle tracking data, one is actually sampling from the velocity distribution in the depth-wise direction at a given horizontal location. In flows with strong velocity gradients, this can be problematic. Even in flows with relatively shallow gradients, such as laminar pipe flow, the measured velocity profile is depressed compared to the radial cross section velocity profile. For laminar pipe flow, where there is an analytical solution, the DAVP can be calculated by taking a depth-wise average, shown in equation 5.5.

$$\langle u(x, y) \rangle_y = \frac{4Q}{3\pi R^2} \left[1 - \left(\frac{x}{R}\right)^2 \right] = \frac{2}{3}u(r = x) \quad (5.5)$$

It is to this profile that we compare our data. Here, $\langle \cdot \rangle_i$ denotes mean in the i direction, Q is the volumetric flow rate, R is the pipe radius, and x is the horizontal coordinate (aligned here with the projected radial coordinate). Measuring from a distribution of velocities that

Experiment	Particle Rotation Blur (d_p)	Particle Motion Blur (d_p)
3D Pipe Flow	1.6	0.8
3D Taylor Bubble	1.2	3.33

Table 5.3: The expected maximum particle blur due to pipe rotation and particle motion in units of nominal particle diameter, d_p (50 μm). Maximum expected flow velocities and radial distances are used to calculate these values. Values greater than one indicate that some blurring is possible.

ranges from $u = 0$ to center line speed, U , means that there will inevitably be a wide spread in the velocity measurement. As with the RME, issues with depth-averaged profiles can be alleviated with 3D tomographic or stereo XPTV measurements.

The final source of uncertainty is buoyancy, which is discussed in section 5.2. Buoyancy effects in vertically aligned flows can create a velocity measurement bias. We showed in section 5.2 that the flow speeds are sufficiently high compared to the particle settling speeds that buoyancy error can be neglected. In many previous proof-of-concept XPTV experiments, this was not the case [31], [33], [44].

Calibration and Uncertainty for TXPTV

For TXPTV we define a voxel-to- mm^3 calibration similar to the 2D calibration. We identify the outer diameter of the pipe – known to be 9.5 mm – and count the number of pixels required to traverse it. Since the pixels are square, a voxel volume is simply the pixel width cubed.

TXPTV is subject to unique sources of uncertainty compared to visible light tomographic PTV. Particles are segmented in a reconstructed volume; that is, they are identified based on their voxel intensity. The particle volume centroid is taken to be its location. The particles and Taylor bubble are moving during the CT scan, though, which can result in a number of artifacts that may smear or warp the particle and bubble reconstruction. In order for the particles and bubble to appear static in the reconstruction, it is crucial that the scan time is much faster than the particle and bubble motion. If the scan time is too fast, though, the particles and bubble may be blurred in the projections by rotational motion. Balancing these effects is important for getting an accurate CT scan. [46] discusses some typical reconstruction artifacts for moving particles. Table 5.3 shows the expected maximum particle rotation blur and particle motion blur during scanning. These blurring phenomena create the motion artifacts in the reconstruction. We use the maximum expected velocities, the rotation speed, and the maximum particle radial distance to calculate the values in table 5.3. Based on these values, we would expect to see some blurring and warping. In the experiments, we find that a sufficient number of particles are not significantly blurred or warped, making particle tracking feasible.

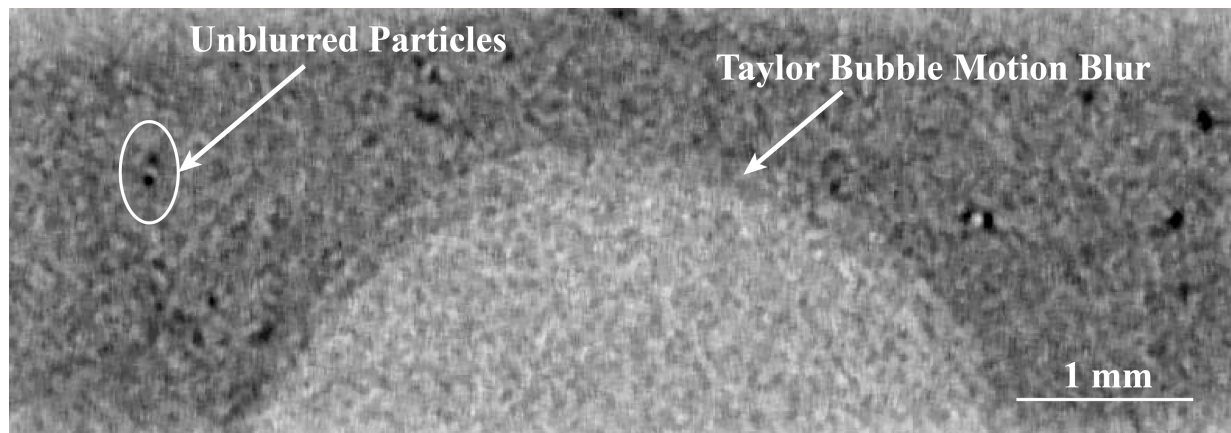


Figure 5.7: A $5 \times 5 \times 5$ median-filtered vertical slice through the central plane from a reconstructed Taylor bubble experiment. Only the pipe inner diameter is shown here for clarity. The Taylor bubble exhibits some motion blur manifested as a roughly $200 \mu\text{m}$ thick “halo” around the bubble, while in reality the interface is sharp. Fast moving particles may similarly warp, but many particles do not. There is a sufficient number of unwarped particles to successfully track them.

Figure 5.7 shows a vertical cross section through a Taylor bubble flow reconstruction. The bubble exhibits a motion blur artifact because it is rising too quickly for our scan speed. Although some particles may appear blurred, there are numerous particles that do not exhibit blurring and warping in the reconstruction, making tracking feasible.

In order to mitigate the error introduced by particle distortions (appendix of [46]), we introduced a radius match to the particle tracking cost function in order to make sure particle volumes of the same size are tracked as one particle. Particles that experience a large change in reconstructed volume are not tracked, mitigating error introduced from particle warping.

5.3 Results and Discussion

Poiseuille Pipe Flow

2D-projected Measurements of Poiseuille Pipe Flow

Figure 5.8 shows the DAVP as measured by CW tracer particles in a 45 mm/s center line speed flow ($Re = 0.40$). The data agree reasonably well with the analytical solution for the depth-averaged velocity profile. This profile is averaged over 850 frames to gather converged particle tracking statistics and assess the 2D-projected variance.

While 2D measurements from a single source-detector pair are of limited value, multiple source-detector pairs can triangulate the location of particles in 3D in what is known as stereo-XPTV (SXPTV). SXPTV would not require the rotation of source-detector pairs or

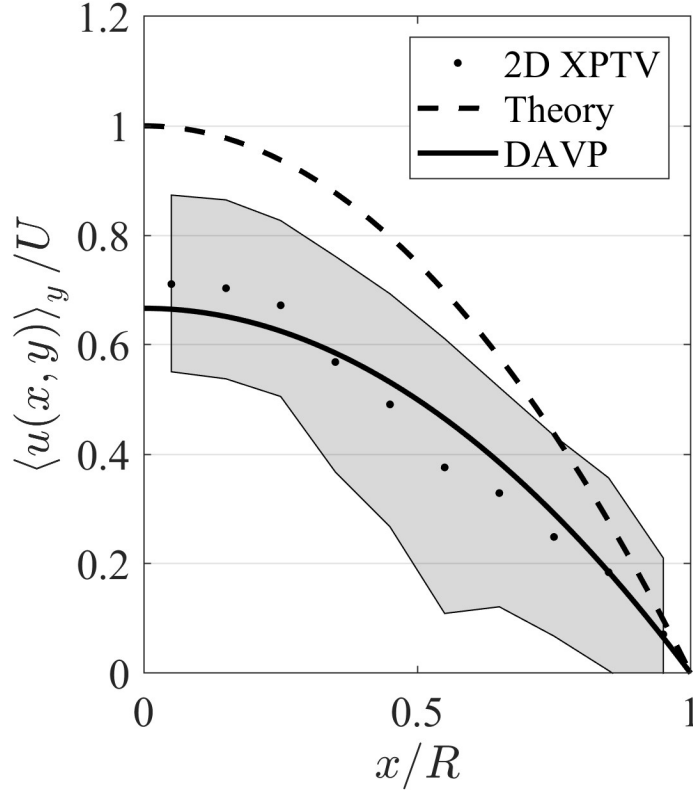


Figure 5.8: The depth-averaged velocity profile of a pipe flow with a 45 mm/s center line speed ($Re = 0.40$) agrees well with the expected profile. The gray region depicts the variation of the velocity measurement between the 16th and 84th percentiles to approximate the 2σ error bounds for a non-Gaussian-distributed variable. The data are normalized by the theoretical center line speed.

the flow experiment and the temporal resolution would only be limited by the detector frame rate. In other words, 1 kHz SXPTV is possible if multiple sources and detectors are used.

TXPTV of Poiseuille Pipe Flow

Figure 5.9 shows the average velocity profile from six reconstructions captured at 2 Hz. This rotation speed is $4.5\times$ faster than previously demonstrated in [46]. The measured profile shows good agreement with the theoretical Poiseuille pipe flow profile, demonstrating that TXPTV can accurately measure fluid flow. Although Poiseuille pipe flow is a simple, well-understood flow, it can serve as a useful benchmark for steadily improving XPV measurements. The relatively large spread in the data is likely due to particle motion artifacts in the reconstruction. [46] explains in detail the root cause of these artifacts. The relative rotation between the moving particles and the source-detector pair smears the particles in

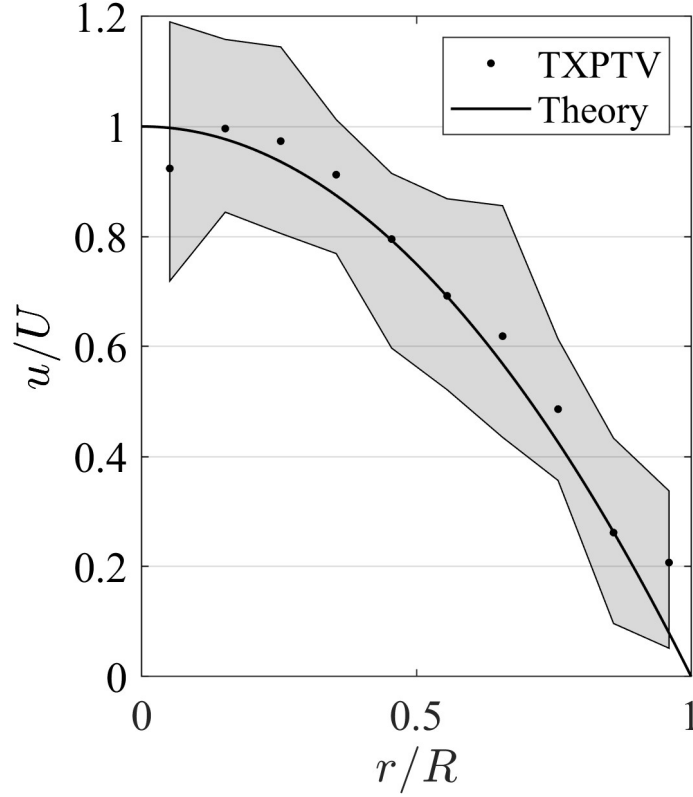


Figure 5.9: The TXPTV radial flow profile from a single scan shows good agreement with the theoretical Poiseuille profile. The 16th and 84th percentiles are shown to approximate 2σ error bounds for a non-Gaussian-distributed variable. Unlike 2D-projected XPTV, TXPTV can measure the true radial flow profile. The data are normalized by the theoretical center line speed, $U = 0.08\text{ mm/s}$.

the reconstruction, making it difficult to locate their true centroids. Reducing rotation time relative to the flow velocity reduces the impact of these artifacts.

TXPTV of Flow Around a Rising Taylor Bubble

One benefit of fast TXPTV is that it becomes possible to reconstruct the flow morphology in addition to capturing the flow velocity. For example, a full CT reconstruction of the Taylor bubble can be seen in figure 5.10 with the pathlines of tracked particles flowing around it. These data are captured at 3 Hz, which is $9\times$ faster than previously achieved by [46]. Furthermore, due to the higher frame rate we use here, we can capture 333 frames per rotation as opposed to the 200 frames per rotation used in [46]. More frames improves reconstruction spatial resolution and reduces noise, making particle detection more accurate and ultimately enabling the use of smaller particles.

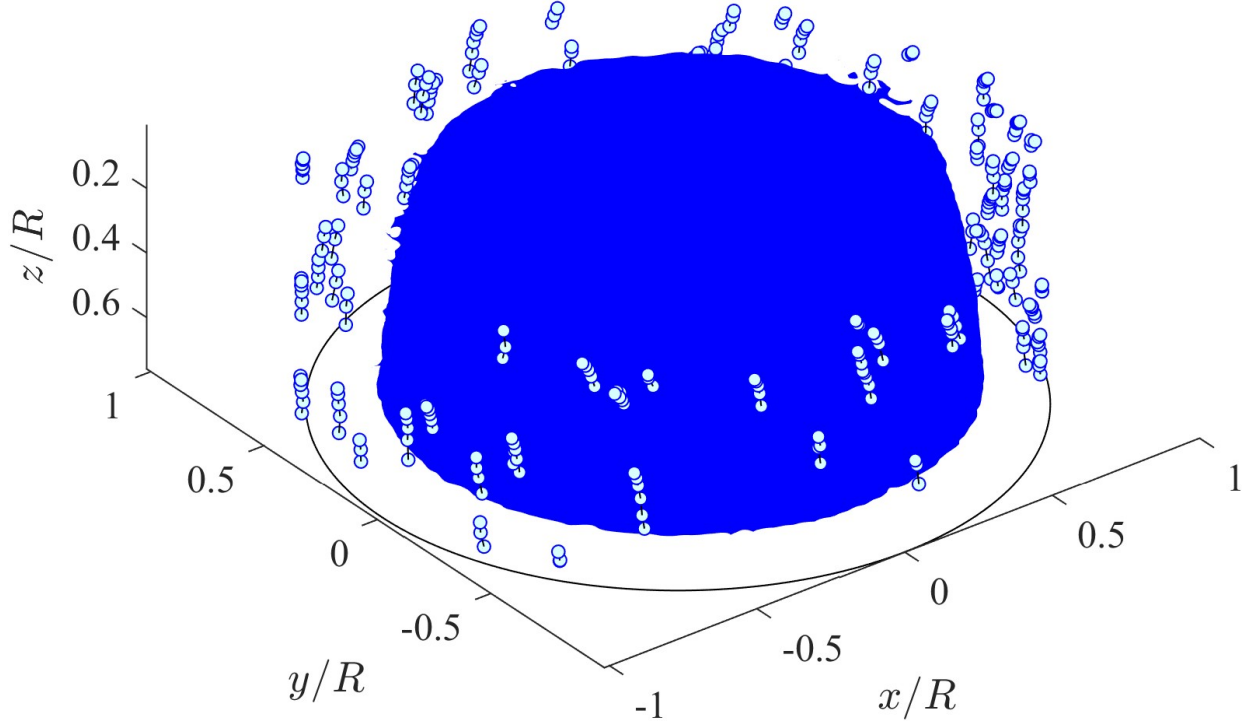


Figure 5.10: The particle pathlines flowing around the Taylor bubble moving through the field of view. The bubble-glycerine interface is shown in blue. A black circle on the bottom indicates the theoretical pipe interface.

A near order of magnitude improvement in the temporal resolution over the previous state of the art exemplifies the rapid development of TXPTV specifically and XPV in general. As new, brighter X-ray sources become available alongside faster detectors TXPTV will become increasingly useful as a technique for studying optically opaque fluid flows.

Scalar Mixing of a KI Jet

Figure 5.11 shows the evolution of the KI jet as it passes through the center slice of the reconstructed volume. The KI-water solution attenuates X-rays more than glycerine, so the KI jet appears darker. Initially, a more diffuse solution becomes visible; after 0.25 s the jet is more sharply visible. In fact, we can calculate the concentration of KI-water solution in glycerine by using the voxel intensity. According to the Beer-Lambert law for a mixture,

$$I = I_0 \exp \left(- \sum \sigma_i n_i(c) x_i \right) \quad (5.6)$$

the photon intensity I is a function of the source intensity I_0 , the attenuation cross section of material i , σ_i , the atomic number density n_i as a function of concentration c_i , and the

material thickness x_i . For a given material – in this case KI-water mixture – the attenuation cross-section and material thickness (i.e., voxel size) are constant. The source intensity is assumed to be effectively constant. Flickering, from either the source or the detector, are accounted for with a correction factor in post-processing. Thus, the detected photon intensity for a given voxel is a function only of the concentration in said voxel. Atomic number density is proportional to concentration, so we can calculate from equation 5.6 the concentration

$$c = A \ln(I) + B, \quad (5.7)$$

where A and B are constants that are determined by examining the voxel intensity with known concentrations. We can identify the voxel intensity at zero concentration by assuming no solution is in the first four reconstructions. Then, by assuming the solution is well mixed in the four final reconstructions, we can identify the intensity at the well mixed concentration. The well mixed concentration, based on a control volume of 1.9 mL and the injected volume in 4 sec of 2.48 mL, is 2.15 g/mL. The control volume here is taken to be the pipe length from the nozzle injection point to the viewing section of the pipe. Data is collected over 4 sec. We find the coefficients $A = -116$ and $B = 1216$. Figure 5.11 shows the concentration of the KI-water solution in the reconstruction as a function of time. As the jet develops in the field of view, the concentration of the solution increases. The qualitative data in each reconstruction is transformed into quantitative data in figure 5.11.

Figure 5.11 demonstrates the potential of high speed CT scans for measuring the time evolution of scalar mixing in 3D. Although not done here, these data could be combined with TXPTV measurements to simultaneously capture the evolution of fluid velocity and concentration fields.

5.4 Conclusions

Measurements of Poiseuille pipe flow, a rising Taylor bubble, and a laminar jet demonstrate the improved capabilities of in-lab high speed 2D XPTV, TXPTV, and CT thanks to the combination of a LMJ source, X-ray imaging-specific tracer particles, and a PCD. The 2D and 3D Poiseuille pipe flow measurements demonstrate the improved accuracy of the technique compared to our prior work. 2D-projected Poiseuille pipe flow data is acquired at 1 kHz, showing that $O(1\text{ kHz})$ acquisition rates are possible in the laboratory with the requisite hardware. The Taylor bubble and KI jet measurements demonstrate the applicability of TXPTV to multiphase flows and the ability of this technique to reconstruct the flow morphology in 3D. The laminar KI jet experiments demonstrate the potential for high speed CT to capture scalar mixing in 3D alongside 3D flow velocity measurements.

The measurements captured in this study are orders of magnitude faster than the previous state of the art thanks to the combined usage of a PCD, X-ray imaging-specific $O(50\text{ }\mu\text{m})$ tracer particles, and a LMJ X-ray source. This combination enabled imaging frame rates of 1 kHz, which is $15\times$ faster than previous comparable studies. Additionally, the bright LMJ source allows us to capture full 360 degree data for TXPTV reconstructions at nearly an

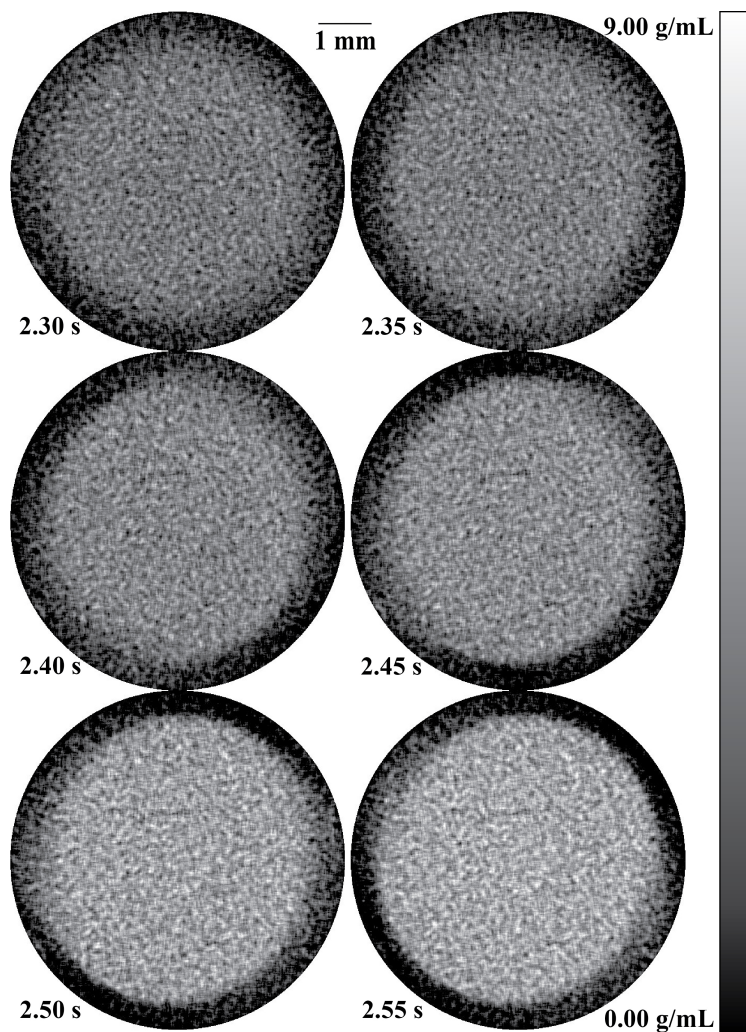


Figure 5.11: The evolution of the jet, but with the intensity values converted to KI-water solution concentration. The pipe wall is masked out for clarity. The reconstructed slices are Gaussian filtered with an $11 \times 11 \times 11$ kernel and a 7 pixel standard deviation.

order of magnitude higher speed than previous studies. These acquisition speeds bring more diverse fluid dynamic applications – from biological flows, to soil mechanics, to boiling flows – into the realm where in-lab XPV may be used.

Many of the limitations in this study are practical in nature, and are easily overcome. The rotation stage used here, for example, is limited to 1080 dps, meaning 3 Hz CT was the fastest achievable scan speed due to the rotation stage. Additionally, there is only a single, static source-detector pair. More source-detector pairs would improve the temporal resolution by requiring fewer angles to be traversed to capture a full CT scan. The detector used in this study also has a relatively small buffer size, which limited the number of frames that could be continuously acquired. This is to say that there is ample room for easy improvement.

A more involved improvement to the X-ray imaging setup could include implementing SXPTV, which uses multiple static source-detector pairs. For example, three source-imager pairs would enable particle or feature tracking at imaging frame rates, i.e., $O(1\text{ kHz})$. Such a system would theoretically only be temporally limited by the detector frame rate and source brightness. In this case, as demonstrated by the 2D-projected pipe flow measurements, that would allow 1 kHz SXPTV. Source and detector technology is rapidly improving, so future source-detector pairs will likely be able to go to even faster speeds. SXPTV is also possible with a single source-detector pair by using limited-angle reconstruction, although that is not attempted in this study and is an area for future work.

XPV is rapidly improving. Already these techniques could be useful for slow flows such as porous media flows and biological flows. The authors expect that the technique will see a dramatic increase in data acquisition speed over the coming years as PCDs become faster, sources become brighter, and newer, better X-ray tracer particles are developed. This study shows that the combination of these three elements yields orders of magnitude improvement in acquisition speed, thereby increasing the usefulness of XPV.

Chapter Supplementary Material: Image Processing Algorithms

2D Pipe Flow

For the 2D pipe flow experiments, the following algorithm was applied in LaVision DaVis version 8.4:

1. Apply a mask such that only the pipe inner diameter is visible.
2. Subtract the average intensity of all the frames from each frame.
3. Apply a strict sliding minimum filter with a scale length of 5 pixels.
4. Subtract a constant 0.01 from each frame.
5. Set all pixel values below zero to zero.

6. Apply a median filter with a 5×5 kernel size.
7. Multiply each frame by a constant 100.
8. Conduct PIV in two iterations:
 - a) One pass of 64×64 pixel window size with 50% overlap.
 - b) Two passes of 48×48 pixel window size with 50% overlap.
 - c) Apply vector removal based on pixel displacement with the following limits: $V_x \in [-5, 5]$, $V_y \in [-12, 0]$.
9. Conduct PTV with particle tracking assisted with PIV. The particle size range is 1 – 8 pixels; the intensity threshold is 0 counts; the correlation window size is 32 pixels; the allowed vector range relative to reference is 20 pixels.
10. Export to MATLAB to apply calibration and plot results. Average the profile over 850 frames since PTV requires statistical convergence to calculate a profile.

3D Pipe Flow and Taylor Bubble Flows

In the tomographic, 3D flow experiments, the following algorithm was used. First, for the CT reconstruction:

1. Convert flat field corrected images to integrated attenuation values by dividing by the empty field of view (FOV) pixel value.
2. Identify the center of rotation.
3. Use normalization ring artifact removal with a standard deviation of 15 pixels.
4. Use ASTRA Toolbox [62], [63] to reconstruct the volume. A standard Ram-Lak filter is used.

For the segmentation, MATLAB is used to segment the reconstructed data based on the voxel intensity in the following manner. For each reconstructed volume:

1. Normalize the reconstructed volume to grayscale.
2. 3D median filter the volume with a $5 \times 5 \times 5$ kernel and replicating boundary conditions.
3. Binarize the volume using `imbinarize`. The threshold is adaptive, the sensitivity is 55%, and the foreground polarity is dark.
4. Invert the binarized image.
5. Use `conncomp` to identify 6-connected components volumes.

6. Use regionprops3 to measure the volume and centroid location of the volumes. Weight the centroid location with voxel intensity from the normalized reconstructed volume.

For particle tracking, a MATLAB code was developed based on a nearest-neighbor algorithm. The code will be publicly available at the FLOW Lab website: flow.berkeley.edu.

KI Jet Processing

The KI jet is captured with high speed CT. These scans were reconstructed using ASTRA Toolbox [62]. Each reconstruction consisted of 250 projections. For better temporal interrogation, we reconstruct each volume with a 90% overlap in the projections used. For example, the first reconstruction is done with projections 1 to 250; the second reconstruction is done with projections 25 to 275, and so on.

Post-reconstruction, MATLAB is used to correct the source flicker and calculate the solution concentration. The algorithm is as follows:

1. The average intensity of the middle slice of each volume is normalized by the average intensity of the middle slices for all volumes. This provides the flicker correction factor.
2. Each volume is divided by the correction factor, then 3D Gaussian-filtered with an $11 \times 11 \times 11$ kernel with a standard deviation of 7 pixels.
3. The first five and the final five volumes are averaged to calculate the zero concentration and fully mix concentration voxel intensities. The average inner pipe voxel intensity is taken to be the intensity for its respective concentration. The coefficients in equation 5.7 are then calculated.
4. Equation 5.7 is applied to each Gaussian-filtered volume to calculate the concentration in each voxel.

Chapter 6

Direct Comparison of Isobaric and Isochoric Vitrification of Two Aqueous Solutions

X-ray image-based measurement techniques have novel applications outside of fluid dynamics. Thermodynamic systems offer unique opportunities and challenges. Many thermodynamic phases are difficult to distinguish visually. In particular, many materials look the same when frozen or in liquid form. Some thermodynamic state variables, such as concentration, are difficult to measure non-invasively. For systems under extreme conditions such as high pressure, low or high temperature, or some combination thereof, measuring concentration and ascertaining phase can be difficult or impossible.

As discussed in previous chapters, X-ray measurements can prove useful in such conditions. X-ray imaging is able to discern density differences, concentrations, material identification, and more. This chapter introduces a use case for X-ray image-based measurements that demonstrate their value in fields such as cryopreservation and thermodynamic state measurements.

This chapter presents ongoing work in collaboration with Anthony Consiglio and Prof. Boris Rubinsky in the Bio-Thermal Laboratory.

6.1 Background

The ability to preserve biological systems such as cells, plants, organs, and even entire organisms for indefinite periods of time has remained an elusive goal of biomedical and conservation communities. Traditional clinical methods only enable the storage of organs for a few hours [64]. Preserving a biological system outside of its normal environment requires either 1) maintaining normal physiological processes that support metabolic activity [65], [66] or 2) suppressing metabolic processes to prevent deterioration. Metabolism, which represents the sum of enzymatic reactions that sustain life, is largely an Arrhenius process,

and as such, is a strong function of temperature. At cryogenic temperatures, molecular processes are arrested to such an extent as to bring about a state of suspended animation.

The greatest challenge facing successful cryopreservation is the avoidance of ice since it is nearly universal lethal to biological systems by mechanical, chemical, and osmotic mechanisms [67]. Vitrification involves the solidification of a liquid not by crystallization but rather to a glassy state by an increase in viscosity that results in arrested molecular motion and suppressed ice nucleation. The nucleation rate of ice in aqueous solutions is a steep function of supercooling, and cooling rates exceeding 10^6 °C/s are required to vitrify water [68], [69]. The addition of solutes known as cryoprotective agents (CPAs) and the application of pressure affect both equilibrium and non-equilibrium freezing processes and lower the cooling rate needed to achieve glass formation [70].

Recently, isochoric (constant volume) confinement has been found to stabilize supercooled water against ice nucleation by providing isolation from external perturbations, removing exposed liquid-air interfaces, and potentially imparting an energetic toll upon incipient ice embryos as they expand relative to the liquid phase [71]–[74]. Isochoric confinement can also alter ice growth dynamics since the formation of ice in a confined volume acts to self-pressurize the system, which shifts the phase equilibrium and can result in arrested crystallization dynamics and multi-phase solid-liquid equilibrium [75]. In light of these findings, isochoric confinement has received attention for its potential to promote less ice in vitrification. This interest is highlighted by two recent studies. The first by Powell-Palm et al. [76] employed an isochoric system to successfully vitrify and revive centimeter-scale coral specimens. The second, a computational study by Solanki and Rabin [77], proposed that contraction of the aqueous solution when cooled in liquid nitrogen would generate vapor cavities and nullify the initial isochoric confinement.

In this chapter, we present photon counting X-ray computed tomography (CT) measurements of two vitrified aqueous solutions under isochoric and isobaric conditions. X-ray CT with conventional X-ray detectors has been used to study CPA perfusion and vitrification in the past [78]. The experiments presented here use a laboratory-scale X-ray system to resolve phase segregation inside rigid metallic chambers at 98 μ m projection spatial resolution. We find that isochoric systems do not usefully promote vitrification relative to isobaric systems, observing nearly equivalent vitrification outcomes when controlling for all other parameters. The formation of vapor cavities is observed, and when cooling is not fast enough to achieve total vitrification, nearly identical volume fractions partial ice crystallization are found to occur under both isochoric and isobaric confinement.

6.2 Methods

Vitrification Solutions

The first solution is a commonly used binary glass-forming aqueous solution consisting of 50 wt% dimethyl sulphoxide (DMSO) in deionized water. This solution has a well charac-

terized and easily achievable critical cooling rate of $< 5^{\circ}\text{C}/\text{min}$ [79], which enables us to isolate the effects of isochoric confinement on solution contraction and evaluate the findings of Solanki and Rabin [77], [80]. The second solution, known as CVS1, was used in isochoric systems in the aforementioned Powell-Palm et al. study [76] to cryopreserve whole coral specimens and is selected in order to provide insights into this promising cryopreservation result. The CVS1 solution consists of molarity 1.05M DMSO, 1.05M propanediol, 1.05M glycerol, and 0.85M trehalose in physiological buffered saline (PBS). Since CVS1 is comprised of multiple traditional CPAs as well as a disaccharide, the critical cooling rate is difficult to estimate *a priori*. Previous studies have not characterized the critical cooling rate of CVS1. However, cooling at $100^{\circ}\text{C}/\text{min}$ was found in [76] to yield positive outcomes during the coral cryopreservation. The experiments presented herein aim to investigate vitrification processes in this newly employed solution.

Vitrification Chambers

Figure 6.1 depicts the three systems employed in this study. The main chamber body, constructed of aluminum-7075, has an inner diameter of 12.7 mm (0.5"), an outer diameter of 34.9 mm (1-3/8"), and a nominal internal volume of 5.3 ml. Due to thermal contraction, the internal chamber volume contracts by approximately 1.5% during cooling in liquid nitrogen. We measure the volume of the chamber after contraction using X-ray CT as described below. An aluminum threaded plug with a metal-on-metal tapered sealing surface provides rigid confinement for the isochoric trials, as shown on the left in figure 6.1. In order to study the solutions under the same thermal conditions, but without rigid (i.e., isobaric) confinement, we introduce two additional configurations. The first configuration (figure 6.1 center) employs an aluminum piston that replicates the thermal and surface boundary condition of the isochoric plug yet is able to move freely in the vertical direction in order to produce isobaric conditions. In order to isolate the effect of isochoric confinement on vitrification success it is critical to match thermal conditions between the two configurations. To ensure that no air is trapped underneath the piston, the face of the piston or plug is wetted with the solution prior to inserting it into the chamber. In the second isobaric configuration (figure 6.1 right) the liquid interface is open to air and the chamber is covered with Kapton tape to prevent the intrusion of liquid nitrogen during cooling. This configuration allows the liquid interface to freely deform and has been employed by previous studies of thermo-mechanical processes during vitrification [80], [81]. We use this configuration to study the formation of a vapor cavity. In preliminary trials this configuration resulted in a lower cooling rate at the top of the chamber compared to the isochoric condition, motivating the introduction of the piston configuration to match the boundary conditions for the high critical cooling rate CVS1 solution.

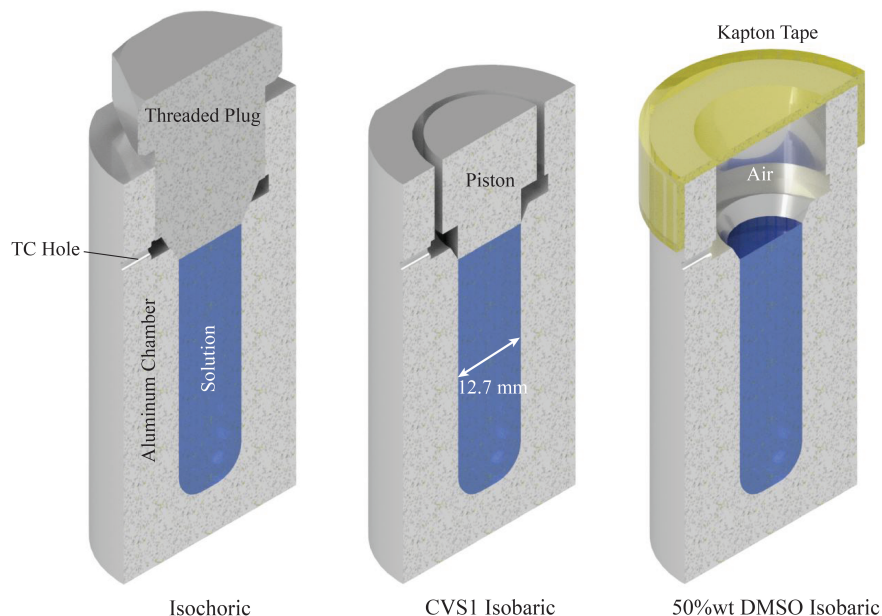


Figure 6.1: Vitrification chambers enabling control of boundary conditions. A two-piece assembly with metal-on-metal sealing surfaces generates the isochoric (rigid) confinement. Replacing the rigid plug with a movable piston produces isobaric conditions while ensuring equivalent thermal and surface conditions as the isochoric system. This configuration was employed in the trials with CVS1. Critical cooling rates of $\leq 5^\circ\text{C}/\text{min}$ are easily achievable with 50 wt% DMSO, and the solution is left uncovered in order to allow the free interface to deform. In this configuration, the chamber is capped with Kapton tape to prevent intrusion of liquid nitrogen during cooling.

Vitrification Procedure

In order to facilitate rapid cooling, we lower the chambers to the bottom of a liquid nitrogen bath in a vertical orientation. The temperature is monitored during this process with a type-T thermocouple inserted into the wall of the chamber, as indicated in figure 6.1, and recorded with an Omega TC-08 thermocouple data logger ($\pm 1^\circ\text{C}$ uncertainty). Due to the large size and thermal mass of the systems, thermal gradients are present during the plunge cooling, and we utilize this temperature measurement as a rough indicator of a chamber's thermal history. The average cooling rate is computed for each trial from the recorded temperature measurement and is reported in table 6.1. We conduct three repetitions for each condition in order to account for potential variability in assembly condition and cooling profile.

Table 6.1: Average cooling rates ($^{\circ}\text{C}/\text{min}$) during plunge cooling in liquid nitrogen of CVS1 and 50 wt% DMSO under isochoric and isobaric conditions.

	CVS1		50 wt% DMSO	
	Isochoric	Isobaric	Isochoric	Isobaric
Trial 1	120	108	112	109
Trial 2	106	117	116	120
Trial 3	100	104	110	127
Average	110	109	113	119

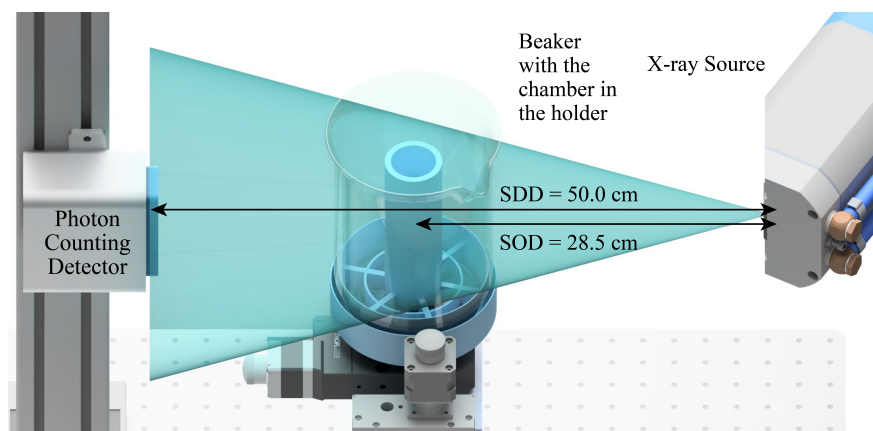


Figure 6.2: The experimental setup consisting of a cone beam X-ray CT source and a photon counting detector. A vitrified chamber is placed into the beaker that sits atop a stack of three linear stages and one rotation stage. The beaker is partially filled with liquid nitrogen below the sample to ensure the chamber does not warm over the duration of the CT scan.

Photon Counting X-ray Computed Tomography

Figure 6.2 depicts the X-ray CT setup, which consists of an YXLON FXE225.48 X-ray source used in concert with a Dectris Pilatus3 100K-M photon counting detector. Photon counting detectors (PCDs) achieve a higher signal-to-noise ratio than conventional X-ray detectors [13], making them advantageous for collecting CT scans of systems with low X-ray contrast such as multiphase vitrified systems. Lower energy photons, which are more likely the result of scatter and thus introduce noise, can also be rejected with PCDs. The energy threshold for these experiments is 20 keV. We collect 114 projections per CT scan, each with a 9.99905 s integration time and a 0.95 ms dead time for a total imaging period of 10 s. The rotation rate is constant at 0.316 degrees per second.

The X-ray source emits a polychromatic cone beam, which produces geometric magnification based on the positions of the object and the detector relative to the source. The

magnification factor, M , is the ratio between the source-to-detector distance (SDD) and the source-to-object distance (SOD). Based on a magnification factor of $M = 1.75\times$ and a pixel size of $172\text{ }\mu\text{m}$ square, we achieve a spatial resolution of $98\text{ }\mu\text{m}$ square for a single projection. Gaining higher spatial resolution sacrifices the field of view, however, reducing the nominal detector area of 8.38 cm high by 3.35 cm long to an effective field of view measuring 4.79 cm high by 1.91 cm long.

We operate the X-ray source at an acceleration voltage of 80 kVp and a target current of 0.8 mA . Based on the manufacturer’s specifications, the focal spot for the source at these settings is approximately $87.5\text{ }\mu\text{m}$ full-width half-max. With geometric magnification, we expect the focal blurring to be approximately $153\text{ }\mu\text{m}$. This is less than the size of a single pixel, so we do not expect notable focal blurring artifacts. Further details on the X-ray imaging setup we use can be found in [23] and [57].

Phase Identification

We use the filtered back projection algorithm in the ASTRA Toolbox [62], [63] to reconstruct the CT scans, then we segment the phases in ORS Dragonfly [82] based on their voxel intensity to calculate their respective volumes and assess morphology. A 3D median filter with a span of 5 voxels is used to enhance the contrast between ice and the vitrified solution. We do not filter the vapor cavity scans because there is sufficient contrast. Figure 6.3 depicts the voxel intensity histograms for the entire reconstructed volume of CVS1 and 50 wt% DMSO under isochoric and isobaric confinement, respectively. We use these as examples to demonstrate the contrast between ice, vapor, and vitrified solution. Multiple peaks are visible. Starting from the left: the first is the aluminum chamber walls; the second is the vitrified solution and ice; the third is a vapor cavity, which is only present in the 50 wt% DMSO solution. The difference in the 50 %wt DMSO and CVS1 vitrified solution peaks is due to the higher X-ray attenuation of the DMSO solution compared to CVS1 across the range of photon energies (see Supplemental Information). The ice voxels in the CVS1 solution are hidden within the vitrified solution peak. Figure 6.4 depicts the histogram of a single unfiltered slice that contains ice in a CVS1 isochoric scan, which reveals the distinction between ice and the surrounding vitrified solution. Each phase is segmented from the others by establishing thresholds at the local minima between peaks.

6.3 Results and Discussion

Vapor Cavity Formation

The 50 wt% DMSO solution is easily vitrified at $\leq 5^\circ\text{C}/\text{min}$ and is selected to investigate thermal contraction behavior. Figure 6.5 depicts representative 3D reconstructions of the vitrified 50 wt% DMSO solution under isochoric and isobaric conditions. The same contracting taper morphology is observed in both confinements. Average vapor volume fractions of

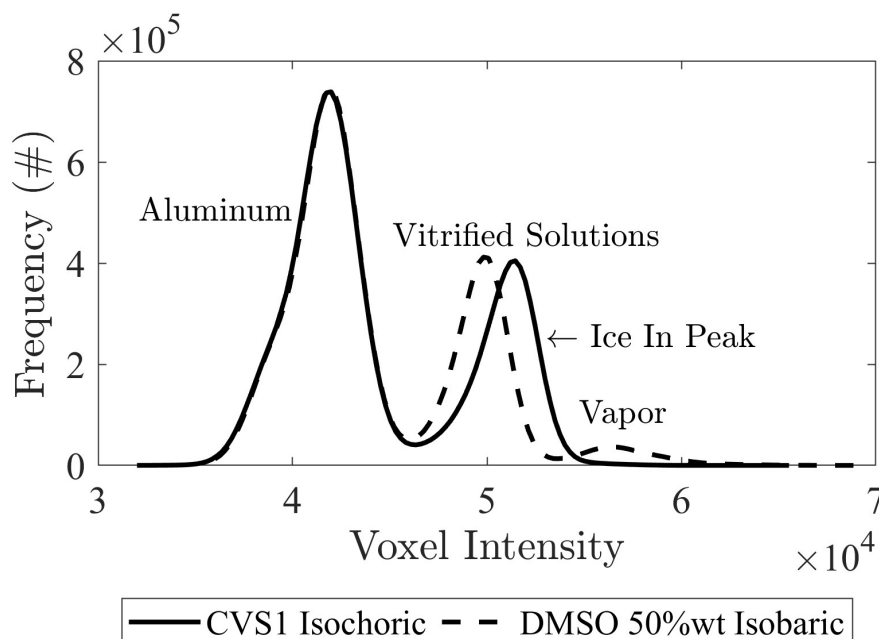


Figure 6.3: CT scan voxel intensity histograms for entire reconstructed volume of CVS1 and 50 wt% DMSO vitrified under isochoric and isobaric conditions, respectively. These examples show the contrast between the aluminum walls, the vitrified solution, and the vapor. The histograms are aligned based on the aluminum peaks.

5.7% and 6.7% are computed for the isochoric and isobaric conditions, respectively. Table 6.2 reports the volume fractions of vapor from each trial. As predicted by the recent computational study by Solanki and Rabin [77], [80], vapor cavities are observed to form during cooling of DMSO solutions in the isochoric chamber. This occurs due to contraction of the fluid, which causes the pressure to drop below the saturation pressure (and possibly even below the cavitation limit) and results in the rejection of dissolved gases or vapor cavitation. The potential presence of these phenomena is discussed below.

Notably, convective flows generated from heat transfer and surface deformation in isochoric confinement are a potential application of the TXPV methods discussed in previous chapters. Rabin [80] numerically models the velocity field in a similar isochoric system to the one studied here, providing an interesting comparison point for experiments. The velocities predicted in that study may be within the measurable range for in-lab TXPV.

Ice Formation

Due to its lower molarity, CVS1 has a higher critical cooling rate and lower coefficient of thermal expansion than the 50 wt% DMSO solution. It is therefore well suited for investigating the potentially beneficial effects of isochoric confinement on vitrification. Figure

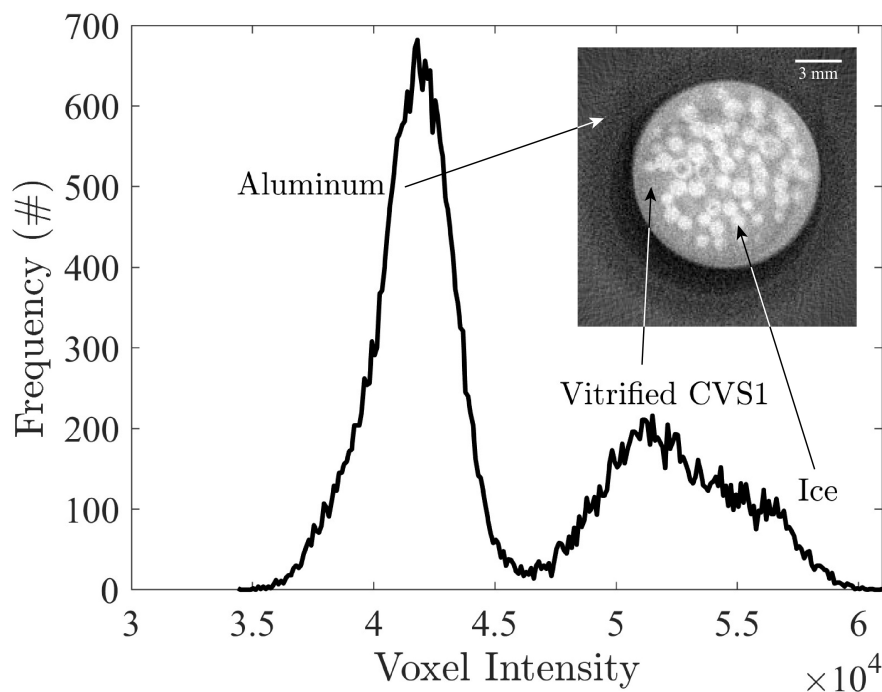


Figure 6.4: CT scan voxel intensity histogram for a single unfiltered reconstructed slice of the CSV1 solution vitrified under isochoric confinement. Peaks are visible for the three present phases: aluminum, vitrified CVS1, and ice. Inset: the unfiltered reconstructed slice showing globular ice formation.

6.6 shows a representative 3D reconstructions of the ice formed under isochoric and isobaric confinement. Despite the isochoric configuration working for the vitrification and revival of coral fragments [76], we find that isochoric and isobaric conditions yield similar amounts of ice (on average 0.56% and 0.78%, respectively) during rapid cooling in liquid nitrogen. The volume fractions of ice formed in each trial are reported in Table 6.2. As discussed in the introduction, one advantage of isochoric vitrification would be if it suppresses ice growth compared to isobaric vitrification. We do not find a useful difference in this respect, however, given this chamber design, this solution, and this cooling rate. The observed volumes of ice could both prove fatal to biological specimens.

Although we find nearly equivalent ice volume fractions, we do observe morphological differences in the ice formed in CVS1 under isochoric and isobaric conditions. Figure 6.7 depicts close up reconstructions of the segmented ice phases in CVS1 for both conditions. The ice formed near the top surface in both conditions. Whereas the ice in the isobaric system formed one continuous phase, the ice in the isochoric system formed multiple millimeter-scale globular crystals. This difference could be the result of outgassing- or cavitation-induced ice nucleation [83], [84]. Under isochoric confinement the pressure in the chamber drops as the solution contracts during cooling. As the pressure drops below the equilibrium vapor

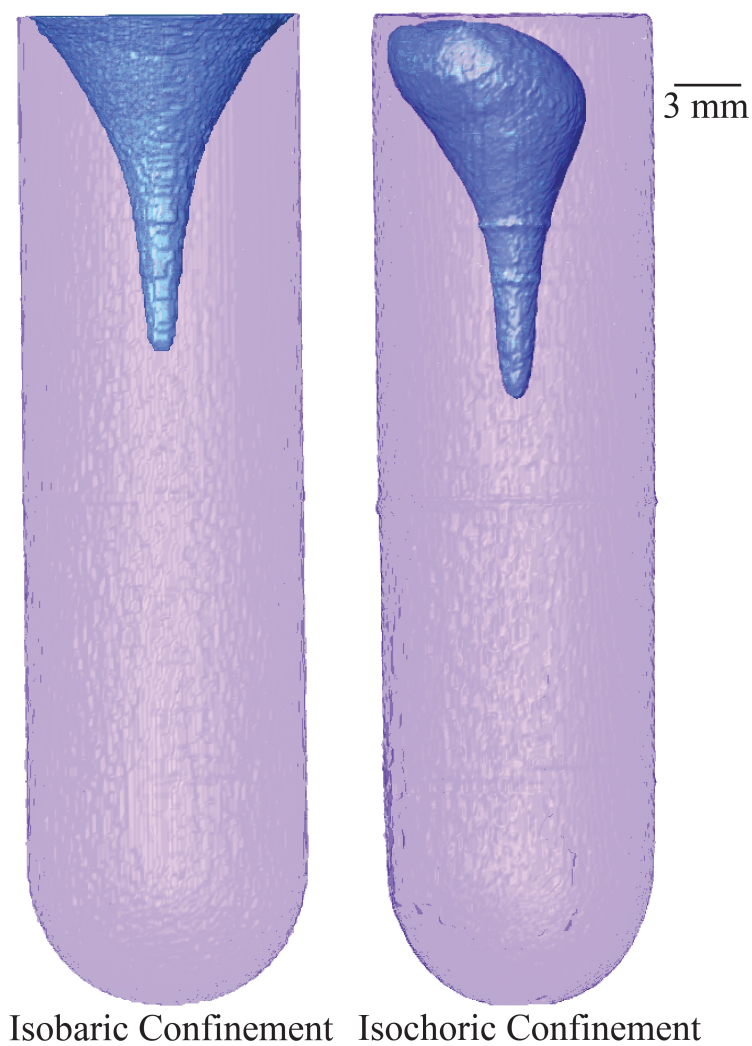


Figure 6.5: Reconstructed CT scans of 50 wt% DMSO in isobaric (left) and isochoric (right) systems showing vapor cavity formation. Vitrified solution and vapor phases are segmented and shown in purple and blue, respectively.

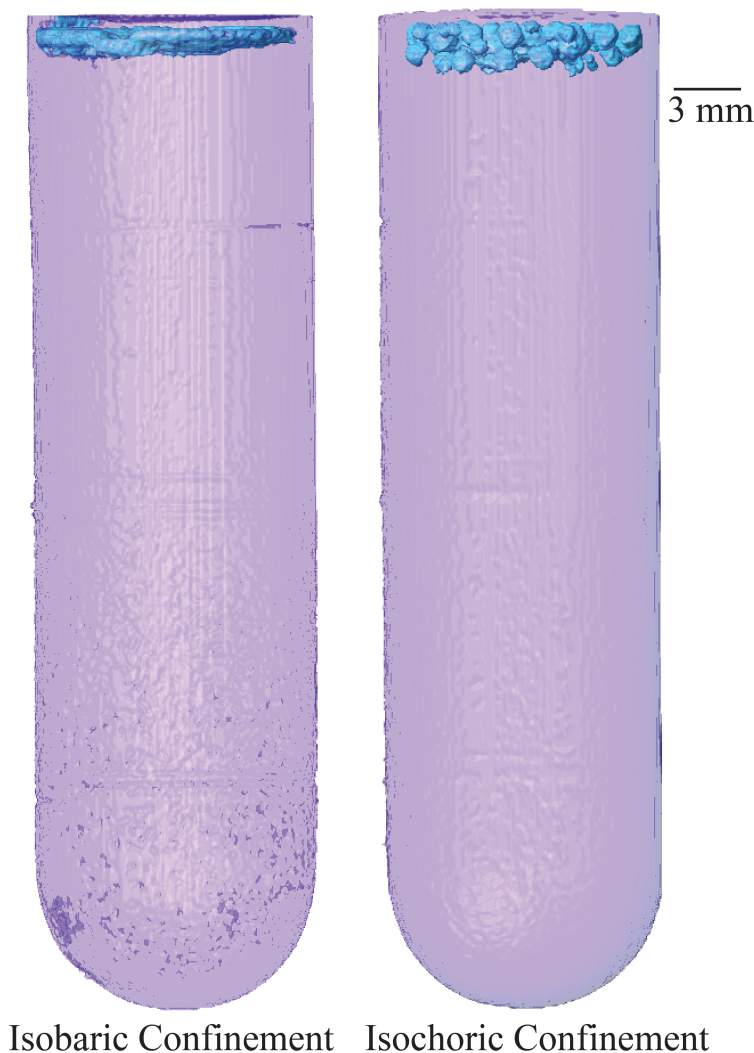


Figure 6.6: Reconstructed CT scans of CVS1 in isobaric (left) and isochoric (right) systems showing partial ice formation. Vitrified solution and ice phases are segmented and shown in purple and blue, respectively.

pressure dissolved gases may be forced out of solution or the solution may cavitate.

If outgassing is occurring, the morphology of rising bubbles in a static fluid is a well understood function of the dimensionless Eotvos and Reynolds numbers [85]. In the case of vitrification, however, the viscosity of the fluid and the surface tension of the gas are changing rapidly with temperature and pressure, making these parameters difficult to estimate. Determining the occurrence of cavitation is also challenging and would require $O(1 \text{ MHz})$ frame rate measurements. Isochoric vitrification occurs in a thick-walled metal chamber, necessitating long exposure periods with in-lab X-ray sources. Cryogenic temperatures in a

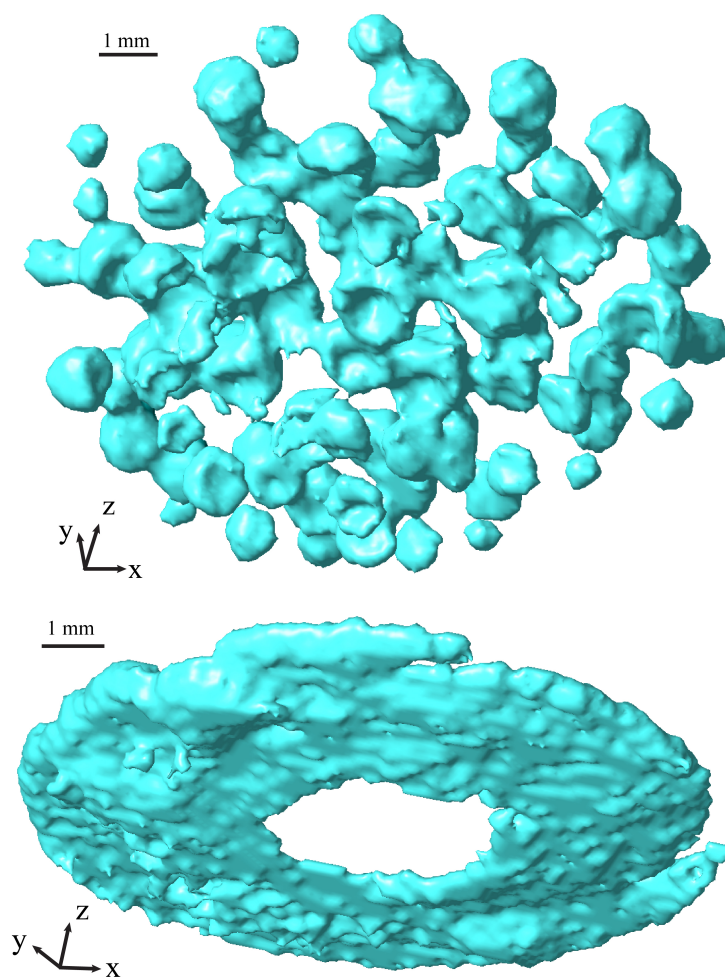


Figure 6.7: Segmented ice phases from reconstructed CT scans of vitrified CVS1 in isochoric (top) and isobaric (bottom). Globular ice formation in the isochoric system is reminiscent of bubbles formed during vapor cavitation.

Table 6.2: Ice volume fractions (%) of ice for the CVS1 and vapor volume fractions (%) for 50 wt% DMSO under isochoric and isobaric conditions.

	CVS1		50 wt% DMSO	
	Isochoric	Isobaric	Isochoric	Isobaric
Trial 1	0.33	0.53	5.8	5.2
Trial 2	0.44	0.75	5.5	8.9
Trial 3	0.9	1.07	5.9	6.1
Average	0.56	0.78	5.7	6.7

boiling nitrogen bath hinder the use of acoustic inspection or accelerometers. High-speed synchrotron X-ray imaging is potentially well-suited for determining the presence of either cavitation or outgassing during vitrification in isochorically confined systems.

The isochoric system employed in this study closely emulates Berthelot tube systems used to study liquids at negative pressure [86]. In such studies, rigid containers are filled with a liquid and cooled, and in the absence of pre-existing bubbles or cavitation, the fluid can be metastably tensioned. At negative pressures and temperatures below 0°C, water is metastable with respect to both ice and vapor [87]. In order to maintain negative pressures, however, these systems must be pre-conditioned by initially heating the sealed container in order to pressurize the liquid and force stubborn vapor trapped in crevices – which can act as a nucleation seed – into solution. Since this pre-conditioning would certainly injure any contained biological material, we did not perform this step in our experiments. Thus, only mild tension is expected to be possible.

These findings also suggest that the critical cooling rate of CVS1 is higher than reported in Powell-Palm et al. [76]. Powell-Palm et al. report the critical cooling rate to be approximately 100°C/min, but at roughly 110°C/min we observe ice formation. That study uses pressure measurements to detect the formation of ice and thus determine if full vitrification occurs or not. We observe ice in identical chambers with similar cooling rates, indicating that pressure measurements may not be sufficient for determining full vitrification.

6.4 Conclusions

Using a laboratory-scale X-ray CT system, we compare vitrification outcomes in two aqueous solutions, CVS1 and 50 wt% DMSO, under isochoric and isobaric conditions. We find that isochoric confinement yields similar volumes of ice and vapor compared to the isobaric configuration. Despite nearly equivalent outcomes for the two confinement configurations in practical terms, we observe that isochoric vitrification of CVS1 produces a unique ice morphology that could be the result of outgassing or cavitation.

By studying two solutions of different thermal contraction and controlling for all other

experimental parameters, we find that isochoric confinement does not usefully promote complete vitrification compared to isobaric confinement under the conditions tested. Observing that higher cooling rates throughout the system are the largest determinant of vitrification outcomes, thinner-walled containers than are achievable with isochoric confinement may be preferred. Our findings also suggest that measurement of pressure may not be a sufficient indicator of vitrification. Thermal contraction of the fluid causes vapor cavities to form through either degassing or cavitation, which can be displaced by ice if the local cooling rate is below the critical rate.

Despite these findings, isochoric systems may yet prove useful for vitrification cryopreservation when ice growth is permitted in portions of the chamber separated from the preserved biomaterial. In this scenario, controlled ice growth would pressurize the system and the rejection of solutes from the growing ice phase would increase the concentration of the unfrozen solution, further modulating its melting point and passively diffusing additional solutes into the biomaterial [88], [89]. The combined effects of higher pressure and increased solute concentration would increase the likelihood of successful vitrification in these portions of the systems, if protected from an advancing ice front.

Lastly, photon counting X-ray CT has proven to be a promising approach for the study of vitrification in aqueous systems. The laboratory-scale system employed in this study enables us to resolve phase morphology at projection resolutions of 98 μm . This resolution scales linearly with magnification factor and detector resolution. The current setup at $M = 10\times$ would yield a resolution of 17.2 μm , for example. Volumes of ice at this scale are below the detectable limits possible with differential scanning calorimetry [90], as well as most visual [91] and X-ray diffraction [79] methods. Furthermore, assuming ideal conditions where phase segmentation is possible from single voxels, we can place a theoretical lower detection limit to volume fraction detection threshold of $O(10^{-8})$ for this detector. Detectors with more pixels (and therefore more reconstructed voxels) would have lower limits. Furthermore, PCDs are energy-reesolving detectors, meaning that they can measure the energy deposited by incoming photons. This feature allows one to use photon-counting X-ray CT to measure the distribution of arbitrary numbers of chemical species in 3D. In other words, one could use photon counting X-ray CT to observe the distribution and transport of CPAs throughout a vitrifying system and biological specimen. This technique could thus prove valuable for investigating vitrification and ice nucleation kinetics in the future.

Chapter 7

Conclusions and Future Steps

7.1 X-ray Particle Velocimetry

X-ray particle velocimetry (XPV) has undergone rapid advancement over the last five years. In the coming five years it will continue to do so. There remain a number of practical and technical challenges that must be overcome, however. In order for XPV to gain wider adoption in the fluid dynamics community it will have to become more accurate, faster, and more easily accessible. I will discuss each of these challenges in turn.

Making XPV More Accurate

Like any particle-based velocimetry technique, the accuracy of XPV is dependent on the hardware used to capture the data. Hardware that can achieve faster collection speeds is important and will be discussed subsequently. Equally important, though, are the tracer particles and the detector.

Tracer Particles

Chapter 4 discussed the benefits of tracer particles designed specifically for XPV. The particles in that study are $O(50 \mu\text{m})$. While small enough to study many flows, faster flows or flows in small channels are still inaccessible to XPV. Visible light particle image and particle tracking velocimetry (PIV and PTV, respectively) can use particles that have sub-micron diameters. Furthermore, there are a plethora of tracer particles available to the visible light techniques, including bubbles, droplets, plastic particles, glass particles, hollow particles, solid particles, and coated particles. Each type of tracer is best suited for certain applications. The equivalent toolbox for XPV does not yet exist.

In large part, the tracer toolbox does not exist for XPV because there are not many users with an accompanying wide array of applications. In addition, tracer particles for XPV face unique design constraints that visible light PIV and PTV do not. Chapter 3 discusses these constraints in detail, but the primary constraint is generating contrast in X-

ray images without making a particle that is too heavy or too light to be a good tracer. The design space for new XPV tracer particles is large and open for innovation. Most XPV tracer particles, for example, have focused on attenuating *more* X-ray light than the surrounding medium. XPV works equally as well when the particle attenuates *less* than the surrounding medium.

XPV tracer particles can also be X-ray source-specific. Different X-ray sources have different spectra, so some photon energies may be more common than others. XPV tracer particles can be uniquely designed to most (or least) attenuate those photon energies, thereby generating the most contrast. Alternatively, X-ray source settings can be adjusted to best suit the tracer particle being used. A tracer particle could, for example, be coated in silver. Silver has a K-edge at 25.5 keV. Such a particle could be well-suited to an X-ray source whose most common photon energy is 25.5 keV because it would be relatively highly attenuated.

Tracer particles can also make use of the special characteristics of X-rays. For example, in many multiphase flows it could be difficult to find a single particle design that creates strong contrast and is an accurate flow tracers in both phases. Using multiple particle designs would be advantageous in that case. Both types of particles would end up in both phases (either by happenstance or design) and could introduce error into the measurement. How, then, to distinguish the particles in each phase to ensure that only those particles designed for that phase are being used for the measurement? Take the silver-coated particle again. Its K-edge photon energy is unique because it is a function of its atomic structure, as discussed in chapter 2. The silver-coated particle could be used to trace the flow in one phase, while another particle coated in a material with a different, distinct K-edge could be used for the other phase. By using a multi-threshold energy-resolving photon-counting detector (PCD), one could distinguish these two particles with K-edge detection. Using PCDs in novel ways for fluid dynamics will be discussed in the following section.

Fundamentally, there needs to be a wider array of tracer particles available for XPV in order for more people to use the technique. Most users are uninterested or incapable of designing and manufacturing their own tracer particles. Using simulations such as those in chapter 3 can help design new tracer particles for specific applications faster. New XPV tracer particles must be smaller, generate good contrast across more applications, and, of course, accurately trace the fluid flow.

X-ray Detectors

X-ray detectors are constantly improving, particularly PCDs. Using PCDs in laboratory-scale XPV is attractive for reasons that I have mentioned throughout the dissertation. PCDs are energy-resolving, so it becomes possible to simultaneously distinguish materials and capture fluid velocity, a powerful combination with many applications. As chapter 2 discusses, PCDs can also achieve a higher signal-to-noise ratio than more traditional CCD, CMOS, and scintillating detectors.

In order to fulfill their potential, PCDs need to improve their spatial resolution. Many PCDs can now capture at rates above 1 kHz, enough to study a number of exciting fluid

flows, yet the spatial resolution is the limiting factor. Pixel sizes for PCDs are $O(100\text{ }\mu\text{m})$, far larger than visible light cameras. Optical components for X-rays are exceedingly rare currently, so X-ray imaging has to rely on geometric magnification in order to improve the effective spatial resolution. This high spatial resolution comes at the cost of a narrower field of view, though.

The fundamental limitation for PCDs is charge sharing, wherein the electric charge created by an incoming photon is shared across multiple pixels. Smaller pixels and a thicker detection plate – both desirable design outcomes for PCDs – increase charge sharing amongst pixels. PCD developers are working to reduce charge sharing and thereby decrease pixel size and increase quantum detection efficiency. New materials, better correction algorithms, and more are being explored to reduce charge sharing.

In the coming years, PCDs should continue to see rapid improvement. Already, there are indications that the next generation of PCDs will be able to capture photons at an order of magnitude higher count rate while simultaneously having a larger field of view and more energy thresholds. As the spatial resolution limitations disappear, PCDs may be the future of in-lab XPV because of their unique energy-resolving capabilities and high signal-to-noise ratio. Mixing flows, multiphase flows, chemical reaction flows, and other active media flows are all interesting areas of exploration that could make use of PCDs to capture measurements that would otherwise be difficult or impossible.

Making XPV Faster

Related, but separate from, making XPV more accurate is making XPV faster. Higher collection speeds improves velocity measurement accuracy and broadens the range of fluid flows that are possible to study. There is more to making XPV faster than simply improving detector frame rates, however.

As discussed in chapters 4 and 5, X-ray images are two projections, i.e. shadowgraphs. Unlike visible light PIV and PTV images, X-ray images do not capture slices of a fluid flow. Instead, methods such as tomographic and stereographic XPV (TXPV and SXPV, respectively) are necessary to reconstruct the fluid flow in three dimensions. Although faster frame rates are an important component, improving the speed of TXPV and SXPV by other means is more impactful for improving the utility of XPV than faster frame rates alone.

Brighter X-ray Sources

Currently, XPV is fundamentally limited by the brightness of the X-ray source. A brighter source means better image quality, or the same image quality in a shorter exposure time. Many in-lab X-ray sources do not exceed the pixel photon count rate limitations of PCDs. Should sources be brighter, higher imaging speeds are possible in the laboratory. With higher imaging speeds one can study faster phenomena or use techniques such as TXPV that require hundreds of images to capture one moment. Improvements in X-ray source brightness will have positive downstream effects for XPV speed.

Chapters 2 and 5 discuss liquid metal jet (LMJ) anode X-ray sources, which are much brighter than conventional solid tungsten anode sources. LMJ X-ray sources are still in the relatively early stages of commercialization. In the coming years, it is expected that LMJs will become cheaper, smaller, brighter, and have smaller focal spots as the technology underpinning them improves. In my opinion, LMJ sources are an important enabling technology for broad-application in-lab XPV.

More X-ray Sources and Fewer Angles

As discussed in previous chapters, X-ray imaging is actually shadowgraphy – a 2D projection of the objects in the field of view. As a result, 3D XPV measurements such as TXPV and SXPV are necessary to overcome the 2D-projected nature of X-ray imaging. In order to get a full 3D reconstruction with a single source-detector pair (SDP) one has to traverse 360° . With two SDPs set $(180 - \alpha)^\circ$ apart, for example, one would only have to traverse $(180 + \alpha)^\circ$, nearly halving the rotation required to acquire a full reconstruction. In effect, this nearly doubles the TXPV acquisition rate. Here, acquisition rate refers to the rate at which one can acquire the data needed to locate the tracer particles in 3D. Moreover, two SDPs enables the use of SXPV; more than two improves its accuracy. With SXPV, the speed limitation would then be the detector frame rate or the source brightness, whichever comes first.

A less costly solution is to maintain a single SDP, but use fewer angles for a reconstruction. The compromise is that a full reconstruction is unachievable, so the system geometry is not likely to be recoverable. Small, spherical objects such as tracer particles can be, however. Using limited-angle tomography one can reduce the number of angles needed to locate the tracer particles in 3D. Similar to adding a SDP, reducing the number of angles traversed by a factor of X increases the acquisition rate by a factor of X . Algorithm development for this technique is an area of active research. Combining multiple SDPs and limited-angle tomography would have a magnifying effect because the acquisition rate increases combine multiplicatively. For example, a $2\times$ increase in acquisition rate from two SDPs and a $3\times$ increase in acquisition rate from limited-angle tomography would result in a $6\times$ increase overall in the acquisition rate.

Making XPV More Accessible

Presently, XPV is a class of measurement techniques that are available only to laboratories that have X-ray imaging equipment, unique tracer particles, and the expertise on X-ray imaging and data processing. Nearly every step along the data collection and processing path is unique to each laboratory; there is very little standardization. Certainly there are no currently available integrated commercial XPV systems like there are for visible light PIV and PTV. Even if XPV improves to the point that it could be used for a broad set of fluid dynamics experiments, it may not be adopted by the fluid dynamics community unless the barrier to entry is lowered.

Two important developments are necessary to make XPV measurements more accessible: integrated hardware and software conducting for XPV measurements and user-friendly, open source software for XPV data processing. The former is a likely candidate for commercialization in the near future since it is an extension of already existing supply chains for other X-ray imaging applications. The latter is an important endeavor that the XPV community must undertake. Ideally, future users should be able to buy off-the-shelf X-ray imaging systems designed for broad-application XPV and use said systems with proprietary or open source software to process their data. Without such systems, XPV will never be widely adopted on the scale of visible light PIV and PTV.

Interesting Applications for XPV

While XPV remains a slow technique at present, there are still a number of interesting fluid flows that could benefit from this unique perspective. For example, reacting flows or active droplet flows present an opportunity for XPV. As shown in chapter 5, X-rays are sensitive to elemental distributions and could observe reactions within or around active droplets as they occur, even if they are refractive index-matched. Photon-counting XPV in particular could leverage energy-resolved imaging to track the mixing and distribution of reactants and products. XPV could also take advantage of its relative indifference to refraction to observe phenomena close to the boundaries of active droplets, potentially where substantial mass and energy transfer is occurring.

Another potential application of XPV with present day limitations is buoyancy-driven bubble plumes. Typically, buoyancy-driven bubble plumes are relatively low speed systems depending on the bubble size and the continuous medium viscosity. Pseudo-turbulence in buoyancy-driven bubble plumes is used for mixing in a wide range of industrial and chemical applications due its low shear stress. In high volume fraction bubble plumes (10-20% or higher), however, it is difficult to characterize the turbulence intensity statistics, mean flows fields, and bubble-bubble interactions due to the refractivity of the flow. Furthermore, there is a dearth of experimental data to validate numerical simulations in these regimes. Direct numerical simulations of high volume fraction bubbly flows are challenging due to the multitude of moving gas-liquid interfaces; as a result, it is challenging to validate other numerical methods without experiments.

Systems such as these that can take advantage of X-ray light to observe changes in composition, fluid interfaces, and fluid motion simultaneously are ideal candidates for exploration with XPV, so long as they are relatively slow – for the time being, at least.

7.2 Thermodynamic State Measurements and Cryopreservation

Although the main focus of this dissertation is developing in-lab XPV, thermodynamics applications are also introduced. In this section I will discuss the potential for photon-

counting X-ray measurement techniques for capturing fundamental thermodynamic state measurements in the laboratory. Additionally, I will touch on the outlook for using photon-counting X-ray CT for cryopreservation applications.

Thermodynamic State Measurements

Photon-counting X-ray imaging has the benefit of being energy-resolving. As discussed in chapter 2, this could be used for material identification. In thermodynamic systems with multiple phases and material species (e.g. solutes, solvents, and precipitates), non-invasive material identification can prove useful in extreme environments. Combined with isochoric chambers, photon-counting X-ray CT (PCXCT) could be used to study the complete state of multiphase equilibria at high pressures and low temperatures. PCXCT would be able to 1) identify the phase material distribution, 2) measure solute concentrations if calibrated as in chapter 5, and 3) measure the volume fraction of each material in the system. Traditional thermocouples and a pressure transducer can measure the chamber temperature and pressure, respectively.

Using such a system, it would presently be feasible to study mL-scale aqueous systems at temperatures ranging from room temperature to approximately -30°C with commercially available Peltier coolers and pressures $O(100\text{ MPa})$ or more. More powerful cooling systems or smaller volumes could study even colder temperatures. These pressures fill an interesting niche wherein differential scanning calorimetry (DSCs) cannot handle them, while diamond anvil cells (DACs), which typically use Raman spectroscopy, would not be able to measure pressure precisely [92], [93]. Typical DSCs can only handle $O(10\text{ MPa})$ [94], [95]. Lastly, Raman spectroscopy measurements couple the temperature and pressure measurements, making precise measurements at pressures $O(100\text{ MPa})$ challenging [96].

A self-pressurizing isochoric system is limited to the examination of aqueous systems at equilibrium since low density ice is needed to generate the pressure. Nevertheless, it could prove useful in studying multi-species aqueous systems for applications in cryobiology, astrobiology, and exoplanetary atmospheric or oceanic studies. These applications all often exist at or near equilibrium and experience low temperatures and high pressures. An in-lab, all-in-one measurement system would allow for simple, rapid characterizations of the thermodynamic states of these systems.

Cryopreservation

Chapter 6 showed how X-ray CT was used to study the vitrification outcomes under different confinement conditions for two different solutions. X-ray CT is relatively simplistic, however, and has been used in other vitrification studies previously [78] to determine solute distribution and vitrification states. PCXCT could introduce new capabilities to the field of cryopreservation. PCXCT could map the distribution of solutes within and outside of the biological specimen. Additionally, since photon-counting detectors can achieve a higher signal-to-noise ratio than traditional X-ray detectors, PCXCT could potentially distinguish

between frozen and unfrozen biological specimens based on small changes in the density. This method could even be used to determine partial freezing of a biological specimen, and locate where that freezing occurs. To the best of my knowledge, direct confirmation of an unfrozen biological specimen in a cryopreservation system has remained elusive. Implicit confirmation such as specimen solute uptake and specimen revivability are commonly used.

7.3 Final Thoughts

Advances in laboratory X-ray imaging such as brighter sources and more sensitive detectors offer unique capabilities for fluid dynamicists and others. These systems will enable us to view both familiar and new systems from a new vantage point. Ultimately, X-ray imaging systems can be used to simultaneously capture multiple state variables (location, velocity, concentration, material, etc.) in both space and time. Such integrated, non-invasive measurements can be a powerful way to reduce errors associated with combining multiple measurement tools.

X-ray imaging in the laboratory will continue to rapidly improve over the coming years, further expanding the range of fluid flows and other dynamic systems that can be studied. The techniques and hardware developed in this dissertation are but one step along the path to improving the utility and accessibility of X-ray imaging-based measurements.

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