Density-modified tracers for positron emission particle tracking

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Abstract. Positron emission particle tracking (PEPT) is a powerful non-invasive technique used to study the underlying dynamics of granular and multiphase flowing systems. The essence of PEPT is to attach (chemically or otherwise) sufficient positron-emitting radioisotope to a particle chosen as representative of the bulk material under study, and hence used as a flow-following tracer. We report on standardised methods for creating density-modified tracer particles to expand the range of applications feasible to research with the PEPT technique.

1. Introduction

Positron emission particle tracking (PEPT) is a nuclear imaging technique capable of non-invasively studying opaque and highly attenuated multiphase systems. The technique was first developed [1] at the University of Birmingham by modifying positron emission tomography (PET) medical scanners. PEPT research requires two main components to study a multiphase system of interest: a tracer particle that emits positrons and matches the physical and chemical characteristics of the material flowing in the system, and a modified positron camera to detect and reconstruct the resulting gamma ray emissions. Positrons emitted by the tracer particle annihilate with surrounding electrons to produce pairs of back-to-back 511 keV photons [2]. These photon pairs are capable of penetrating through the system of interest and can be detected in time-coincidence by the positron camera. By extrapolating the virtual lines between individual detection sites, the location of the tracer particle position can be a determined within millimetre accuracy at millisecond intervals or better [2]. The position and time data describing the tracer particle trajectory allows for deriving velocity, occupancy, and acceleration of a radioactive tracer particle. Tracer production for PEPT is thus a critical component in producing accurate, high quality and representative data of the system under study. There are a wide series of applications suitable for PEPT investigations of granular and fluid flow in opaque systems, typically with the intention of system optimisation or model validation of fundamental studies. Many of the systems studied at PEPT Cape Town are related to engineering and mining applications [3,4]. We address the current state of density-modified tracers and their range of applications possible using the PEPT technique.

2. Tracers for positron emission particle tracking

The accuracy of the data obtained using the PEPT technique is directly related to the representation of the physical and chemical properties of the tracer particle to the material of the system materials that can be replicated. There are two primary methods of tracer production for PEPT (Table 1). The first method requires oxygen-bearing materials that are thermally stable and on the order of millimetre

scale or larger. The radioisotope ^{18}F is produced by direct activation through appropriate nuclear reaction channels creating a near perfect non-invasive representation of the bulk material [5]. The second method relies on ion exchange chromatography by displacing the existing counter-ions on a selected ion exchange material with a suitable positron emitting radionuclide from aqueous solution. The ion exchange materials most suited for PEPT are organic strong acid cationic and strong base anion exchange resins [6]. The structure of both resin types rely on an amorphous styrene polymer back-bone that is cross-linked with divinylbenzene that is then either bound with functional groups that serve as the fixed exchange sites for a target element. As the names imply, cationic resins have a negatively charged functional group to allow for positive counter-ions to attach to them in solution and allow for balanced ionic charges and similarly for anion resins having positive functional groups fixed onto the resin back-bone to allow for anionic exchange.

A bespoke ion exchange method used for tracer production at PEPT Cape Town uses ion exchange resins radiolabelled with ^{68}Ga as the positron-emitting core, which is further modified to produce an analogue of the material of interest. The method has the benefit of not requiring direct access to a particle accelerator by producing a ^{68}Ge ($t_{½}$= 270 days) parent radionuclide that continuously decays to the well-suited daughter radionuclide ^{68}Ga ($t_{½}$= 67 min). Stable gallium is bombarded with a 66 MeV proton beam supplied by the *k* = 200 cyclotron at iThemba LABS to produce ^{68}Ge [7]. The ^{68}Ge is separated and then chemically bound onto a SnO_2 column in dilute hydrochloric acid to create a $^{68}Ge/^{68}Ga$ generator, which is supplied by the Radionuclide Production Department at iThemba LABS. The ^{68}Ga is eluted from a $^{68}Ge/^{68}Ga$ generator with dilute hydrochloric acid while retaining the ^{68}Ge which continuously decays into ^{68}Ga and thus replenishing the required ^{68}Ga multiple times day. The eluted ^{68}Ga is first added to concentrated hydrochloric acid to increase the concentration of the ^{68}Ga solution to at least 7 M. The solution is then passed through a ±1 mL Amberchrom CG-71m adsorption resin and further rinsed with concentrated hydrochloric acid to remove any contaminants from the column before eluting the purified ^{68}Ga with de-ionised water. The ^{68}Ga is then evaporated to dryness to remove the excess hydrochloric acid before being re-dissolved in 100 – 200 µL de-ionised water. The solution is added into a small glass vial that has at least three measured NRW-100 cation exchange resins beads and mechanically shaken for 15-30 minutes [8]. The size of the resin beads and initial activity of ^{68}Ga in the vial will determine the shaking time duration is adjust for each resin tracer core production. Analogue tracers are representative of a range of industrially relevant materials in size and densities. The current range of size and densities produced at PEPT Cape Town are produced over a density range between 1.00 and 2.85 g/cm^3 with final particle diameters from 450 to 900 microns (Table 1). This range of density is well suited for many industrially relevant materials including silica, iron ore, quartz and ceramics.

3. Results

The ability to directly activate materials routinely for PEPT is limited at PEPT Cape Town by the access to the particle accelerator used for bombardment of the oxide bearing minerals. Particles are often required in size ranges less than the 1 mm minimum diameter for direct activation and raise the need for producing tracer analogues with sufficient activity for tracking. The ^{68}Ga is radiolabelled onto strong acid cation exchange resin beads to form the core of the tracer material and act as the positron supply required for tracking its position. To accurately track the position of the tracer, >10 ~100 Mbq of activity is needed to maintain a tracking frequency greater than 1 kHz and depends on the amount of attenuation within the system of interest [8]. In order to consistently radiolabel this activity range on a single resin bead, the diameter of the particles need to be greater than 200 µm. The physical and chemical modifications made to these cores are typically achieved using coating methods developed with epoxy resins and acrylic polymers. The particle physical properties are tailored towards the bulk material of interest by adding powdered metals to the inner layers to typically increase the overall tracer particle density to match that of the material. If the chemical properties of the bulk material are significant to the phenomena under study, an additional layer of material is coated onto the surface of the density modified tracer core with a fine powder (< 50 µm diameter). Further surface properties

including roughness, hydrophilicity or hydrophobicity and particle shape can be manipulated at this stage of the method.

At PEPT Cape Town, recent research has been conducted on the feasibility of appropriate reaction channels for producing ^{18}F from oxide bearing materials using a 100 MeV proton beam at iThemba LABS, with substantial success using glass beads to confirm that direct activation is possible for oxide bearing materials larger than 1 mm in diameter. For smaller particles, ion exchange resins are typically produced in size range from 50 µm to 1.2 mm and have shown to be viable for radiolabelling with either ^{18}F or ^{68}Ga. Ion exchange resins are available in a narrow density ranging from 1.0 to 1.2 g/cm^3, making them good candidates for further modification to increase the density range to those of the minerals and metals of interest. The modifications are done by developing various coatings for the resin cores and although having a higher minimum particle diameter of 500 µm, it is possible for further optimisation to produce particles as small as 400 µm in diameter. These particles can be produced in density range starting at the density of the selected resin core (1-1.2 g/cm^3) and up to 2.9 g/cm^3. It is possible to make coated particles of a higher density but a reliable method to accurately measure the density of individual particles is still under development. For bulk materials that are resilient to drilling and larger than 3 mm in diameter, any size or density particle can be made into a PEPT tracer.

Table 1. Tracer particles produced at PEPT Cape Town and their properties.

	Size Ø [mm]	**Properties**	**Density [g/cm^3]**	**Applications**
Direct activation	1 -10	Physically identical, oxide bearing and resistance to thermal stress dependent	Varies	Non-invasive β+ activation of particles of interest for modelling studies, milling
Ion exchange resins	0.05-1.2	Naturally hydrophilic, most efficient radiolabelling per particle volume	1.0-1.2	β+ emitting source for coating and insertion for drilled tracers
Coated tracers	>0.5	Hydrophilic, Hydrophobic, range of densities, shapes, and surface chemistry	1.0-2.9	Analogue of particles of interest for flotation, hydrocyclones
"Drill and fill" tracers	>3.0	Dependent on material, near identical properties to material of interest	Varies	Invasive insertion of exact particles typical use in modelling studies, milling

As an example, a fundamental study into the interactions between bubbles and particles entrained in a three-phase particle laden fluid flow was produced [10]. The particles of the system used were polymerized methyl methacrylate (PMMA), which were chosen for their high fluorescence enabling optical methods to be used to investigate the flow streamlines. These particles were used as tracers in optical particle image velocity (PIV) studies – which is an alternative tracer-based method of visualizing the flow. The range of interest in the study exceeded the capability of the as PIV fails to accurately track particles because of the flow becoming optically distorted and so a positron-emitting particle analogue of PMMA was needed to investigate the more aggressive and industrially relevant conditions. The resin core was produced by radiolabelling ^{68}Ga onto NRW-100 cation exchange resin core and left to dry briefly to improve adhesion of the epoxy. A two-part epoxy was mixed along with powdered nickel to increase the density of the resin from 1.08 to approximately 1.5 g/cm^3 as required. As this study required matching the chemical properties of the resin with the PMMA bulk material, a fine layer of PMMA powder (1-20 µm Ø) was added to the surface. The final tracer size required was 300-400 µm in diameter and an average tracer activity of 31.5 ± 7.5 MBq per particle was achieved to

allow for at least 2 hours of high precision tracking. The final coating thickness was required to be no more than 100 μm thick to sufficiently match the bulk PMMA material size range of 200 – 400 μm in diameter. The final density of the tracer analogue of 1.5 ± 0.2 g/cm^3 was similar to the desired ~1.2 g/cm^3 of the bulk PMMA.

If the bulk material under study is around 3 mm in size or greater and can be bored into, a different application of the ion exchange resin beads becomes viable for tracer production. The most commonly used material for this tracer type are glass spheres modified by drilling a small hole (300-500 μm diameter) into the centre and inserting a radiolabelled resin core. The cavity is then back-filled with an epoxy resin loaded with a similar density material to maintain the original particle density. The technique is also well suited for metals and metal oxides such as iron ore and ceramics and produce tracer analogues that are nearly identical to the bulk material following the same procedure. When controlling the shape of the particle analogue is required, the coating can be molded by means of a silicon mold of the negative form of a desired shape and combining this with density and surface property manipulation to match the properties of the bulk material of interest.

4. Conclusion

This paper describes several methods for modifying tracer particles developed at PEPT Cape Town. They all require slight variations in the method to deliver a suitable analogue of the bulk materials under study, often requiring balance between the final desired physical and chemical properties, the activity of positron emitter attached to the tracer and the time taken to complete the production. By extending the limit of these methods, the density range between 1.00 and 2.85 g/cm^3 with final particle diameters from 450 to 900 microns range expand the number of applications feasible to research with the PEPT technique.

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