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Technique for Hydrodynamic Characterization of Industrial-Scale
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affiliée à l'Université de Montréal

**Development of a Model-Free Radioactive Particle Tracking (RPT) Technique
for Hydrodynamic Characterization of Industrial-Scale Reactors**

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Thèse présentée en vue de l'obtention du diplôme de *Philosophiæ Doctor*
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**Development of a Model-Free Radioactive Particle Tracking (RPT) Technique
for Hydrodynamic Characterization of Industrial-Scale Reactors**

présentée par **Ghazaleh MIRAKHORI**

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DEDICATION

To Tara and Parham,
my niece and nephew,
whom I left behind
and I was nothing more than
an absence to them.

I hope, in one way or another,
they will know what it means
to live freely.

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RÉSUMÉ

Les systèmes polyphasiques sont présents dans de nombreuses industries chimiques et de transformation, notamment la pétrochimie, l'agroalimentaire, les produits pharmaceutiques, les biotechnologies, l'exploitation minière et les pâtes et papiers. La compréhension de la mécanique des fluides et des configurations d'écoulement au sein de ces systèmes constitue la base de la conception et du développement de nouveaux procédés ou l'amélioration des procédés existants. Cependant, malgré les progrès scientifiques, notre compréhension de nombreux procédés polyphasiques est limitée.

La conception et la mise à l'échelle des systèmes polyphasiques nécessitent une compréhension fiable des configurations d'écoulement dans des réacteurs de différentes tailles. En particulier, la compréhension des échelles de laboratoire, pilote et industrielle est requise. L'incapacité à identifier ces configurations peut entraîner de mauvaises performances des réacteurs à grande échelle. Il est donc essentiel d'avoir accès à des outils expérimentaux fiables pour obtenir des données de haute qualité.

Dans l'étude des systèmes polyphasiques, un outil expérimental quasi idéal devrait être non invasif, capable de fournir des données sur l'ensemble du champ d'écoulement et applicable aux milieux et réacteurs optiquement opaques. Idéalement, un tel outil serait aussi applicable aux systèmes industriels sans nécessiter une implication humaine importante dans la collecte des données. Cependant, la plupart des techniques expérimentales disponibles pour caractériser le champ d'écoulement des réacteurs polyphasiques présentent au moins une de ces limitations communes : elles ne s'appliquent qu'aux milieux et réacteurs transparents, ne peuvent pas s'adapter aux systèmes industriels du monde réel ou nécessitent des protocoles d'étalonnage laborieux.

La technique de suivi des particules radioactives (RPT) est un outil expérimental bien établi pour étudier le comportement des différentes phases dans les systèmes polyphasiques. Cette méthode non invasive utilise une série de détecteurs à scintillation placés autour du système et une particule traceuse radioactive qui émet des rayons γ . La RPT suit la particule traceuse qui est conçue pour suivre l'écoulement d'une phase spécifique du système, capturant sa trajectoire au fur et à mesure qu'elle se déplace. La RPT est performante dans les environnements à forte proportion de bulles et de solides, où les techniques optiques traditionnelles échouent en raison de la réfraction de la lumière et de l'opacité du milieu. Actuellement, la RPT s'appuie sur des modèles complexes de rayonnement nucléaire pour prédire l'intensité de rayonnement attendue au niveau de chaque détecteur en fonction de l'emplacement de la

particule dans le système.

Le modèle de rayonnement nucléaire nécessite le réglage de plusieurs paramètres, tels que le temps mort du détecteur, l'intensité de la source radioactive et le coefficient d'atténuation du milieu, qui sont spécifiques à chaque système. Ces paramètres sont généralement obtenus par un processus appelé calibration. La calibration consiste à positionner manuellement la particule à des centaines de points connus et à collecter des données pour résoudre un problème d'optimisation afin de déterminer les paramètres inconnus du modèle. Cependant, la mise en œuvre du modèle de rayonnement nucléaire présente des limites considérables. Tout d'abord, la complexité du modèle oblige à des simplifications qui introduisent des imprécisions. De plus, il s'agit d'un long processus, ce qui limite le nombre de points de données pouvant être collectés. La collecte d'un nombre suffisant de points de données d'un système peut nécessiter plusieurs jours, ce qui rend la procédure de calibration peu pratique.

Ce travail se concentre sur le développement d'une technique RPT avancée qui intègre un robot collaboratif (cobot) pour générer un ensemble de données expérimentales, reliant le rayonnement détecté par chaque détecteur à des centaines de milliers de positions dans le domaine de l'écoulement. Ce vaste ensemble de données est utilisé pour former un réseau neuronal artificiel (RNA) afin de reconstruire la trajectoire d'une particule de traceur dans des expériences à écoulement libre. Contrairement aux méthodes RPT traditionnelles, cette technique avancée ne repose pas sur des modèles mathématiques ou physiques, ce qui permet d'éviter les artefacts associés.

Outre le modèle RNA pour la reconstruction de la position, une nouvelle méthode appelée reconstruction de la position basée sur la méthode des éléments finis (FEM-PR) a été mise au point. Cette technique utilise l'inverse de la méthode des éléments finis pour trianguler la position de la particule traceuse à partir des données de rayonnement. La méthode FEM-PR résout un problème de moindres carrés pour évaluer les fonctions de forme dans les éléments simples. Elle compare les valeurs de l'expérience en écoulement libre avec les valeurs attendues, qui peuvent être fournies par le modèle de rayonnement nucléaire ou collectées par le cobot à partir de la position de chaque nœud dans les éléments. Elle identifie la cellule qui satisfait à l'interpolation inverse des éléments finis comme la région contenant la position du traceur. La méthode FEM-PR utilise l'emplacement précédent du traceur pour effectuer une recherche locale autour de celui-ci, ce qui rend l'algorithme extrêmement rapide. Comme le temps nécessaire à la reconstruction de chaque position est inférieur à celui requis pour l'échantillonnage des données de rayonnement, la méthode FEM-PR devient un outil puissant pour le suivi des particules en temps réel. La précision de cette technique de reconstruction de positions est vérifiée et validée à la fois par un cas de référence et par un test expérimental.

La capacité de la RPT sans modèle à capturer les valeurs de vitesse moyenne est vérifiée par l'étude de l'écoulement à l'intérieur d'un réservoir agité dans des régimes laminaires et transitoires. Cette vérification est effectuée en comparant les résultats de la RPT avec ceux des simulations de la mécanique des fluides numérique (CFD). Pour le régime d'écoulement laminaire, les résultats de la RPT sont superposés aux les données CFD. Pour le régime transitoire, l'écoulement complexe dans une cuve agitée non couverte et non brassée est analysé en comparant les simulations monophasiques et polyphasiques. La RPT sans modèle capture efficacement les principales caractéristiques du tourbillon central autour de l'arbre, y compris sa profondeur et le rayon critique séparant le tourbillon libre du tourbillon forcé. Les résultats pour les composantes de la vitesse moyenne sont en accord avec la CFD, en particulier dans la simulation polyhasique.

En conclusion, les limites de la RPT sans modèle sont discutées, ainsi que des suggestions pour des travaux futurs potentiels.

ABSTRACT

Multiphase systems are standard components in many chemical and process industries, including petrochemicals, food and beverages, pharmaceuticals, biotechnology, mining, and pulp and paper. Understanding the fluid dynamics and flow patterns within these systems is the foundation for designing and developing new processes or troubleshooting existing ones. However, despite the advancements in science and technology, our understanding of many multiphase processes is incomplete.

Design and scale-up of multiphase systems require a trustworthy understanding of flow patterns across different reactor sizes: laboratory, pilot, and industrial scales. The inability to identify these patterns can lead to poor performance in scaled-up reactors. Therefore, access to reliable experimental tools for obtaining experimental data is essential.

A near-ideal experimental tool for investigating multiphase systems should be non-invasive, capable of providing data across the entire flow field, applicable to optically opaque media and reactors, and ideally applicable to real-world industrial systems without an extensive need for human involvement in data collection. However, most available experimental techniques for characterizing the flow field within multiphase reactors have at least one of these common limitations: they only apply to transparent media and reactors, cannot accommodate real-world industrial-scale systems or requiring labor-intensive calibration protocols.

Radioactive Particle Tracking (RPT) is a well-established experimental tool for investigating the behavior of different phases within multiphase systems. This non-invasive method utilizes a series of scintillation detectors placed around the system and a radioactive tracer particle that emits γ -rays. RPT tracks the tracer particle designed to flow with a specific phase in the system, capturing its trajectory as it moves. RPT works well in environments with a high fraction of bubbles and solids, where traditional optical techniques fail due to light refraction and the medium's opacity. In current practice, RPT relies on complex nuclear radiation models to predict the expected radiation intensity at each detector based on the particle's location within the system.

The nuclear radiation model requires tuning several parameters, such as detector dead time, radioactive source strength, and the medium attenuation coefficient, which are specific to each system. These parameters are typically obtained through a process called calibration. Calibration involves manually positioning the particle at hundreds of known points and collecting data to solve an optimization problem for determining the model's unknown parameters. However, implementing the nuclear radiation model has considerable limitations.

First, the model’s complexity forces simplifications that introduce inaccuracies. Additionally, it is a time-consuming process, which limits the number of data points that can be collected. Collecting sufficient data points from a system might require several days, making the calibration procedure impractical.

This work focuses on developing an advanced RPT technique that integrates a collaborative robot to generate an experimental dataset, linking the radiation detected by each detector to hundreds of thousands of positions within the flow domain. This extensive dataset is employed to train an Artificial Neural Network (ANN) for reconstructing the trajectory of a tracer particle in free-flowing experiments. Unlike traditional RPT methods, this advanced technique does not rely on mathematical or physical models, thus avoiding associated artifacts.

In addition to the ANN model for position reconstruction, a novel method called Finite Element Method-based Position Reconstruction (FEM-PR) is developed. This technique uses the inverse of FEM to triangulate the tracer particle’s position from radiation data. FEM-PR solves a mean square problem to evaluate shape functions within simplex elements. It compares values from the free-flowing experiment with expected values, which can either be provided by the nuclear radiation model or collected by the cobot from the position of each node in the elements. It identifies the cell that satisfies the inverse finite element interpolation as the region containing the tracer position. FEM-PR uses the previous location of the tracer to search locally around it, which makes the algorithm extremely fast. Since the time to reconstruct each position is less than that required for sampling radiation data, FEM-PR becomes a powerful tool for real-time particle tracking. The accuracy of this position reconstruction technique is verified and validated through both a benchmark case and an experimental test.

The model-free RPT’s ability to capture average velocity values is verified by studying the flow inside a stirred tank across laminar and transitional regimes. This verification is done by comparing RPT results with those from Computational Fluid Dynamics (CFD) simulations. For the laminar flow regime, RPT results closely align with CFD data. In the transitional regime, the complex flow in an uncovered, unbaffled stirred tank is analyzed by comparing single-phase and multiphase simulations. Model-free RPT effectively captures key features of the central vortex around the shaft, caused by the absence of baffles, including its depth and the critical radius separating the free and forced vortex. Its results for average velocity components are in strong agreement with CFD, particularly in the multiphase simulation.

In conclusion, the limitations of the model-free RPT are discussed, along with suggestions for potential future work.

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LIST OF SYMBOLS AND ACRONYMS

AI	Artificial Intelligence
ANN	Artificial Neural Network
ASM	Algebraic Stress Model
CFD	Computational Fluid Dynamics
FID	Free Induction Decay
FEM	Finite Element Method
FEM-PR	Finite Element Method-based Position Reconstruction
ILES	Implicit Large-Eddy Simulation
LDA	Laser Doppler Anemometry
LoR	Line of Response
MAE	Mean Absolute Error
MCM	Monte Carlo Method
MCNP	Monte Carlo N-Particle transport
MEDE	Mean Euclidean Distance Error
MRI	Magnetic Resonance Imaging
MPT	Magnetic Particle Tracking
NaI	Sodium Iodide
NIB	Nitsche Immersed Boundary
PBT	Pitched Blade Turbine
PEPT	Positron Emission Particle Tracking
PET	Positron Emission Tomography
PIV	Particle Image Velocimetry
PSPG	Pressure-Stabilizing/Petrov-Galerkin
PTV	Particle Tracking Velocimetry
RF	Radio Frequency
RPT	Radioactive Particle Tracking
SD	Standard Deviation
SUPG	Streamline-Upwind/Petrov-Galerkin
SVR	Support Vector Regression
VOF	Volume of Fluid

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CHAPTER 1 INTRODUCTION

In 2019, Pyrowave, a Canadian company based in Montreal, approached us. They were developing a microwave-assisted reactor that used microwave irradiation to heat a mixture of melted polystyrene and silicon carbide (SiC) particles to convert polystyrene to styrene. The reactor was a complex solid-liquid mixing unit, in which the liquid was non-Newtonian. Agitation played a critical role in the process. To maximize the surface area for the reaction, the stirring rate had to be high enough to keep the solid particles suspended, preventing them from settling on the tank's base for more than a second or two. Heat generated in the SiC particles needed to dissipate to avoid hot spots, while the bulk temperature had to remain low to fully take advantage of selective microwave heating. Therefore, optimizing the design required an experimental study of the solid-liquid hydrodynamics within the reactor. Most research at the time focused on solid-liquid mixing of Newtonian fluids under turbulent conditions, with limited studies on laminar mixing of non-Newtonian fluids. A better understanding of the hydrodynamics in the early transitional regime was necessary for the operation of Pyrowave's reactor.

We were specifically approached because of our access to the Radioactive Particle Tracking (RPT) facilities at Polytechnique Montréal. Given that the Pyrowave reactor was optically opaque and could not be operated in a transparent tank due to the nature of microwave energy, RPT, which is known for its applicability to optically opaque media, was the ideal tool for this investigation.

In any RPT experiment, a radioactive tracer particle emitting γ -rays flows with the phase being studied. Each particle is specifically designed for the experiment. For example, if the aim is to investigate a solid phase, the particle must match the shape, size, and density of the solid particles. If it's a liquid phase, the particle should ideally be neutrally buoyant in the liquid [1]. This makes the careful design and manufacturing of the particle essential. Scintillation detectors, strategically placed around the experimental volume, continuously track the tracer particle's movement.

RPT relies on complex nuclear radiation models to connect the tracer particle's position to the radiation detected by scintillation detectors. One such model is the phenomenological model developed by Beam et al. [2]. This model has three unknown parameters: the detector's dead time, the radioactive source strength, and the medium's attenuation coefficient; these parameters are specific to each experiment.

Typically, RPT is conducted in two stages: calibration and the free-flowing experiment. During the free-flowing phase, the tracer particle moves freely within the specific phase, while the RPT hardware records the radiation received by each detector at specified sampling intervals. Afterward, a designated algorithm triangulates the positions of the tracer particle based on the recorded radiation data. The triangulation process, also known as the position reconstruction algorithm, is complex and requires additional information beyond what is collected during the free-flowing experiment. It requires access to the unknown parameters of the phenomenological model associated with the system of interest.

Calibration is a manual process where the particle is placed at specific calibration points, and the radiation emitted from the tracer is recorded by each scintillation detector at those locations. After calibration, an optimization problem is solved to determine the unknown parameters. With these parameters, the phenomenological model predicts the radiation each scintillation detector receives from the tracer at any position within the domain.

As a preliminary step in studying the Pyrowave reactor, the initial goal was to improve the existing position reconstruction algorithms, as there was significant potential to reduce reconstruction errors and develop faster algorithms capable of real-time particle tracking.

From the beginning, it became clear that performing manual calibration inside a cylinder with dimensions similar to Pyrowave’s—1 meter in diameter and 1 meter in height—was impractical. While RPT could technically be applied to study such a tank, the size made manual calibration unfeasible. This scale required a fundamental change in how calibration was approached.

Automating the calibration process was identified as the solution to this limitation, making it much more feasible to use RPT for industrial-scale reactors. A collaborative robot (cobot) was chosen to replace manual calibration, offering precise control in moving the particle to calibration points. The cobot could improve accuracy, eliminate the labor-intensive process, and drastically reduce calibration time from several days to just a couple of hours. More importantly, it could minimize the operator’s exposure to the radioactive source, addressing critical health concerns.

Integrating the robot with the RPT technique offers far more than just automating the calibration process; it could fundamentally enhance the technique itself and open up new possibilities. The cobot could move the particle from one point to another at any predefined velocity while recording data from the particle’s position in space at short time intervals, close to the time resolution of the RPT sampling intervals. Linking the tracer’s positions to the radiation detected by the detectors could generate a dataset with millions of points. This integration had the potential to eliminate the long-standing reliance of RPT on nuclear

radiation models, replacing the so-called dictionary generated by those models. The nuclear radiation models has always had many unavoidable sources of error, which have been accepted as inherent limitations of the technique.

First, these models depend on multiple measurements with respect to an imaginary coordinate system defined by the operator. This includes two designated points from each detector along a line perpendicular to the face of the detectors, the position of the calibration points, and the exact boundaries of the domain. Any inaccuracies in these measurements can significantly contribute to errors.

Additionally, the models assume a homogeneous medium, which means a uniform attenuation coefficient throughout the entire domain. This assumption is often invalid for many multiphase systems. Furthermore, if there are internal objects, such as baffles or shafts, that attenuate the γ -rays, the model does not account for these variations. Therefore, this can introduce artifacts, making the reconstruction algorithm prone to significant errors.

The use of the cobot to generate an experimental dataset, replacing nuclear radiation models, was a turning point that shifted the focus of this work toward developing a new generation of the RPT technique. As a result, this work now aims to develop an advanced RPT technique that is completely independent of nuclear radiation models, requiring no additional spatial measurements beyond what the cobot provides. The new RPT technique is fully automated, allowing it to be used in industrial-scale reactors. Data collection for training the ANN model can happen under conditions that are more realistic and similar to actual operating environments.

CHAPTER 2 LITERATURE REVIEW

The literature review is structured to introduce various measurement techniques for evaluating hydrodynamics in single-phase and multiphase systems, with particular emphasis on non-invasive methods that do not disturb the natural behavior of the flow. It also reviews the application of some of these techniques in the specific context of single-phase mixing in stirred tanks.

Section 2.1 introduces some non-invasive measurement techniques applicable to transparent and opaque systems. It summarizes the fundamental principles of each method, the information they provide, and their advantages and disadvantages.

Section 2.2 provides an in-depth analysis of the Radioactive Particle Tracking (RPT) technique, explaining how this method captures the trajectory of a radioactive tracer particle's motion. The section covers two critical components of RPT: the calibration methods applied before an free-flowing experiment and the position reconstruction techniques used afterward, discussing various approaches and their limitations.

Section 2.3 revisits the experimental techniques outlined in Section 2.1, focusing on their application to stirred tank hydrodynamics. It reviews the hydrodynamic data provided by each technique, addresses considerations for data post-processing, and evaluates the reliability of each method in producing accurate results. Additionally, the section reviews the experimental work done to study the specific case of unbaffled, uncovered stirred tanks, as well as the studies that numerically simulated this unit operation using Computational Fluid Dynamics (CFD).

2.1 Experimental Techniques for Hydrodynamic Characterization

The design and scale-up of many unit operations often depend on empirical correlations mostly validated within the range of operating conditions, making their reliability uncertain outside of those ranges. Moreover, applying fundamental fluid dynamics models to a new system requires experimental verification to ensure their reliability. Therefore, the experimental measurement of fluid dynamic quantities becomes essential to determine the range of validity of current correlations and verify the performance of hydrodynamic models for the new applications. Additionally, these measurements are necessary for understanding, interpreting, and predicting the performance of existing reactors, ultimately driving more efficient operation [1]. This section reviews experimental techniques that provide three-dimensional

flow field information, categorizing them based on whether they use tomographic or particle tracking methods.

2.1.1 Tomographic Techniques

Tomographic techniques are a group of methods used to create detailed images of the internal structure of a system through a series of cross-sectional slices. The imaging can be performed using electromagnetic radiation, such as γ -rays and X -rays, optical light, or radio waves.

Positron Emission Tomography (PET)

Positron Emission Tomography (PET) is an imaging method that uses radioactive tracers which decay by emitting positrons. This decay results in the emission of two γ -rays that travel in nearly opposite directions, separated by about 180 degrees. The line of decay can be identified by detecting these γ -rays simultaneously with position-sensitive detectors. Analyzing numerous such decay events through post-processing allows for creating a three-dimensional image [3].

PET is particularly effective for studying the behavior of a specific phase within a multiphase system. For instance, a particular phase can be tagged with a positron-emitting tracer, enabling the observation of its movement and behavior throughout the system [1].

Figure 2.1a illustrates a simplified representation of a back-to-back photon pair emitted from a radioactive source at random angles. Figure 2.1b highlights the photon pairs that interact with the detector along a perpendicular line.

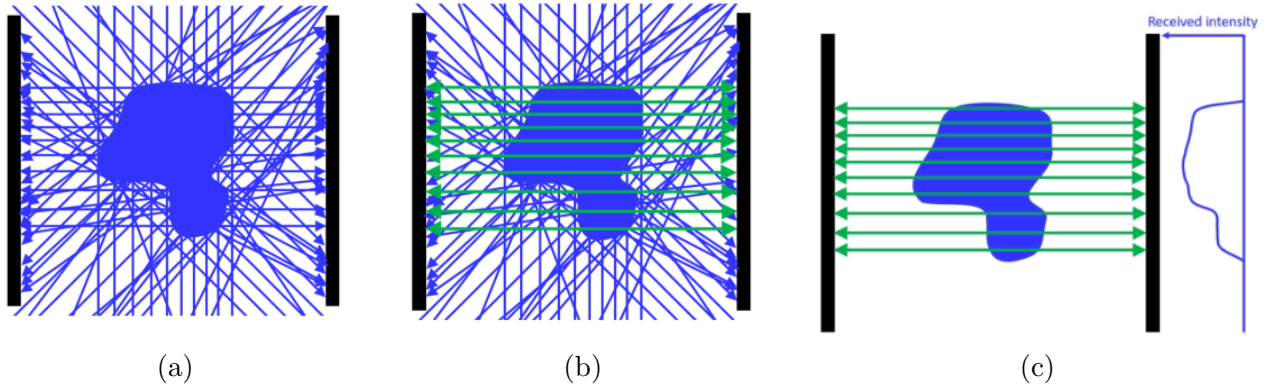


Figure 2.1 Simplified representation of a single projection via PET using a dual-headed gamma camera [3].

An intensity map like the one shown in Figure 2.1c can be constructed using these interaction lines. The detectors must be rotated to capture additional projections from various angles to generate a complete tomographic image.

Magnetic Resonance Imaging (MRI)

Magnetic Resonance Imaging (MRI) operates based on the principles of nuclear magnetic resonance (NMR). Atoms with an odd number of protons and/or neutrons possess a non-zero nuclear spin angular momentum, and these nuclei are often referred to as spins. In the absence of an external magnetic field, the spins are randomly oriented, resulting in no net magnetic moment due to the cancellation of individual magnetic moments. However, in the presence of a strong magnetic field, such as the one in an MRI scanner, the spins align with the magnetic field, creating a net magnetic moment—this state is referred to as the equilibrium state of magnetization [4].

The Radio Frequency (RF) coil inside the scanner emits an RF magnetic pulse to generate a magnetic resonance signal. This pulse applies a torque to the spins and disturbs their equilibrium state. When the RF pulse is turned off, the spins gradually return to their equilibrium alignment with the primary magnetic field of the scanner, releasing the energy absorbed from the RF pulse in the process. The system’s receivers detect this emitted signal, known as Free Induction Decay (FID) [4].

The process described above only produces a single-valued signal for the entire system. Additional magnetic fields known as linear gradient magnetic fields are applied to achieve spatial localization and construct detailed images. The primary magnetic field is aligned along the z -axis, while gradients are applied in the x - and y -directions. These gradients enable the construction of a three-dimensional image by encoding spatial information into the signal [3].

MRI captures detailed three-dimensional images from optically opaque systems. However, it has two main limitations: the scanner’s size restricts the systems that can be imaged, and there are limitations on the material present in the system; for instance, ferromagnetic materials disrupt MRI’s performance. The material restriction makes the choice of tracer particle also difficult, as some of the materials with nuclei that exhibit appropriate spin properties interfere with the process [3].

2.1.2 Particle Tracking Techniques

Particle tracking techniques work by following the motion of one or multiple tracer particles. They can be either Lagrangian techniques, which track the motion of a single particle over

time, or Eulerian techniques, which observe various particles within a specified space. Depending on the type of tracer used, for instance, if they use γ -ray emitters, these techniques can be applied to optically opaque systems. Conversely, if they are optical techniques, they may only be practical for transparent systems.

Particle Tracking Velocimetry (PTV)

Particle Tracking Velocimetry (PTV) is a Lagrangian technique that obtains particle trajectories by capturing and analyzing multiple images. This relatively inexpensive technique requires an illumination system, a camera, tracer particles, and a suitable image processing algorithm [5].

The illumination system consists of a high-frequency pulsed light source with a short pulse duration synchronized with the camera frame rate. Tracer particles seeded in the fluid scatter the light with each pulse. The image processing algorithm identifies the pixels associated with the particles and reconstructs their positions in each image. A proper algorithm reconstructs the particle's trajectory by connecting the positions captured across successive images.

The main limitation of PTV is its use of electromagnetic radiation, which cannot penetrate optically opaque objects. Therefore, it is unsuitable for systems with opaque walls and is best suited for transparent, shallow, or highly dilute systems [3].

Particle Imaging Velocimetry (PIV)

Like PTV, Particle Image Velocimetry (PIV) relies on four essential components: tracer particles, a light source, a camera, and image processing techniques. However, unlike PTV, PIV is an Eulerian measurement technique. Instead of following individual particles through space and time, PIV captures the motion of seeded particles within a fixed region of space using laser illumination. Multiple snapshots are collected to analyze how particles move through the fluid in these regions.

The recording phase in PIV can be carried out using two main methods: single-frame double exposure and double-frame single exposure. Two consecutive images are captured in both methods with an interval of Δt . The single-frame double exposure method captures two illuminations in one frame, while the double-frame single exposure method captures one image per illumination pulse [6].

Various techniques are employed to process the images from a PIV experiment. One common approach, as illustrated in Figure 2.2 involves dividing each frame into smaller regions called interrogation windows.

For a sufficiently small time step Δt and a large enough window size, the algorithm tracks the movement of tracer particles within each window between the two frames. It calculates the average displacement of particles within each window and associate their mean velocity to that specific window. The algorithm performs this operation for all windows, ultimately generating a comprehensive picture of the flow field [3].

Similar to PTV, PIV is also unsuitable for optically opaque systems and is best suited for shallow and dilute systems. It also provides Eulerian information, making it appropriate only for systems where particle motion is strongly spatially correlated [3].

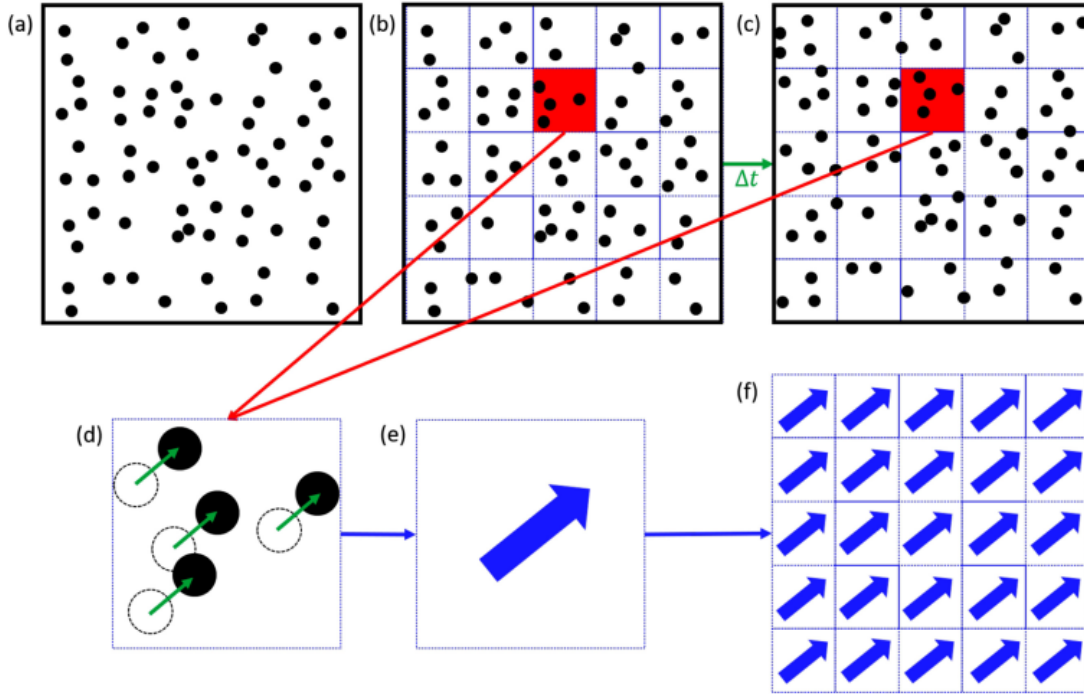


Figure 2.2 Schematic diagram of a PIV workflow. a) Digital image of the system, corresponding to a single frame from a video. b) Image is subdivided into interrogation windows. c) Subdivided image for the subsequent frame in the video. d) Single interrogation window containing information from two consecutive frames. e) Mean velocity for the window obtained from an appropriate correlation function. f) Flow field map generated by applying the described algorithm to all windows [3].

Positron Emission Particle Tracking (PEPT)

The Positron Emission Particle Tracking (PEPT) has been developed as a variant of Positron Emission Tomography (PET). PEPT is a particle tracking method that uses a non-radioactive material labelled with positron-emitting radioisotopes, similar to those used in PET as a

tracer. The tracer emits a pair of back-to-back γ -rays [1].

To track the tracer particle, the system must be placed within the field of view of a detection system, typically called a gamma or positron camera. The interaction position is recorded when a gamma photon strikes one of the gamma camera's detectors. Suppose two gamma photons are detected within a very short time window, such as on the order of nanoseconds. In that case, it is assumed they originated from a positron annihilation event from the tracer (see Figure 2.3(a)). Since the annihilation event produces γ -rays that travel in opposite directions, the recorded points can be connected by a straight line (see Figure 2.3(b)). This line represents the Line of Response (LoR), along which the tracer was located at the time of annihilation. The system records all detected interactions and stores their positions for future reconstruction of the LoR. By analyzing multiple pairs of emitted γ -rays and reconstructing their LoRs, the intersection of these lines can be used to triangulate the tracer's position (see Figure 2.3(c)) [3].

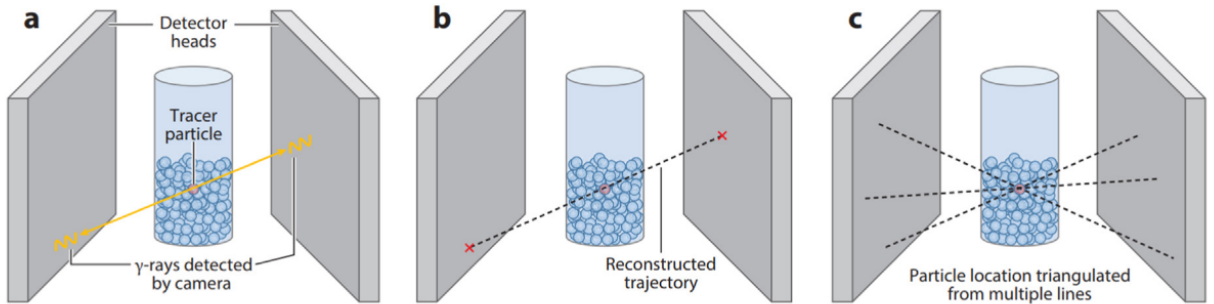


Figure 2.3 Schematic diagram of PEPT locating a single tracer particle in a granular system. (a) γ -ray camera detecting a pair of emitted γ -rays by the radioactively-labelled tracer particle. (b) Trajectory of the pair reconstructed by a suitable algorithm. (c) The intersection point of multiple reconstructed trajectory can be used to reconstruct the position of the tracer's center [7].

Radioactive Particle Tracking (RPT)

Radioactive Particle Tracking (RPT) works based on tracking one or more radioactive tracers emitting γ -rays. In an RPT setup, as illustrated in Figure 2.4, scintillation detectors are strategically placed around the volume of interest to detect gamma radiation emitted by the radioactive tracer particle. When the particle emits radiation, each detector picks up the signal, which is normally weak. This signal is sent to an amplifier, where it is strengthened. The amplified signal then passes through a discriminator, which filters out noise by allowing

only signals within the energy range of the tracer's radiation to pass through, effectively discarding lower-energy events like Compton scattering [1].

Once valid signals are identified by the discriminator, they trigger the generation of pulses, with each pulse representing the detection of a gamma photon by a specific detector. These pulses are fed into a counting system, where high-speed counters record the number of pulses detected by each scintillation detector over a specific sampling interval. This collection of counts across all detectors provides data that, when processed, allows the precise localization of the tracer particle within the system [1].

RPT tracks the trajectory of a radioactive particle over an extended period. By differentiating successive positions over time, Lagrangian velocity data is obtained. Further post-processing of this data produces Eulerian velocity fields. RPT can be applied in non-transparent and large-scale systems, making it a proper tool for studying complex reactors [3].

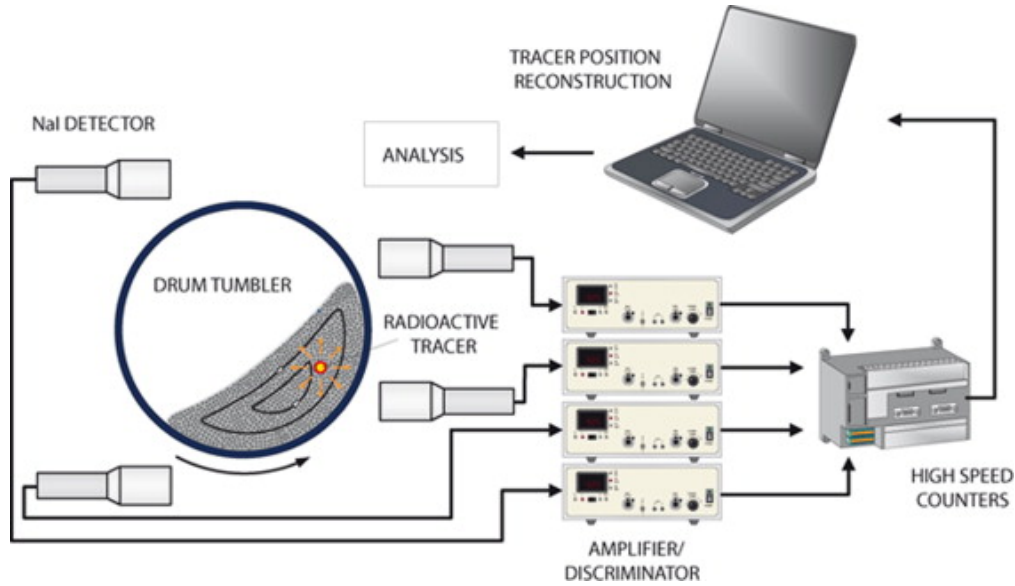


Figure 2.4 Schematic diagram of an RPT setup [8].

Magnetic Particle Tracking (MPT)

Magnetic Particle Tracking (MPT) functions on the same principle as RPT but measures magnetic field intensity rather than γ radiation. Like RPT, MPT utilizes a set of detectors to capture signals from a magnetic source, typically a magnetic dipole. A proper algorithm to reconstruct the magnetic field to obtain the magnet's position and orientation [9].

MPT accommodates optically opaque systems and offers a significant advantage over other particle tracking techniques, such as RPT and PEPT. MPT provides comprehensive infor-

mation by tracking both position and orientation of the tracer. This feature is especially interesting for systems involving granular materials, where understanding the particle's orientation and rotation is essential for accurate analysis [10].

Despite its unique advantages over other techniques, MPT has several drawbacks. A significant limitation is the interference of external magnetic sources with information from the tracer particle, which makes the data unreliable. Additionally, MPT requires sophisticated algorithms for data processing, making it complex and expensive to implement [3].

2.1.3 Advantages and Disadvantages of Each Method

Figure 2.5 provides an overview of the key strengths and limitations of each technique, offering insight into why one approach might be chosen over another. This comparison helps to clarify the factors that influence the selection of a particular method, such as its advantages in certain applications and the trade-offs involved.

	PET	MRI	PTV	PIV	PEPT	RPT	MPT
Temporal resolution	Yellow	Green	Green	Green	Green	Green	Green
Spatial resolution	Yellow	Green	Green	Green	Green	Yellow	Yellow
Opaque system	Green	Green	Red	Red	Green	Green	Green
3D information	Green	Green	Yellow	Yellow	Green	Green	Green
Orientation information	Red	Red	Yellow	Yellow	Yellow	Red	Green
Lagrangian information	Red	Green	Green	Green	Green	Green	Green
Limitation on involved materials	Green	Red	Yellow	Yellow	Green	Green	Yellow
Need for a detailed calibration	Green	Yellow	Yellow	Yellow	Green	Red	Yellow
Cost	Red	Red	Green	Green	Red	Yellow	Green

Figure 2.5 Summary of the main strengths and limitations of the methods reviewed in the previous sections. Green indicates the strengths of each method, while yellow and red indicate the levels of weakness, with yellow representing a minor weakness and red indicating a significant weakness. This table is adapted from a book by Windows-Yule et al. [3].

Among all the non-invasive methods reviewed in the previous section, RPT stands out due to several significant advantages that make it unique:

- It is applicable to optically opaque systems.
- The materials involved in the process do not conflict with each other.
- Background radiation does not interfere with the radiation detected by the detectors.
- RPT is suitable for large-scale reactors as its detectors can be placed externally, unlike other techniques, where the study volume is limited by the size of the equipment, which the reactor must fit within.

These factors make RPT an excellent candidate for studying real-world reactors; the only restriction is the need for extensive calibration [11], which is more challenging to perform as the size of the domain increases or the geometry becomes more complex.

2.2 Deep into Radioactive Particle Tracking (RPT)

Before developing a modern radioactive particle tracking technique, various methods were employed to quantitatively measure flow by tracking a radioactive tracer. However, at the time, the old techniques struggled with the size of the considerably massive tracer particles, and their motion was hardly representative of the natural flow. In addition, a bulky equipment responsible for receiving the radiation was also inside the flow, disturbing its natural behavior [12].

In 1985, Lin et al. [12] introduced the RPT hardware in its modern format. They developed photon-counting instrumentation and established a monotonic relationship between the count rate received by each detector and the distance between the tracer and the detector through an in-situ calibration. They fitted all calibration data to a third-degree polynomial as follows:

$$r_i = f(1/I_i) = a_{0i} + a_{1i}(1/I_i) + a_{2i}(1/I_i)^2 + a_{3i}(1/I_i)^3 \quad (2.1)$$

where r_i indicates the distance from the i th detector, I_i represents the photon count received by this detector, and a_{0i} , a_{1i} , a_{2i} , and a_{3i} are the coefficients derived by the least squares method. This curve-fitting approach was only suitable for systems where the counts are solely a function of the detector-particle distance [13].

In 1994, Larachi et al. [14], considering the complex relationship between the position of the tracer particle and the detectors, proposed implementing a phenomenological model to calculate the number of photon counts received by an individual detector from a tracer

particle in its vicinity. This model associates the tracer position and the recorded photopeak counts by a given detector as:

$$C = \frac{T\nu R\phi\xi(\vec{X})}{1 + \tau\nu R\phi\xi(\vec{X})} \quad (2.2)$$

where T is the sampling time (in seconds), ν is the number of γ -rays emitted by each disintegration, R is the activity of the tracer (in Bq), ϕ is the peak-to-total (photopeak) ratio, τ is the dead time of the detector (in seconds) per accepted pulse, and $\xi(\vec{X})$ is the efficiency of the detector associated with the tracer position which can be expressed as [2]:

$$\xi(\vec{X}) = \oint_{\Omega} \frac{\vec{r} \cdot d\vec{A}}{||\vec{r}||^3} f_a(a, \theta) f_d(a, \theta) \quad (2.3)$$

where $\vec{r}$ is a vector from the position of the tracer to a point which γ -rays enter to the detector, $d\vec{A}$ is the external surface vector normal to the surface at the contact point on the detector crystal, and Ω is the solid angle subtended by the radiation. Since there is no analytical solution to calculate the efficiency (except for some simple problems), a numerical method based on the Monte Carlo technique can compute Eq. (2.2) [15]. This method generates thousands of random photon path directions towards a detector to estimate the surface integral of Eq. (2.3). Beam et al. [2] employed the Monte Carlo technique, which estimates this equation as:

$$\xi(X) = \frac{1}{N} \sum_{i=1}^N \omega_i f_a(\alpha, \theta) f_d(\alpha, \theta) \quad (2.4)$$

where N is the number of generated photon paths, the angles α and θ specify the direction of each traced ray from the tracer position, ω_i determines the solid angle (details regarding the solid angle calculation can be found in the Appendix A) for the particular selection of α and θ , and $f_a(\alpha, \theta)$ and $f_d(\alpha, \theta)$ are:

- f_a is the probability of non-interaction between the γ -rays emitted within Ω and the material inside the column:

$$f_a(\alpha, \theta) = \exp(-\mu_r e(\alpha, \theta)) \quad (2.5)$$

- f_d is the probability function of the γ -rays interaction with the detector:

$$f_d(\alpha, \theta) = 1 - \exp(-\mu_d d(\alpha, \theta)) \quad (2.6)$$

In Eq. (2.5), μ_r is the total linear attenuation coefficient of the medium inside the reactor and $e(\alpha, \theta)$ is the length of the path travelled by γ -ray inside the reactor, see Appendix B for details of calculation. In Eq. (2.6), μ_d is the total linear attenuation coefficient of the detector, and $d(\alpha, \theta)$ is the effective distance travelled by γ -ray inside the crystal along the direction of α and θ angles [2].

Some parameters of this model, including the detector's dead time, source strength, and the medium attenuation coefficient, are unknown and unique to each experimental system and operating conditions. Therefore, a calibration process for each individual reactor is necessary. Using the data from the calibration process and solving an optimization problem would determine these unknowns.

2.2.1 Calibration Techniques

In the calibration procedure, an operator places the particle at various known positions within the domain. At each position, the tracer remains stationary for a sufficient time, and the RPT data acquisition system records the γ -rays received at each detector. The calibration process generates a three-dimensional map of the photon counts recorded by each detector from the stationary positions.

However, in a free-flowing RPT experiment, the acquisition system records a single data point at each sampling time while the particle moves freely with the flow. This contrasts with the calibration protocols, where data is averaged over a longer period. The difference between time-averaged counts recorded during calibration and the single data point from a free-flowing experiment introduces a problem known as dynamic bias. This difference affects the position reconstructed from the instantaneous data, leading to an error in the particle trajectory and subsequent hydrodynamic information derived from it [13].

To address the issue arising from dynamic bias, Khane et al. [13] introduced a hybrid dynamic approach for the RPT calibration procedure. They designed a setup allowing manual vertical movement of the particle toward the bottom of the tank while continuously recording photon count data; Figure 2.6 is a schematic of their design.

Khane et al.'s work marked the first step toward overcoming the biases from static calibration. However, they acknowledged limitations in their setup, particularly the challenge of recording

adequate data points using their design. To compensate for the lack of information, they had to integrate the dynamic dataset with a static dataset to generate the photon count-position dictionary.

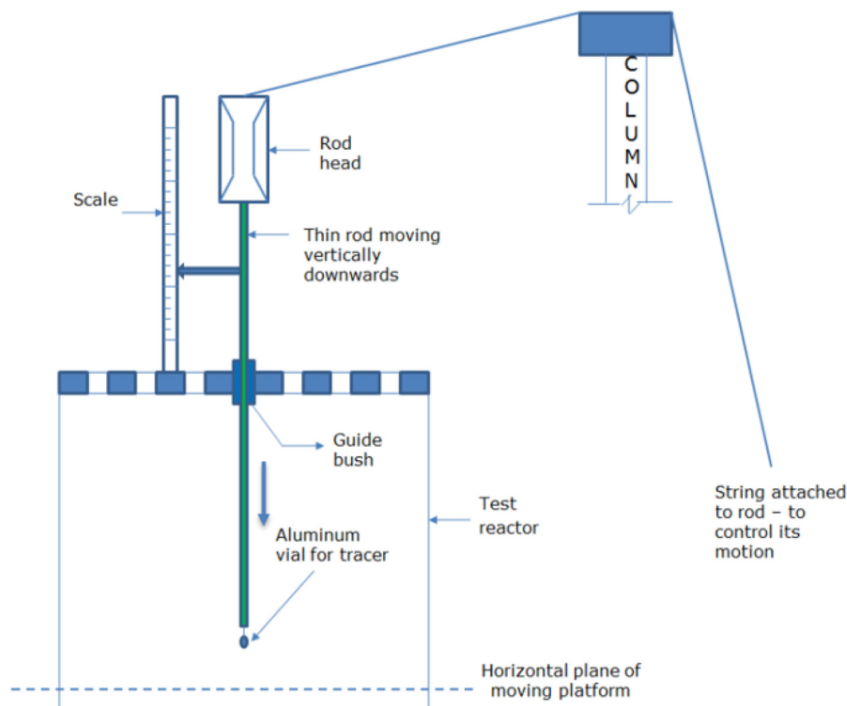


Figure 2.6 Schematic diagram of experimental arrangement for dynamic data acquisition [13].

Existing calibration techniques face several other challenges in addition to the fundamental issue of dynamic bias. The process is typically manual, monotonous, and time-consuming, particularly when dealing with large reactor volumes. The manual approach restricts the use of RPT to laboratory-scale reactors, making it impractical for application in industrial-scale systems. Furthermore, manual calibration is not immune to human errors, which can propagate through subsequent calculations. Additionally, close contact with the radioactive particle during calibration poses potential health risks to the operator.

2.2.2 Position Reconstruction Algorithms

A position reconstruction algorithm converts recorded photon counts by the array of detectors into instantaneous particle coordinates (x, y, z) . These algorithms are typically prone to errors, which can impact the spatial resolution of the technique [16], and are computationally expensive, as they involve solving inverse problems with millions of data points per experiment. Over the past two decades, researchers in the RPT domain have developed various

techniques to achieve faster and more accurate position reconstruction.

These techniques are categorized into physics-based approaches and machine learning algorithms. This section focuses on the methods most frequently cited in the literature for their performance in position reconstruction. The first group includes approaches such as the least squares search algorithm combined with Monte Carlo simulations and iterative methods paired with the Monte Carlo N-Particle transport (MCNP) code.

The second group covers two widely implemented machine learning algorithms in RPT problems: Artificial Neural Network (ANN) and Support Vector Regression (SVR). Additionally, some studies that comprehensively compare these techniques while applying them to the same problem are reviewed to better understand their relative performance.

Physics-Based Algorithms

In combination with Monte Carlo simulation, conventional reconstruction methods typically rely on a dictionary that associates positions within the volume of interest with the photon counts received by the array of detectors from those positions. The process begins by identifying a small control volume that contains the particle's location and then refining this volume one or more times to estimate the particle's exact position.

Larachi et al. [14] introduced the least-squares search algorithm for particle position reconstruction in 1994. This grid search algorithm compares the experimental photon counts with the calculated photon counts from the Monte Carlo simulation at each time step and aims to minimize χ^2 :

$$\chi^2 = \sum_{i=1}^n \frac{(C_i - M_i)^2}{\sigma_i^2} \quad (2.7)$$

where n is the number of detectors, C_i is the count calculated by the Monte Carlo simulation for detector i , M_i is the experimentally measured count, and σ_i is the statistical uncertainty.

Due to the computational expense of generating a dictionary with a relatively high number of points from the outset, the algorithm first identifies a region where the particle falls based on the existing points. Then it continues searching within the neighborhood and, if necessary, considers new points not already in the dictionary. For these new points, the photon counts associated with their positions must be calculated.

Larachi et al. stated that it is not necessary to perform a Monte Carlo simulation for each new point, opting instead to balance accuracy with computational cost. They proposed that, rather than calculating the detector efficiency through Monte Carlo simulations for each

point, the efficiency could be approximated as varying with the inverse square of the distance between the particle and the center of the detector. Consequently, instead of using Equation (2.2), they used the following equation to calculate the count for the new points:

$$C^* = \frac{TC \left(\frac{r}{r^*}\right)^2 \exp(\mu_r(e - e^*))}{T - \tau C + \tau \left(\frac{r}{r^*}\right)^2 C \exp(\mu_r(e - e^*))} \quad (2.8)$$

The symbols with asterisks refer to the new positions, while the other symbols denote existing points in the dictionary. Here, r represents the distance between the particle and the geometric center of the detector, and e denotes the length traveled by the γ -rays.

In 2011, Mosorov et al. [17] used the MCNP5 instead of the commonly applied Monte Carlo simulation based on the phenomenological model to calculate the radiation received from the tracer at each position. They proposed this replacement because the standard Monte Carlo simulation was a simplified model of radioactive transport. Additionally, they introduced their reconstruction algorithm based on the so-called dictionary generated by MCNP5.

They proposed an iterative approach that does not compare single points for position reconstruction, unlike the traditional algorithm. Instead, it incorporates multiple data points to improve position reconstruction. This algorithm aims to eliminate the dependency on a single reference position, which may contain statistical fluctuations and inaccuracies from measurement errors.

Their proposed algorithm operates based on the similarity σ_j between the experimentally measured counts C_i^m and the calculated counts $C_{i,j}^c$ from MCNP5, defined as:

$$\sigma_j = \sum_{i=1}^n \left| \ln(C_i^m) - \ln(C_{i,j}^c) \right|, \quad j = 1, \dots, J_{\max} \quad (2.9)$$

where $j_{\max}$ is the number of simulated positions for the particle using MCNP5. Then, for each individual group of counts received by the detectors, they sort the similarities and pick the three most similar groups of simulated counts and calculate the positions as follows:

$$\mathbf{X}_e = \frac{\mathbf{X}(\sigma_{\min 1})w_1 + \mathbf{X}(\sigma_{\min 2})w_2 + \mathbf{X}(\sigma_{\min 3})w_3}{w_1 + w_2 + w_3} \quad (2.10)$$

$\mathbf{X}_e$ denotes the particle position calculated by the algorithm, and $w_i = \sigma_{\min 1} / \sigma_{\min i}$ for $i = 1, 2, 3$.

Artificial Intelligence (AI) Algorithms

The model-based reconstruction methods are limited by the number of locations in the initial dictionary and rely on nuclear radiation models to generate photon counts for new points in the search region. In contrast, AI algorithms, after the training phase, do not depend on nuclear radiation models for new calculations. AI for position reconstruction was introduced early in the development of modern RPT, gaining attention for its ability to enable real-time flow visualization.

In 1997, Godfroy et al. [18] were the first to introduce a data-driven technique for position reconstruction. They replaced the conventional least-squares method with a three-layer feed-forward neural network for particle tracking in a three-phase fluidized bed reactor. Over the following decades, ANN models became researchers' most widely used technique for position reconstruction.

ANNs are machine learning models inspired by the structure and function of biological neural networks. They consist of several layers: an input layer, one or more hidden layers, and an output layer. The fundamental components of ANNs are units called neurons, which closely resemble biological neurons. These neurons are connected by weighted links that function similarly to synapses in biological systems [19].

During the training phase, the weights of these connections are adjusted based on training data. This adjustment process continues until the output of the ANN model closely matches the desired target or training instances.

For position reconstruction, ANNs are trained using a large number of examples. The input to the ANN consists of well-distributed positions inside the volume of interest, whereas the output corresponds to the amount of radiation received by the detector from tracer particles at those positions. By training the model with this dataset, it learns to predict particle positions based on radiation measurements accurately.

Another machine learning algorithm used for position reconstruction is the Support Vector Regression algorithm. SVR is an extension to the Support Vector Machine (SVM) classification algorithm. The main principle of SVM is to find a hyperplane in a high-dimensional space that best classifies the data. SVM aims to maximize the margin, which is the distance between the hyperplane and the nearest data points from each class, known as support vectors. By maximizing this margin, SVM aims to ensure the best possible generalization to new, unseen data [20].

Unlike SVM, which provides binary classification responses, SVR is designed for regression tasks. The core concept in SVR is to fit the data within a specified margin of tolerance,

known as ϵ . Instead of finding a hyperplane that separates the training samples, SVR aims to find a function that approximates the target values within this margin. The loss function used in SVR ensures that the predicted values for training samples fall within ϵ of the actual observed values, meaning that deviations from the true values are limited to ϵ or less [20].

In 2020, Yadav et al. [21] utilized SVR alongside other machine learning algorithms to perform the position reconstruction procedure. Figure 2.7 illustrates their approach for defining the position reconstruction function $f(\mathbf{I})$ and the ϵ -sensitive regions within the context of RPT position reconstruction [21].

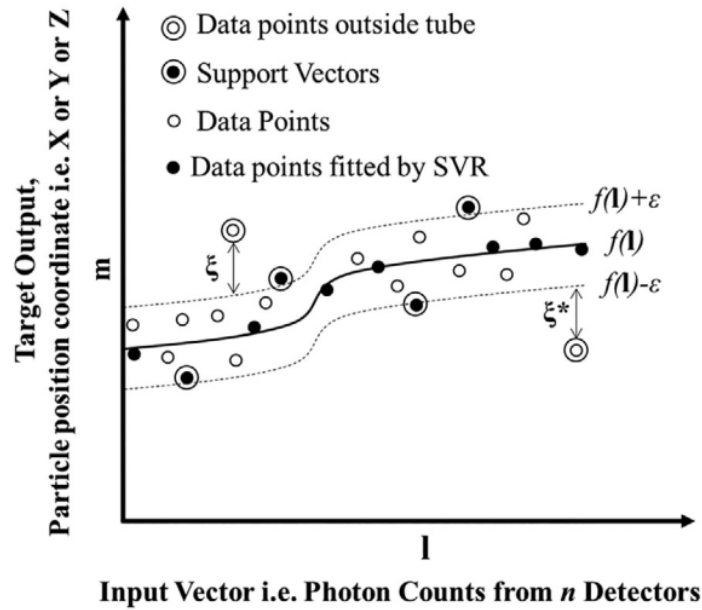


Figure 2.7 A schematic illustration of the SVR reconstruction model using ϵ -sensitive loss function [21].

Comparison of Reconstruction Algorithms Performance

Several studies in the literature report the errors of their position reconstruction algorithms. However, individual reports can be complex to compare due to differences in experimental setups and methodologies. For this reason, this section focuses on works that compare the performance of various techniques for the same case study. By using consistent evaluation criteria and conditions, these comparative studies provide more reliable insights into the relative strengths and weaknesses of different algorithms in position reconstruction.

The first case study is the work conducted by Yadav et al. [21], who performed an RPT

experiment inside a bubble column with an internal diameter of 15 cm and a static liquid height of 30 cm, surrounded by eight strategically positioned detectors. They compared the accuracy of several machine learning algorithms with a least-squares search algorithm combined with the Monte Carlo method, referred to as the MCM.

They performed the calibration at 276 known positions inside the column. Out of these, they randomly selected 57 points to test the accuracy of the algorithms, while they used the remaining 219 points to train the machine learning algorithms and, for the MC technique, to tune the model parameters.

Table 2.1 lists various statistical quantities, including Mean Absolute Error (MAE), Standard Deviation (SD), and Mean Euclidean Distance Error (MEDE), for some of the methods implemented for position reconstruction. They also reported that SVR took 70s to reconstruct 1 million particle locations while MCM took days to reconstruct the same amount of data [21].

Table 2.1 Reconstruction performance metrics from the study by Yadav et al. [21].

	MAE (mm)			SD (mm)			MEDE (mm)
	X	Y	Z	X	Y	Z	
MCM	2.74	2.82	3.01	2.01	1.78	3.56	4.41
ANN	2.25	3.97	6.25	2.26	4.21	5.70	6.58
SVR	1.45	1.41	2.66	1.27	1.38	2.51	3.84

The second case study is the work by Lindner et al. [22], which is not experimental but rather involves comparing different techniques in a virtual case. They studied a rectangular fluidized bed reactor with the following dimensions: $0.23 \times 0.076 \times 1.2$ m, surrounded by 16 strategically positioned detectors. The size of the virtual calibration dataset in their study was 10,527 points, and for validation, they used 100 points on a spiral path. Table 2.2 shows their results.

Table 2.2 Reconstruction performance metrics from the study by Lindner et al. [12].

	MAE (mm)			SD (mm)			MEDE (mm)	Time (s)
	X	Y	Z	X	Y	Z		
MCM	0.49	0.25	0.71	0.5	0.19	0.75	1.43	38.76
ANN	0.7	0.4	0.81	0.65	0.44	0.94	1.16	59.12
SVR	0.46	0.2	0.43	1.46	0.38	0.56	1.05	215.35

Comparing Table 2.1 and 2.2 shows a considerable gap between the performance of all the algorithms in the real experiment and the virtual test. Evidently, as expected, several sources of error exist in the experiments that, if eliminated, could bring the reconstruction results closer to those obtained from the virtual test.

2.3 Mixing in Stirred Tanks

The earliest documented example of a stirred tank reactor is found in Georg Bauer's book *De Re Metallica* (Latin for On the Nature of Metals), published in 1556 [23]. Since then, mixing has advanced and is now recognized as an engineering science. The initial application of engineering principles to quantify stirring technology occurred in 1855 through the work of James Thompson [24]. He measured the power required to rotate a disc in water and identified a relationship now known in its modern form as the power number or Newton dimensionless number [25].

In the twenty-first century, mixing and stirred tanks are crucial in a wide range of industries, including pharmaceuticals, petrochemicals, biotechnology, food processing, and water and wastewater treatment, among others [26].

A classic mixing operation aims to achieve at least one of the following goals [26]:

- Blending of miscible liquids
- Solid-liquid mixing
- Gas-liquid mixing
- Mixing to reach chemical reactions
- Solid-solid mixing
- Heat transfer

Mixing tanks can either be equipped with baffles or operate without them. Among these two types, baffled tanks have drawn considerably more attention due to their higher industrial importance as they are known for having better mixing performance [27]. In the absence of baffles, the tangential flow caused by the impeller moves the entire fluid around the tank like a solid body rotation, creating a central vortex around the shaft [28]. Baffles inside the stirred tank prevent this phenomenon and provide additional benefits to mixing. For instance, they change the flow pattern by transforming part of the radial velocity component into an axial velocity component, improving the overall mixing [29].

However, some chemical and industrial processes prefer unbaffled tanks for various reasons. For example, processes involving shear-sensitive components, such as cells in bioreactors or fragile particles in crystallization, benefit from the absence of baffles. Another consideration is the sanitation of equipment: in industries like food and pharmaceuticals, the presence of baffles can complicate cleaning, making unbaffled tanks more suitable [30–32].

In processes requiring solid suspension or dissolution, the flow pattern in unbaffled tanks helps draw particles into the liquid [33]. Lastly, the central vortex in unbaffled tanks can function as a sparger for processes requiring air, transferring air into the liquid [34, 35].

The following sections will revisit some of the experimental techniques discussed earlier and take a deeper look at the type of hydrodynamic information each technique can provide from the flow, specifically within stirred tanks. They will also review the current state of research on unbaffled stirred tanks, focusing on both experimental techniques and CFD simulations.

2.3.1 Flow Characterization in Stirred Tanks Using Experimental Approaches

Since the main focus of this thesis is flow measurements and hydrodynamic characterization, this section explores different techniques to characterize the flow field in stirred tanks, specifically those used to study the hydrodynamics of single-phase mixing in stirred tanks. Previous sections introduced these techniques under the umbrella of tomographic or particle-tracking methods. Here, a different approach is taken by categorizing them according to whether they work in Eulerian or Lagrangian reference frames.

For the Eulerian methods, this section focuses on PIV, one of the most widely used approaches in stirred tanks. Regarding Lagrangian techniques, the section discusses two particle tracking methods, RPT and PEPT.

Eulerian Techniques

The Eulerian velocity frame focuses on specific points in space through which the fluid flows. Some Eulerian techniques continuously monitor a single point over time, known as single-point observation. Others capture data from a spatial region, providing an instantaneous velocity distribution across a plane.

Pitot tubes [36], hot-wire anemometry [37], laser Doppler velocimetry [38], and phase Doppler anemometry [39] are examples of single-point flow measurement techniques used in stirred tanks. Pitot tubes and hot-wire anemometry are considered invasive techniques because they disrupt the flow by directly interacting with it. In contrast, laser Doppler velocimetry and its extension, phase Doppler anemometry, are non-invasive techniques, as they measure flow

characteristics without interfering with the flow pattern.

Two key criteria must be considered when comparing point measurements to full-field measurements: spatial and temporal resolution. Spatial resolution refers to the smallest physical scale a technique can capture, which must be finer than the flow details and eddies it seeks to resolve. Temporal resolution, on the other hand, dictates the frequency with which measurements are taken over time. The higher temporal resolution allows the technique to capture rapid changes in the flow with greater accuracy [28]. Although some single-point measurement techniques provide high spatial and temporal resolution results, there is generally more interest in methods offering full-field velocity patterns.

The PIV technique provides full-field velocity patterns. PIV was first designed to capture two-dimensional data, enabling time-averaged flow patterns after post-processing. As discussed, PIV determines velocity by correlating tracer positions between two sequential images. The calculation is performed within the interrogation windows, where each window provides a single velocity vector, representing the spatially averaged displacement of particles.

Since PIV resolves time-averaged velocities rather than instantaneous velocities at single points, the actual velocity vector, $\mathbf{u}_{actual}$, can be defined as a combination of the PIV-resolved measurements, $\mathbf{u}$, and a correction term as follow [40]:

$$\mathbf{u}_{actual} = \mathbf{u} + O\left(\frac{\Delta^2}{l^2}\right) \quad (2.11)$$

where l is the integral length and Δ is the size of the interrogation window. If the width of the interrogation window is small compared to the integral length, the following approximation can be made [40]:

$$\mathbf{u}_{actual} \approx \mathbf{u} \quad (2.12)$$

$$\mathbf{u} = \bar{\mathbf{u}} + \mathbf{u}' \quad (2.13)$$

where $\bar{\mathbf{u}}$ refers to the time-averaged velocity vector, and $\mathbf{u}'$ represents the velocity fluctuation [41]. Turbulent kinetic energy, using the three velocity components, can be approximately defined as [40]:

$$k = \frac{1}{2} \left(\langle u_1'^2 \rangle + \langle u_2'^2 \rangle + \langle u_3'^2 \rangle \right) \quad (2.14)$$

PIV can go further in deriving other hydrodynamic information, such as the turbulence dissipation rate.

One approach to calculate the turbulence dissipation rate, ϵ , is through the direct computation of the strain rate tensors, as shown below [42]:

$$\epsilon = 2\nu \langle s'_{ij} s'_{ij} \rangle = \frac{\nu}{2} \left\langle \left(\frac{\partial u'_i}{\partial x_j} + \frac{\partial u'_j}{\partial x_i} \right)^2 \right\rangle \quad (2.15)$$

Direct computation of ϵ as given by Equation (2.15), requires PIV resolves velocity field down to the Kolmogorov scale, λ_k , which might be impossible. According to Saarenrinne et al. [43], a resolution of approximately $2\lambda_k$ is necessary to capture around 90% of the dissipation rate, while a resolution of roughly $9\lambda_k$ is needed to capture 65% of ϵ . Since PIV is limited by the size of its interrogation window, it cannot capture the velocity fields at this small scale directly. As a result, accurately determining ϵ from PIV measurements is challenging.

To reduce the impact of PIV's low spatial resolution on the calculation of ϵ , various techniques have been employed. Sheng et al. [42], inspired by the large eddy and Sub-Grid Scales (SGS) from Large Eddy Simulation (LES), introduced a large eddy PIV to approximate the energy dissipation at scales smaller than the resolution of PIV in a stirred tank.

Another approach to enhance spatial resolution for better understanding of the integral length scale is the multi-block technique. In this method, the area of interest is divided into smaller blocks. Time-averaged and angle-resolved quantities (mean velocity, k , ϵ , integral length scale) are calculated for each block and then combined with data from other blocks. This method maintains high spatial resolution on a larger area of the vessel [44].

Lagrangian Techniques

The Lagrangian velocity frame tracks individual particles or fluid elements moving through space and time. Plotting the position of a single particle over time shows its trajectory. Most Lagrangian studies of stirred tanks have employed RPT or PEPT, where a neutrally buoyant tracer particle is tracked.

The accuracy of the hydrodynamic measurements with tracer particles is highly dependent on the particles' characteristics. While the tracer particles must be neutrally buoyant, this is not the sole criterion they must satisfy. Another criterion is the particle's ability to follow the streamlines of the flow, which is characterized by the Stokes number (St):

$$\text{St} = \frac{\tau_p}{\tau_f} \quad (2.16)$$

where τ_p is the particle response time and τ_f is the fluid response time to an external distur-

bance, such as the impeller movement within a stirred tank. Assuming a Stokesian flow, τ_p and τ_f are defined as [45]:

$$\tau_p = \frac{\rho_s d_p^2}{18\mu} \quad (2.17)$$

$$\tau_f = \frac{D}{\pi N D} \quad (2.18)$$

where ρ_s , d_p , μ , and D respectively are particle density, particle diameter, fluid viscosity, and impeller diameter. The calculated St number occurs at the impeller tip, which has the highest velocity, meaning that the particle further away from the impeller experiences lower Stokes numbers.

When calculating hydrodynamic information from RPT or PEPT data, tracer particle size is another critical factor for data accuracy. PEPT uses micrometer-scale tracers, while RPT employs larger particles, typically a few millimetres in size. Smaller particles can better capture finer flow structures.

Lagrangian data from RPT are collected over an extended period at specified sampling intervals. During post-processing, these data are converted into Eulerian form to obtain time-averaged values.

In a stirred tank, the fluid predominantly moves with a cylindrical trajectory around the tank's axis, making cylindrical coordinates the preferred choice for reporting data. Velocities can be transformed from Cartesian coordinates or calculated directly in the cylindrical frame. Chiti et al. [45] found that while both approaches may produce similar results, calculating velocities in the cylindrical frame yields more accurate estimates, particularly for higher radial velocity components.

For two consecutive positions, (r_i, θ_i, z_i) and $(r_{i+1}, \theta_{i+1}, z_{i+1})$, observed at times t_i and t_{i+1} , the cylindrical midpoint coordinates can be determined as [1]:

$$\begin{pmatrix} x_m \\ y_m \\ z_m \end{pmatrix} = \begin{pmatrix} \frac{x_{i+1} + x_i}{2} \\ \frac{y_{i+1} + y_i}{2} \\ \frac{z_{i+1} + z_i}{2} \end{pmatrix} \quad \text{or} \quad \begin{pmatrix} r_m \\ \theta_m \\ z_m \end{pmatrix} = \begin{pmatrix} \sqrt{x_m^2 + y_m^2} \\ \text{atan}\left(\frac{y_m}{x_m}\right) \\ z_m \end{pmatrix} \quad (2.19)$$

Then the local instantaneous velocity can be defined as follows:

$$\mathbf{u}(r_m, \theta_m, z_m) = \begin{pmatrix} u_r \\ u_\theta \\ u_z \end{pmatrix} = \begin{pmatrix} \frac{r_{i+1}-r_i}{T} \\ \frac{r_m(\theta_{i+1}-\theta_i)}{T} \\ \frac{z_{i+1}-z_i}{T} \end{pmatrix} \quad (2.20)$$

where T is the time difference between two successive positions. The local time-averaged velocity is defined as:

$$\bar{\mathbf{u}}(r_m, \theta_m, z_m) = \frac{1}{P(r_m, \theta_m, z_m)} \sum_{j=1}^P \mathbf{u}_j(r_m, \theta_m, z_m) \quad (2.21)$$

where P is the number of occurrences. The Eulerian fluctuating velocity would be:

$$\mathbf{u}'(r_m, \theta_m, z_m) = \mathbf{u}(r_m, \theta_m, z_m) - \bar{\mathbf{u}}(r_m, \theta_m, z_m) \quad (2.22)$$

and the correlation between fluctuating velocities would be:

$$\langle u'_k u'_l \rangle = \frac{1}{P(r_m, \theta_m, z_m)} \sum_{j=1}^P u'_{kj}(r_m, \theta_m, z_m) u'_{lj}(r_m, \theta_m, z_m) \quad (2.23)$$

The normal and shear stresses (Reynolds stress tensor) can be calculated as:

$$\tau_{kl} = -\rho \langle u'_k u'_l \rangle \quad (2.24)$$

where ρ denotes density.

Once the midpoint positions and their corresponding instantaneous velocities are calculated in cylindrical coordinates, they are averaged over an imposed grid to provide azimuthally averaged time-averaged velocities. As noted earlier, due to the axial symmetry of stirred tanks, azimuthal averaging is a common approach in their analysis. Figure 2.8 illustrates the process of transforming three-dimensional data into two-dimensional representations for stirred tanks.

A key limitation of the data obtained from RPT experiments, as identified by Rammohan et al. [46], is that RPT measurements are not angle-resolved. As a result, they contain contributions from the periodic motion of the impeller, which is referred to as pseudo turbulence. In the impeller region, RPT results deviated from literature by 30% to 50%, and farther from the impeller, values remained lower than reported data. The discrepancies were partly attributed to sampling frequency but were largely associated with the size of the tracer particle

(2.3 mm). The finite size of the tracer particle makes it different from the liquid element and results in limiting the particle to capture the small scale eddies [47].

Recently, in 2022, Patil et al. [48] conducted a similar RPT study to that of Rammohan et al. and compared their data with the same literature (LDA measurements). They also reported an underestimation of mean velocities in the range of 2% to 18% from RPT data.

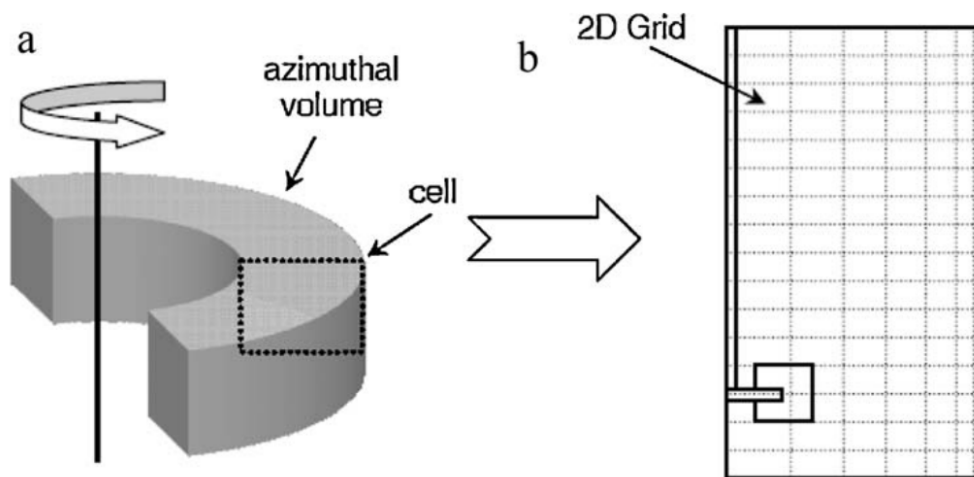


Figure 2.8 Schematic of azimuthal grid for spatially averaging data. [45]

PEPT, like RPT, is a Lagrangian technique that provides time series of particle positions. In stirred tank reactors, post-processed PEPT data are typically azimuthally averaged, following the same principles as RPT.

Tracer velocity can be determined using the same method as applied in RPT by calculating the velocity between consecutive points. However, for slow-moving particles or tracers with low activity, the central difference method produces more accurate results [3].

PEPT has been used in the literature to study stirred tank reactors across single, two, and three-phase systems, although single-phase mixing has received the least attention. Most single-phase mixing studies by PEPT have focused on mean velocity measurements in turbulent regime [45, 49–53], while only a few have explored certain turbulent flow parameters [54, 55].

The lack of reliable data on turbulent properties has been attributed to several factors, including the large ratio of tracer particle size to turbulence structure size, the short half-life of tracer particles, which limits experiment duration and hinders achieving statistical significance in calculated kinematic values, as well as errors associated with the PEPT camera.

While the experimental factors previously mentioned are crucial for obtaining meaningful hydrodynamic data, post-processing algorithms and the treatment of the data afterwards also significantly impact the quality of the information derived from RPT and PEPT raw data.

In one of the most recent studies on stirred tanks using PEPT, Savari et al. [55] applied discrete wavelet transforms to decompose the three-dimensional Lagrangian PEPT flow trajectories into their deterministic (mean or background flow motion) and fluctuating components (small-scale fluid structure). This post-processing approach provided a pointwise description of mean and fluctuating velocities, turbulent kinetic energy, its dissipation rate, and turbulent diffusion coefficients. They have validated their results via 2D PIV measurements .

2.3.2 Current State of Experimental Research on Unbaffled Stirred Tanks

As mentioned earlier in this section, significantly fewer experimental studies have focused on unbaffled tanks. Table 2.3 reviews several studies on unbaffled stirred tanks, noting the technique used, Reynolds number, and whether a central vortex was present. When the vortex was absent, it was due to the top surface being covered with a lid.

It is important to highlight two points: first, most of the studies were conducted in the turbulent regime; and second, all these techniques are only applicable to transparent media and reactor bodies.

Table 2.3 Previous Experimental Work on Unbaffled Stirred Tanks.

Ref.	Method	Central vortex	Reynolds number
Plusa et al. [30]	LDV	Present	162,000
Armenante et al. [56]	LDV	-	71,000, 111,000
Mahmud et al. [57]	LDV	Present	31,922, 53,204, 85,127
Haque et al. [58]	LDV	Present	19,500
Lamarque et al. [59]	LDV	Present	70,000
Dong et al. [60]	LDV	-	1,000
Sharp et al. [61]	PIV	-	4,580
Montante et al. [62]	PIV	Present	41,000
Alcamo et al. [32]	PIV	-	30,000
Fan et al. [63]	PIV	-	13,000

2.3.3 Numerical Simulation of Flow

Computational Fluid Dynamics (CFD) is a sub-field of both fluid mechanics and mathematics, dealing with the numerical solution of mathematical equations that describe fluid flow and related phenomena [64]. The continuous displacement of fluid is described by a combination of mass, and momentum conservation equations.

Providing an analytical solution to the equations governing the physical phenomena of a fluid system is unfeasible; therefore, these equations need to be transformed into a system of algebraic equations that can be solved computationally using CFD. Most CFD software or codes utilize one of the following approaches: the finite difference method (FDM), the finite volume method (FVM), or the finite element method (FEM). All the simulation in this work has been done using an open-source library for solving partial differential equations using the FEM. The assumption here is that the fluid is incompressible and that changes in the fluid's density are negligible.

In an Eulerian reference frame, on a control volume (CV) through which the fluid passes, deriving the mass and energy conservation equations lead to the formulation of governing transport equations known as incompressible Navier-Stokes equations. Mass conservation presented by continuity equation and conservation of momentum is presented by equation known as Cauchy equation [65]:

$$\nabla \cdot \mathbf{u} = 0 \quad (2.25)$$

$$\frac{\partial \rho \mathbf{u}}{\partial t} + \rho(\mathbf{u} \cdot \nabla) \mathbf{u} = -\nabla p + \nabla \cdot \boldsymbol{\tau} + \rho \mathbf{g} \quad (2.26)$$

where ρ is the fluid's density, $\mathbf{u}$ is the fluid's velocity, $\mathbf{g}$ is the gravitational acceleration, p is pressure, and $\boldsymbol{\tau}$ is the deviatoric stress tensor:

$$\boldsymbol{\tau} = \mu (\nabla \mathbf{u} + (\nabla \mathbf{u})^T) \quad (2.27)$$

where μ is the dynamic viscosity of the fluid. If the fluid's viscosity is not too high or its speed is moderate to high, the flow becomes irregular, chaotic, or turbulent, making it unpredictable, unlike laminar flow, which deals with highly viscous, slow-moving, and predictable fluids. In turbulent flow, the velocity field fluctuates randomly over time and is highly disordered in space [66].

Turbulence indeed exists in most engineering and naturally occurring flows, and it plays a significant role by facilitating a more rapid distribution of mass, heat, and momentum compared to laminar flow, which is often advantageous for the process [67]. The dimensionless number Reynolds (Re) defined as the ratio of inertial to viscous forces characterizes the

transition from laminar to turbulent regime and is defined as:

$$\text{Re} = \frac{\rho \mathbf{u} l}{\mu} = \frac{\mathbf{u} l}{\nu} \quad (2.28)$$

where l is the characteristic length of the geometry, $\mathbf{u}$ is the mean flow velocity, ρ and μ are the density and dynamic viscosity of the fluid, respectively, and ν is the kinematic viscosity of the fluid. For example, when calculating the Reynolds number in the impeller region of a stirred tank, l is the diameter of the impeller, and $\mathbf{u}$ would be the impeller velocity [68].

In stirred tanks, the laminar flow regime typically occurs at very low impeller Reynolds numbers ($\text{Re} < 10 - 100$), while the fully turbulent flow regime in the impeller region is generally considered to begin at $\text{Re} \geq 20,000$ [68].

In case of the presence of the turbulence, at each location in a steady-on-average flow, the velocity would be:

$$\mathbf{u}(\mathbf{x}, t) = \bar{\mathbf{u}}(\mathbf{x}) + \mathbf{u}'(\mathbf{x}, t) \quad (2.29)$$

where $\mathbf{u}$ is the velocity, and $\mathbf{u}'$ is the deviation from the average velocity, representing a fluctuation. Therefore, the mean average velocity is given by $\bar{\mathbf{u}} = \mathbf{u} - \mathbf{u}'$ [66]. At any instant, turbulence consists of a broad spectrum of eddy sizes, it can vary as big as the characteristic length scale of the mean flow to much smaller one. The turbulence range of length scale can be estimate by:

$$\frac{l}{\eta} \sim \text{Re}_l^{3/4} \quad (2.30)$$

where l is the integral scale, η represents the Kolmogorov microscale of turbulence, and Re_l is calculated based on l , with the velocity related to the largest eddies [66]. In turbulent flows, various methods can be employed to solve the incompressible Navier-Stokes equations, with the most common being Direct Numerical Simulation (DNS), Reynolds-Averaged Navier-Stokes (RANS), and Large-Eddy Simulation (LES). These techniques primarily differ in the extent of modeling involved and their associated computational cost.

Direct Numerical Simulations

DNS directly solves the Navier-Stokes equations without relying on any turbulence model, using suitable initial and boundary conditions to resolve all scales of motion down to the Kolmogorov scale. As a result, each simulation gives a single representation of the flow, and flow statistics can be obtained by averaging across multiple simulations [41, 69]. DNS, while offering the most accurate solution to the problem, is also the most computationally

expensive method. It becomes impractical at high Reynolds numbers and is typically limited to low or moderate Reynolds numbers [41].

Explicit Large Eddy Simulations

LES addresses the Navier-Stokes equations by resolving larger scales of motion while typically modeling the smaller scales, referred to as subgrid-scales (SGS). This approach makes LES computationally cheaper than Direct Numerical Simulation (DNS), as it involves some degree of modeling. LES applies a filtering operation that decomposes the velocity, $\mathbf{u}$, into the sum of resolved (filtered) scales, $\bar{\mathbf{u}}$, and unresolved scales (SGS), $\mathbf{u}'$. The filtered velocity is three-dimensional and time-dependent, representing the motion of large eddies. The equations for the evolution of the filtered velocity field are derived from the Navier-Stokes equations, with the momentum equation containing the SGS stress tensor from residual motions. The obtained system is open since the apparent stress tensor is missing information of the unfiltered velocity field. The closure to this is obtained by modeling the residual-stress tensor using an eddy-viscosity model. Finally, the model filtered equations are solved numerically for $\bar{\mathbf{u}}$ providing an approximation for the large-scale motions in one realization of the turbulent flow [41].

In this thesis, simulations are done using Implicit Large Eddy Simulations (ILES), implemented through the use of stabilized finite element techniques. In ILES which is often referred to under resolved DNS, there is no modeling of subgrid-scales. In ILES the refinement of mesh determines the resolved length scale. The mesh is finer in the regions with large flow variation so that smaller eddies can be resolved. In stabilized approaches, when the cell size is not sufficiently small to resolve all eddies up to the scale in which viscous dissipation occurs, additional dissipation is numerically introduced through a stabilization term. This term includes a stabilization parameter and the strong residual of the momentum equation [70].

Reynolds-Averaged Navier-Stokes Simulations

The RANS approaches solve the Navier-Stokes equations for the mean velocity field, while the Reynolds stresses are modeled using a turbulence model, typically through the turbulent viscosity hypothesis. In this hypothesis, eddy viscosity is introduced to represent the Reynolds stresses, and is typically determined using $k - \epsilon$ or $k - \omega$ models, which calculate the eddy viscosity based on the turbulent kinetic energy and its dissipation rate [41].

RANS is less expensive than DNS and LES; however, it has a higher degree of modeling, as

it models the entire turbulent cascade, unlike LES, which only models the smaller scales of turbulence. Due to its lower cost, RANS is commonly used in industry; however, it is known to perform poorly for transient problems [71].

Simulation of flow in unbaffled tanks

This section focuses on the current state of the art in numerical simulations of flow in unbaffled stirred tanks. It reviews the approaches taken in each study to simulate turbulence, discusses the Reynolds number and the operating regime of the tank, and describes whether the central vortex was present in the simulations or if it was not considered. In cases where the vortex was simulated, it mentions the methods used to capture its effects.

The work of Guadarrama-Pérez et al. [72] presents an example of an unbaffled tank used for mixing Newtonian and shear-thinning fluids in the laminar regime. They investigated the impact of various geometric configurations and fluid rheology on power consumption, pumping capacity, shear rates, and viscous dissipation, using both numerical and experimental approaches. Similarly, Márquez-Baños et al. conducted a numerical study of an unbaffled tank operating in the laminar regime. They examined ten different volumes around the impeller to determine the region where Metzner-Otto’s constant could be directly extracted from non-Newtonian simulations [73].

In simulations of unbaffled tanks operating in the turbulent regime, most studies have employed Reynolds-Averaged Navier–Stokes (RANS) turbulence models. Table 2.4 summarizes these works. Many of these studies have combined the Volume of Fluid (VOF) method to capture liquid deformation at the air-liquid interface. Additionally, some have simplified the simulations by covering the top surface with a lid to prevent vortex formation around the shaft [56, 74], as the presence of a top vortex adds complexity to the simulation.

Based on the findings from the mentioned works, most of the RANS models are unable to accurately solve the secondary flows, the impeller discharge area, and the region close to the side wall in unbaffled tanks. The Unsteady Reynolds-Averaged Navier–Stokes (URANS) method has also been explored to overcome RANS’ inability to resolve the unsteady behavior introduced periodically by the motion of the impeller. Zamiri et al. simulated an unbaffled tank at $Re = 7.1 \times 10^4$, using the $k - \omega$ SST model. They reported that, unlike RANS, URANS successfully resolved this unsteady behavior and predicted the flow field. However, Tamburini et al. [74] stated that this turbulence model achieved good agreement only for low to intermediate Re values. They suggested the adoption of LES for simulation of unbaffled tanks for fully turbulent regime.

Table 2.4 Summary of works carried out using RANS turbulence models.

Ref.	Turbulence model	Central vortex	Reynolds number
Ciofalo et al. [75]	k- ε	An iterative procedure	10,000
Armenante et al. [56]	k- ε , ASM	-	71,000, 111,000
Montante et al. [62]	k- ε , RSM	Failed to predict	41,000
Glover et al. [76]	SST	VOF	10,000
Haque et al. [58]	k- ε , SST, RST	VOF	19,500
Mahmud et al. [57]	SST, RST	VOF	31,922, 53,204, 85,127
Li et al. [77]	SST k- ω	-	50,400-202,000
Tamburini et al. [74]	SST k- ω , k- ε , SSG	-	33,000
Plusa et al. [30]	k- ε , SST k- ω CC, RSM SO	VOF	162,000

Several studies have utilized Large Eddy Simulation (LES) to model unbaffled tanks. Table 2.5 provides a summary of some of this research. These simulations demonstrated a reliable prediction of both mean and fluctuating velocities and revealed significant turbulent anisotropy throughout the tank. This anisotropy is a key reason why many RANS models, such as the k- ε model, are not considered reliable in this context [59].

Table 2.5 Summary of works carried out using LES.

Ref.	Central vortex	Reynolds number
Alcamo et al. [32]	-	30,000
Lamarque et al. [59]	Front-tracking method	70,000
Yoon et al. [78]	-	4,000, 16,000, 64,000
Ramírez-Cruz et al. [27]	Level-Set method	20,055, 30,008, 35,990

A review of the literature reveals that the majority, if not all, studies conducted on unbaffled tanks focus primarily on either laminar or fully turbulent flow regimes. Several of

these investigations also employed a covering on the top surface to mitigate the formation of a central vortex. One notable exception addressing transitional flow at $Re = 600$ is the study by Tamburini et al. [74]. However, this study also utilized a surface cover to prevent vortex formation. The authors highlighted that the combination of low Reynolds numbers and swirling flow makes flow simulation very challenging, resulting in the failure of many turbulence models.

CHAPTER 3 THESIS OBJECTIVES AND ORGANIZATION

This chapter identifies gaps in the literature to assess RPT's limitations within the current state of the art. It then introduces the motivation and objectives of this thesis, followed by an overview of its structure, which will guide the reader through the subsequent chapters.

3.1 Gaps in the Literature

Current RPT experiments rely on complex nuclear radiation models such as the well-known phenomenological model that define the relationship between the expected radiation received by each detector and the tracer particle's position within the domain. Phenomenological model requires detailed calibration to identify three unknown parameters: the detector's dead time, the radioactive source strength, and the medium attenuation coefficient.

The calibration process requires an operator to manually position the particle at numerous locations within the domain. Therefore, the bottleneck in implementing RPT for systems larger than laboratory scale is the manual calibration process, which can take several days, making it impractical for large-scale applications.

Moreover, in this classical calibration process, the tracer particle is held stationary at each position for a fixed duration, and the irradiation counts are time-averaged. However, during a free-flowing RPT experiment, the RPT hardware records the instantaneous photon counts at each sampling interval while the particle is in motion. The inconsistency between the time-averaged counts when the particle is stationary, and the instantaneous counts during movement introduces an issue known as dynamic bias. As a result, the position calculated by the algorithm using the time-averaged photon count might differ from the particle's actual position in the free-flowing experiment.

In addition to possible sources of error from static calibration, the phenomenological model has its own inherent errors and biases. The model assumes a uniform attenuation coefficient, which may not be a reliable representation of the multiphase systems with large density differences between phases (e.g., gas bubbles and solid particles). It also requires multiple spatial measurements, including two positions from each detector along a line perpendicular to the detector face, the position of the calibration points, and the boundaries of the domain. The model is highly sensitive to the precision of these measurements; inaccuracies of even a few millimeters can lead to errors in the reconstruction of the particle's trajectory and, consequently, in the derived hydrodynamic information.

The comparison of various position reconstruction methods from the literature reveals a significant difference between the theoretical and actual errors observed in real experiments. The actual error is much larger than predicted, which may be caused by several factors mentioned earlier, including dynamic bias, assumptions made in the phenomenological model, and the accuracy of the calibration process. The actual error is at least three times higher than the expected error, highlighting the need for improvements to overcome all or part of the aforementioned sources of error.

3.2 Research Objectives

The main objective of this research is to develop an advanced RPT technique that is independent of nuclear radiation models and capable of accurately reconstructing the position of the radioactive tracer particle in a free-flowing experiment.

Although the primary objective of this work is to develop a model-free RPT technique that accurately reconstruct the trajectory of a tracer particle, it is important to recognize that the ultimate aim of implementing the RPT technique is to derive reliable hydrodynamic information from the reconstructed trajectory. Therefore, the hydrodynamic information obtained from model-free RPT within a stirred tank will be compared with CFD simulation results.

To accomplish the above mentioned objectives, they are broken down into the following sub-objectives, each contributing to their achievement. The sub-objectives are defined as follows:

- Develop a fast and robust position reconstruction algorithm using the principal of inverse finite element method.
- Develop a process to implement a cobot capable of precisely maneuvering the tracer particle within the domain while the RPT hardware simultaneously records the radiation levels from the surrounding detectors. This aims to generate an extensive experimental dataset that correlates particle positions with the radiation levels measured by the detector array.
- Develop a data-driven ANN model for particle position reconstruction. Evaluate the model's performance by comparing the reconstructed trajectory of a particle in motion, as controlled and recorded by the cobot, to the actual trajectory.
- Verify the model-free RPT's ability to predict hydrodynamic information, such as averaged velocity profiles, within a stirred tank by comparing its results to CFD simu-

lations. This will be accomplished by investigating the hydrodynamics in laminar and transitional regimes.

3.3 Organization of the Thesis

The upcoming chapters of this thesis address the sub-objectives outlined in Section 3.2, following the order in which they were accomplished.

Chapter 4 presents the general methodologies applied in this research.

Chapter 5 (Article 1) introduces a novel position reconstruction algorithm that applies the inverse of the Finite Element Method (FEM) for position reconstruction. It also presents the implementation of NOMAD, a derivative-free black-box optimizer, to calculate the unknown parameters of the phenomenological model. Finally, the validation of the proposed algorithm is conducted through a benchmark case and experimental test.

Chapter 6 (Article 2) presents a novel approach that bypasses the need for calibration in RPT experiments. It describes the process of integrating a collaborative robot to strategically guide the tracer particle within the volume of interest, generating an experimental dataset that links positions to the radiation measured by the detectors. It explains how this experimental dataset is used to train an ANN model to predict the particle's trajectory. The accuracy of the proposed method has been validated through experiments. Additionally, the averaged velocity profiles inside a stirred tank operating in a laminar regime have been calculated from the data provided by RPT and verified by comparing them to results from CFD simulations.

Chapter 7 (Article 3) aims to verify the performance of the model-free RPT technique in predicting hydrodynamic information inside an unbaffled, uncovered stirred tank operating in a transitional regime. This information is compared against single-phase and multiphase CFD simulations to assess not only the capability of the model-free RPT technique but also the effect of the free surface on the results obtained from the CFD simulations.

Chapter 8 provides a general discussion on the results of this research. It explains the logical connection of each section of this work and mentions the challenges and obstacles related to each part.

Chapter 9 summarizes the work, highlights its originality and contribution to the existing state of the literature. It also discusses the limitations of the newly developed technique. At the end, it provides suggestions for future studies to overcome these limitations as well as for further improvements.

Appendices A, B, and C present the mathematical calculations used for implementing the Monte Carlo simulation to generate the photon count linked to position using the phenomenological model.

CHAPTER 4 METHODOLOGY

This section provides an overview of the methodology used in this work. While each chapter details its specific approach, this chapter covers the general methodologies applied throughout.

4.1 Experimental Methodology

The Radioactive Particle Tracking experiments were conducted using the facilities available in the RPT laboratory at Polytechnique Montreal. This section outlines the hardware used, the preparation of tracer particles, and introduces the collaborative robot employed for automated data collection, along with an overview of the post-processing and analysis of the experimental data.

4.1.1 RPT Equipment

The RPT system includes a series of Sodium Iodide (NaI) scintillation detectors mounted on aluminum pedestals surrounding the volume of interest. An ORTEC ACE Mate amplifier/discriminator (925-Scint) is connected to each detector to amplify the signals. The discriminators generate a pulse when they detect radiation falling within the energy range of the specified isotope. These pulses are counted by high-speed counters using an Allen-Bradley CompactLogix PLC, which integrates the counts from each detector over a sampling duration of 10 ms. The recorded radiation data time series is subsequently saved in a file for further analysis.

4.1.2 Collaborative Robot

A Doosan cobot (A0912), mounted on a fixed pedestal close to the RPT setup (Figure 4.1a), maneuvers the tracer particle within the study volume. The tracer is attached to the tip of a rod held by a custom-made gripper (4.1b). The cobot records the tracer particle's position along with the corresponding time intervals, saving this data in separate files for further processing.

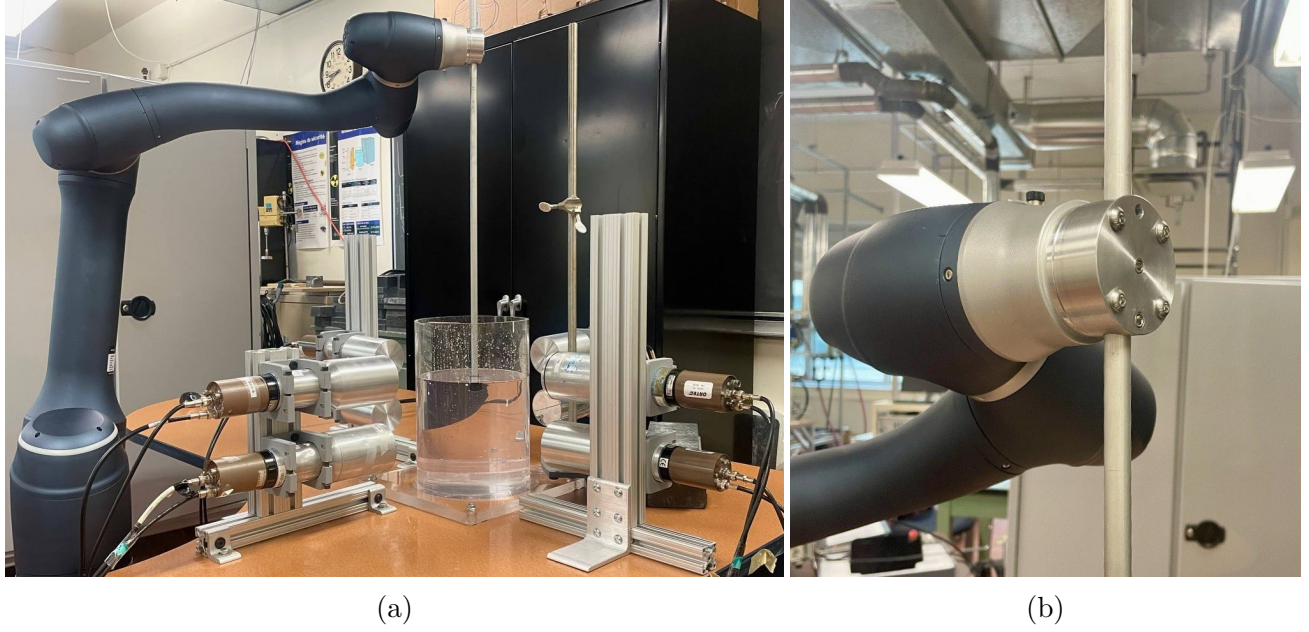


Figure 4.1 Experimental setup: a) sampling the stirred tank by the cobot, b) custom-made gripper.

4.1.3 Tracer Particle Preparation

Scandium-46 (^{46}Sc) was used as the radioactive source to manufacture tracer particles in all experiments. The scandium, initially in sheet form, was cut into small pieces (1 x 1 mm) with a total weight of 0.01 g. These pieces were bonded together and, once dried, coated with a layer of epoxy glue, intentionally including air pockets to compensate for the higher density of scandium relative to the liquid. The amount of glue and air was fine-tuned by testing the tracer in the liquid to ensure it achieved neutral buoyancy. Once the final composition was achieved, the particles were sent to the Slowpoke Laboratory at Polytechnique Montreal for activation. All tracers used in this work had an activity of approximately $125\ \mu\text{Ci}$.

4.2 Post-processing Experimental Data

The time series of photon counts obtained from the RPT system, along with the position data sampled by the cobot, were post-processed using various Python tools. This processed dataset was then used to train an ANN model for particle position reconstruction. The ANN model was developed with Keras, a high-level neural networks API running on top of TensorFlow, an open-source machine learning framework. Using the trained ANN, the time series of photon counts from the free-flowing experiment was reconstructed to determine the

particle’s trajectory over time. This trajectory data was subsequently post-processed and transformed into hydrodynamic information using Python-based tools. Finally, the extracted hydrodynamic data was visualized using Matplotlib, and all plots presented in this work were generated with this library.

4.3 CFD Simulation

The CFD simulations in this work were carried out using Lethe, an open an open-source implicit large-eddy simulation (ILES) software [79]. Lethe is based on deal.II, an open-source library to solve partial differential equations using finite element method [80]. Lethe solves incompressible Navier-Stokes equations using a continuous Galerkin Finite-Element formulation with an SUPG/PSPG (Streamline-Upwind/Petrov-Galerkin and Pressure-Stabilizing/Petrov-Galerkin) stabilization approach [79].

CHAPTER 5 ARTICLE 1 : AN IMPROVED POSITION RECONSTRUCTION METHOD FOR RADIOACTIVE PARTICLE TRACKING

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Declaration: The author of this thesis has contributed to the literature review, code development, performing the experiments, analysis of the results and the writing of the article.

Abstract: This work introduces a novel reconstruction method exploiting the inverse of Finite Element Method (FEM) to reconstruct positions from ray tracing experimental measurement such as those obtained by radioactive particle tracking. FEM based Position Reconstruction (FEM-PR) relies on shape function evaluation within simplex elements (i.e. tetrahedron) for position reconstruction using a mean-square problem, which has proven to be robust, accurate, and fast. Tracer positions are reconstructed under the condition that least-squares compatibility is maintained between experimental counts and interpolated values within inverse elements. This approach is implemented in a module within the open-source software, Lethe-RPT, enabling user to carry out radioactive particle tracking (RPT) experiments from calibration to tracer position reconstruction. The test benchmark and experimental setup are described. For the same test cases, the reconstruction error is investigated using an increasing number of inverse elements and Monte Carlo (MC) iterations. The accuracy and robustness of FEM-PR concerning the inevitable measurement errors due to calibration, measurement systems, and the quantized nature of γ -rays is demonstrated. This approach provides a new view for immediate and precise position reconstruction. FEM-PR reconstructs the tracer position with an accuracy of 0.08 mm. Approximately 3.37 ms is required to reconstruct one position on a desktop computer, which is less than the sampling time for experimental data collection.

5.1 Introduction

Developing a reliable experimental approach for designing chemical reactors is one of the key challenges in multiphase flow reactor engineering. Scaling up chemical reactors requires a reliable description of the hydrodynamics. The inability to properly describe the flow pat-

terns will inevitably lead to poor performance of scaled-up reactors [1, 21, 81]. To address this need, several experimental techniques have been developed to study the dynamics of multiphase flows. These techniques are categorized as intrusive and non-intrusive methods. As the name suggests, intrusive methods measure fluid velocity by interacting with the fluid. Such methods include Pitot tube, Hot Wire Anemometry (HWA), and optical probes. Non-intrusive techniques characterize the flow without disturbing the flow. Laser Doppler Anemometry (LDA), Particle Image Velocimetry (PIV), Positron Emission Particle Tracking (PEPT), Magnetic Resonance Imaging (MRI), and Radioactive Particle Tracking (RPT) are examples of non-intrusive techniques [82].

Unlike other non-intrusive methods, RPT copes well with opaque systems [83]. Other methods which use light or laser fail in systems with a large volume fraction of the dispersed phase. Due to the extensive demands of RPT for computational resources, researchers use this technique offline. It is nevertheless desirable to monitor the hydrodynamics of pilot scale and industrial reactors in real time with high accuracy [21, 84, 85].

The objective of this paper is to present a highly accurate and noise-insensitive method that would apply to any domain geometry and experimental configuration. To accomplish this, our work presents a novel particle position reconstruction algorithm called FEM-PR. This technique follows the principle of the inverse Finite Element Method. The inverse problem of FEM was first introduced to enable real-time tracking of the three-dimensional displacement field of a structure, often called shape sensing. iFEM reconstructs structural deformations using experimental strains and minimizing a weighted least-squares function [86]. The FEM-PR method, also derived from the inverse problem of FEM, can monitor the dynamics of a multiphase flow system in real-time. FEM-PR uses the experimental data of photon counts received from a radioactive particle and solves a least-squares function between experimental and numerical values. The implementation of this technique makes RPT method more practical for industrial applications and accelerates research and development of more advanced reactors. This novel position reconstruction strategy has been implemented in Lethe-RPT, a module of Lethe [79], an open-source Computational Fluid Dynamics (CFD) library. We also present the implementation of the NOMAD blackbox optimizer to proceed with the model parameter tuning of the RPT experiments. Blackbox optimizer overcomes the Monte Carlo model's stochastic nature, and it obtains a precise set of unknown parameters for each detector from the calibration data.

In the following sections, we will introduce the mathematical framework of the RPT. Then, we present a derivative-free, blackbox optimizer that implements the Mesh-Adaptive Direct Search (MADS) algorithm for tuning the model parameters. Afterward, we introduce

the FEM-PR approach. Finally, using benchmark cases and experimental validation, we demonstrate the capability and accuracy of the proposed algorithms for tracer position reconstruction.

5.2 Methodology

5.2.1 Monte Carlo model for photon counts calculation

RPT aims to reconstruct the motion of a radioactive particle by capturing the γ -rays it irradiates. Each RPT experiment includes the following steps: calibration, experiments, and position reconstruction [83].

A phenomenological model proposed by Beam *et al.* [2] associates the tracer position and the received photon count by a given detector as:

$$C = \frac{T\nu R\phi\xi(\vec{X})}{1 + \tau\nu R\phi\xi(\vec{X})} \quad (5.1)$$

where T is the sampling time (in seconds), ν is the number of γ -rays emitted by each disintegration, R is the activity of the tracer (in Beq), ϕ is the peak-to-total (photo peak) ratio, τ is the dead time of the detector (in seconds) per accepted pulse, and $\xi(\vec{X})$ is the efficiency of the detector associated with the tracer position which can be expressed as [2]:

$$\xi(\vec{X}) = \oint_{\Omega} \frac{\vec{r} d\vec{A}}{\|\vec{r}\|^3} f_a(\alpha, \theta) f_d(\alpha, \theta) \quad (5.2)$$

where, $\vec{r}$ is a vector from the position of the tracer to a point which γ -rays enter to the detector, $d\vec{A}$ is the external surface vector normal to the surface at the contact point on the detector crystal, and Ω is the solid angle subtended by the radiation. Since there is no analytical solution to calculate the efficiency (except for some simple problems), a numerical method based on the Monte Carlo technique can compute Equation (5.2) [15]. This method generates thousands of random photon path directions towards a detector to estimate the surface integral of Equation (5.2). Beam *et al.* employed the Monte Carlo technique, which estimates this equation as:

$$\xi(X) = \frac{1}{N} \sum_{i=1}^N \omega_i f_a(\alpha, \theta) f_d(\alpha, \theta) \quad (5.3)$$

where N is the number of generated photon paths, the angles α and θ specify the direction of each traced ray from the tracer position, ω_i determines the solid angle for the particular

selection of α and θ , and $f_a(\alpha, \theta)$ and $f_d(\alpha, \theta)$ are:

- f_a is the probability of non-interaction between the γ -rays emitted within Ω and the material inside the column:

$$f_a(\alpha, \theta) = \exp(-\mu_r e(\alpha, \theta)) \quad (5.4)$$

- f_d is the probability function of the γ -rays interaction with the detector:

$$f_d(\alpha, \theta) = 1 - \exp(-\mu_d d(\alpha, \theta)) \quad (5.5)$$

In Equation (5.4), μ_r is the total linear attenuation coefficient of the medium inside the reactor and $e(\alpha, \theta)$ is the length of the path travelled by a photon inside the reactor. In Equation (5.5), μ_d is the total linear attenuation coefficient of the detector, and $d(\alpha, \theta)$ is the effective distance travelled by γ -ray inside the crystal along the direction of α and θ angles [2].

However, some parameters of this model, such as the detector's dead time, source strength, and the medium attenuation coefficient, which is a parameter involved in calculating the efficiency, are unknown and unique to each experimental system.

The calibration phase plays a central role in finding the model unknowns. During the calibration procedure, the tracer is placed at many known positions inside the domain. The RPT system hardware records the γ -rays for a specified time to reach acceptable counting statistics at each position. The calibration process generates a 3D map of the photon counts received by each detector from the static positions. Finally, a parameter tuning algorithm compares the counts from the experimental measurement and the obtained counts from the Monte Carlo algorithm to find the model's unknown parameters. The algorithm obtains these unknowns by finding the optimum value of the Equation (5.6).

$$\min \sum_{i=1}^N \left(\frac{C_i - M_i}{C_i + M_i} \right)^2 \quad (5.6)$$

Where N is the number of sampling points, C is the calculated counts and M is the measured counts.

At the final step, a reconstruction algorithm converts the recorded photon counts to the instantaneous tracer positions. Over the past two decades, researchers have developed various techniques for more immediate and accurate position reconstruction, including the least-squares search algorithm, real coded genetic algorithm, and machine-learning algorithms

[14, 18, 21, 85]. Support Vector Regression (SVR) was the most adequate technique among the proposed ones. Based on the experimental validation, this algorithm's Mean Absolute Error (MAE) was 1.45, 1.41, and 2.66 mm in the x, y, and z coordinates, respectively [21].

5.2.2 Phenomenological model parameter optimization using MADS

Description

Obtaining the parameters of the phenomenological model (see Eq. 5.1) using gradient-based optimization is difficult. The stochastic nature of the Monte Carlo model makes the calculation of the derivatives of error between the model-predicted counts and the experimental counts challenging. Therefore gradient-based optimization methods become rapidly ineffective. Due to all the limitations and complexity of implementation of the existing optimization methods, we propose to combine MADS, a non-gradient-based direct search optimization algorithm with the Monte Carlo technique to optimize the objective function. We achieved this by combining the NOMAD blackbox optimizer, which is a C++ implementation of the MADS algorithm, [87, 88] into the open source Lethe-RPT code.

This class of algorithms includes three steps: search, poll, and update. To begin the process, the algorithm determines an initial mesh M_0 of size Δ_0^m which represents a discretization of the solution domain. The MADS algorithm uses an initial point at each iteration to calculate the objective function at a finite number of points and put away the infeasible points. MADS compares the objective function values at feasible trial points with the current best possible value that it has found for the objective function so far. Each of the trial points lies on the current mesh with size Δ_k^m . By finding an improved mesh point the algorithm stops the iteration and the next iteration would start with a new incumbent solution $x_{k+1} \in \Omega$ with $f_\Omega(x_{k+1}) < f_\Omega(x_k)$. If the search algorithm fails to find a mesh point that decreases the objective function it calls for the poll step. At this step MADS evaluate the objective function at neighboring trial points which exist on a frame P_k with larger or equal size Δ_k^P from the previous mesh $\Delta_k^m \leq \Delta_k^P$. At the end, if both search and poll steps fail to find a better solution, the algorithm would decrease the mesh size and redo the first step using the current incumbent solution x_k . If either the search step or the poll step becomes successful, the algorithm will increase the mesh size and perform step 1 with the new incumbent solution x_{k+1} [8, 88, 89]. In the next section we discuss the integration of MADS algorithm with Lethe-RPT to optimize the detector's parameters.

Implementation in Lethe-RPT software

To start the optimization, NOMAD receives the input variables and their constraints presented in Table 5.1.

Table 5.1 Input, output, and constraints for optimization of detector parameters.

Input Variables	Output Variables	Constraints
Position of the detector face center	Tracer activity	Tracer activity > 0
Position of a point on the detector axis	Detector dead time	Detector dead time > 0
Position of the calibration points	μ_r	$\mu_r > 0$
Experimental photon counts at calibration points	-	-
Initial guess for unknown variables	-	-

Prior to the optimization we set the parameters for the detectors (i.e., the photopeak-to-total ratio), the source (the crystal's attenuation coefficient, the number of γ -rays emitted), and the experimental parameter (i.e., the sampling period) as well as those for optimization algorithm (e.g., optimization constraints and the maximum number of evaluation).

To evaluate the objective function (Equation (5.6) at each step), NOMAD uses the parameters from the previous optimization step and it recalculates the counts implementing the Monte Carlo code. The algorithm stops once it reaches the maximum number of iterations. Figure 5.1 shows the algorithm of the parameter tuning process.

5.2.3 FEM-PR for back calculating the tracer position

Position reconstruction algorithm

First, FEM-PR filters the noise resulting from the Monte Carlo model by using an $\mathcal{L}^2$ orthogonal projection of the Monte Carlo model onto the domain. Then, the algorithm searches through the cells and it minimizes the least-squares error between the photon count obtained from the FEM interpolation and the experimental count for all the detectors in each cell. Only the cell that satisfies the inverse of the finite element interpolation contains the tracer position. This novel approach uses the information from the previous position of the tracer and searches locally within the space around the last found position.

The first step in approximating a property using the finite element method is to build up a mesh over the domain of interest. Therefore, the 3D domain is discretized using linear

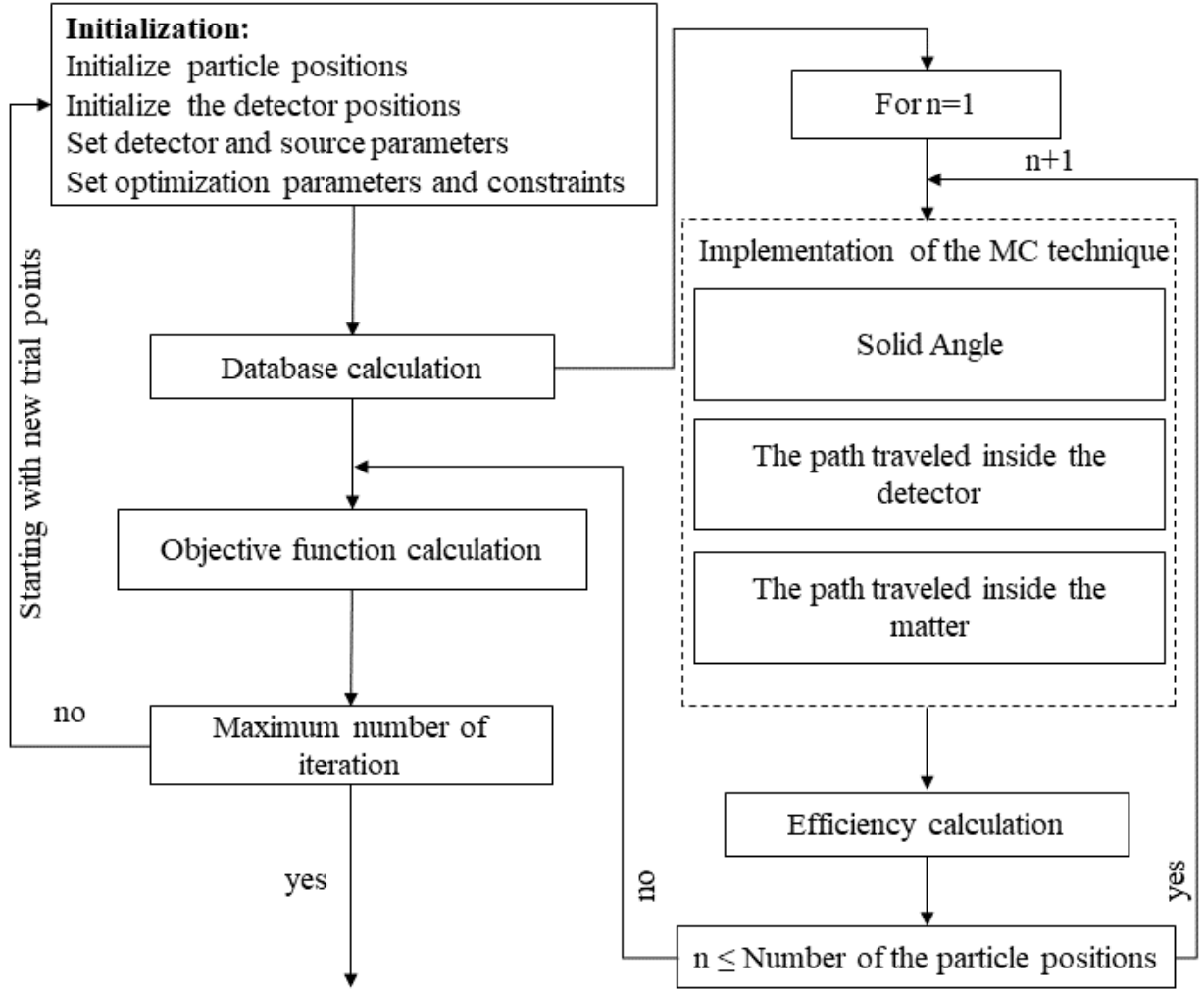


Figure 5.1 Optimization strategy algorithm.

tetrahedral elements. In the case of continuous approximations, isoparametric formulations can be used to approximate physical properties. Isoparametric elements are defined using natural and global coordinates. Each element's domain is mapped onto the global coordinate system using the shape functions constructed on its reference element. Therefore, an approximation of the physical values inside an element can be written in terms of shape functions and natural coordinates as:

$$\varphi_{x,y,z} = \sum_{i=1}^n N_i \varphi_i \quad (5.7)$$

where φ is a continuous physical value, $\varphi_{(x,y,z)}$ is the value at the global coordinate point (x,y,z) , φ_i are the values at the nodes of the element and n is the number of nodes of the element ($n = 4$ for a linear tetrahedral element), and N_i are the shape functions defined as

follows [90]:

$$N_1 = 1 - \eta - \xi - \zeta, \quad N_2 = \eta, \quad N_3 = \xi, \quad N_4 = \zeta \quad (5.8)$$

In case of RPT, the isoparametric approximation combined with a search algorithm can calculate the tracer's position using a set of photon counts received at each detector. The search algorithm goes through each cell inside the domain and solves a least-squares cost-function defined as the difference between experimental and numerical values:

$$F = \sum_{i=1}^{N_D} (C_{num}^i - C_{exp}^i)^2 \quad (5.9)$$

where C_{num} and C_{exp} are the numerical and experimental counts, respectively, N_D is the number of detectors, and i is the number associated with the detector. The main search hypothesis is that the tracer is located inside the search element. The numerical count value within the finite element framework can be calculated using the isoparametric approximation previously presented:

$$C_{num} = \sum_{i=1}^4 N_i C_i \quad (5.10)$$

The minimization of the least-squares function can be written by setting its partial derivatives to zero, which in vector-form results in:

$$F' = \begin{bmatrix} F_1 \\ F_2 \\ F_3 \end{bmatrix} = \begin{bmatrix} \frac{\partial F}{\partial \eta} \\ \frac{\partial F}{\partial \xi} \\ \frac{\partial F}{\partial \zeta} \end{bmatrix} = \begin{bmatrix} 0 \\ 0 \\ 0 \end{bmatrix} \quad (5.11)$$

Since the elements are linear, this problem can be solved using the following system of equation:

$$\begin{bmatrix} \frac{\partial F_1}{\partial \eta} & \frac{\partial F_1}{\partial \xi} & \frac{\partial F_1}{\partial \zeta} \\ \frac{\partial F_2}{\partial \eta} & \frac{\partial F_2}{\partial \xi} & \frac{\partial F_2}{\partial \zeta} \\ \frac{\partial F_3}{\partial \eta} & \frac{\partial F_3}{\partial \xi} & \frac{\partial F_3}{\partial \zeta} \end{bmatrix} \begin{bmatrix} \eta \\ \xi \\ \zeta \end{bmatrix} = -F' \quad (5.12)$$

where the matrix corresponds to the Jacobian of the element. Solving this system of equations, the values of natural coordinates can be obtained. To decide whether the current parent cell can be considered as an appropriate cell for the particle position, the following conditions are checked and both need to be satisfied:

1. $\eta, \zeta, \xi \in [0, 1]$
2. $1 - \eta - \zeta - \xi \in [0, 1]$

Afterwards, the shape functions on the reference element are used to map the tracer position to the global coordinate system, $\mathbf{x}$:

$$\mathbf{x} = N(\varepsilon)\tilde{\mathbf{x}} \quad (5.13)$$

where $N(\varepsilon)$ denotes a matrix composed of the shape functions, and $\tilde{\mathbf{x}}$ corresponds to position in natural coordinates.

$\mathcal{L}^2$ orthogonal projection of the Monte Carlo model

Photon counts fluctuate in space due to the stochastic nature of the Monte Carlo technique, which can mislead the algorithm in position reconstruction. The algorithm uses the photon counts at the nodes of each element; therefore, the estimate of the tracer's position may be inaccurate if the nodal values are offset. Thus, rather than directly calculating photon counts at the nodes using the beam model, we approximate the value of photon counts at the nodes of the mesh by finding the coefficients c_j which minimize the following equation [91]:

$$\min_{c \in \mathbb{R}} ||f_i - \sum_j c_j \varphi_j|| \quad (5.14)$$

where c_j is the nodal photon counts and f_j is the photon counts calculated using the beam model (see Eq. 5.1). Finding the value of c_j is done by assembling and solving the following finite element system in the domain Ω :

$$\sum_j \int_{\Omega} \varphi_i c_j \varphi_j d\Omega = \int_{\Omega} \varphi_i f_i d\Omega \quad (5.15)$$

Through the solution of this system of equations, we can filter the oscillation of photon counts and have smooth photo counts as a function of position in space.

5.3 Results and discussion

5.3.1 Model parameter optimization using MADS

The case-study we use to validate our approach is a two-dimensional rectangular geometry having a 0.22 m length and 0.16 m width, and 0.02×0.02 m square mesh size (108 points). Figures 5.2a and 5.2b represent the experimental setup for the RPT tests and the configuration of the two-dimensional geometry, respectively. First, we evaluated the proposed optimization strategy using a simple case of one-dimensional tracer placement towards the detector. Next, we evaluated the algorithm implementing a two-dimensional arrangement of tracer placement. All the parameters used to obtain the results presented in this section

are shown in Table 5.2. The detector saturation test inspired the one-dimensional validation

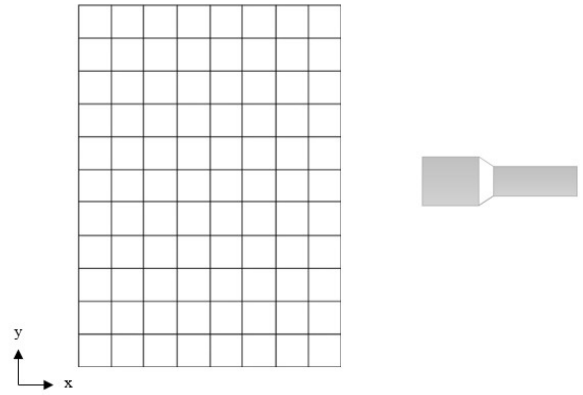
Table 5.2 Parameters for optimization strategy.

Parameter	Value
Source	Scandium-46
Number of γ -rays (ν)	2
Total linear attenuation coefficient of crystal (μ_d)	2.477 m^{-1}
Photopeak-to-total ratio (ϕ)	0.4
Detector's crystal type	Sodium iodide (NaI)
Crystal length	7.62 cm
Crystal diameter	7.62 cm
Sampling time (τ)	1 s

approach. In this test, the radioactive tracer is moved along a line perpendicular to the detector's face. The detector is placed at a distance of 0.26 m from the center of the geometry. This test clarifies the space in which the detector cannot sense the actual radiation, called the blind zone.



(a) Experimental RPT system

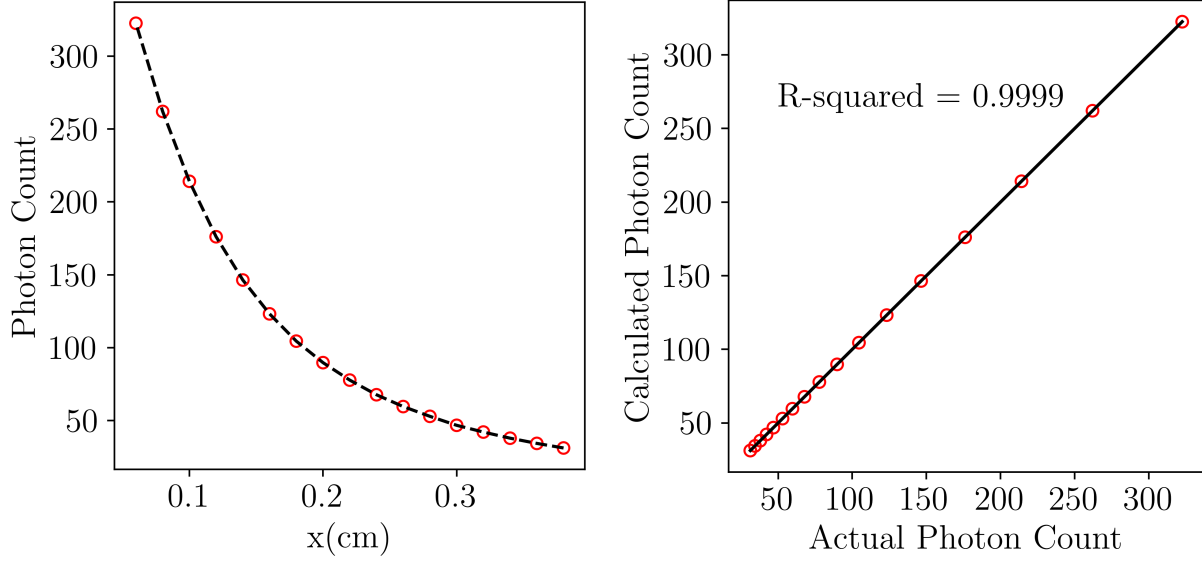


(b) Pattern used for recording the photon counts

Figure 5.2 Experimental setup for validation of optimization strategy.

Figure 5.3a illustrates the experimentally measured and the predicted counts at several points on a straight line perpendicular to the detector face (one-dimensional validation). All the parameters optimized by NOMAD used in calculating the predicted photon counts are given in Table 5.3. Figure 5.3b shows a parity plot of the calculated photon counts by Lethe-RPT and the measured photon counts. The simulated data are in agreement with the

measurements, which indicates the validation of the NOMAD blackbox optimizer in tuning the detector parameter using one-dimensional experimental data.



(a) Predicted and measured counts comparison (b) Parity plot: predicted vs. measured counts

Figure 5.3 1D validation of optimization strategy, with the black dashed line indicating the calculated photon count and the red dots showing the experimental photon count.

For a two-dimensional assessment of the algorithm the received photon counts by the detector from 108 points, and their associated positions feed the algorithm. Table 5.3 displays the results of the 2-dimensional detector's parameter tuning procedure. Figure 5.4 illustrates the parity plot of the calculated count using the optimized parameters and the experimentally measured counts. The 5% and -5% limits are also displayed in the figure. The simulated counts correspond well with the measured counts. The maximum deviation was about 2%, and the minimum was less than 1%. This indicates the validation of the Lethe-RPT code to calculate counts received by the detectors in the RPT calibration process.

Table 5.3 Tuned parameters by NOMAD for 2D experiment.

Parameter	Value
Detector dead time	$9.74 \times 10^{-6} \text{ s}$
Tracer activity	$7.39 \times 10^6 \text{ Bq}$
μ_r	0.457388

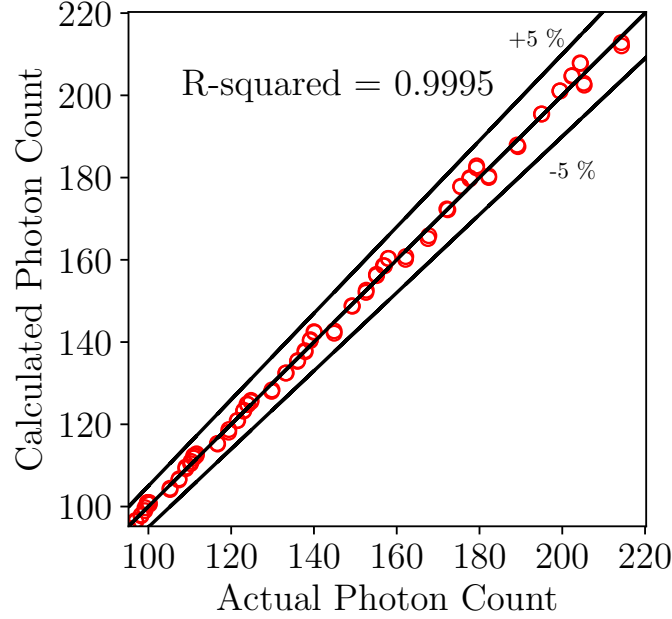


Figure 5.4 2-D validation of optimization strategy.

5.3.2 FEM-PR for position reconstruction

Theoretical study and sensitivity analysis

We carry out sensitivity analysis to evaluate the impact of the number of Monte Carlo iterations and number of cells on the accuracy of the FEM-PR algorithm. This virtual test determines the inherent error of the method independent of the experimental noise. The geometry is a cylinder with a 0.02 m radius and a 0.1 m height surrounded by six detectors, Figure 5.5 shows the detector configuration around the cylinder.

We skip the calibration step and take default values for the three unknown parameters considering that all the detectors have the same physical properties. The mesh domain is generated through `deal.II GridGenerator` subdivided cylinder with tetrahedron elements [92].

We generated a spiral trajectory of 100 points and calculated the photon counts at each of the 6 detectors to verify FEM-PR's performance for 3D position reconstruction. Using the calculated photon counts, we reconstructed their positions. The algorithm was evaluated for 28 cases, which consisted of 7 mesh sizes and 4 different number of Monte Carlo iterations. We performed the simulations with different meshes ranging from 1920 to 30720 cells.

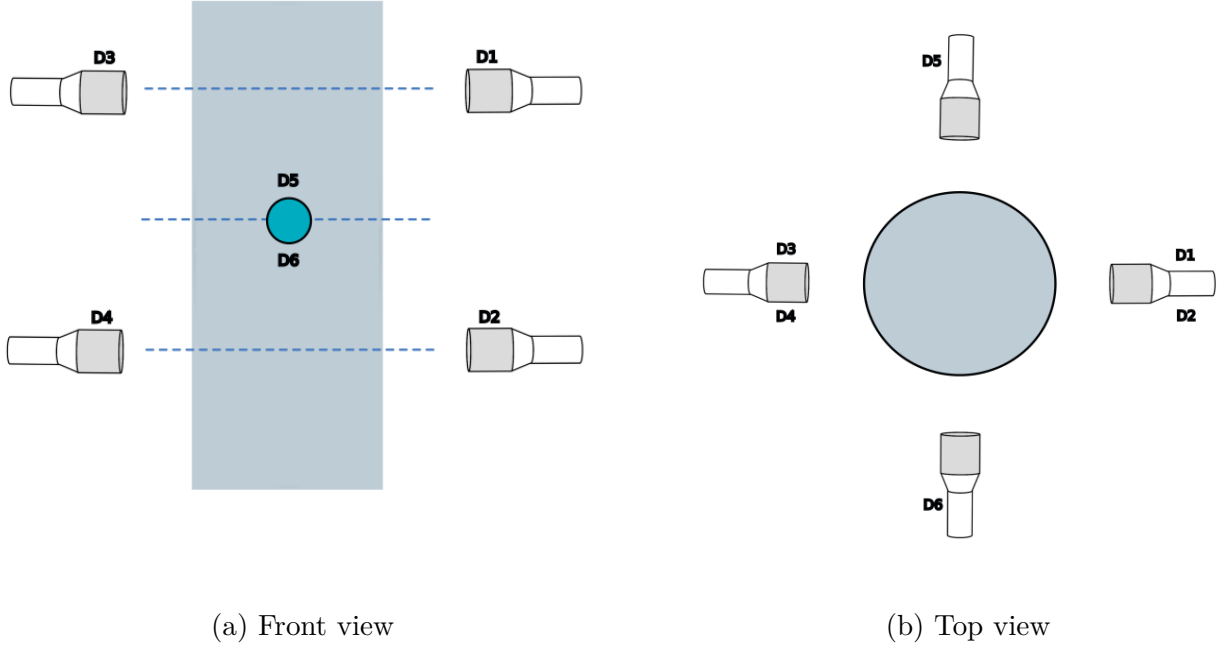


Figure 5.5 RPT system simulated with Lethe-RPT.

We use performance indices such as Mean Absolute Error (MAE), Standard Deviation (SD), and Mean Euclidean Distance Error (MEDE) to assess the accuracy of the FEM-PR algorithm on the spiral trajectory. Figure 5.6 illustrates two key points:

- A sharp drop in error by increasing the number of cells from 3840 to 7680: the coarser meshes do not accurately represent the original geometry since they approximate curved boundaries using linear elements. Consequently, points close to the wall fall outside the mesh, making it difficult to reconstruct their location. Refining the mesh allows us to fit the boundary and avoid reconstructing positions outside it. Because of this, there is a drop in reconstruction error by increasing the number of cells from 3840 to 7680.
- At a Monte Carlo iteration of 1000, which has the highest relative error in photon counts calculation (see Fig. 5.7), refining the mesh results in a slight improvement in the algorithm performance. In contrast, at higher numbers of Monte Carlo iterations, more levels of refinement considerably decrease the error. In the first case, when the error of the Beam model (see Eq. 5.1) is high, all the points that the $\mathcal{L}^2$ projection uses for estimating the nodal count are also contaminated to a significant amount of error. Therefore, mesh refinement and increasing the available data points for the

$\mathcal{L}^2$ projection does not filter the stochastic error of the Monte Carlo model and the reconstruction remains inaccurate. Conversely, at higher Monte Carlo iterations, when the data points in the domain are less impacted by the stochastic nature of Monte Carlo technique in the Beam model, and in fact, they are a valid representation of γ -rays radiation, increasing the number of cells allows the $\mathcal{L}^2$ projection to cancel the noise at full capacity. Consequently, searching through a clean data set reduces reconstruction error.

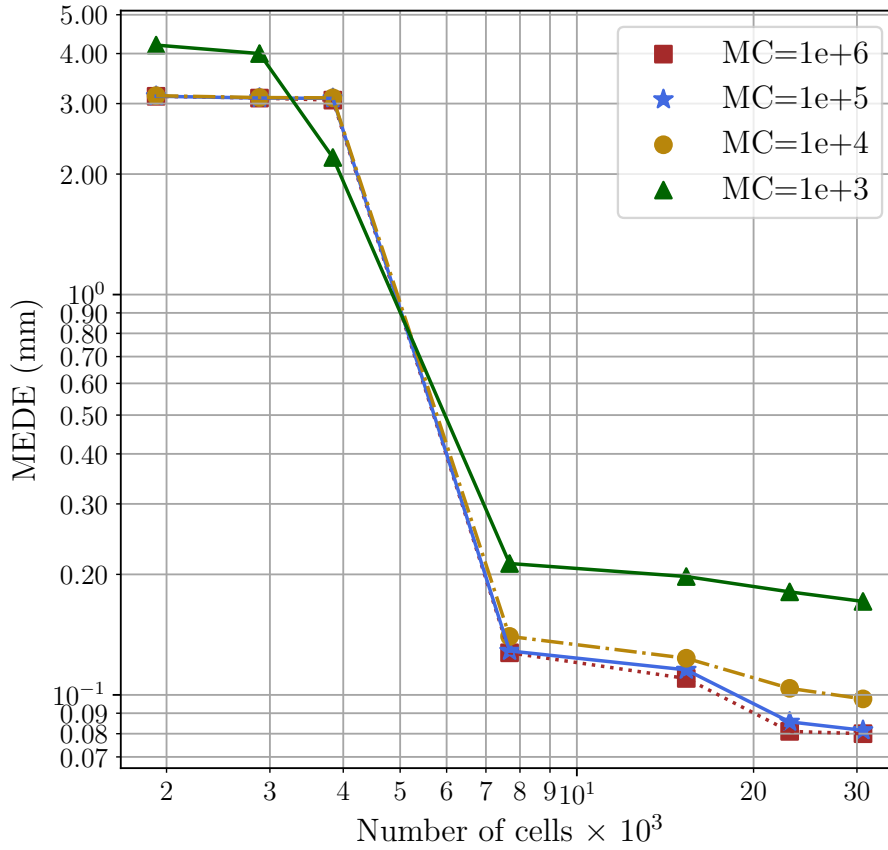


Figure 5.6 Comparison of MEDE and its dependency on cell numbers and Monte Carlo iterations for the spiral trajectory reconstruction.

Values in Table 5.4 are the performance indices of the spiral trajectory reconstruction using Monte Carlo iterations equal to $1 + e6$. These data show a higher and more uneven error distribution for a coarser mesh. Figure 5.8 visualizes the spiral trajectory in XY and XZ planes.

Table 5.4 Reconstruction performance indices for $1 + e6$ Monte Carlo iteration.

Number of cells	MAE (mm)			SD (mm)			MAED (mm)
	X	Y	Z	X	Y	Z	
1920	0.4	0.48	3	2.2	2.5	16.7	3.13
2880	0.29	0.52	3	1.9	2.9	16.7	3.1
3840	0.4	0.46	3	2.2	2.5	16.7	3.06
7680	0.04	0.06	0.09	0.08	0.13	0.06	0.13
15360	0.03	0.04	0.07	0.03	0.04	0.07	0.11
23040	0.02	0.04	0.06	0.02	0.04	0.05	0.08
30720	0.02	0.04	0.05	0.02	0.04	0.04	0.08

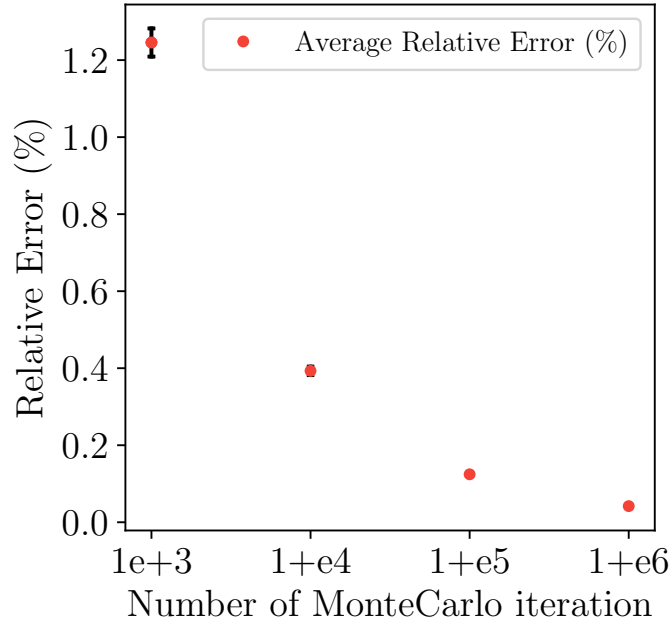


Figure 5.7 Evaluation of error confidence interval for photon count generation regarding the number of Monte Carlo iterations for 2865 points inside the geometry.

The required computational time for reconstructing one position is around 3.37 ms, it is lower than the sampling time of the RPT hardware, 10 ms, which shows the capability of the FEM-PR algorithm for real time position reconstruction. This time does not include the detector parameter tuning and $\mathcal{L}_2$ orthogonal projection of the Monte Carlo model since they must be done before running the experiment.

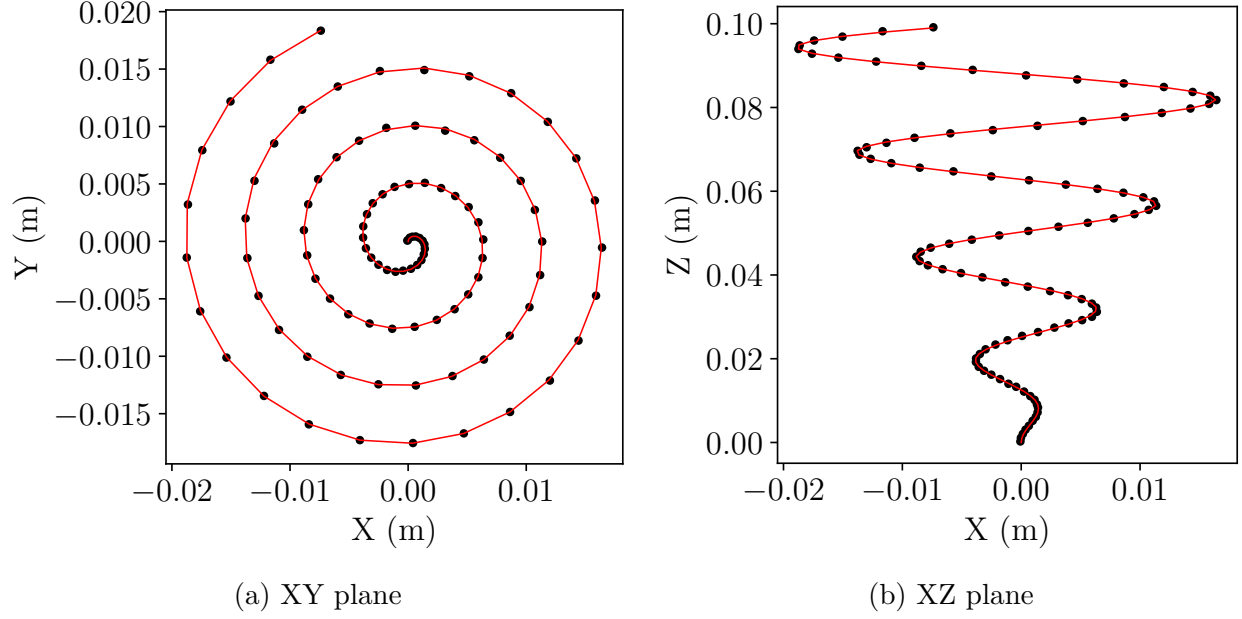


Figure 5.8 Trajectory of the radioactive particle

Assessment and validation of the reconstruction algorithm

Finally, we assess the validity of the reconstruction algorithm with a reconstruction of a set of experimental points. The RPT system to check the validity of the proposed reconstruction algorithm consists of a square with dimensions of 0.12×0.12 m. We used a sealed Sc-46 tracer with a size of approximately 3 mm. We placed four ($2'' \times 2''$) NaI scintillation detectors around the plane to monitor the γ -rays radiation. The detectors were in one plane perpendicular to the z-axis and the distance between the detectors and the center of the plane was 0.24 m. The photon count recorded by all detectors was saved on a computer through a data acquisition interface. The acquisition time at each position was 10 ms (data acquisition frequency of 100 Hz). We performed the calibration at 169 different locations. The registered counts of each of the four detectors, and their associated positions are the inputs of the detector's optimization algorithm. Using the tuned parameters for each detector we generate the dictionary for the corresponding geometry.

Figure 5.9b shows the absolute error in predicting x and y coordinates over the entire dataset, and Figure 5.10 shows the parity plot of experimental and calculated coordinates for each point. The coefficient of determination (R^2) is 0.9997 for the x coordinate and 0.9993 for the y coordinate. The MAE error observed in x and y is 0.49 and 0.76 mm, with standard deviations of 0.68 and 0.98 mm, respectively. Figure 5.9a compares positions (x,y) predicted

by Lethe-RPT and measured by experiment. According to these results, the algorithm can reconstruct the position with excellent precision, which is better than the error reported by Yadav *et al.* (1.45 and 1.41 mm in x and y coordinates) [21].

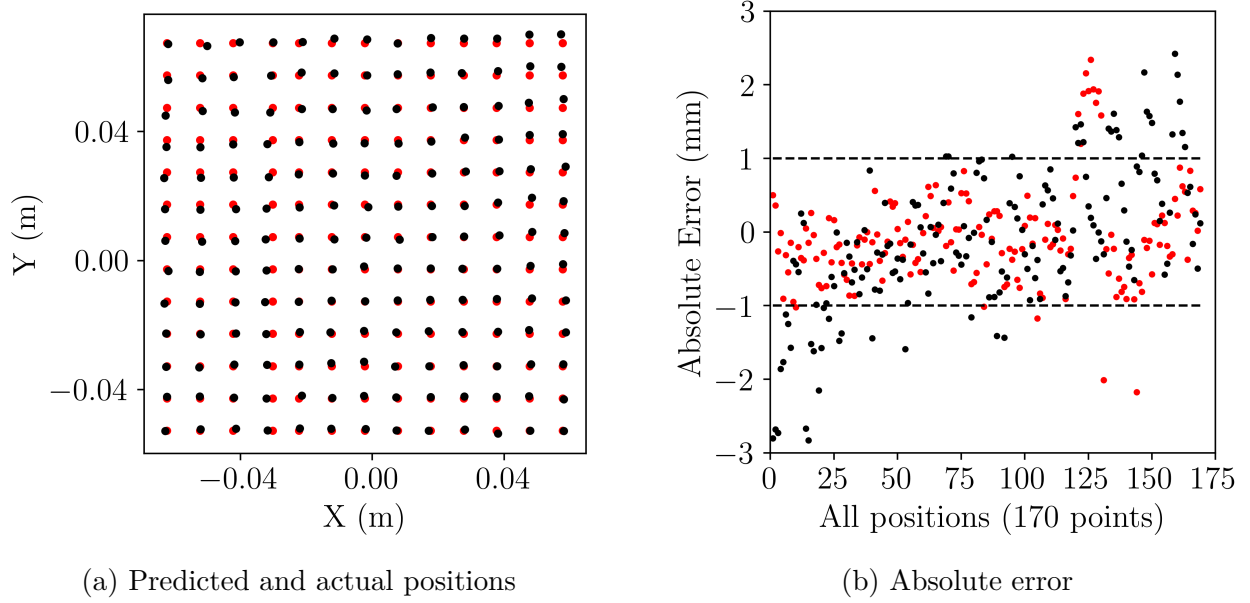


Figure 5.9 Predicted and actual positions error.

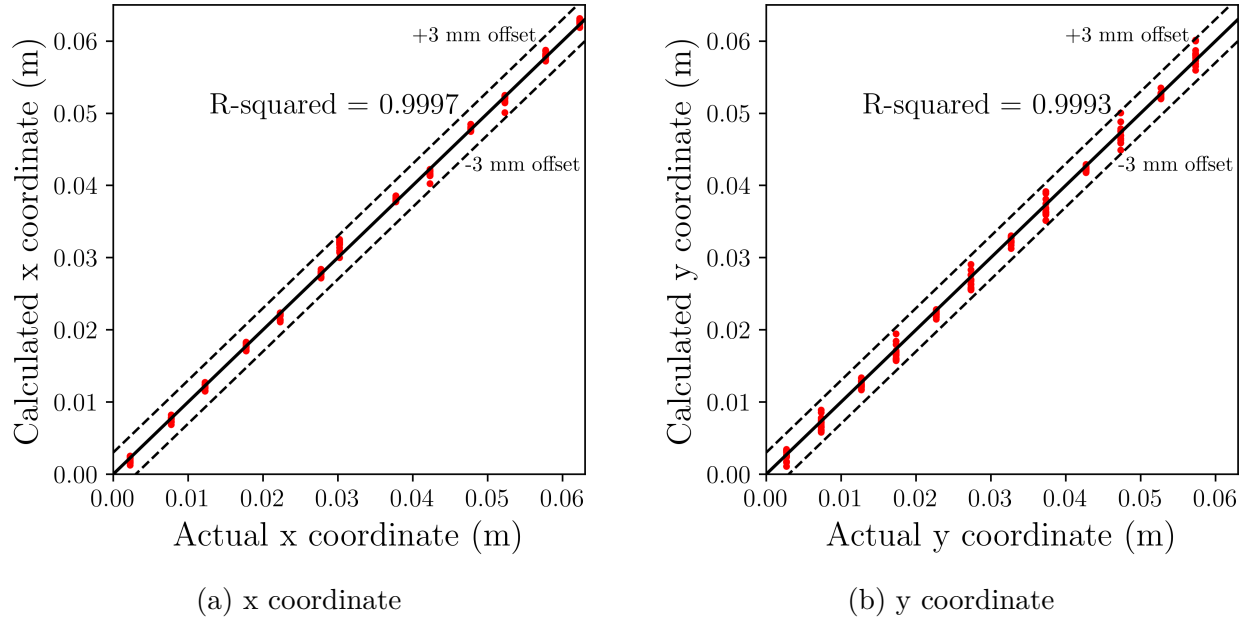


Figure 5.10 Parity plots of the predicted and experimental x and y coordinates.

5.4 Conclusion

This paper presented a novel technique for reconstructing RPT experiments. We first presented a method to improve the model parameter optimization using a non-gradient based optimization algorithm. We then presented a reconstruction method using an inverse finite element method to retrieve the tracer's position using the model output.

We integrated the NOMAD blackbox optimization software into Lethe-RPT to calibrate the detector parameters. The entangled performance of the Monte Carlo engine and NOMAD optimizer for tuning the detector parameters was experimentally verified.

We compared the performance of the FEM-PR algorithm for various case studies in a "virtual" cylinder. An analysis of the sensitivity of simulation parameters, such as number of Monte Carlo iterations and number of cells inside the meshed geometry, was presented. Using the mean Euclidean distance between the reconstructed tracer positions and the original ones, we determined the accuracy of the proposed technique. Our result indicates that the accuracy of the reconstruction increases as the number of Monte Carlo iterations and the number of cells in the mesh increase. Using a square surrounded by four detectors, we experimentally validated the FEM-PR approach accuracy by reconstructing tracer positions in two dimensions. We reconstructed the tracer position using a high number of Monte Carlo iterations to as much as possible minimize the inherent error of the model for photon count calculations. A combination of a high number of Monte Carlo iterations and a high number of cells inside the mesh enables the FEM-PR to eliminate Monte Carlo noise and provides smooth data sets of photon counts. Therefore, the algorithm searches through a clean dataset and finds the unique cell that meets the condition of the inverse finite element method. FEM-PR drastically reduces the reconstruction error, it reconstructs the position with an accuracy of 0.08 mm. Additionally, FEM-PR is a promising method for real time position reconstruction since the computational time is less than the sampling time of the traced photons from the radioactive source.

The applicability of this reconstruction algorithm is not limited to RPT as it could be readily extended to other fields in which inverse reconstruction problems have to be solved, such as medical imaging techniques, shape sensing for structural deformation, reconstruction of the sound pressure field, and other particle tracking techniques like Positron Emission Particle Tracking (PEPT).

5.5 Acknowledgements

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CHAPTER 6 ARTICLE 2 : AI-ENHANCED RADIOACTIVE PARTICLE TRACKING: A GAME CHANGING METHODOLOGY FOR ACCELERATING INDUSTRIAL PROCESS DEVELOPMENT

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Declaration: The author of this thesis has contributed to the literature review, code development, performing the experiments, analysis of the results and the writing of the article.

Traditionally, Radioactive Particle Tracking (RPT) relies on nuclear mathematical models to triangulate the position of a radioactive tracer, a process that involves significant computational resources and manual calibration. This work presents a novel methodology leveraging artificial intelligence and collaborative robotics to overcome these limitations.

In this approach, a collaborative robot is employed to maneuver a radioactive tracer, generating a precise and extensive dataset that correlates physical positions with radiation levels measured by surrounding detectors. This dataset is then used to train an Artificial Neural Network (ANN) to reconstruct the particle positions.

Results demonstrate that this approach provides superior accuracy in position reconstruction and flow field prediction, compared to traditional methods. The proposed technique offers significant advantages, including reduced computational complexity, faster and more accurate data acquisition, and the ability to handle complex geometries and flow conditions. These improvements make RPT method a promising tool for industrial applications.

6.1 Introduction

Radioactive Particle Tracking (RPT) is an experimental, non-intrusive technique used to measure the position of a free-flowing radioactive tracer in multiphase systems. RPT is well-suited for opaque systems and performs effectively in environments with a high fraction of bubbles and solids since gamma radiation can be measured easily through such material. This capability offers a significant advantage over other optical techniques, which require transparent media and various methods to compensate for light refraction [83,84].

The RPT method aims to triangulate the position of a radioactive, free-flowing, buoyantless particle in a fluid by using radiation measurements from an array of scintillation detectors. The particle's position is obtained by solving an optimization problem to identify the most probable position of the particle based on predictions from a nuclear radiation model. This triangulation occurs at each sampling interval, typically on the order of milliseconds. After completing an experiment, the output consists of a time series representing the particle's position at each sampling time, usually every 10 ms. Various quantities can be derived from this time series, providing insights into fluid dynamics and other transport phenomena, particularly instantaneous Lagrangian velocities and average Eulerian velocity fields [47].

Historically, triangulation relied on determining the most probable position of the tracer compared to predictions from a complex nuclear mathematical model. This model established the relationship between expected radiation intensity at each detector based on its position within the domain using ray tracing. The model required tuning multiple parameters through a calibration procedure, which involved manually placing the particle at known positions and solving an optimization problem to determine the most suitable model parameters [93]. However, the complexity of this mathematical model often necessitated simplifications, introducing potential inaccuracies and artifacts into the results.

In addition to the model's limitations, the calibration process was labor-intensive and prone to inaccuracies due to the manual positioning of detectors and calibration points. The manual nature of the calibration also limited the number of calibration points, as extending the calibration over several days was impractical [94].

To address these limitations, a novel approach has been developed that eliminates the need for a nuclear mathematical model. This new method combines the capabilities of artificial intelligence and collaborative robotics, allowing a cobot to move the tracer particle with high spatial precision within the volume of interest. The cobot can extensively sample the studied volume with unprecedented accuracy, generating a large dataset that correlates the precise positions of the tracer with real-time radiation levels measured by the surrounding detectors.

This approach does not rely on assumptions about the system or the fitting of parameters into a model to predict radiation levels. Instead, real measurements are used and fed into an Artificial Neural Network (ANN) model, which reconstructs the particle's position without relying on assumptions about nuclear parameters.

This methodology increases the accuracy of RPT by sampling more points with greater precision, while also reducing the time required to obtain reliable data by avoiding manual operations and complex mathematical model reconstruction. By automating the data acquisition process and eliminating the need for complex mathematical models, this approach opens the door to applying RPT in more complex reactor geometries and industrial-scale applications.

6.2 Methodology

The proposed method involves three main steps: the construction of the experimental dataset, the generation of the AI-based model, and the free-flowing experiment.

6.2.1 Construction of the Experimental Dataset

The proposed method utilizes a collaborative robot (cobot) to create an experimental dataset. The cobot is chosen for its high repeatability and precision, of 0.05 mm with a payload of 9 kg at 1200 mm, ensuring a high degree of confidence in the position of the sampled points. This dataset is essential for training an ANN model, which will be used to predict the position of a radioactive tracer based on the radiation measured by an array of scintillation detectors surrounding the system.

Hardware

To generate this dataset, two distinct hardware systems are employed—one for recording radiation and another for measuring the spatial coordinates of the tracer position.

The radiation detection hardware consists of an array of Sodium Iodide (NaI) scintillation detectors arranged and mounted on aluminum pedestals around the volume of study. Each detector is connected to an ORTEC ACE Mate amplifier/discriminator (925-Scint), which amplifies the signals. The discriminators generate a pulse whenever the detector radiation falls within the energy range of the targeted isotope, discarding lower energy radiations resulting from Compton scattering. The pulses are counted by high-speed counters using an Allen-Bradley CompactLogix PLC that integrates the counts measured by each detector over

a sampling time determined by the experimenter. The recorded radiations are synchronized and stored in a file for later processing.

The hardware used to measure the spatial coordinates of the tracer position during the sampling step consists of a Doosan cobot (A0912) mounted on a fixed pedestal close to the RPT setup. The cobot carries the tracer particle attached to the tip of a metallic rod, secured by a custom, in-house-developed gripper. The operation of the cobot is managed by a controller, which is programmed and executed by a computer using a Python script.

Sampling Strategy

The sampling strategy involves covering selected points using the cobot within the volume of interest, with the goal of maximizing the information extracted from the system while minimizing sampling time [95]. Extensive literature on different sampling approaches is available, with Garud et al. providing a comprehensive comparison of existing methods [96]. Among these techniques, Latin Hypercube Sampling (LHS) is widely used to sample space in various engineering domains [95, 97–99]. LHS subdivides the sampling space into non-overlapping intervals and selects a single random variate within each interval, resulting in space-filling, well-distributed points across the entire volume [97, 100, 101].

The open-source Design of Experiments (pyDOE) Python library is used to generate sampling points. To evaluate the performance of the LHS technique, regular Cartesian grid sampling (GS) is also performed for comparison with the results from the LHS technique.

Synchronization

Synchronization between the radiation detection system and the cobot’s position logging is crucial because these systems operate on different internal clocks. To ensure accurate data alignment, an external synchronization process, known as a handshake mechanism, is implemented.

This synchronization involves three key steps:

- **Delay Calculation:** A designated detector is used to synchronize the clocks of both systems. The cobot positions the tracer particle in front of this detector and moves it towards and away from the detector at a constant speed. The timing differences between the recorded positions and radiation counts are analyzed to calculate the time delay between the systems.
- **Smoothing:** Gamma radiation measurements often fluctuate, introducing noise. To

reduce this noise and improve the accuracy of position reconstruction, a second-order Savitzky–Golay filter is applied to smooth the radiation signal.

- **Time Step Alignment:** The data from the cobot and the radiation detection system are collected at different time intervals. Linear interpolation is used to align the datasets so that corresponding positions and radiation counts are matched accurately.

6.2.2 Construction of the AI-Based Model

ANN model consists of interconnected neurons organized into layers: the input layer, hidden layers, and the output layer. The input layer incorporates radiation counts received by the array of detectors at each time step. The hidden layers consist of nodes designed to capture the non-linearity between the input and output, which corresponds to the spatial position of the tracer particle at each time step (the coordinates x , y , and z).

Each set of nodes $\mathbf{n}^{[i]}$ is determined through a linear combination, as shown in Equation (6.1). This involves summing the input neurons from the previous layer $\mathbf{a}^{[i-1]}$, each multiplied by its corresponding weight $\mathbf{W}^{[i]}$. Additionally, a bias term $\mathbf{b}^{[i]}$ is added as a constant to this linear function [19].

$$\mathbf{n}^{[i]} = \mathbf{W}^{[i]} \mathbf{a}^{[i-1]} + \mathbf{b}^{[i]} \quad (6.1)$$

The output of each node $\mathbf{a}^{[i]}$ is obtained by applying a non-linear activation function σ to the weighted sum of inputs and biases [102]:

$$\mathbf{a}^{[i]} = \sigma(\mathbf{n}^{[i]}) \quad (6.2)$$

To evaluate the performance of a trained ANN model, the model's output $\hat{y}^{(i)}$ is compared with the expected results $y^{(i)}$. Depending on the nature of the problem, the evaluation algorithm employs a cost function—in this case, the Mean Square Error (MSE) is utilized:

$$\frac{1}{m} \sum_{i=1}^m \left(\hat{y}^{(i)} - y^{(i)} \right)^2 \quad (6.3)$$

The objective during the training of the ANN model is to minimize the loss function. To fine-tune the hyper-parameters, a grid search with k-fold cross-validation (CV) is performed. Grid search, coupled with CV, systematically explores various hyper-parameter combinations. For each combination, the dataset is divided into k folds. The model is trained on $k - 1$ folds and validated on the remaining fold, with the process repeated k times, using a different fold as the validation set in each iteration. Grid search provides the optimum set of hyper-parameters by averaging the model's validation MSE across all folds.

6.2.3 Free-Flowing Experiment

Once the ANN model is generated, the tracer is set free in the volume of interest, and the cobot is no longer used. At this stage, the radiation acquisition hardware begins recording radiation from the source as it moves within the domain. The duration of the experiment varies depending on the size of the domain to be covered, typically ranging from 4 to 24 hours. The required time is influenced by the hydrodynamic conditions inside the domain.

Figure 6.1 shows the stirred tank used for the free-flow experiment. The tank is a cylindrical vessel with a flat bottom and no baffles, equipped with a pitched blade turbine (PBT) agitator and filled with glycerol. The dimensions of the experimental rig are summarized in Table 6.1.

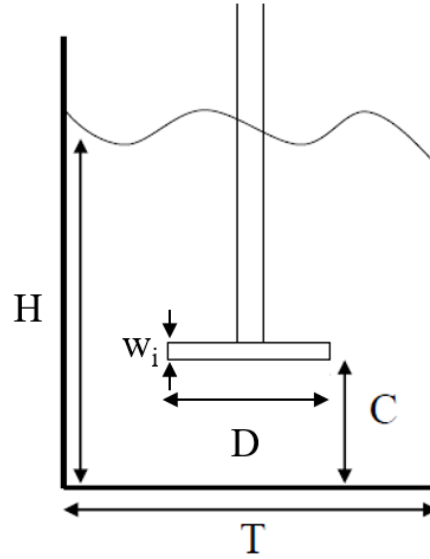


Figure 6.1 Schematic of the experimental rig.

Table 6.1 Parameter of the experiment.

Property	Value
Density (Glycerol)	1270 kg m ⁻³
Viscosity (Glycerol)	1.18 Pa s
N (rpm)	180
Re	63
Tank diameter (T)	20 cm
Impeller diameter (D)	14 cm
Liquid level (H)	20 cm
Off-bottom clearance (C)	6.8 cm
Blade width (W_i)	3.8 cm

6.2.4 Computational methodology

The CFD simulations of the stirred tank are conducted under the same operating condition as the experiment. Simulations are made using Lethe, an open-source implicit large-eddy simulation (ILES) software [79]. Lethe is based on *deal.II*, an open-source library to solve partial differential equations using finite element method [80]. Lethe solves the Navier-Stokes equations inside a domain Ω :

$$\nabla \cdot \mathbf{u} = 0 \quad (6.4)$$

$$\frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u} \cdot \nabla) \mathbf{u} = \nabla p^* + \nabla \cdot \boldsymbol{\tau} + \mathbf{f} \quad (6.5)$$

where $\mathbf{u}$ is the velocity, p^* is the pressure p divided by density ρ , $\mathbf{f}$ is a source term and $\boldsymbol{\tau}$ is the deviatoric stress tensor:

$$\boldsymbol{\tau} = \nu \left(\nabla \mathbf{u} + (\nabla \mathbf{u})^T \right) \quad (6.6)$$

where ν is the kinematic viscosity of the fluid. Equations (6.4) and (6.5) are solved using a continuous Galerkin Finite-Element formulation with an SUPG/PSPG (Streamline-Upwind/Petrov-Galerkin and Pressure-Stabilizing/Petrov-Galerkin) stabilization approach [79]. The moving impeller and the flow generated by its rotation are simulated using the Nitsche immersed boundary (NIB) method developed by Joachim et al. [103].

The fluid domain is meshed using the *cylinder* grid generator from the *deal.II* library, which is a cylinder and composed of hexahedral elements [104]. Using a Kelly error estimator on the velocity field, mesh adaptation is carried out in areas with significant velocity variation, such as those near the impeller, while avoiding unnecessary refinement elsewhere. This approach helps to lower the overall simulation cost.

The volumetric mesh of the impeller is generated using the open-source software

Gmsh [105]. The mixing experiments are simulated for 5 seconds, which is equivalent to 15 revolutions of the impeller. This was found to be sufficient to obtain time-independent average velocity profiles.

6.3 Results and Discussion

In this section, several experiments are conducted to validate the proposed method and evaluate its precision and accuracy. First, a simple case is evaluated by comparing the actual position of a tracer moved by the robot with its reconstruction predicted by the model. Then, a free-flowing particle experiment is compared to a CFD simulation.

6.3.1 Simple case Reconstruction

In this case, the ANN model was trained using the experimental dataset generated by the cobot. After training, the cobot was instructed to perform a spiral movement in air (without fluid or boundaries), following a path programmed with known coordinates. The trajectory of the robot is then compared to the trajectory reconstructed by the method. The performance of the model was assessed using metrics such as Mean Absolute Error (MAE), Standard Deviation (SD), and Mean Euclidean Distance Error (MEDE).

Table 6.2 shows the reconstruction errors for both the LHS and Cartesian Grid Sampling (GS) techniques, with datasets of 260,000 and 293,610 sampling points, respectively. The sampling patterns are fully described in Appendix D. The results indicate that the ANN model trained with the LHS dataset achieved significantly lower reconstruction errors compared to the model trained with the GS dataset. This suggests that LHS provides a more comprehensive and effective training dataset for the ANN model.

Table 6.2 Reconstruction performance indices: MAE, SD, and MEDE.

	MAE (mm)		SD (mm)		MEDE (mm)	
	GS	LHS	GS	LHS	GS	LHS
x	1.7	0.4	1	0.3	2.8	1.2
y	0.8	0.4	0.6	0.3		
z	1.7	0.9	1.2	0.7		

Figure 6.2 illustrates the actual spiral trajectory followed by the cobot and the reconstructed positions using the ANN model. The results demonstrate that the ANN model, trained with

the LHS dataset, accurately reconstructed the particle's trajectory with minimal deviations, highlighting the effectiveness of the LHS approach in generating a robust training dataset.

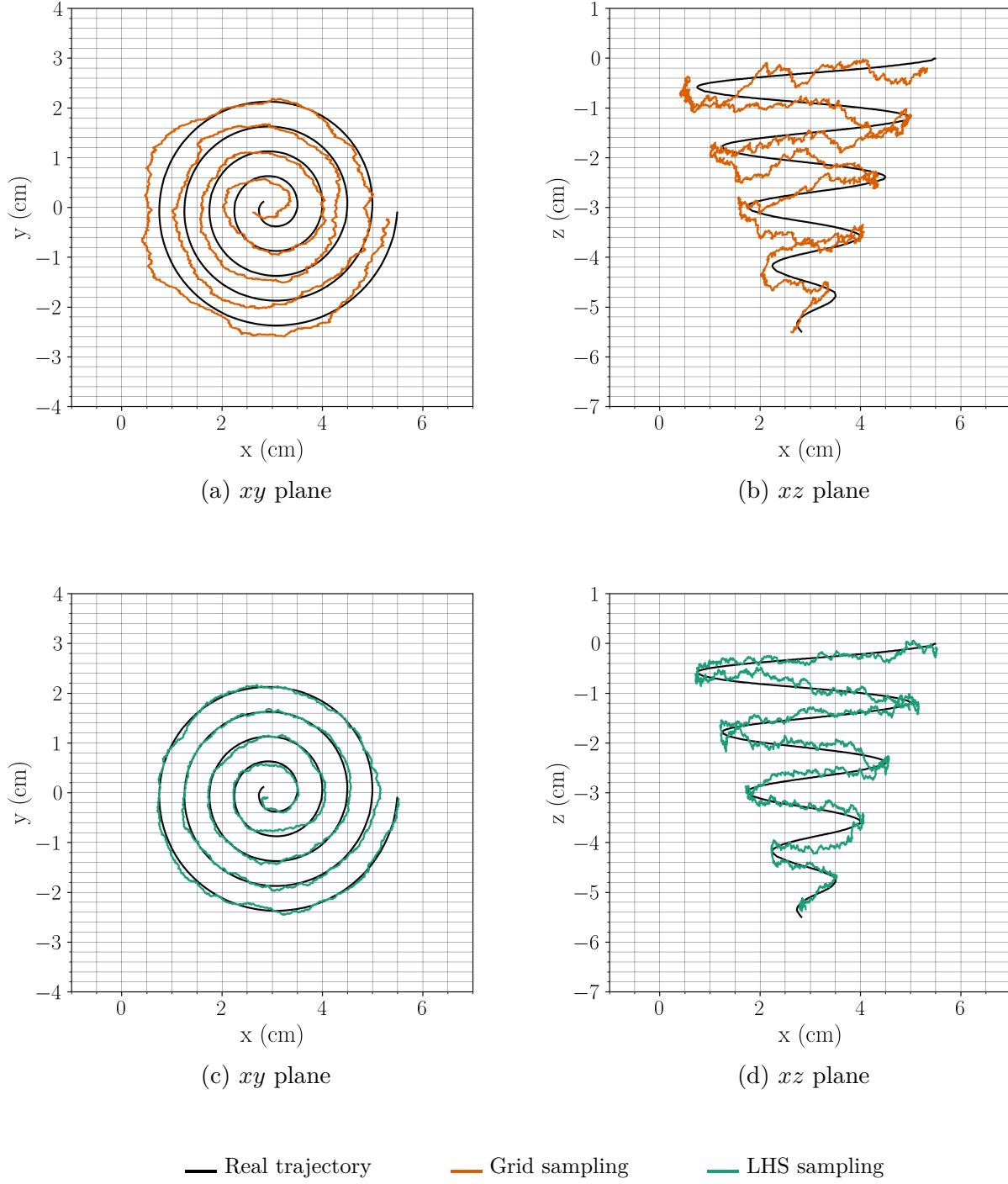


Figure 6.2 Reconstruction of spiral path trajectory.

6.3.2 Free-Flowing Case

To evaluate the capability of the ANN model in predicting flow fields, free-flowing experiments were conducted in a stirred tank filled with glycerol. A total of 854,677 sampling points were recorded during the calibration step. The reconstructed trajectories of the tracer particle were used to calculate the time-averaged velocities, which were then compared with the results from Computational Fluid Dynamics (CFD) simulations.

Figure 6.3b shows the frequency distribution of particle passages on the 2D grid and the azimuthally averaged velocity vector, revealing the presence of double recirculation loops typically associated with the PBT impeller [106]. These features were effectively captured by the RPT experiment.

Figure 6.3a illustrates the frequency distribution of particle passages on the 2D grid. To preserve the accuracy of extracted information from RPT results, cells through which the particle passed fewer than a specified threshold (400 times) are excluded. Reporting focuses solely on areas where the particle spent sufficient time, ensuring meaningful velocity profiles.

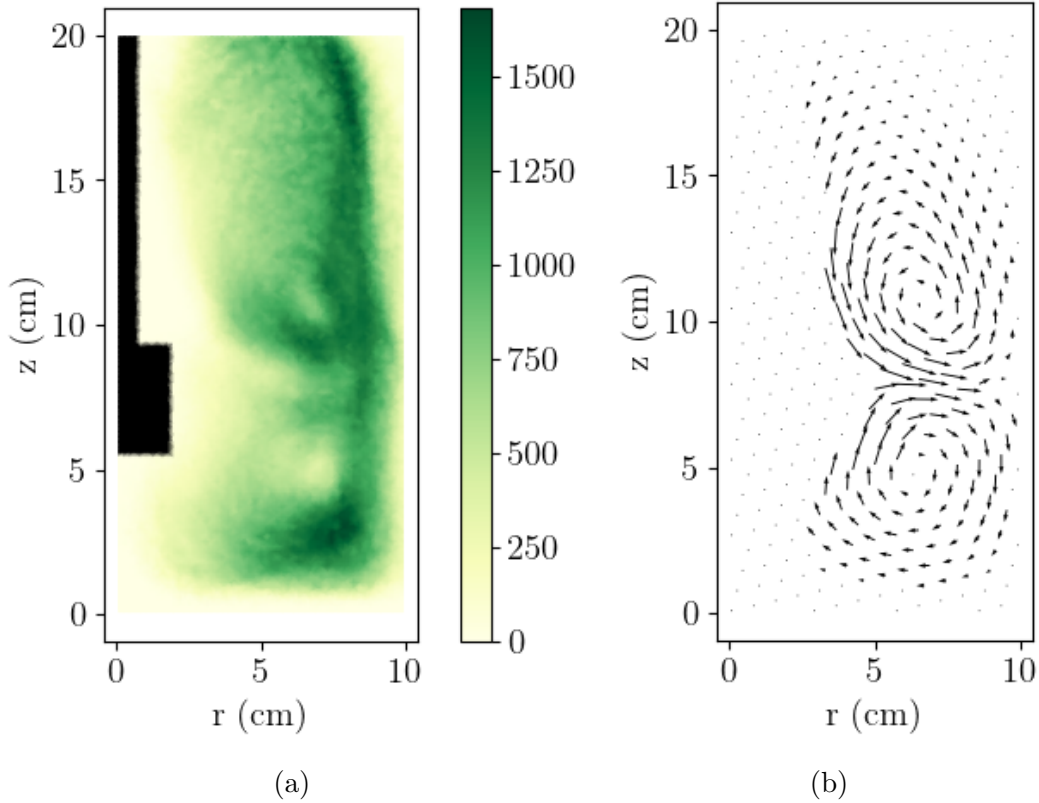


Figure 6.3 a) occurrence distribution of particle; b) azimuthally averaged velocity vector.

A comparison between the outcomes of CFD and RPT, as depicted in Figure 6.4, shows that the radial, axial, and tangential components of the azimuthally averaged velocity from the RPT experiment accurately reflect the qualitative trends observed in the CFD simulation.

Figure 6.5 presents axial profiles of radial, axial, and tangential velocity at two distinct radial positions, including the impeller tip ($r/R = 0.7$), derived from both CFD and RPT. The greatest difference between RPT and CFD results for radial velocity occurs at the radial velocity peak near the impeller tip region, with the results differing by approximately 16%, where RPT measurements are lower than those obtained from CFD.

For axial velocity, RPT accurately predicts the size and location of the two recirculation loops. Data from RPT and CFD show the highest agreement in the region farther from the impeller. For example, at a radial distance of $r/R = 0.85$, the deviation at the peak location in both upper and lower recirculation loops is about 2%. However, the most significant difference in axial velocity occurs in both recirculation loops near the impeller tips. At the peak, around $r/R = 0.7$, RPT estimates are approximately 29% higher than those obtained from CFD.

For tangential velocity at a radial distance of $r/R = 0.7$, corresponding to the impeller tip, the velocity peaks of RPT and CFD in the region around the impeller's blades ($6 \text{ cm} \leq z \leq 10 \text{ cm}$) deviate by about 44%. However, farther from the impeller, in the same axial region, the deviation is approximately 26%. In the regions with $z \leq 6 \text{ cm}$ or $z \geq 10 \text{ cm}$, the difference between RPT and CFD data ranges from 1% to 10%.

The higher discrepancy around the impeller and in regions at the same level as the impeller in all three components of velocity might be related to the rapid acceleration of the velocity and the 90-degree changes in direction as vertical motions shift to radial motions, as Chiti et al. also reported similar observation using positron emission particle tracking (PEPT) to study stirred vessels [45].

The high velocity and acceleration prevent the particle from spending sufficient time in the region around the impeller. Additionally, uncertainties due to the observed vibration of the shaft and the impeller led to poorer estimations in that region. Farther from the impeller, where velocities are lower and there are no rapid changes, the particle spends sufficient time in the region to fully adhere to the fluid motion. As the Stokes number (0.006) suggests, the particle follows the fluid stream with high fidelity and is able to track the fluid motion with high precision, as discussed above.

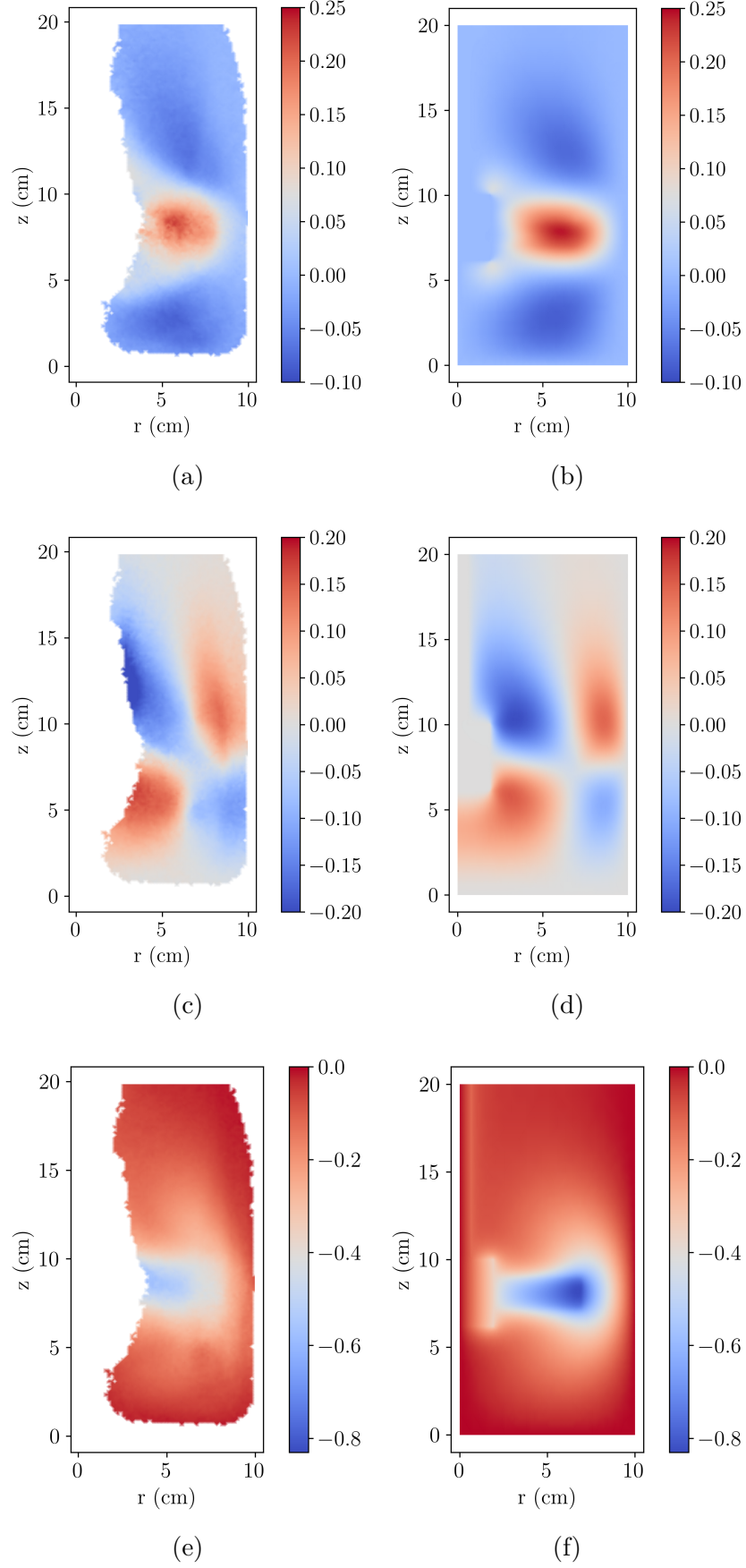


Figure 6.4 Azimuthally averaged velocity (m/s): [left] RPT, [right] CFD components; a,b: radial, c,d: axial and e,f: tangential.

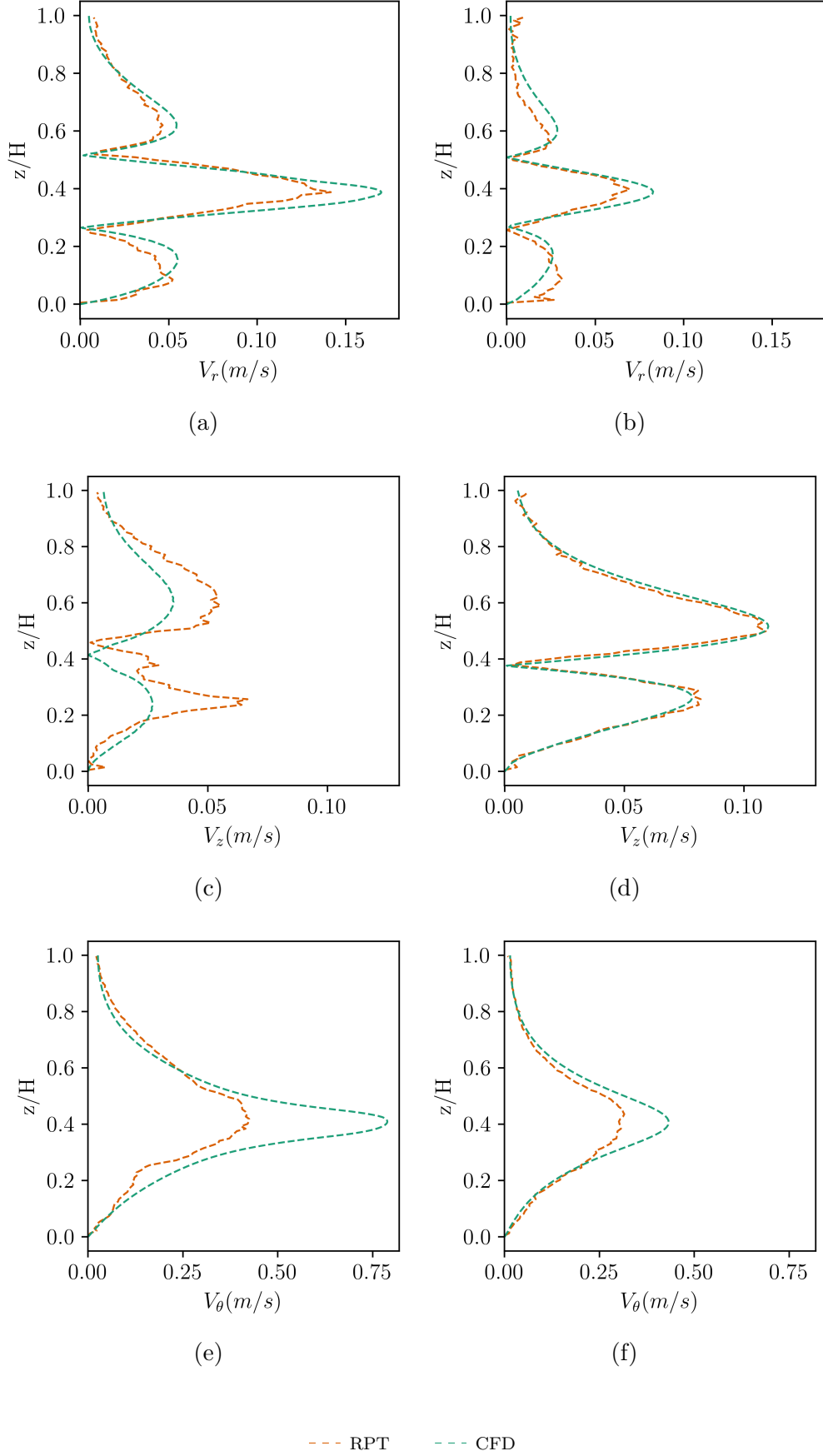


Figure 6.5 Axial profile of normalized radial, axial, and tangential components of azimuthally averaged velocities; a,c,e at radial distance of $r/R = 0.7$ and b,d,f at $r/R = 0.85$.

6.3.3 Discussion

The ANN model trained with LHS data provided accurate position reconstructions in the air medium and reliable velocity profiles in the stirred tank experiment. The slight discrepancies near the impeller in the velocity field predictions can be attributed to the rapid changes in flow direction, the high velocities present in these regions, and vibrations of the shaft and impeller. Despite these challenges, the ANN model demonstrated its capability for positions reconstruction with a high degree of accuracy, positioning it as a viable alternative to traditional RPT methods.

The simulation of the flow at $Re = 63$ in the laminar regime takes 12 hours using 4 nodes on the Digital Research Alliance of Canada Beluga cluster. Sampling the tank with the cobot requires 4 hours, and the free-flow experiment takes 10 hours, nearly the same duration as the CFD simulation.

For the same geometry, as flow complexity increases (such as in the transient regime at higher Re numbers) CFD simulations can take significantly longer to obtain time-independent average velocities, potentially up to multiple days. In contrast, the time required for an RPT experiment remains relatively unchanged, regardless of flow conditions, fluid rheology or the presence of multiple phases, making CFD more time-consuming.

The results show that this advanced RPT technique can now deliver more accurate information in a short amount of time compared to previous RPT methods and compared to CFD methods. It will be useful to those seeking to understand and optimize the complex interactions in single and multiphase systems more rapidly and more accurately than before, especially when addressing the effects of scale involved in process design.

6.4 Conclusion

This study introduced a novel methodology for RPT that leverages artificial intelligence and collaborative robotics to overcome the limitations of traditional RPT techniques. By employing a cobot to generate a comprehensive experimental dataset and using an ANN to predict particle positions, the proposed approach significantly enhances the accuracy and efficiency of RPT.

The methodology was validated through two sets of experiments: a controlled, simple case of reconstructing a spiral trajectory in air, and a more complex free-flowing particle experiment in a stirred tank. The ANN model trained with LHS data demonstrated superior performance in accurately reconstructing particle trajectories and predicting flow patterns compared to models trained with GS data. These findings highlight the effectiveness of LHS in creating

robust training datasets for ANN models in RPT applications.

The new RPT approach offers significant advantages, including reduced computational complexity, the capability to handle complex geometries, and notable time and cost savings in process development. By bypassing the need for simplifications and phenomenological models, this new approach not only competes with numerical methods but also delivers fast and reliable insights into systems with complex interactions, such as mass, heat, and momentum transfer, as well as chemical reactions and phase changes.

In conclusion, this advanced RPT method represents a significant step forward in the field of process analysis and optimization, particularly for industrial applications where conventional methods fall short. The integration of this technique into industrial settings has the potential to enhance process understanding, facilitate more informed decision-making, and ultimately drive improvements in efficiency and performance across a wide range of systems.

6.5 Acknowledgements

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CHAPTER 7 ARTICLE 3 : RPT FLOW MEASUREMENTS AND CFD SIMULATION OF TRANSITIONAL FLOW IN AN UNBAFFLED STIRRED TANK

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Abstract: The flow in an unbaffled stirred tank operating in the transitional regime was studied using Radioactive Particle Tracking (RPT) and Computational Fluid Dynamics (CFD). Both single- and multiphase models were used to simulate the flow. The simulation used implicit large-eddy simulation (ILES) to capture turbulence. The multiphase simulations used a Volume of Fluid (VOF) method to capture the free-surface behavior. Comparisons between multiphase and single-phase simulation were used to evaluate the free surface's impact on the velocity profile. Correlations from the literature were utilized to assess the capability of the RPT technique and multiphase simulations in predicting the central vortex characteristics. The multiphase simulation provided predictions closer to the RPT measurements in terms of pattern and magnitude, particularly in the upper third of the tank. While the average velocity profiles from RPT showed good agreement with simulations, RPT data in the impeller region underestimated the average velocities compared to simulations.

7.1 Introduction

Unbaffled tanks are less common and less studied due to the better mixing performance associated with baffled tanks. However, some processes, like those in the pharmaceutical and food industries, require the absence of baffles for sanitation purposes. Other examples include bioreactors for shear-sensitive cells, crystallization of fragile particles, or mixing of highly viscous fluids, where baffles can cause flow disruptions and dead zones [30–32].

In uncovered tanks without baffles, the swirling effect can cause a liquid surface depression, known as vortex formation [107]. Some processes benefit from this vortex, such as solid suspension or particle dissolution, where it helps draw solids into the liquid [33]. In other cases, the central air vortex helps transfer the required oxygen to the solution [34,35].

Most experimental studies on the hydrodynamics of unbaffled stirred tanks have used Laser

Doppler velocimetry (LDV) [57–60] or Particle Image Velocimetry (PIV) [32, 61–63]. However, these methods require transparent systems, and refraction compensation mechanisms, making them difficult to set up and unsuitable for optically opaque reactors and fluids. Consequently, it is relevant to develop Computational Fluid Dynamics (CFD) tools capable of accurately simulating the flow in those conditions. These tools can reduce the cost, time, and challenges associated with experimental studies. However, CFD requires additional sets of constitutive equations to capture additional flow features and the combination of these equations may lead to inaccurate results and therefore requires careful experimental validation [108]. This highlights the need for the experimental techniques that work in such conditions.

CFD simulations and their validations for unbaffled tanks are fewer due to their complex flow patterns. Unlike baffled tanks, where turbulence is mainly anisotropic near the impeller and isotropic elsewhere, unbaffled tanks exhibit anisotropic turbulence throughout the vessel [75].

Several studies, such as those by Márquez-Baños et al. [73] and Guadarrama-Pérez [72], focus on laminar flow, while others investigate fully turbulent regimes. Most numerical studies employ the Reynolds-averaged Navier–Stokes (RANS) approach for turbulence modeling, with some incorporating the Volume of Fluid (VOF) method to capture liquid-air interface dynamics [30, 57, 58, 76]. An alternative approach using surface pressure to calculate surface deformation has also been applied [75]. In certain cases, studies used a lid to cover the top surface to suppress vortex formation or were unable to resolve the vortex structure accurately [56, 62, 74, 77]. Since RANS models cannot capture the periodic flow behavior caused by impeller motion, some works have implemented the Unsteady Reynolds-Averaged Navier–Stokes (URANS) method, while others have used Large Eddy Simulation (LES) to address this limitation [27, 32, 59, 78].

The low Reynolds numbers and central vortex in unbaffled, uncovered tanks during the early transitional regime complicate flow simulations and often cause turbulence model failures [108]. Tamburini et al. [74] studied unbaffled tanks in the transitional regime at $Re = 600$, but covered the top surface to prevent vortex formation, noting that simulating the flow in uncovered tanks in transitional regime is highly complex.

This study presents experimental results from unbaffled, uncovered stirred tanks operating in the transitional regime, using the Radioactive Particle Tracking (RPT) technique. RPT is a three-dimensional velocimetry method that tracks radioactive tracer particles, making it suitable for optically opaque systems and applicable to real-world industrial-scale reactors. As a Lagrangian technique, RPT generates time series data of particle positions throughout the experiment, which can be post-processed to obtain averaged velocity profiles.

Additionally, this work includes results from CFD simulations using implicit large-eddy simulation (ILES). The study features single-phase and multiphase simulations employing the VOF method to assess the free surface's impact on the velocity profile. Furthermore, correlations from the literature for estimating vortex characteristics are used to verify the findings from both the RPT measurements and the CFD simulations.

The objective of this work is to show that RPT is capable of capturing the characteristics of the free surface and predicting the average velocity profile with good agreement with CFD simulation results. The outcome demonstrates the potential of this rapid and accurate experimental technique to capture complex flow behavior, meaning it can be applied to other systems, such as solid-liquid systems involving non-transparent complex fluids that are difficult to study experimentally or simulate numerically with the current state of the art.

7.2 Methodology

7.2.1 Stirred Tank Geometry and System Description

The stirred tank has a diameter of $T = 20$ cm and a height of $H = 1.05T$. The agitator is a Pitched Blade Turbine (PBT) with a diameter of $D = 0.7T$, mounted at a height of $C = 0.33T$ from the bottom of the tank. The fluid is an aqueous solution of glycerol (75% wt. concentration) at room temperature, with a density of 1210 kg m^{-3} and a viscosity of 0.062 Pa s . The impeller rotates at 210 rpm, resulting in an impeller Reynolds number of $Re = 1370$.

7.2.2 Radioactive Particle Tracking (RPT)

In this work, nine Sodium Iodide (NaI) scintillation detectors were used around the stirred tank, arranged in two planes. Four detectors were placed at each plane, spaced 90° apart and oriented so that their faces were perpendicular to the tank. Additionally, one detector was positioned at the bottom of the tank, at a distance of 15 cm.

An artificial neural network (ANN) was used to reconstruct the particle trajectory from the radiation measured by the detectors. The ANN model functions in two stages. Initially, a dataset is utilized to train the ANN and determine the optimal hyperparameters for the model. Once the hyperparameters are established, the trained model is used to reconstruct particle positions based on the photon count data collected during the free-flowing experiment.

To obtain the training dataset, we used the approach outlined in our previous work [109]. A

cobot maneuvered a tracer particle attached to the tip of a metallic rod within the stirred tank. As the particle moved, the cobot recorded its position at specified time intervals, while the RPT hardware simultaneously captured radiation intensity at each detector at intervals of $T = 10$ ms. This dataset, which links the particle's position to the received radiation at each detector, will be used as the training dataset.

After collecting data for training the ANN model, the free-flowing experiment was performed. Using the trained ANN model, the time series of the particle trajectory was reconstructed. From the reconstructed positions, the particle's instantaneous velocity was calculated by determining the displacement between consecutive positions and dividing by the time interval between each measurement.

Tracer Particle

A radioactive tracer with a diameter of 3.5 mm and an activity of 125 μ Ci was designed and manufactured for the water-glycerol solution. Scandium-46 (^{46}Sc) was used as the radioactive source. The Scandium was mixed with epoxy glue, and, after the mixture dried, it was coated with another layer of epoxy containing entrapped air bubbles. This adjustment was made to fine-tune the overall density of the particle and to ensure that the particle is neutrally buoyant.

A key factor influencing fluid velocity measurements with a tracer particle is its ability to follow streamlines, which is characterized by the Stokes number (St):

$$St = \frac{\tau_p}{\tau_f} \quad (7.1)$$

where τ_p is the particle response time and τ_f is the fluid response time to an external disturbance, such as the impeller movement within a stirred tank. Assuming a Stokesian flow ($Re \ll 1$), τ_p and τ_f are defined as [45]:

$$\tau_p = \frac{\rho_s d_p^2}{18\mu} \quad (7.2)$$

$$\tau_f = \frac{1}{\pi D} \quad (7.3)$$

where ρ_s , d_p , μ , and D respectively are particle density, particle diameter, fluid viscosity, and impeller diameter. When $St < 0.1$, the tracing error is negligible [110]. However, in non-Stokesian flow, Stokes' law underestimates the drag force on the particle and overestimates the value of τ_p , leading to an overestimation of the St number [111]. For the particle designed

to follow the fluid inside the stirred tank, the overestimated St number is equal to 0.15. This value occurs at the impeller tip, which has the highest velocity. Consequently, the Stokes number is lower away from the impeller. This value of the Stokes number indicates that the particle will closely follow the flow with fidelity in regions far from the impeller, where the Stokes number is lower. However, it may not adhere as well in the region of the impeller discharge.

The Stokes number helps assess the particle's ability to follow the streamlines to some extent. However, the particle's capability to respond to the smallest turbulent scales must also be considered. This can be evaluated by comparing the particle size to the smallest scale in turbulent flow, known as the Kolmogorov scale (η_k). Assuming isotropic turbulence, the Kolmogorov length scale can be estimated as [112]:

$$\eta_k = L \times Re^{-3/4} \quad (7.4)$$

where L represents the integral scale, taken to be equal to the impeller diameter (D). With a Reynolds number of 1370, this approximation yields a Kolmogorov scale of approximately 621 μm . Since the particle is much larger than this smallest turbulent scale ($d/\eta \approx 5.6$), it doesn't follow any structures smaller than its size down to the Kolmogorov scale.

Furthermore, when the particle size significantly exceeds the Kolmogorov length scale, the interactions between the particle and turbulence become complex. The introduction of such a particle into the turbulent flow alters the existing flow patterns and can either augment or attenuate the turbulence within the flow [113]. Therefore, it is essential to consider that the presence of such tracer particles will inevitably influence the flow pattern.

7.2.3 Data Processing

Following the approach of Chiti et al. [45], the particle locations were transformed from Cartesian coordinates into cylindrical coordinates before the velocity components are calculated in cylindrical coordinates. Given the axial symmetry of the stirred tank, the velocity was then azimuthally averaged on a rectangular grid, figure 7.1 presents a schematic of the 2D grid used for averaging.

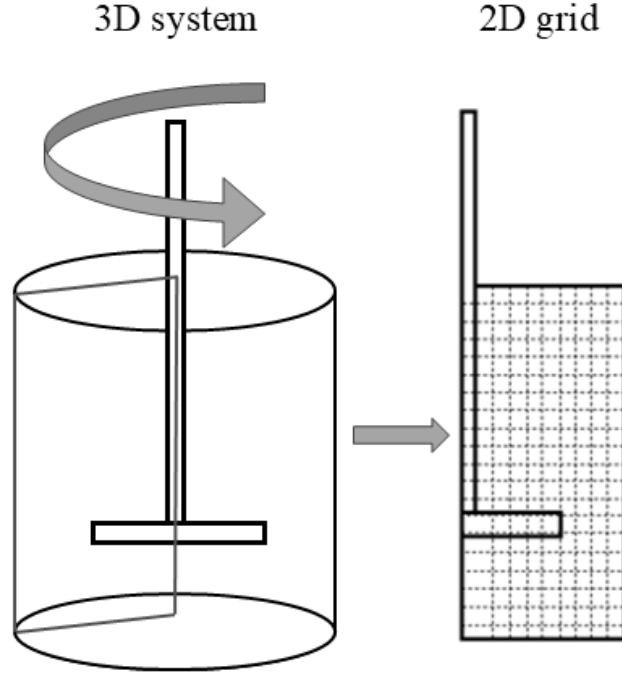


Figure 7.1 Schematic of the 2D grid used for the azimuthal average of the RPT data; the graphic has been modified from [114]).

7.2.4 CFD Setup

The flow in the agitated vessel was simulated under the same conditions as the experiments. The simulations were carried out using Lethe, an open-source implicit large-eddy simulation (ILES) software [79]. Lethe is based on *deal.II*, an open-source library for solving partial differential equations using the finite element method [80].

Both single-phase and multiphase simulations were conducted to evaluate the effect of the liquid-air interface (free-surface) on the simulated velocity profile. In the single-phase simulation, the liquid-air interface was modeled using a slip boundary condition at the top boundary, while in the multiphase simulation, the VOF method was applied to capture the dynamics of the interface.

Lethe solves the Navier-Stokes equations using continuous finite element discretization inside the domain Ω :

$$\nabla \cdot \mathbf{u} = 0 \quad (7.5)$$

$$\rho \left(\frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u} \cdot \nabla) \mathbf{u} \right) = -\nabla p + \nabla \cdot \boldsymbol{\tau} + \rho \mathbf{f} \quad (7.6)$$

where $\mathbf{u}$ is the velocity, p is the pressure, ρ is density, $\mathbf{f}$ is a source term and $\boldsymbol{\tau}$ is the viscose stress term:

$$\boldsymbol{\tau} = \mu \left(\nabla \mathbf{u} + (\nabla \mathbf{u})^T \right) \quad (7.7)$$

where μ is the dynamic viscosity of the fluid. While solving a multiphase problem, the domain of interest Ω is the union of the gas and liquid subdomains:

$$\Omega = \Omega_l \cup \Omega_g \quad (7.8)$$

where Ω_l and Ω_g respectively represent the liquid and gas phase. To identify the volume of each phase in a given computational cell, the VOF method uses a phase indicator that takes a value of 0 for air and 1 for the liquid liquid, and goes rapidly and smoothly from 0 to 1 at the liquid-gas interface. The fluid properties ρ and μ are calculated as:

$$\chi = (1 - \phi)\chi_g + \phi\chi_l \quad (7.9)$$

where χ , χ_g , and χ_l respectively represent the value of the given property in each computational domain, air, and liquid.

Equations (7.5) and (7.6) are solved using SUPG/PSPG (Streamline-Upwind/Petrov-Galerkin and Pressure-Stabilizing/Petrov-Galerkin) stabilization approach [79]. To model the moving impeller and the flow generated by its rotation, the Nitsche immersed boundary (NIB) method developed by Joachim et al. [103] was used.

The fluid domain was meshed using hexahedral elements with the *cylinder* grid generator from the *deal.II* library. Dynamic mesh adaptation was applied in regions with significant velocity variations, using the Kelly error estimator.

7.3 Results and Discussion

The flow fields within the stirred tank were analyzed by comparing data from RPT and CFD predictions. A time-independence test was conducted to confirm the stability of the time-averaged velocity profiles over time. Additionally, a mesh sensitivity test was performed to ensure mesh independence in the CFD results, while a grid sensitivity test was carried out to verify that the RPT results remained unaffected by the size of cells in the grid used for averaging purposes.

The velocity profiles were compared at two axial positions: one at the impeller tip ($r/R =$

0.7), where the radial and tangential velocity components reach their maximum values, and another near the wall, farther from the impeller ($r/R = 0.93$), where the axial velocity component is at its peak.

7.3.1 Grid and Mesh Sensitivity Analysis

For the RPT data, after following the post-processing routine outlined in previous sections, the azimuthally averaged velocities were calculated on a two-dimensional grid. Grid sensitivity was evaluated for three different grid sizes. Table 7.1 provides the details of the RPT grid sizes, where N_r and N_z represent the grid resolution in the radial and axial directions, respectively.

Table 7.1 Details of the grid and mesh sizes used for the RPT technique and CFD simulations.

	RPT		CFD
	N_r	N_z	Number of elements
Grid, Mesh I	25	50	470,294
Grid, Mesh II	70	140	3,240,978
Grid, Mesh III	100	200	22,789,479

Figure 7.2 illustrates the impact of grid resolution on the axial profile of the normalized radial velocity near the impeller tip, as measured using the RPT technique. Grid I recorded higher values at certain positions, including at the axial position of the impeller. In contrast, Grid II and III produced comparable values across most positions.

For the multiphase simulations, three different mesh sizes were evaluated in the mesh sensitivity test. Table 7.1 presents the number of elements for each mesh. The velocity profile at the same axial position used in the RPT study was also analyzed for the CFD. As illustrated in Figure 7.3, in the upper half of the tank there is a noticeable change of pattern from Mesh I to Mesh II. Meshes II and III however, show highly similar patterns. Based on the mesh sensitivity analysis, Grid III and Mesh III, corresponding to the RPT and CFD results respectively, were selected for further analysis as they offer mesh-independent solutions.

For the single-phase CFD results are reported from the finest possible mesh (3,240,978 elements), Mesh II, as simulations with more refined meshes became unstable and diverged after a short time (0.1 impeller revolutions).

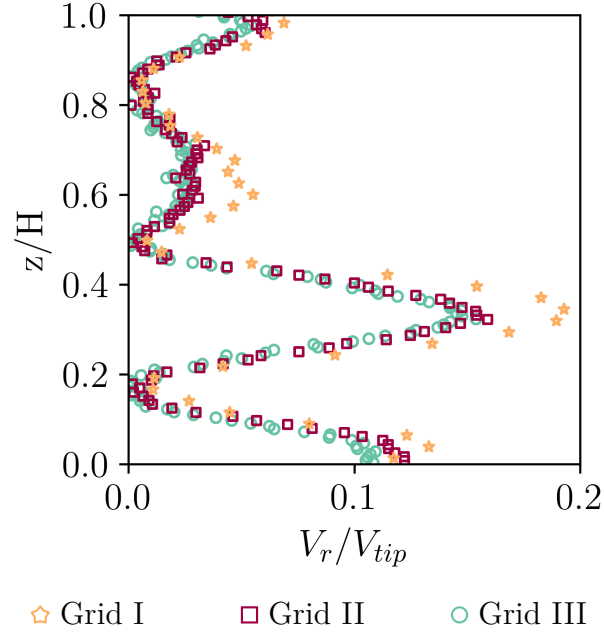


Figure 7.2 Effect of grid resolution on the axial profile of normalized radial velocity at $r/R = 0.7$ obtained from RPT.

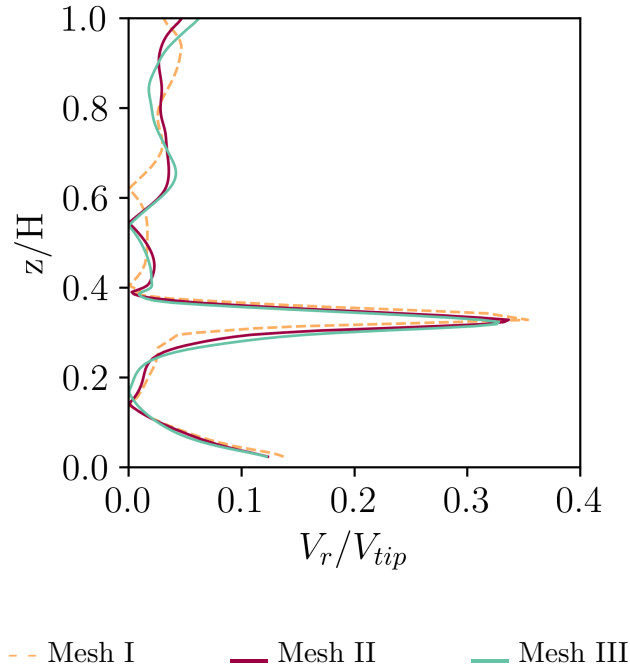
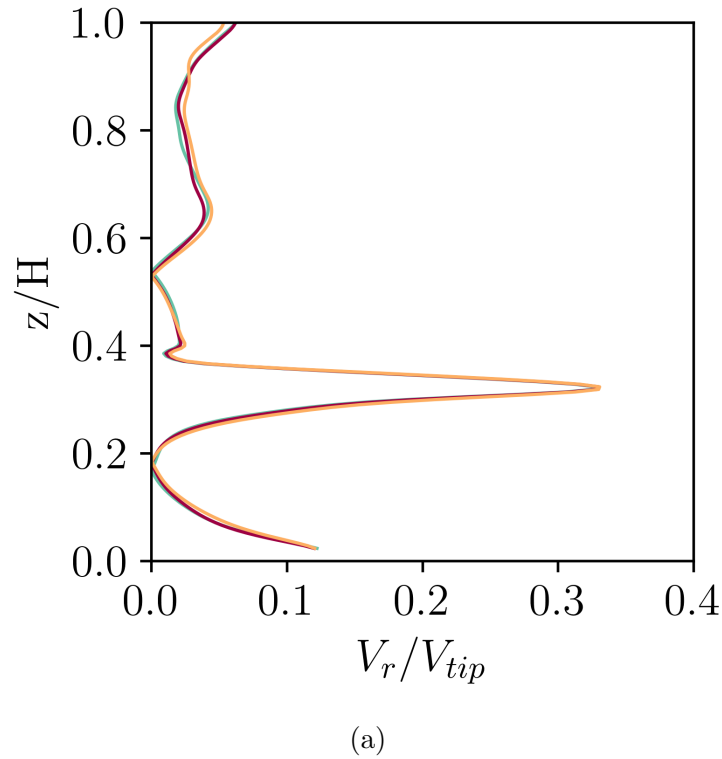


Figure 7.3 Effect of mesh resolution on the axial profile of normalized radial velocity at $r/R = 0.7$ obtained from CFD.

7.3.2 Time Independence Test

Since the velocity profiles derived from RPT are time-averaged, the CFD data are also presented as time-averaged for consistency. To verify the time-independence of the time-averaged values, the time-averaged velocity profiles are compared at three different stages of the simulation: 10.5, 14, and 17.5 impeller revolutions using Mesh III. Figure 7.4 presents the axial profile of normalized radial velocity at $r/R = 0.7$ for each simulation stage, with the near-identical results indicating that the time-averaged values are indeed independent of the time-averaging window.



— 10.5 impeller revolutions — 14 impeller revolutions — 17.5 impeller revolutions

Figure 7.4 Effect the length of the time-averaging window on the axial profile of normalized radial velocity at $r/R = 0.7$ obtained from CFD.

7.3.3 Free Surface Prediction

RPT captures the trajectory of the tracer particle. Figure 7.5a displays the number of particle location over the course of a 15-hour experiment. From this data, it is possible to identify locations where the particle was never located. Figure 7.5b presents a binary map of the

particle pass. By analyzing the highest and lowest axial positions the particle reached at the liquid surface, the vortex depth can be estimated.

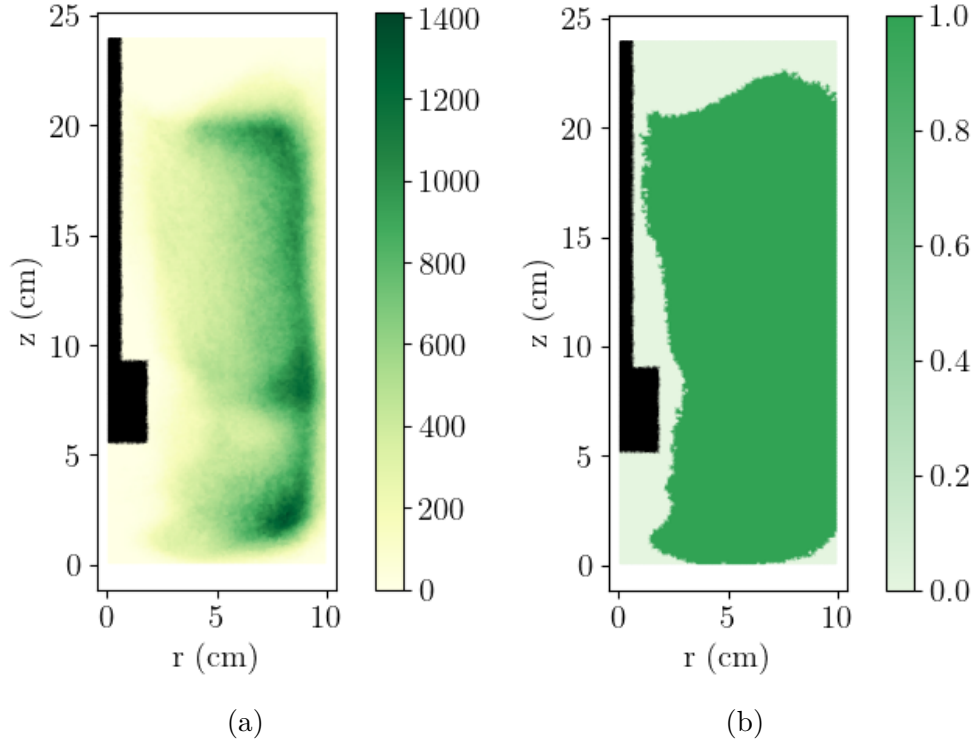


Figure 7.5 a) map of particle distribution, b) map of binary particle existence

The results obtained with both RPT and CFD were compared with Nagata's model [115] from the literature. Figure 7.6 illustrates the relationship between the normalized vortex depth $\Psi = z/d$, where d represents the impeller diameter, and the normalized tank radius $\xi = 2r/d$. According to Nagata's model, the normalized depth is:

$$\Psi = \begin{cases} \Psi_w - \frac{(\alpha\pi\xi_c)^2}{2} Fr \left(2 - \frac{\xi_c^2}{\xi_D^2} - \frac{\xi^2}{\xi_c^2} \right), & \text{if } \xi \leq \xi_c \\ \Psi_w - \frac{(\alpha\pi\xi_c)^2}{2} Fr \left(\frac{\xi_c^2}{\xi^2} - \frac{\xi_c^2}{\xi_D^2} \right), & \text{if } \xi > \xi_c \end{cases} \quad (7.10)$$

where Ψ_w is the vortex depth at the tank wall and is given by [116]:

$$\Psi_w = \Psi_0 + \frac{(\alpha\pi\xi_c)^2 \xi_c^2}{2\xi_D^2} Fr \left(\frac{1}{2} - \ln \left(\frac{\xi_c^2}{\xi_D^2} \right) \right) \quad (7.11)$$

where $Fr = N^2 d/g$ is the Froude number, and $\Psi_0 = H/d$. The value of Fr is 0.17, while α

and ξ_c are 0.739 and 0.53, respectively [117].

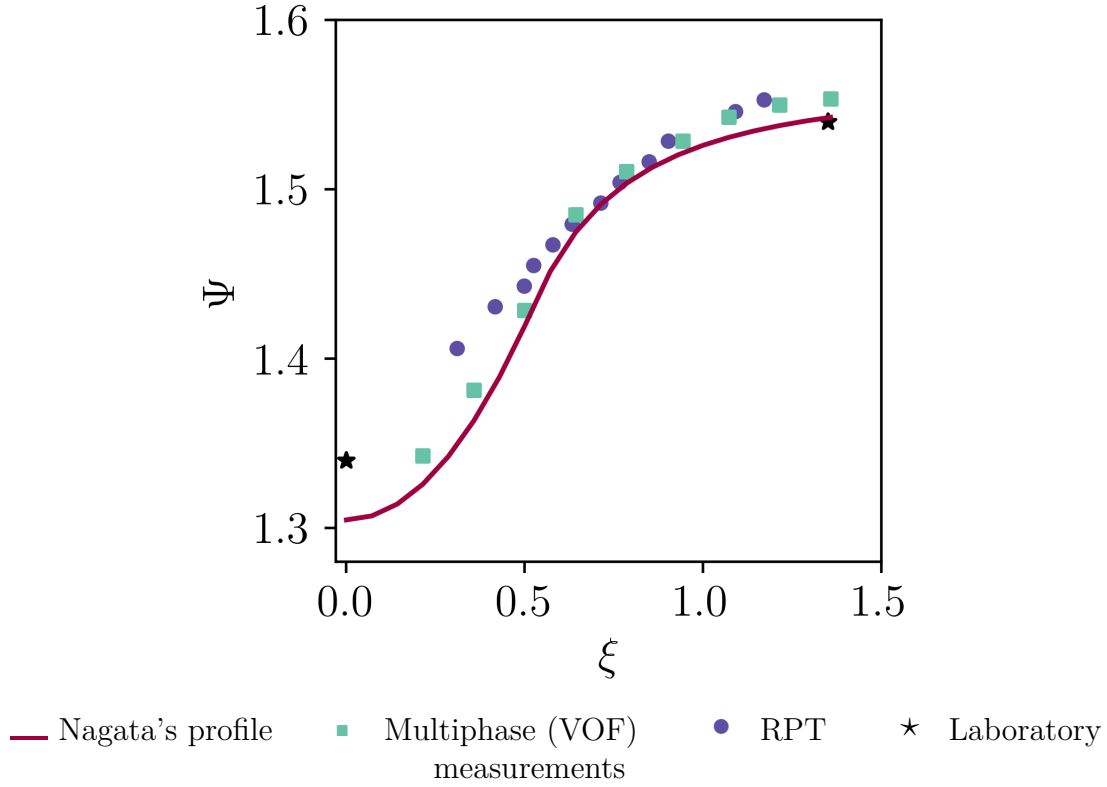


Figure 7.6 Comparison of Vortex Shapes: Nagat's profile, RPT, CFD multiphase simulation, and laboratory measurements.

The vortex depth at the wall predicted by Nagata's model closely matches the experiments. The multiphase CFD simulation also predicts the vortex depth with a relative deviation of less than 1%. RPT successfully captured the vortex shape starting from a distance of 2 cm from the wall. As the data moves toward the vortex center, the three datasets—Nagata, CFD, and RPT—remain in agreement, with a maximum deviation of 1.5% from Nagata's model. However, as the data approach the shaft, within a range of 2 cm, the RPT data become sparse.

Closer to the shaft, RPT data are either absent or unreliable due to the presence of the shaft, which leads to γ -ray attenuation and may result in inaccurate position predictions. Meanwhile, the lowest part of the vortex predicted by the CFD deviates by 3% from Nagata's model, although it aligns with the experimental measurements.

Next, the critical radius (r_c) was compared between the RPT and CFD simulation data. According to Rankine's vortex model, r_c represents the boundary separating the forced vortex

region from the free vortex [116].

Figure 7.7 shows the radial profile of tangential velocity at the top of the tank ($z^* = 0.92$), comparing RPT and CFD results, both of which align with the Rankine vortex model. As shown in Table 7.2, the r_c values from RPT and CFD closely match each other and the Nagata's model [117].

Table 7.2 Comparison of $2r_c/D$ values from different methods.

	Nagata	CFD	RPT
$2r_c/d$	0.53	0.55	0.58

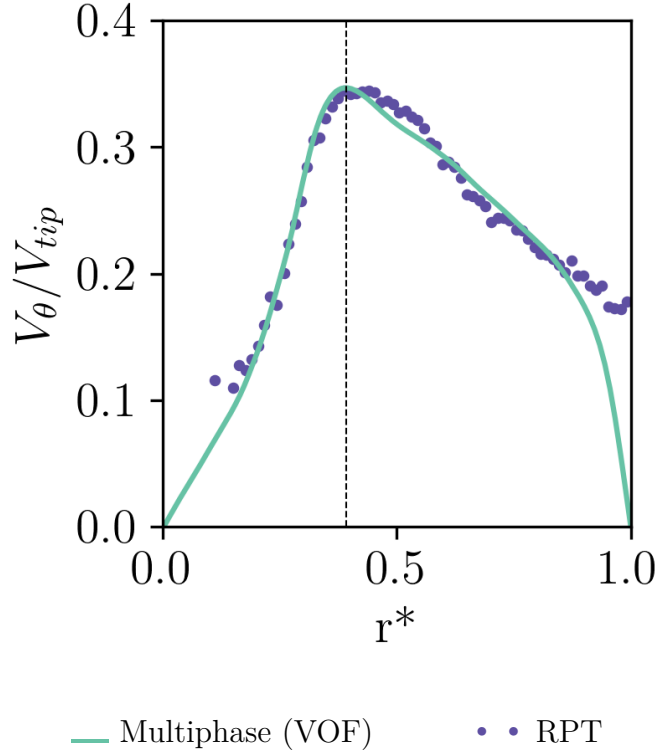


Figure 7.7 Time-averaged azimuthally averaged radial profile of the tangential velocity component at $z^* = 0.92$ for quantification of the critical radius separating the fixed and free vortex.

7.3.4 Prediction of flow pattern

Figure 7.8 presents the averaged velocity components obtained from RPT and multiphase simulation results. The radial component of the azimuthally averaged velocity in $r - z$ plane,

as illustrated in Figures 7.8, shows qualitative agreement between the RPT and CFD results in most regions, with minor discrepancies observed in some areas.

Both RPT and CFD show negative radial velocity values near the base of the tank, suggesting the existence of a recirculation zone beneath the impeller. The radial velocity reaches its maximum in the impeller's discharge jet, propagating from the impeller towards the sidewall. While both techniques capture this discharge jet, RPT consistently presented lower velocity magnitudes than CFD.

In the axial component of velocity, near the wall, there is an evident radial jet splitting. It shows up as a decomposition into two distinct circulation loops of flow toward the upper and lower parts of the tank. While both methods captured the recirculation pattern, CFD simulations show a higher axial velocity near the wall region.

The azimuthal velocity component from RPT and CFD consistently exhibits negative values throughout the entire height, indicating alignment with the impeller's rotation. In the upper third of the tank, both RPT and CFD results demonstrate a trend that confirms the presence of a vortex near the top around the impeller. This observation, as discussed in the previous section, agrees with Rankine's vortex theory. In this region, both methods closely correspond in terms of trend and magnitude. The most significant divergence between RPT and CFD occurs on the impeller plane, in a region approximately 1.7 cm wide, where the RPT results exhibit lower velocity values.

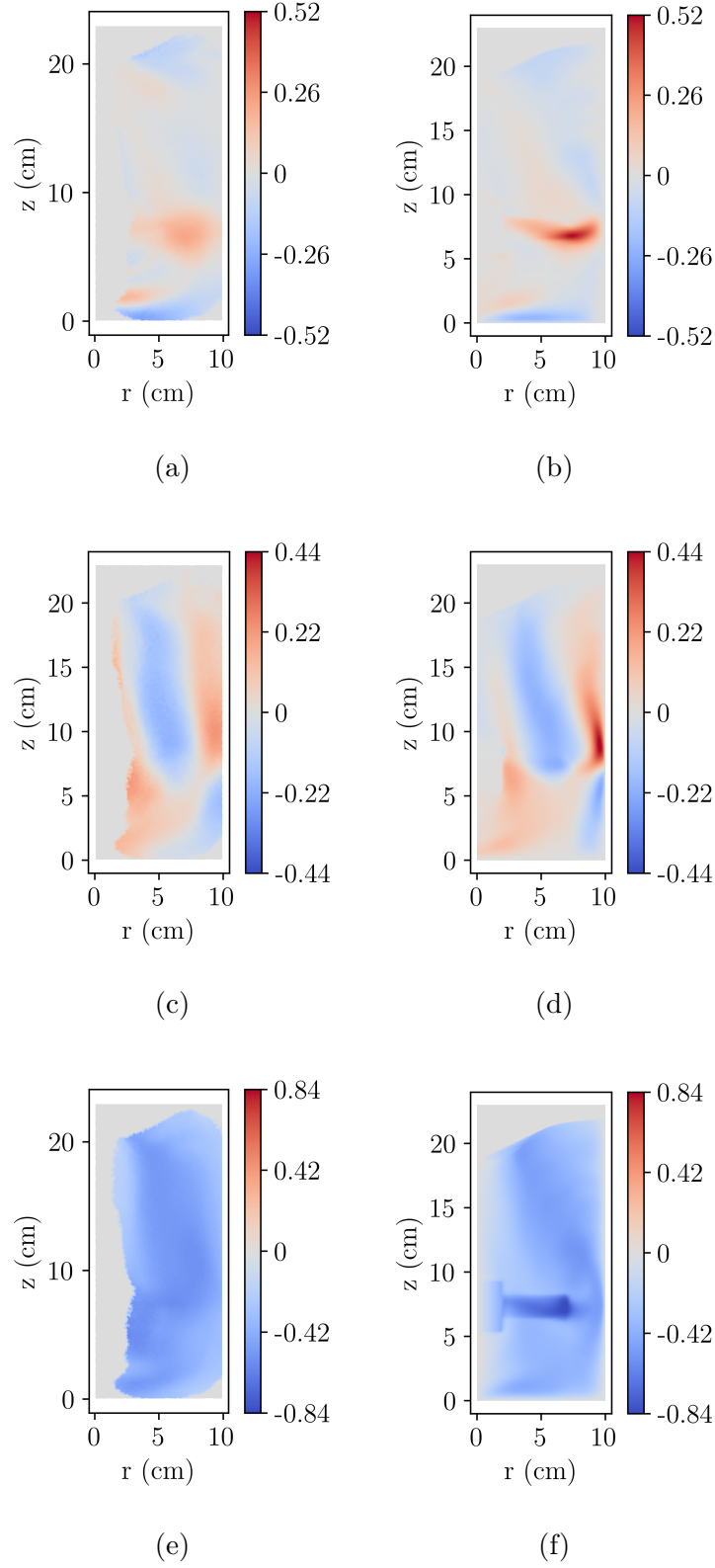


Figure 7.8 Azimuthally averaged velocity (m/s): [left] RPT, [right] CFD components (multiphase simulation); a,b: radial, c,d: axial and e,f: tangential.

7.3.5 Prediction of averaged Velocities

Figure 7.9 presents the axial profile of azimuthally averaged, time-averaged velocity components at two radial positions: one at the impeller tip ($r/R = 0.7$) and another near the sidewall ($r/R = 0.93$). It compares the results obtained from the RPT technique with those from single-phase and multiphase CFD simulations.

There is a significant difference between the RPT results and both CFD approaches in all three velocity components within a very narrow region around the impeller. This discrepancy is especially pronounced in the axial profile of the radial velocity. As highlighted in Figure 7.10, the high-velocity region at the radial jet is less than 5 mm wide, and this difference diminishes substantially as the radial distance from the impeller increases.

At the impeller height where the velocity is highest in Figure 7.9a, the single-phase and multiphase simulation results show less than 1% discrepancy, while the RPT results deviate by 50%. In contrast, at the same height close to the sidewall in Figure 7.9b, the multiphase simulation results are 33% lower than the single-phase results and 53% higher than the RPT measurements.

Another clear inconsistency in the radial velocity component is observed between the single-phase and multiphase CFD simulations in the upper third of the tank ($0.6 < z/H < 1$), where the results follow completely different trends. In contrast, the multiphase simulation agree with RPT measurements.

Considering axial velocity at the impeller tip (Figure 7.9c), the results from all three approaches generally align in both trend and magnitude, except in the region $0.7 < z/H < 1$, where the RPT velocity moves in the opposite direction to the CFD simulations. Additionally, near the sidewall at $z/H = 0.45$ (Figure 7.9d), the differences are most pronounced, with the multiphase simulation being 13% lower than the single-phase simulation and 34% higher than the RPT results.

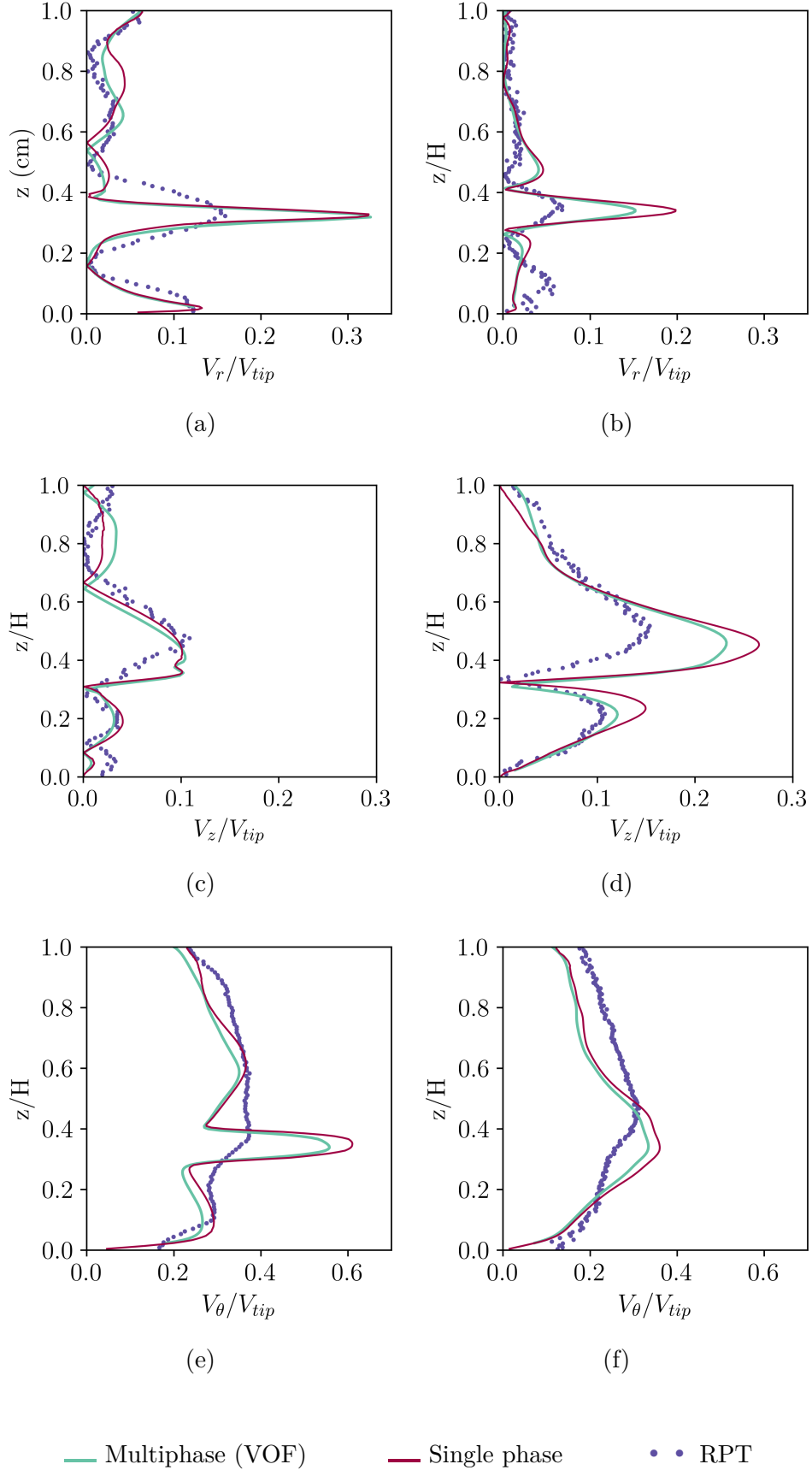


Figure 7.9 Axial profile of normalized radial, axial, and tangential components of azimuthally averaged velocities; a,c,e at radial distance of $r/R = 0.7$ and b,d,f at $r/R = 0.93$.

For the azimuthal velocity component, which has the highest magnitude of all velocity components, the results from the single-phase and multiphase simulations are comparable for the most part. However, at the radial discharge of the impeller, the multiphase simulation yields lower values compared to the single-phase simulation, with a 9% difference at both the impeller tip and the sidewall profile. In the high-velocity region of the impeller discharge jet, RPT results at the impeller tip are 36% higher than the multiphase simulation, but this difference decreases as the axial distance from the impeller increases.

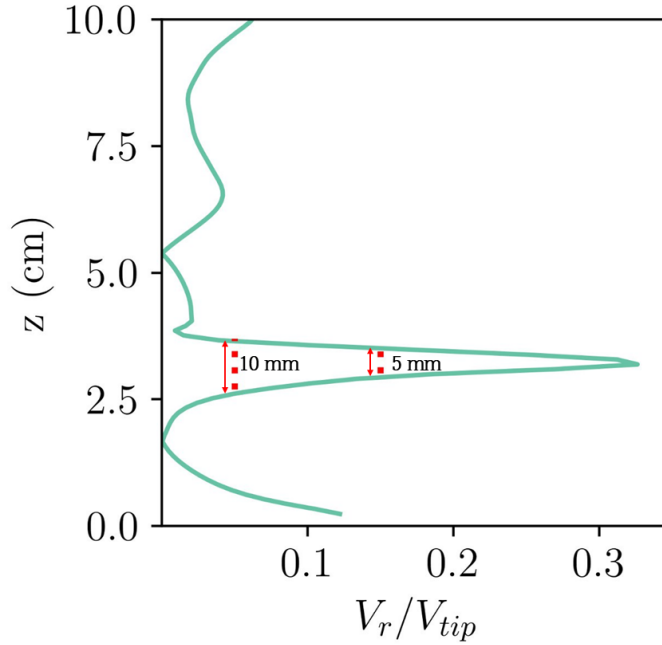


Figure 7.10 Visualization of the high-velocity region and its width at the impeller tip radial position.

The discrepancy between the RPT and CFD results in the narrow region around the impeller may primarily be attributed to the particle size (3.5 mm), which is relatively large and not fully capable of resolving flow structures in high-velocity regions. In these regions, where velocity acceleration is high and the flow direction shifts by 90° from vertical to radial, the importance of the Stokes number becomes apparent. With a Stokes number of 0.15, the particle's ability to follow rapid velocity changes is lower in the impeller region.

Similar to observations in this work, Plusa et al. [30], in their study comparing velocity profiles in unbaffled, uncovered stirred tanks using Laser Doppler Anemometry (LDA) and multiphase CFD simulations, noted an overestimation in the multiphase simulation results in the impeller region compared to LDA measurements. Moreover, results from Alberini et

al. [118] indicate that flow in the impeller region is particularly unstable in the transitional regime, which could further contribute to the discrepancies observed.

Additionally, other sources of error should not be overlooked. For instance, shaft vibrations could further contribute to this discrepancy. Furthermore, given that the tracer particle size exceeds the Kolmogorov scale ($d/\eta \approx 5.6$), it would interact with the turbulence inside the experimental tank. Consequently, the conditions in the experiment and simulation are not perfectly identical.

7.4 Conclusion

Experimental measurements using the RPT technique and computational simulations were conducted for an uncovered, unbaffled stirred tank. Established correlations from the literature for calculating the central vortex depth were applied to validate the results from both the experiments and CFD simulations. The depth of the vortex and the critical radius, which separates the forced and free vortex regions, predicted by RPT and multiphase simulations, were in good agreement with these correlations.

The performance of all three approaches was compared by analyzing time-averaged, azimuthally averaged velocity components. Near the central vortex in the upper third of the tank, single-phase and multiphase simulations showed different trends, particularly in the radial velocity component. Elsewhere in the tank, they followed similar patterns. Across all velocity components, multiphase simulations consistently yielded lower values than single-phase simulations, with the largest deviation, around 33%, observed near the impeller.

Multiphase simulations required significantly more computation time than single-phase simulations. The single-phase simulation took 40 hours on 2 nodes of the Digital Research Alliance of Canada's Beluga cluster, while the multiphase simulation extended to 374 hours on 4 nodes of the Narval cluster.

The multiphase simulations aligned more closely with the RPT results, particularly in capturing the radial velocity structure near the central vortex and the upper third of the tank. In contrast, the single-phase simulations failed to replicate these key features. Overall, the lower velocity values from the multiphase simulations were a better match to the RPT experiment compared to the higher values observed in the single-phase simulations.

The differences observed between the RPT and simulation results can likely be attributed to the limitations of the particle size in fully resolving flow structures and its larger size compared to the Kolmogorov scale, which influences turbulence. Additionally, the Stokes number indicates that the particle does not perfectly follow the streamlines, along with the

high instability in the impeller discharge zone during the transitional regime. Although the CFD simulations are time and mesh independent, they may not fully capture all the physical phenomena present in the experiment, leading to further discrepancies between the experimental observations and simulation results.

Despite the differences, single-phase simulation was considerably less computationally expensive, making it a faster option for studying unbaffled tanks. Additionally, both RPT and multiphase simulation proved they are reliable in capturing complex characteristics of flow in the transitional regime, and their use could be extended to more complex systems, such as solid-liquid unbaffled tanks operating in transitional regime.

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CHAPTER 8 GENERAL DISCUSSION

The main objective of this thesis can be reformulated as :

Development of an RPT technique independent of nuclear radiation models for the characterization of industrial-scale reactors.

This main objective was broken down into several sub-objectives, each contributing to the overall goal.

This work initially aimed to use conventional RPT to investigate the hydrodynamics of large-scale industrial reactors. Therefore, the first sub-objective was the development of a position reconstruction algorithm with more accuracy than the existing ones. In Chapter 5 (Article 1), a novel reconstruction method, FEM-PR, was introduced. It divides the domain into smaller cells and reconstructs the tracer particle positions by comparing the expected photon count at each cell's nodal points with the experimental values. FEM-PR combines a phenomenological model with the Monte Carlo method to estimate the expected photon counts. The details of the calculations required by the phenomenological model are discussed in Appendices A, B, and C. Additionally, to prepare the method for real-time application, FEM-PR was designed to use information from the particle's previous location in the free-flowing experiment. By narrowing the search area and focusing on localized processing, this approach also reduced the overall computational cost of the process.

As the project progressed, the idea of using the cobot to collect experimental data emerged. Initially, the data gathered by the cobot was intended as input for the FEM-PR model. The cobot was expected to sample certain points, and the radiation data collected from these points would subsequently be interpolated to calculate nodal counts. This strategy would allow the model to operate in a hybrid application, using either experimental data or data from the phenomenological model.

The decision to use experimental data for FEM-PR was developed due to the challenges that existing position reconstruction techniques face with optically opaque objects like shafts or baffles. Yadav et al. [21] demonstrated that conventional reconstruction techniques, which depend directly on nuclear radiation models, are highly inefficient in such scenarios. Additionally, machine learning algorithms require larger training datasets when obstacles are present. FEM-PR was expected to manage the presence of optically opaque objects when fed experimental data, as this data would be independent of any models. Furthermore, by sampling the volume with obstacles, the cobot would ensure that the dataset naturally captured

this complexity.

Contrary to our expectation, in the final phase, this work encountered significant complications related to the RPT hardware system, and as a result, the concept was never validated.

The core work of this thesis is presented in Chapter 6 (Article 2), where the integration of the cobot into the data collection process is explained. As the cobot moves, it records the position and time of the particle, while the RPT hardware simultaneously registers the photon counts detected by each detector at specified sampling intervals. The first step in using these two distinct datasets was linking the positions to the counts, using time as a common parameter. However, these systems operated on different internal clocks, which required an external synchronization (handshake) mechanism. Proper protocols were developed to synchronize these two systems and provide clean dataset for training the ANN model.

Dynamic sampling was selected over static sampling to mitigate errors associated with dynamic bias. The extended stationary period in static sampling allows the averaged photon count to stabilize, reducing noise from the stochastic behavior of γ -rays. In contrast, dynamic sampling provides data more representative of instantaneous conditions seen in free-flowing experiments. However, data obtained through dynamic sampling, similar to that from free-flowing experiments, contains noise that, if not properly addressed, can affect the accuracy of reconstructed positions. Degaleesan et al. [47] suggested that noise in free-flowing photon counts may not significantly impact mean quantities, such as average velocities. In contrast, this work revealed that inadequate filtering of dynamic sampling data or free-flowing counts can indeed affect the averaged velocities. Various filtering techniques were explored, the Savitzky-Golay filter showed to be effective in reducing noise from γ -rays stochasticity.

The model-free RPT technique was verified and validated through various experiments, demonstrating that it is the most accurate approach for reconstructing particle positions based on radiation from a radioactive tracer. However, the following aspects were not investigated within the context of this thesis and should be considered in future work:

- The cobot records information from trajectories connecting a set of points. This work tested only one set of trajectories at a constant velocity while moving through the points. Thus, determining the optimal set of trajectories and the cobot's velocity is crucial for deriving the most information in the shortest time.
- Only one orientation of detectors around the volume was tested. Therefore, the detectors' orientation must be optimized to maximize sensitivity and resolution.

To showcase its ability to provide hydrodynamic information, mixing in an unbaffled, un-

covered tank was characterized. These tests were conducted under two different regimes: laminar and transitional. Mixing in the laminar regime was addressed in Chapter 6, while the characterization of the transitional regime is the core of Chapter 7 (Article 3). The key variables in these experiments were the higher impeller rotational speed, as well as the lower viscosity and density of the fluid in the transitional regime case compared to the laminar regime. RPT results were in close agreement with CFD results for mixing in the laminar regime. When comparing RPT and CFD for mixing in the transitional regime, it is evident that both replicate the same flow pattern. Although they are generally in close agreement, some deviations occur in the narrow region around the impeller. The probable reasons for this discrepancy are discussed below.

Preparing the tracer particle for the transitional regime was particularly challenging. The particle must meet specific criteria, most notably having a density that matches the fluid to achieve neutral buoyancy. This was especially difficult with a low-density fluid. Since Scandium is nearly three times denser than the fluid, neutral buoyancy was achieved by trapping air bubbles inside the epoxy used to encapsulate the Scandium. However, reducing the amount of Scandium was not feasible, as this would require significantly longer activation times in the nuclear reactor. Although increasing the amount of epoxy and air helped achieve neutral buoyancy, it also substantially increased the particle size.

Another key criterion for a tracer particle in any particle tracking method is its Stokes number, which must be less than 0.1 to ensure the particle closely follows the flow streamlines without detaching. In the transitional regime, increasing the particle diameter and reducing the fluid's viscosity resulted in a higher Stokes number compared to the laminar regime. This may explain why the particle near the impeller struggled to respond to sudden changes in both velocity and flow direction, leading to the disappearance of the narrow high-velocity region around the impeller tip.

The discrepancies between the RPT and CFD results in the transitional regime are not only due to the particle's size affecting how it follows the streamlines. The particle, being 5.6 times larger than the Kolmogorov length scale, cannot follow flow structures smaller than itself and also interacts with turbulence, disturbing the natural flow. This complex interaction, which exists in the real experiment, was not considered in the CFD simulations, opening the door for future work to characterize the effect of the particle on the flow using numerical simulations.

Furthermore, due to the complexity of programming the cobot to sample the tank in the presence of the shaft and impeller, sampling was conducted in their absence. However, the presence of these objects leads to the attenuation of γ -rays, and if not considered, this effect can impact the results, particularly in regions closer to these objects.

CHAPTER 9 CONCLUSION

This section summarizes the work accomplished in this thesis to present its contributions to the existing body of literature. Furthermore, the limitations of the proposed technique are discussed, along with potential future work.

9.1 Summary of Works

In the first part of this thesis, a novel position reconstruction method, FEM-PR, is developed to triangulate the position of a radioactive tracer particle. FEM-PR works based on the principle of the inverse of finite element method; it divides the domain into simplex elements and, within each element, triangulates the position by comparing the photon counts at each node between the experimental and expected values. In this work, a phenomenological model provides the expected nodal values for photon counts. FEM-PR uses information from prior time steps of the tracer particle's trajectory to perform a local search around previous positions. This approach reduces computational cost and enables faster convergence, making the technique suitable for real-time experiments. These capabilities are verified through a benchmark, and an experimental test is conducted to validate the technique and determines its accuracy.

Next, a model-free RPT technique is developed. In this novel approach, experimental data replaces the data calculated by the phenomenological model. A collaborative robot precisely carries a radioactive tracer within the domain, registering the tracer's position at specified time intervals. Linking these positions to the radiation levels received by the array of scintillation detectors around the domain provides a substantial dataset. An ANN model receives the radiation information as input and the corresponding positions as output during the training phase. Subsequently, the trained ANN model will be capable of predicting the radioactive tracer's trajectory in a free-flowing experiment using the instantaneous photon counts received by surrounding detectors. Model-free RPT is validated through a controlled case of reconstructing a spiral trajectory in air. The ANN model, trained with data collected through the motion of particles connecting a near-random set of data points generated by the LHS technique, demonstrated significantly more accurate results compared to the data collected through the movement of particles on a Cartesian grid covering the volume.

The adequacy of the model-free RPT in predicting hydrodynamic parameters is evaluated in an unbaffled, uncovered stirred tank equipped with a PBT, operating in both laminar

and transitional regimes. A suitable post-processing algorithm transformed the Lagrangian flow fields derived from RPT into azimuthally averaged Eulerian flow fields, which are then benchmarked against CFD simulations. Despite the uncertainties inherent in each technique, good agreement is observed, particularly in the laminar regime, where both techniques delivered very close results. In the transitional regime, the swirling effect led to vortex formation around the shaft. To address this, in addition to single-phase mixing, a multiphase simulation combined with the VOF method is conducted to capture characteristics associated with the vortex presence. Implicit LES is applied in the CFD simulation to capture turbulence. This work demonstrated that RPT can predict both the vortex depth and the critical radius separating the forced and free regions of the vortex. Results from RPT are compared with a multiphase simulation and Nagata's model from the literature, all showing strong agreement. It is found that RPT results closely align with those from multiphase simulation, whereas single-phase simulations generally overpredict average velocities in all three components compared to RPT and multiphase simulation. Additionally, it is observed that the radioactive tracer particle designed for the transitional regime could not capture the narrow high-velocity region surrounding the impeller.

The main contribution of this work is the replacement of nuclear radiation models with experimental data in RPT experiments. This novel approach improves the technique's accuracy and resolves its most significant limitation, the need for extensive calibration. RPT is now a reliable tool for characterizing the hydrodynamics of large-scale multiphase reactors.

9.2 Limitations

Although the model-free RPT developed in this thesis offers several advantages over model-dependent RPT, and addresses significant limitations of the latter discussed in Section 2.1.3, it still has some limitations and weaknesses.

The major remaining weakness is the size of the radioactive tracer particle. As discussed in Section 2.3.1, tracer particles manufactured for RPT experiments are typically a few millimeters in size. This limits the information that can be obtained from their trajectories: first, they cannot track flow structures smaller than their own size; second, as particle size increases, interaction with turbulence becomes significant, potentially disturbing the natural flow; and finally, a larger particle size increases the Stokes number, reducing the particle's ability to respond to sharp flow changes, such as sudden shifts in direction and velocity near the impeller in a stirred tank.

9.3 Future Research

To address the aforementioned limitation and further enhance and optimize the model-free RPT technique, several directions for future work are possible:

- Verifying the performance of the FEM-PR position reconstruction technique using experimental data collected by the cobot, independent from the phenomenological model.
- Modifying the cobot's tool for particle movement to enable data collection in the presence of external objects, such as the shaft and impeller, and to allow the cobot to maneuver the particle closer to the geometry walls for collecting information in those areas.
- Collecting data in the presence of optically opaque objects and verifying the accuracy of the ANN model and FEM-PR with these objects included.
- Optimizing the set of cobot's trajectory for to reduce sampling time while improving data quality.
- Optimizing the velocity at which the cobot moves the particle.
- Developing an advanced technique for manufacturing smaller tracer particles, while maintaining critical physical properties like particle density.
- Optimizing the orientation of the detectors.
- Considering the tracer particle in CFD simulation to assess its effect on the flow and maintain conditions closer to the experiment.

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APPENDIX A SOLID ANGLE CALCULATION

A.1 Calculation of the parameters h and ρ

We determine the *particle-detector* coordinate based on the relative position of the particle with respect to the detector [14]. Considering point $\vec{X}_P$ as the source position, point $\vec{X}_d$ as the position of the detector face center and $\vec{X}_a$ as a point inside the detector on the axis. The vector between points $\vec{X}_d$ and $\vec{X}_a$ determine the detector orientation vector, $\vec{e}_z$, which is parallel to the z' axis of the particle-detector coordinate:

$$\vec{e}_z = \frac{\vec{X}_d - \vec{X}_a}{\|\vec{X}_d - \vec{X}_a\|} \quad (\text{A.1})$$

In the *particle-detector* reference frame, particle location on the z' axis and its coordinate z'_p can be obtained by the projection of $\vec{X}_{dp}$ on the z' axis as:

$$z'_p = \vec{X}_{dp} \cdot \vec{e}_z \quad (\text{A.2})$$

where $\vec{X}_{dp}$ is given by:

$$\vec{X}_{dp} = \vec{X}_p - \vec{X}_d \quad (\text{A.3})$$

As Figure A.1 shows, z'_p is the distance between the particle and detector face, which is equal to h . The X' coordinate of the particle-detector frame can be defined using $\vec{X}_{dp}$ and $\vec{Z}'$ as:

$$\vec{X}' = \vec{Z}' - \vec{X}_{dp} \quad (\text{A.4})$$

$$\vec{e}_x = \frac{\vec{X}'}{\|\vec{X}'\|} \quad (\text{A.5})$$

where:

$$\vec{Z}' = z'_p \times \vec{e}_z \quad (\text{A.6})$$

As Figure A.1 shows ρ is equal to:

$$\rho = \|\vec{X}'\| \quad (\text{A.7})$$

Consequently, the last basic axis, $\vec{e}_y$, can be defined using the cross product of two other coordinates as:

$$\vec{e}_y = \vec{e}_z \times \vec{e}_x \quad (\text{A.8})$$

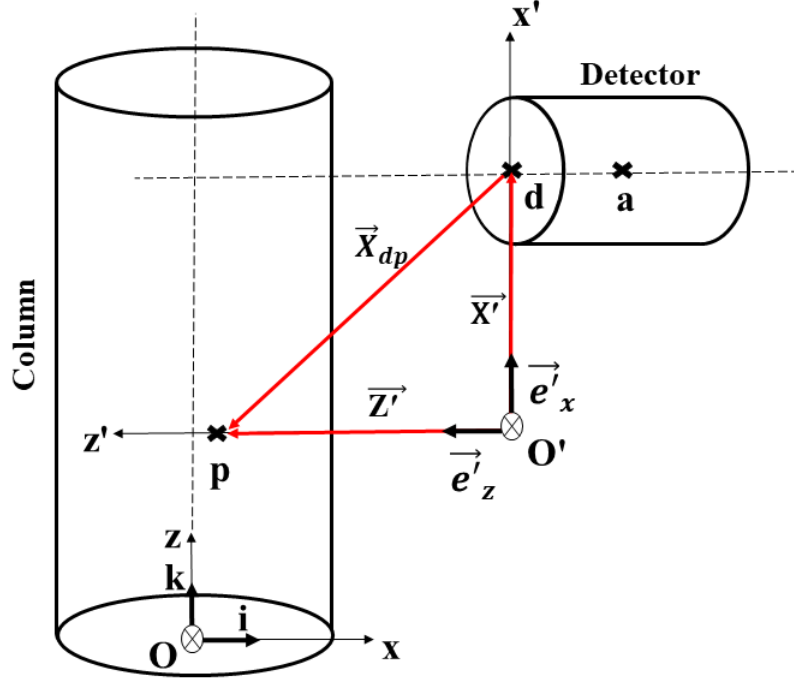


Figure A.1 Particle-detector frame configuration.

γ -ray can enter the detector from the lateral side of the detector, the top of the detector, or both the top and the lateral side. The entrance location depends on the region in which the particle is located with respect to the detector (see Figure A.2 and Figure A.3). These regions can be identified using the values of h and ρ :

- $h \geq 0$ and $\rho > r$ (S_1): The γ rays emitted by the tracer can enter the detector from both top and the lateral side of the crystal.
- $h < 0$ and $\rho > r$ (S_2): The γ rays emitted by the tracer enters the detector from the lateral side of the crystal.
- $h > 0$ and $\rho < r$ (S_3): The γ rays emitted by the tracer enters the detector only from the top surface.

The following sections discuss the calculation of $\omega(\alpha)$ and $\omega(\theta)$.

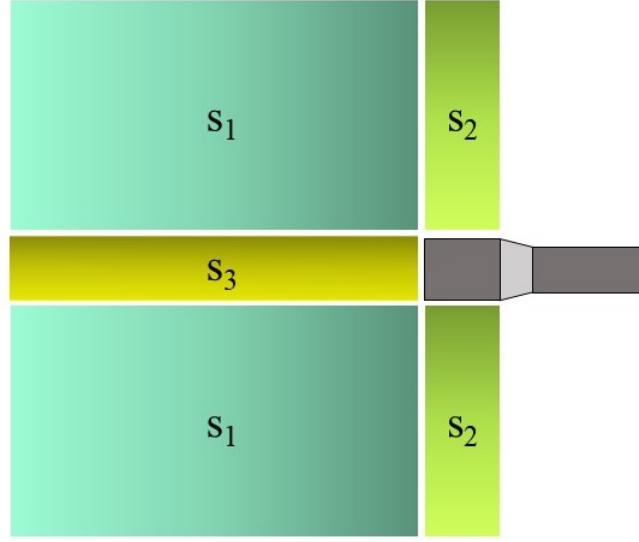


Figure A.2 Different regions around the detector based on h and ρ values.

A.2 S_1, S_2 : Calculation of $\omega(\alpha)$

When particles are located in regions S_1 and S_2 , α is:

$$\alpha = \alpha_{\max}(2n_1 - 1) \quad (\text{A.9})$$

with

$$\alpha_{\max} = \arcsin\left(\frac{r}{\rho}\right) \quad (\text{A.10})$$

and where $n_1 \in [0, 1]$, is a random number sampled from a uniform distribution. Thus $-\alpha_{\max} \leq \alpha \leq \alpha_{\max}$.

The weighting factor associated with this selection of α , $\omega(\alpha)$, is:

$$\omega(\alpha) = \frac{\alpha_{\max}}{\pi} \quad (\text{A.11})$$

A.3 S_1 : Calculation of $\omega(\theta)$

The angle θ which determines the angle along which the photon strikes the detector is calculated using $n_2 \in [0, 1]$, another random number sampled from a uniform distribution:

$$\theta = \arccos(\cos(\theta_{\min}) - n_2[\cos(\theta_{\min}) - \cos(\theta_{\max})]) \quad (\text{A.12})$$

The associated weighting factor, $\omega(\theta)$ is:

$$\omega(\theta) = \frac{\cos(\theta_{\min}) - \cos(\theta_{\max})}{2} \quad (\text{A.13})$$

When $h > 0$, $\theta_{\min}$, $\theta_{\max}$ and θ_{cri} are:

$$\theta_{\max} = \arctan\left(\frac{\overline{OA}}{h}\right) \quad (\text{A.14})$$

$$\theta_{\min} = \arctan\left(\frac{\overline{OB}}{h+l}\right) \quad (\text{A.15})$$

$$\theta_{\text{cri}} = \arctan\left(\frac{\overline{OB}}{h}\right) \quad (\text{A.16})$$

where l is the length of the detector.

For $h = 0$, the tracer is located at O in Figure A.3a and the angles $\theta_{\max}$, $\theta_{\min}$ and θ_{cri} are:

$$\theta_{\max} = \frac{\pi}{2} \quad (\text{A.17})$$

$$\theta_{\min} = \arctan\left(\frac{\overline{OB}}{l}\right) \quad (\text{A.18})$$

$$\theta_{\text{cri}} = \frac{\pi}{2} \quad (\text{A.19})$$

A.4 S_2 : Calculation of $\omega(\theta)$

For a particle inside the S_2 region we calculate θ and $\omega(\theta)$ using Equations A.12 and A.13 and the angles $\theta_{\max}$, $\theta_{\min}$ and θ_{cri} are:

$$\theta_{\max} = \frac{\pi}{2} + \arctan\left(\frac{|h|}{\overline{OB}}\right) \quad (\text{A.20})$$

$$\theta_{\min} = \arctan\left(\frac{\overline{OB}}{l - |h|}\right) \quad (\text{A.21})$$

$$\theta_{\text{cri}} = \frac{\pi}{2} + \arctan\left(\frac{|h|}{\overline{OB}}\right) \quad (\text{A.22})$$

A.5 S_1, S_2 : Calculation of $\overline{OA}$ and $\overline{OB}$

Referring to Figure A.3a, the line segment $\overline{OA}$ and $\overline{OB}$ are defined as:

$$\overline{OA} = \rho \cos \alpha + \left(r^2 - \rho^2 \sin^2 \alpha \right)^{\frac{1}{2}} \quad (\text{A.23})$$

$$\overline{OB} = \rho \cos \alpha - \left(r^2 - \rho^2 \sin^2 \alpha \right)^{\frac{1}{2}} \quad (\text{A.24})$$

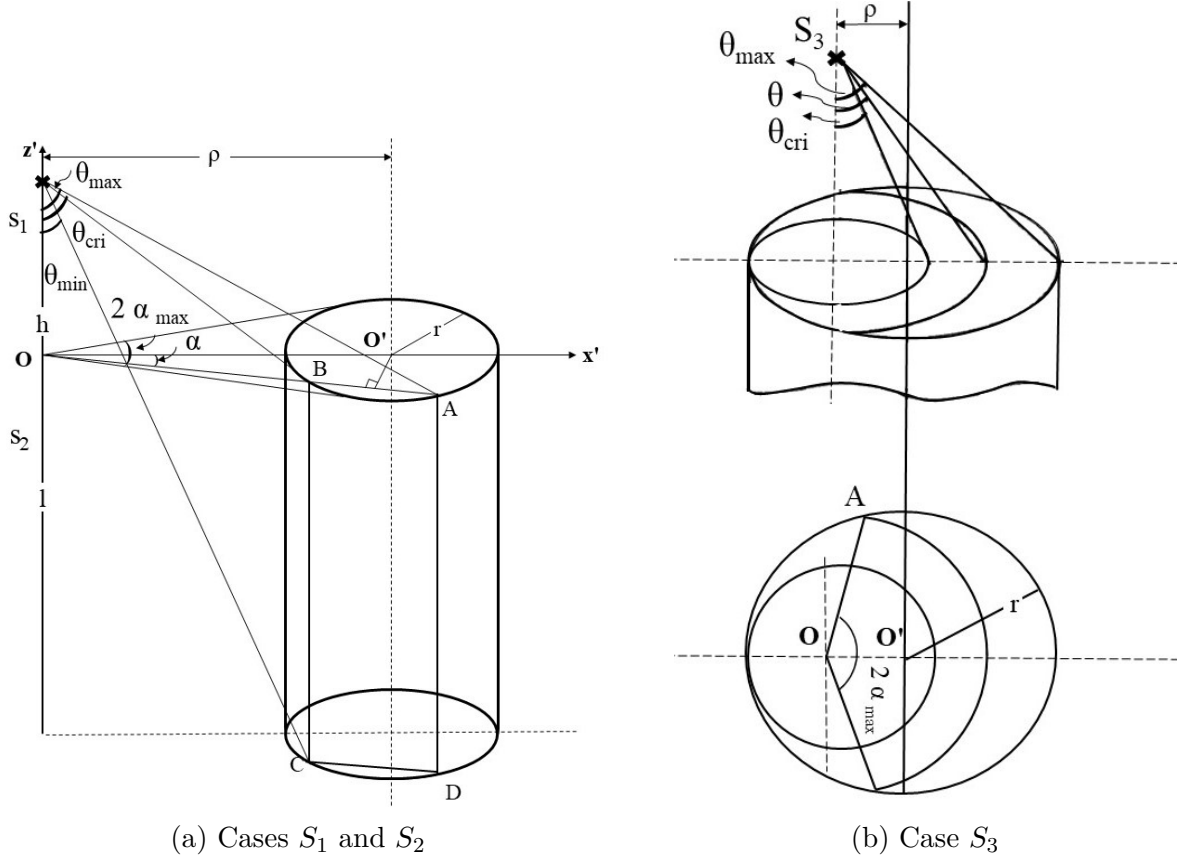


Figure A.3 Different scenarios for the relative position of the particle and the detector (adapted from [119]).

A.6 S_3 : Calculation of $\omega(\theta)$ and $\omega(\alpha)$

For this case, according to the geometric configuration, $\theta_{\max}$ is independent of α . Consequently, θ is calculated first and based on that α will be determined. In this case, when $\theta < \theta_{\text{cri}}$, α can be any angle between zero and 2π . However, for $\theta > \theta_{\text{cri}}$, α will be restricted

by $2\alpha_{\max}$. Inside the S_3 region, $\theta_{\min}$ is always equal to zero and $\theta_{\max}$ and θ_{cri} are defined as:

$$\theta_{\max} = \arctan\left(\frac{r + \rho}{h}\right) \quad (\text{A.25})$$

$$\theta_{\text{cri}} = \arctan\left(\frac{r - \rho}{h}\right) \quad (\text{A.26})$$

Equation (A.12) expresses the angle θ and the Equation (A.9) defines the angle α . If $\theta < \theta_{\text{cri}}$, $\omega(\theta)$ is equal to one. Otherwise for $\theta > \theta_{\text{cri}}$, Equation (A.11) determines it and in this equation $\alpha_{\max}$ is equal to:

$$\alpha_{\max} = \arccos\left[\frac{(\rho^2 + h^2 \tan^2 \theta - r^2)}{2h\rho \tan \theta}\right] \quad (\text{A.27})$$

APPENDIX B CALCULATION OF TRAVELLED DISTANCE BY γ -RAY INSIDE THE REACTOR

The length of the path that a photon travel inside the reactor is given by:

$$x(\alpha, \theta) = \|\vec{X}_{\text{particle}} - \vec{X}_{\text{intersection}}\| \quad (\text{B.1})$$

where $\vec{X}_{\text{intersection}}$ is the intersection point of the line and the circle. The circle equation representing the perimeter of the column in the *laboratory* coordinate framework is:

$$x^2 + y^2 = R^2 \quad (\text{B.2})$$

where R is the reactor radius. The equation of a straight line in parametric form is given by:

$$x = x_1 + t \cos \alpha \quad (\text{B.3})$$

$$y = y_1 + t \cos \beta \quad (\text{B.4})$$

$$z = z_1 + t \cos \gamma \quad (\text{B.5})$$

where $P_1(x_1, y_1, z_1)$ is a point on the line and $\cos \alpha$, $\cos \beta$, $\cos \gamma$ are the direction cosines of the line's direction vector, the parameter t can be any real number. At each position based on the illustrated geometry in Figure B.1, the traced ray has the angle $\pi - \theta$ with z' -axis and the unit direction vector's components are:

$$r = (\cos \alpha, \cos \beta, \cos \gamma) \quad (\text{B.6})$$

where:

$$|r|^2 = \cos^2 \alpha + \cos^2 \beta + \cos^2 \gamma = 1 \quad (\text{B.7})$$

The three direction cosines are not independent and one of them is taken to be implicitly dependent on the other two. The traced ray angle with the x' -axis, γ , is given by:

$$\cos \gamma = \frac{\overline{EF}}{\overline{EG}} \quad (\text{B.8})$$

$$z'' = z_p'' + t \cos(\pi - \theta) \quad (\text{B.15})$$

To transfer the coordinate of a point into a new coordinate framework, we move the origin of the old framework in each direction toward the origin of the new one. The new coordinate of each point (x, y, z) can be obtained from:

$$x_t = x - x_o \quad (\text{B.16})$$

$$y_t = y - y_o \quad (\text{B.17})$$

$$z_t = z - z_o \quad (\text{B.18})$$

where (x_t, y_t, z_t) is the translated position and (x_o, y_o, z_o) is the origin of the *particle-detector* coordinate in the *laboratory* framework given by:

$$\vec{X}_o = \vec{X}_{Particle} - \vec{h} \quad (\text{B.19})$$

where:

$$\vec{h} = (X_{dp} \cdot \vec{e}_x) \vec{e}_x + (X_{dp} \cdot \vec{e}_y) \vec{e}_y + (X_{dp} \cdot \vec{e}_z) \vec{e}_z \quad (\text{B.20})$$

After implementing the translation, using the following rotation matrix, the new coordinate of each translated point in *particle-detector* coordinate (x'', y'', z'') are:

$$\begin{bmatrix} x'' \\ y'' \\ z'' \end{bmatrix} = \begin{bmatrix} \vec{e}_x \\ \vec{e}_y \\ \vec{e}_z \end{bmatrix} \begin{bmatrix} x_t \\ y_t \\ z_t \end{bmatrix} \quad (\text{B.21})$$

Where $\vec{e}_x$, $\vec{e}_y$ and $\vec{e}_z$ are the unit vectors forming the *particle-detector* frame which Equations (A.5), (A.8), and (A.1) determine them. Therefore by multiplication of the Equation (B.21) in inverse of the transformation matrix $[\vec{e}_x \ \vec{e}_y \ \vec{e}_z]^T$ x_t , y_t and z_t will be obtained as:

$$\begin{bmatrix} x_t \\ y_t \\ z_t \end{bmatrix} = \begin{bmatrix} \vec{e}_x \\ \vec{e}_y \\ \vec{e}_z \end{bmatrix}^{-1} \begin{bmatrix} x'' \\ y'' \\ z'' \end{bmatrix} \quad (\text{B.22})$$

By substituting x_t , y_t and z_t in Equations B.16 to B.18 we can transfer any position from *laboratory* coordinate to *particle-detector* coordinate. Therefore by substituting the equivalent of x , y and z in Equations B.2, B.3, B.4 and B.5 the algorithm finds the intersection point and calculates the traveled distance inside the reactor using Equation B.1.

APPENDIX C CALCULATION OF TRAVELLED DISTANCE BY γ -RAY INSIDE THE DETECTOR

For cases S_1 and S_2 , the photon can enter the detector either from the lateral wall or from the top. If $\theta < \theta_{\text{cri}}$, the photon strikes the lateral surface of the crystal, and may leave the detector from bottom or lateral wall. The angle θ_1 determines the point that the photon exit from the detector, which is shown in Figure C.1, and is equal to:

$$\theta_1 = \arctan \frac{\overline{OA}}{h + l} \quad (\text{C.1})$$

$$x(\alpha, \theta) = \frac{(\overline{OA} - \overline{OB})}{\sin \theta} \quad (\text{C.2})$$

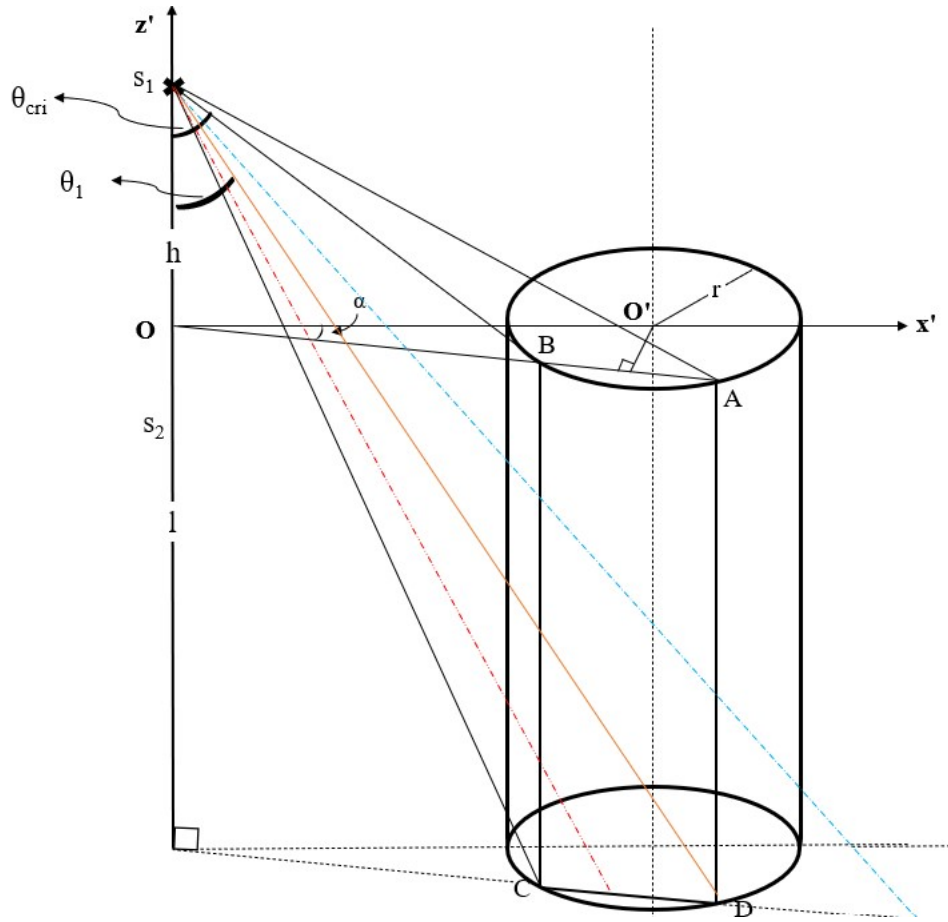


Figure C.1 $\theta < \theta_{\text{cri}}$.

if $\theta < \theta_1$, the photon leaves the detector from the bottom of the cylinder and the path length is:

$$x(\alpha, \theta) = \frac{(h + l)}{\cos \theta} - \frac{\overline{OB}}{\sin \theta} \quad (\text{C.3})$$

where $\overline{OA}$ and $\overline{OB}$ can be obtained from Equations (A.23) and (A.24).

For $\theta > \theta_1$, $x(\alpha, \theta)$ is given by:

$$x(\alpha, \theta) = \frac{(\overline{OA} - \overline{OB})}{\sin \theta} \quad (\text{C.4})$$

When $\theta > \theta_{\text{cri}}$, the photon strikes the detector's top and it can leave from the bottom or the lateral wall of the detector. For $\theta < \theta_1$ as it is illustrated in Figure C.2, the photo enters from the top and leaves from the bottom of the detector, the path length is equal to:

$$x(\alpha, \theta) = \frac{l}{\cos \theta} \quad (\text{C.5})$$

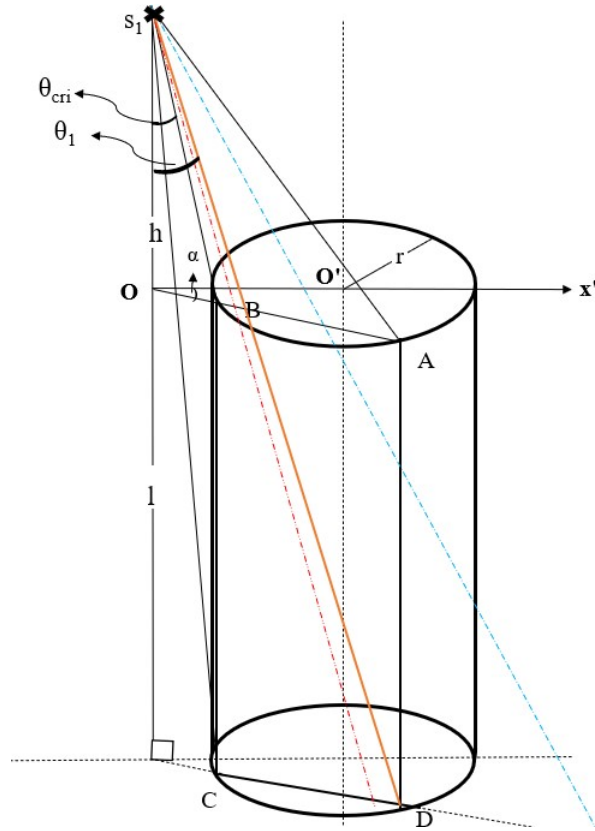


Figure C.2 $\theta > \theta_{\text{cri}}$.

otherwise, for $\theta > \theta_1$, $x(\alpha, \theta)$ is given by:

$$x(\alpha, \theta) = \frac{\overline{OA}}{\sin \theta} - \frac{h}{\cos \theta} \quad (\text{C.6})$$

When the radioactive particle is inside the S_3 region, the angles θ_1 and θ_2 determine the point that photon escapes the detector. As Figure C.3 shows, these angles are equal to:

$$\theta_1 = \arctan \left(\frac{\overline{TI}}{h+l} \right) \quad (\text{C.7})$$

$$\theta_2 = \arctan \left(\frac{\overline{TI}}{h} \right) \quad (\text{C.8})$$

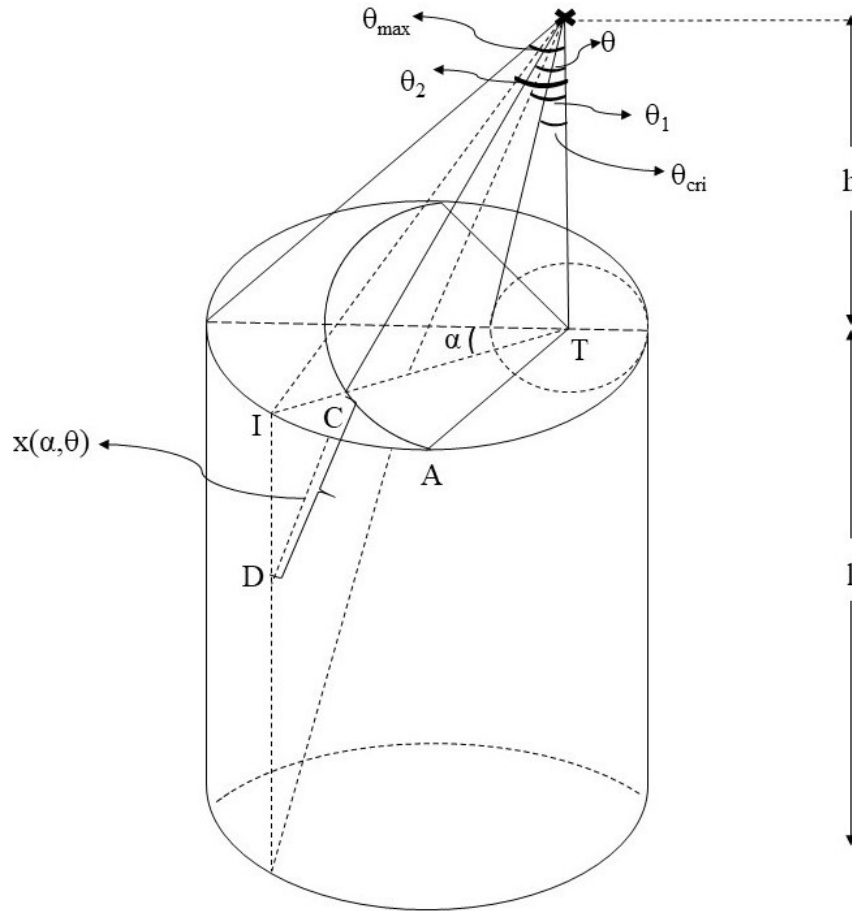


Figure C.3 Geometrical configuration for the photon's path inside the detector when particle locates in S_3 area.

and $\overline{TI}$ (Figure C.4) is given by:

$$\overline{TI} = \rho \cos \alpha + (r^2 - \rho^2 \sin^2 \alpha)^{0.5} \quad (\text{C.9})$$

When $\theta \leq \theta_1$, the photon enters from the top and leaves from the bottom and the path length is:

$$x(\alpha, \theta) = \frac{l}{\cos \theta} \quad (\text{C.10})$$

When $\theta_1 < \theta \leq \theta_2$, the photon enters from the top and leaves from the lateral wall and the path length is:

$$x(\alpha, \theta) = \frac{\overline{TI}}{\sin \theta} - \frac{h}{\cos \theta} \quad (\text{C.11})$$

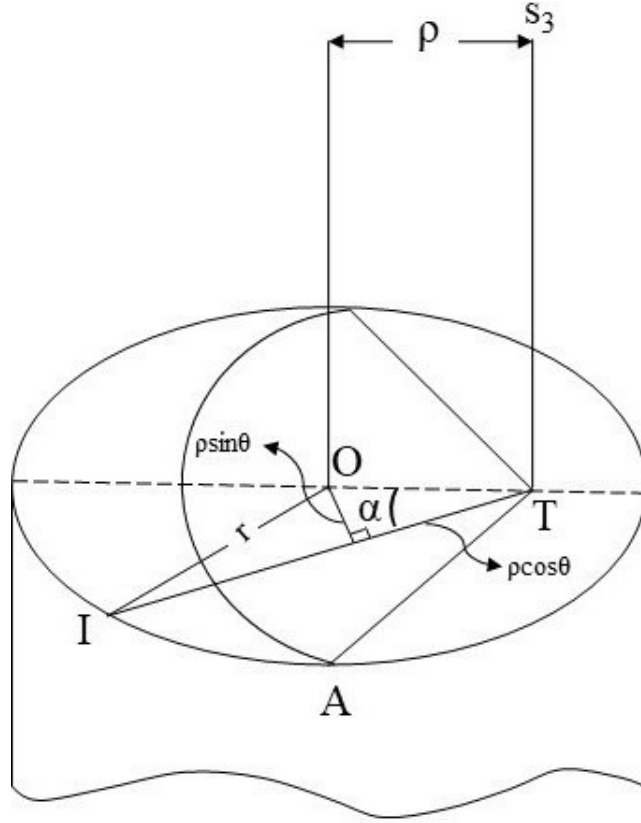


Figure C.4 Geometrical configuration for calculation of $\overline{TI}$.

APPENDIX D SAMPLING TRAJECTORY

Figure D.1 displays the Cartesian grid pattern.

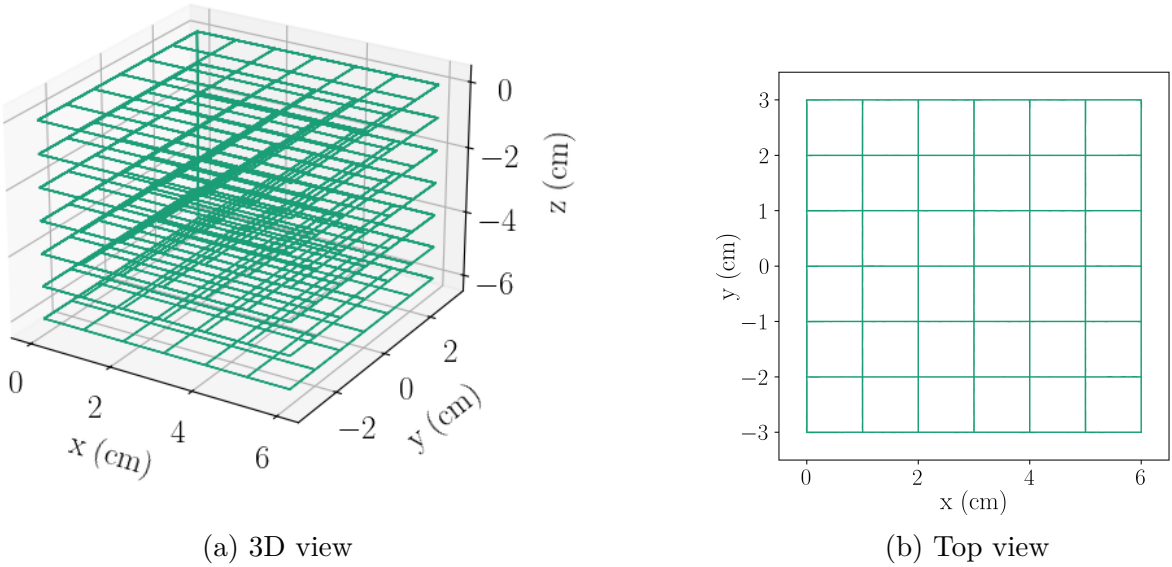


Figure D.1 Pattern of Cartesian grid sampling.

Figure D.2 illustrates the LHS pattern.

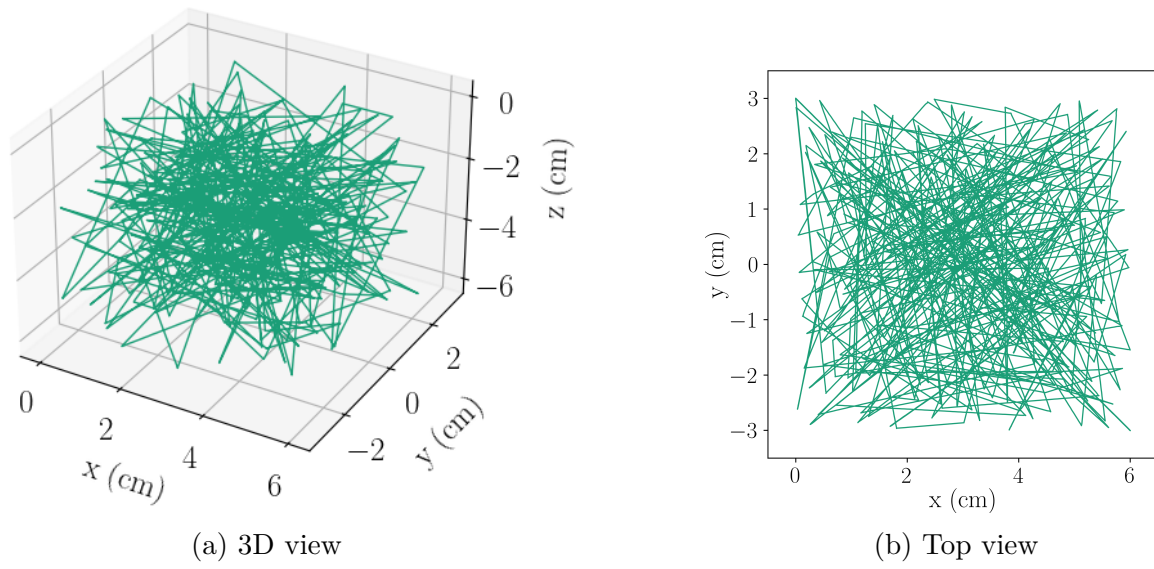


Figure D.2 Pattern of LHS.