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Dynamics of physical flows measured by positron emission techniques

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Abstract. Short lived positron emitting species are used to produce flow-following tracer particles to study flow dynamics in a technique known as positron emission particle tracking (PEPT). The photon pairs produced by positron annihilation are detected in time coincidence by arrays of high-speed position sensitive detectors. Reconstruction of consecutive annihilations are used to determine the near-instantaneous position of the tracer particle. Hence, the resulting bulk flow dynamics are derived, including residence times, velocities, accelerations, and related kinematic properties. The Department of Physics at the University of Cape Town uses PEPT to study dynamic physical processes, turbulent, and multiphase flows. Studies aim to address global challenge topics including problems in water scarce environments, reducing industrial wastes, and enhancing developments towards sustainable economies through improved process efficiencies and design led approaches. The PEPT Cape Town enterprise is discussed, including the development of flow metrology systems and complementary nuclear measurement techniques. Research encompasses four key themes: radioisotope tracer production, instrumentation & detector development, data acquisition & processing, and flow metrology.

1. Nuclear measurement applications

The practical applications of nuclear research and resultant spin off technologies have had high impact for the benefit of human society. To this end the radioisotope tracer method has become an indispensable tool in practically every field of the physical sciences. The use of nuclear techniques applied to diagnosis and treatment in medical healthcare is illustrative [1]. Diagnostic imaging, combining advantages of distinctly different nuclear technologies, has revolutionised personalised healthcare. Of note, transmission imaging using X-ray CT has enabled internal structures of the body to be non-invasively visualised, being sensitive to density and material (Z) variations. Emission imaging (SPECT and PET) has equal relevance, with an injected substance subject to variations within the chemical and molecular environment informing on metabolic processes and function. Modern diagnostic tomography systems combining PET and X-ray CT allow function and structure to be simultaneously visualised and quantified at unprecedented precision. The use of similar techniques for the study of physical systems and those of industrial interest has been explored by a limited number of research groups [2,3] with potential impact appealing to global challenge issues including minimising energy and resource use, enhancing recycling, and enabling a knowledge led approach to industrial design. The University of Cape Town Department of Physics (UCT) has been active in using nuclear techniques to investigate fundamental, real-world, and industrially relevant flows in a wide variety of applications.

2. Emission imaging

The individual particles or fluid elements, in granular, fluid, and mixed-phase flows, cannot be individually distinguished. Radioisotope tracer techniques offer the best approach to differentiating particles from the bulk, which enables a particle-level study of their dynamic behaviour. The measurement process begins with a source(s) of suitable radioisotope (section 3), necessarily short-lived with correspondingly high specific activity. These must be easy to produce, have suitable emissions for detection (with positron emitters being the focus here), and be quickly and easily chemically synthesised within the short timeframe of their half-lives. The radioisotope must be attached to, or otherwise form, the material of interest for the study, being the equivalent of a radioisotope vector molecule in the medical sciences. Representation is critical, for the process to be non-invasive the material properties of the tracer must match those of the bulk. In studying liquids, a neutrally buoyant particle of small scale is used to represent a differential fluid element. Tracer particles are designed to be directly traceable to their mathematical counterparts in continuum mechanics, thus describing the bulk dynamic behaviour.

Photon emissions from a suitable tracer particle(s) are measured using arrays of position sensitive detectors external to the system under study (section 4). These photons carry information correlated with the tracer particle position and are measured with a form of collimation used to determine the direction of incidence. In positron imaging the photon pairs arising from electron-positron annihilation are detected in time coincidence to determine their flight path. The instrumentation and data acquisition systems are required to have low deadtime and high bandwidth to simultaneously acquire data from many thousands of individual detectors. The combined radioisotope, tracer particle, and detection system, enable a suite of novel experimental techniques suitable for investigating flow dynamics (section 5) [4]. A typical PET imaging protocol acquires emission data, applies a set of corrections, and back-projects (or otherwise reconstructs) the resultant integral image. In principle this approach can be used in studying physical systems, with the resulting image representing a time average of density or gated to represent a specific time sequence within the process. As dynamic quantities (including velocities, accelerations, and etc.) are of interest, a differential approach is preferred: following a freely moving tracer particle traversing the system under study. Over long timescales the average motion is equivalent to the integral approach, with the (near-)instantaneous timescale used to investigate dynamic behaviours.

The imaging system is 3-dimensional and regarded as non-intrusive (provided tracer particle representation). The annihilation photons can penetrate opaque materials and physical containment structures, enabling measurements under industrially relevant conditions including high temperatures and/or pressures. Under optimum conditions, with a tracer particle of sufficient activity and detector systems of high intrinsic efficiency and suitable geometry, the recorded coincidence rates are of the order MHz. From the raw data, locations along the tracer particle trajectory can be determined at 10 - 250 kHz rates, allowing high precision tracking with uncertainty proportional to the inverse square-root of the event rate, and ideally smaller than the tracer particle diameter [3]. For moving particles (benchmarked at 1 ms^{-1}), the displacement between successive locations is typically smaller than the measurement uncertainty. Through validation against standard motions, and including a well understood uncertainty budget, the accuracy of tracking is of the same order as the precision.

3. Radioisotope production and tracer particle fabrication

At the iThemba Laboratory for Accelerator Based Sciences (iThemba LABS), UCT has pioneered the use of $^{68}Ge/^{68}Ga$ radioisotope generators as a source of the high specific activity, short-lived, positron emitters required for PEPT. Germanium-68 (half-life 270.95 days) is produced by $^{nat}Ga(p, xn)^{68}Ge$ reactions using a 66 MeV proton beam impinging on a natural gallium target. Following separation and purification, the resulting ^{68}Ge is loaded onto SnO_2 based columns. The long-lived ^{68}Ge decays via electron capture to the short-lived positron emitting isotope ^{68}Ga (half-life 67.71 minutes) in liquid phase. The two isotopes initially exist in secular equilibrium, with a dilute hydrochloric acid solution used to elute the ^{68}Ga when required for tracer particle production via radiochemical techniques. The Radionuclide Production Department at iThemba LABS produces ^{18}F (half-life 109.8 minutes) using the $^{18}O(p, n)^{18}F$ reaction with enriched ^{18}O water targets, irradiated by an 11 MeV proton beam. The isotope is synthesised to the ^{18}F-fluorodeoxyglucose molecule (^{18}FDG) via an automated rapid production cell. Both ^{68}Ge and ^{18}F (obtained from water targets or indirectly from ^{18}FDG) ions can be radiochemically manipulated to extract and concentrate the radioisotopic species. Similar (but distinctly different) ion exchange chromatography techniques are used to produce tracer particles with the radio-species exchanging with counter ions initially attached to a long chain resin polymer backbone [5]. The resin base-layer particle (200 – 500 μm diameter) can achieve a typical uptake of 400 μCi – 2 mCi depending on the chemical conditions in the solution. Once an active resin substrate is produced, further processing refines the physical (size, shape, density, elasticity, hardness, friction coefficient, etc.) and chemical (wettability, hydrophobicity or hydrophilicity, chemical potential, solubility, etc.) properties to match the characteristics of the desired bulk media, thereby attaining tracer particle representation.

With naturally occurring materials being of industrial interest, the ability to activate natural oxygen bearing materials (99.757% ^{16}O) to ^{18}F offers the supreme advantage over radiochemical tracer particle production. Possible reactions include the competing ^{16}O(^{3}He, p)^{18}F and ^{16}O(^{3}He, n)^{18}Ne → ^{18}F using a 35 MeV ^{3}He beam directed onto solid or water targets [2]. At iThemba LABS the competing ^{16}O(α, pn)^{18}F, and ^{16}O(α, d)^{18}F channels, with indirect production from ^{16}O(α, 2n)^{18}Ne → ^{18}F, have been explored [6]. Figure 1 illustrates possible reaction channels on oxygen bearing targets leading to ^{18}F, noting that production from natural oxygen is paramount in producing phase representation.

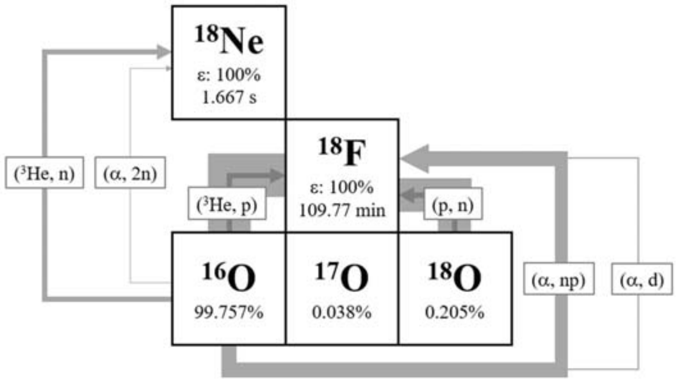

Figure 1. ^{18}F producing reaction channels from oxygen targets. The reaction pathways are scaled to the maximum microscopic cross-section, presented as millibarns / pixel. Data from reference [7].

4. Instrumentation & data acquisition

Large arrays of pixelated detectors are placed surrounding the system under study. The PEPT Cape Town laboratory at iThemba LABS hosts a modified ADAC Vertex dual-headed parallel plane gamma camera (NaI(Tl) scintillators) offering a large field of view and the ability to acquire in single photon (physical collimation) or coincidence (electronic collimation) modes with variable energy windows. The flagship device is a modified Siemens HR++ wide-bore tomograph of diameter 82.0 cm and 23.4 cm axial field of view (Figure 2, left). The HR++ uses detector blocks of 8 × 8 segmented bismuth germinate oxide (BGO) scintillator crystals, viewed by 4 photomultiplier tubes. Groups of 12 blocks are serviced by a detector controller functioning as a front-end data acquisition system, with blocks arranged to give 48 rings of crystal elements 4.1 mm transaxially × 4.0 mm axially × 30 mm radially. The front-end consists of variable gain pre-amplifiers, summing integrators and a single channel analyser (slow channel: determining event energy and crystal position) per block, and CFD timing (fast channel) at nanosecond resolution. Event data from each detector controller are transmitted in parallel to a lossless coincidence processor up to a deadtime limited rate of around 2 MHz per controller, where a coincidence gate of 12 ns is used to assign detected photons to annihilation pairs. Data describing the positions of the endpoints are transmitted over a high-speed fibre optic cable, timestamped, and written to disk storage in listmode format at a maximum rate of 16 MHz.

In the Department of Physics at UCT a dedicated nuclear physics research laboratory has been established. Modular detection systems utilising detector blocks as described above, and of differing dimensions, have been developed for small-scale PEPT applications. A modular positron camera currently consists of 4 modules paired in opposing banks, with horizontal separation of 440 mm (Figure 2, middle). Detectors are controlled through serial communications allowing setting of energy windows, CFD parameters, and energy/spatial/temporal calibrations. Raw singles data are acquired as a serial listmode stream operating at 32.5 MHz, with data from each detector module acquired onto a 32-bit parallel bus. Data streams from up to 10 modules can be acquired simultaneously in parallel. The use of pixelated cadmium-zinc-telluride room-temperature semiconductor arrays (9680 pixels of $1.8 \times 1.8 \times 0.5$ mm^3), is being explored with a hybrid modular camera system consisting of both detector types offering the highest potential for precision measurements of small-scale flows. The design forms a field of view of $150 \times 200 \times 100$ mm^3, with a central high spatial resolution region [8], being described in detail in these proceedings [9]. A Siemens Biograph 16 combined PET/CT tomograph (Figure 2, right) capable of measuring structure (via X-ray attenuation) and function (via emission imaging) has recently been acquired (funded through the UCT Equipment Committee) [10]. The tomograph consists of a rotating X-ray source and opposing pixelated detector system, coupled to a ring of lutetium oxyorthosilicate (LSO) detector blocks of a similar design as described above, with 13 × 13 pixelation. Blocks are arranged into 39 rings of crystal elements of dimension 4.0 mm transaxially × 4.0 mm axially × 20 mm radially giving a uniform field of view of diameter 83.0 cm × 15.6 cm axially. The increased light output and faster decay time of the LSO scintillator improves the energy resolution and timing properties over the BGO based system, offering lower deadtime and higher data acquisition rates resulting in increased measurement precision. A novel fibre optic network-based data acquisition system supporting these devices in arbitrary geometrical configurations has been developed, enabling acquisition of singles and coincidences at the deadtime limited rates produced by the detectors. Real-time acquisition and processing enable the display of singles, coincidence, and tracking data, by sharing the computational load between multiple network nodes and utilising GPU vector processing.

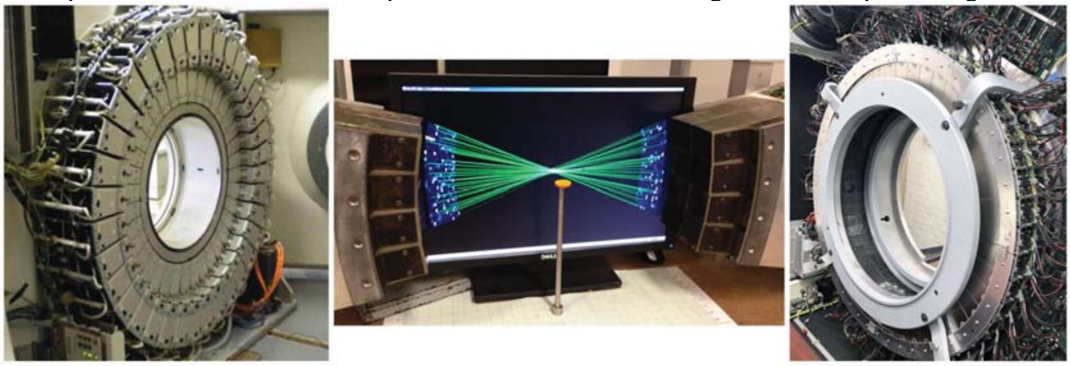

Figure 2. (Left) The HR++ scanner. (Middle) A modular positron camera of opposing banks of block detectors. The live display shows singles events per crystal (blue/white) and coincidence pairs (green lines) converging on the 3D representation of a centrally placed ^{22}Na calibration source (yellow disc). (Right) The Biograph 16 positron tomograph.

5. Positron emission & particle tracking

By regarding the tracer particle as stationary between consecutive coincidence events, the intersection of two reconstructed annihilation pairs is sufficient to localise the instantaneous position. In practice, due to contributions from random coincidences, scattering, and finite volume detector effects (which limit the overall spatial resolution in PET), a small number of consecutive events are used. An iterative minimisation approach rejects outlying events, and position is localised to a precision higher than the intrinsic spatial resolution due to the sampling statistics. The single particle Lagrangian viewpoint is therefore directly measured, with the ability to characterise dynamic processes and rapid changes unconstrained by the steady state or time averaged approaches usually used for flow visualisation. From the instantaneous trajectory data (time, position), first and second order time differentials can be calculated to examine the velocity and acceleration vector fields, noting that these parameters cannot be measured from integral approaches. In a single phase, the local residence time (defined as the relative duration the tracer spends in each image voxel) is equivalent to the local material density. For more complex mixtures, the local densities of individual flow components are determined, being equivalent to steady state PET measurements of material concentration. Advantageously these analyses do not require the usual corrections for attenuation, scattering, or dead-time, and can be resolved to a higher precision than the equivalent integral measurements. Figure 3 illustrates the context of measurement.

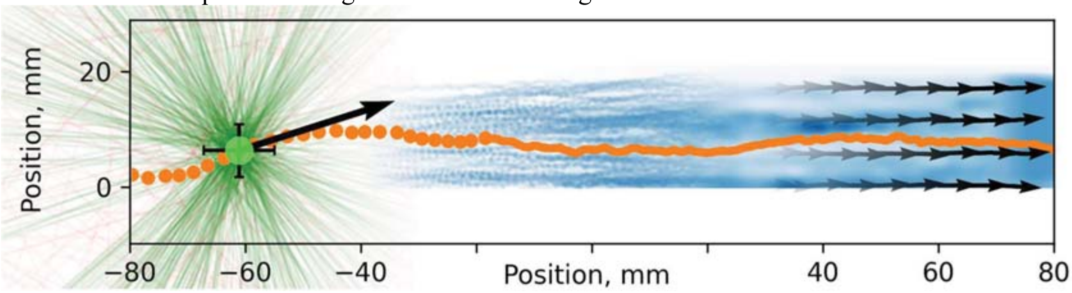

Figure 3. Measurements for a tracer moving in a laminar water channel of square cross-section. From left to right: A stylised single position measurement and associated velocity vector. A single pass through the channel is highlighted. The number of overlaid repeat passes increases until the full integral based description of local density (colour scale) and velocity field (vectors) is displayed.

6. Uncertainty, benchmarking, and validation towards metrology

The PEPT uncertainty budget is well understood, validated against known and established motions, and used to define the valid range(s) of applicability [3]. Three classes of motion can be considered, with equivalent, and increasingly sophisticated, analysis approaches. In the first, motion at constant velocity is used to determine uniformity of relative and absolute responses to position measurements, including stationary points and uniform linear motion produced using a robotic motion stage. In the second, motion at constant acceleration is used to determine the velocity response using uniform circular, or equivalent constant force, motion. Here, the laws of motion can be directly applied in validating the measurement against theoretical expectations. In the third, tracer particles moving within standard "textbook" flow conditions are investigated, with experimental data compared to those produced by theoretical modelling. Examples include unconstrained particles moving within convection cells or pipeline flows, with direct theoretical solutions enabling validation of all dynamic properties.

A simple validation process is outlined for uniformly accelerated motion, with tracer particles accelerating from rest under the influence of gravity. Upon release the particle accelerates with vertical displacement following the parabola $\frac{1}{2}gt^2$, with g the local gravitational acceleration and time t. Dividing the displacement by elapsed time resolves a linear relation proportional to the particle velocity, with respect to time the gradient of which measures the acceleration [11].

PEPT experiments were performed with a ^{68}Ga resin tracer inserted into a small steel ball placed within the field of view of the HR++ camera. An electromagnet supported the ball, which was allowed to drop by disconnecting the magnet current. The drop height was 40 cm, with the mass reaching a linear speed of ~1.5 ms^{-1} during the ~0.3 s flight time, before rebounding from the lower surface. Figure 4 shows the measured vertical trajectories (left) for repeated measurements alongside the analytical expectation. The same data, divided by elapsed time is shown (right), with the gradient of the parameterised line equal to $g/2$. A curve fitted to the linear section enables the gradient to be obtained (ignoring secondary effects such as air resistance), with g measured to be $9.798 \pm 0.001_{stat}$ ms^{-2}, correlating favourably with the accepted literature value of 9.796 ms^{-2} for our locale. This simple experiment, with a relatively high acceleration, is suitable to demonstrate that PEPT measurements are highly representative of the typical motions under consideration and can be extended to steady-state and transient flows without loss of accuracy [3].

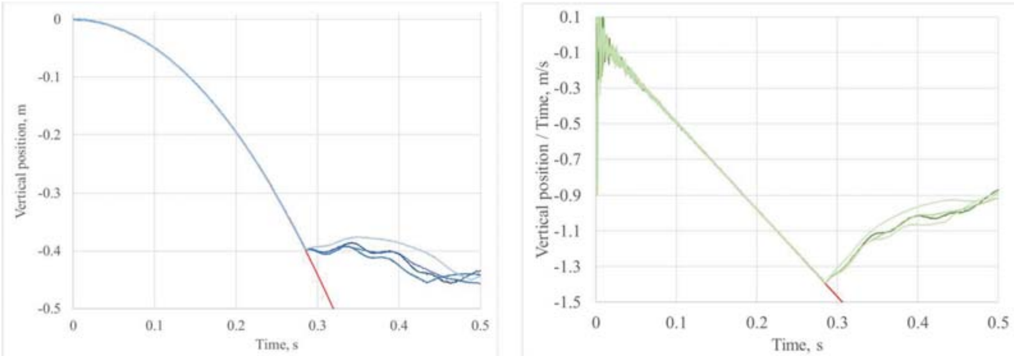

Figure 4. Analytical (red), measured vertical position (left, blue), and corresponding vertical velocity (right, green), measured for a small object falling from rest under gravitational acceleration.

7. Output and impact

Building on the validation of the measurement technique, a significant output from PEPT measurements has been in benchmarking direct measurement to theoretical models and simulations of flow processes. The Navier-Stokes equations describe a set of coupled partial derivatives used in modelling the motion of viscous substances with wide ranging applications. Due to their coupled nature integrable solutions do not exist, necessitating the use of precision measurement techniques such as PEPT to benchmark against, and to validate towards, the mathematical description. Measurement can be used to inform on dimensionless constants describing flow properties and/or provide numerical input data to theoretical models. PEPT has been extensively used in the study of mining and minerals applications, with experimental data informing underlying models of system behaviour, and used to optimise performance in terms of reducing waste streams, energy input, and raw materials requirements. From a design led approach, PEPT data have enabled greater understanding of material transport within flowing systems. In the case of flotation recovery cells, this approach has led to retrofitted adaptions of commercial units potentially saving millions of litres of water per year [12]. PEPT data are used to validate computational and/or theoretical models of turbulent flows and granular dynamics, perhaps offering the best experimental approach to non-invasively measuring flow in three dimensions.

References

[1] Jones T, Townsend D, 2017, J. Med. Imag. 4(1), 011013 (2017), doi: 10.1117/1.JMI.4.1.011013.
[2] Parker D, Broadbent C, Fowles P, Hawkesworth M and McNeil P 1993 Nucl. Instr. Meth. A326.
[3] Buffler A, Cole K, Leadbeater T W, van Heerden M, 2018, Int. J. Mod. Phys: Conference Series, 48, 1860113, 10.1142/S2010194518601138.
[4] Buffler A, Govender I, Cilliers J, Parker D, Franzidis J P, Mainza A, Newman R, Powell M and Van der Westhuizen A, 2009,Nuclear Research Applications and Utilization of Accelerators
[5] Camroodien A, van Heerden M, Nair S, Leadbeater T, 2021, Proc. of SAIP21, 65th Annual Conf. of the South African Institute of Physics (July 22-30 Potchefstroom).
[6] Leadbeater T W, Buffler A, Cole K, van Heerden M, Reich J, Camroodien A, Steyn D, 2023 Nuclear Science and Engineering, 10.1080/00295639.2023.2171234.
[7] Koning A, *et al.*, 2019, Nuclear Data sheets, 155.
[8] van der Merwe R, Peterson S, Buffler A, van Heerden M, McKnight A, Leadbeater T, 2022 Proc. Of SAIP2022, 66th Annual Conf. of the South African Institute of Physics (July 1-8 Gqeberha)
[9] van der Merwe R, Peterson S, Buffler A, van Heerden M, Hyslop N, Hutton T, and Leadbeater T, 2022, Proc. Int. Conf. on Nuclear Physics (September 12-16 Cape Town).
[10] McKnight A, Leadbeater T, van der Merwe R, 2022 Proc. of SAIP2022, 66th Annual Conf. of the South African Institute of Physics (July 1-8 Gqeberha).
[11] Wick K, Ruddick K, 1999, American J. of Physics 67, 962-965.
[12] Cilliers J, Cole K, 2013, UK Intellectual Property Office 1319600.1 (*Patent*).