**Target for the in-beam activation of tracer particles for positron emission particle tracking**

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**Abstract.** Positron emission particle tracking measures the trajectory of a single radioactively labelled tracer particle by coincident detection of emitted annihilation photons. The technique enables the non-invasive study of dense opaque flows, with the tracer acting as a small neutrally buoyant flow-follower. The University of Cape Town has established a PEPT facility at iThemba LABS, utilising tracer particles produced through radiochemical methods, and measured using adapted positron tomographs. An activation approach producing the positron emitter $^{18}$F inside glass target spheres of diameter between 5.0 and 10 mm using accelerated beams of alpha-particles has been explored. The reaction $^{16}$O$(\alpha,x)^{19}$F is used, exploiting the high concentration of natural oxygen and the correspondingly high cross-sections for $^{19}$F formation. A standard target holder for the batch production of radionuclides at iThemba LABS was modified, reducing the entrance window thickness, allowing ingress of circulating cooling water, and adapted for a primary $\alpha$-particle beam of 100 MeV energy delivered by the separated sector cyclotron (SSC) of iThemba LABS. Two-hour bombardment at nominal beam current 0.8 $\mu$A produced activities up to ~110 MBq (3 mCi), with over 95% of the activity being $^{18}$F.

1 Context

Positron emission particle tracking (PEPT) is a technique for measuring the trajectory of a single radioactively labelled tracer particle moving within an apparatus placed inside a PET scanner or positron camera [1]. For typical industrial applications, the tracer particle is usually a glass or ceramic bead, spherical in shape with a diameter between 5.0 and 10 mm, labelled with one of the medically significant positron emitters such as $^{18}$F or $^{68}$Ga. The coincident detection of 511 keV annihilation photons emitted by the tracer particle enables the non-invasive study of dense opaque flows and rapidly changing dynamic systems, including conditions of high turbulence [2]. The tracer particle acts as a small, neutrally buoyant, flow-follower in a viscous fluid, suitable to study fluid or granular behaviour in packed beds, tumbling mills and/or other flow systems [3-5]. The PEPT technique is particularly useful in studying well-mixed systems where tracking a single particle over an extended duration provides essentially the same information as simultaneously observing the instantaneous motion of all the particles in the fluid – formally referred to as ergodic systems [6]. Analysis of the single-particle trajectory over long timescales (minutes to hours) can provide time-averaged maps of velocity fields, the shape of dense flow regions, identification of regions of low-velocity flow, observation of gravity-dominated flows, and etc. [6].

The Department of Physics at the University of Cape Town (UCT) established a PEPT facility at iThemba LABS in 2009, based around a Siemens ECAT HR++ PET scanner modified for PEPT acquisition and analysis [7]. Alternative positron camera systems have since been added, including large area planar detectors (an ADAC Vertex dual-headed gamma-camera) and modular positron camera systems with customised detection geometries designed for specific applications, based on the block detector design [8]. Positron cameras formed from modified positron emission tomography (PET) scanners are used to detect and reconstruct the annihilation photon flight path, by detecting photon pairs in time coincidence. Genuinely reconstructed paths (which have not been produced by random coincidence, or corrupted by attenuation and scattering) converge upon the instantaneous position of the tracer particle. An iterative least squares minimisation approach is used to determine the 3D position where the photon paths converge, and hence the most likely instantaneous position of the tracer particle which can typically be determined at sub-millimetre precision at kilohertz rates. Tracking of particles moving with velocities up to 10 m/s is therefore achievable [7].

The tracer activity required for optimum tracking depends upon the amount of attenuating and scattering materials, and on the flow conditions, which vary considerably between applications. Typical tracer particles are required to range in activity from around 10 to 150 MBq (250 $\mu$Ci to 3 mCi), be predominantly positron emitters, with ideally a short positron range. The principal tracer isotope used at PEPT Cape Town has been $^{68}$Ga (half-life 68 minutes, 88.91% $\beta^+$)

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extracted chemically from the standard medical \(^{68}\text{Ge}/^{68}\text{Ga}\) radioisotope generators produced by iThemba LABS. The \(^{68}\text{Ga}\) is regularly eluted from the \(\text{SnO}_2\) generator column, purified and concentrated, and used to radiochemically label suitable particles to be used as tracers within the study application. If a particle from the bulk material under study cannot be labelled chemically, an analogue particle is produced by attaching sufficient \(^{68}\text{Ga}\) activity onto a resin substrate which is further modified to match the physical properties of the material in question. The representation of the tracer particle to the bulk material under study is critical, where systematic differences in physical properties can alter the dynamic behaviour and hence interpretation of PEPT results. The physical properties that are carefully controlled depend on the application and associated research questions, and include the physical (size, shape, surface roughness, density, etc.) and chemical (surface potential, wettability, hydrophobicity / hydrophilicity, etc.) attributes. When studying liquids, the radioactivity is sealed onto the substrate, and the overall particle density modified to match that of the fluid and achieve neutral buoyancy [9].

In the context of the accelerated beams and infrastructure available at iThemba LABS, an experimental campaign was initiated to activate silicon dioxide (\(\text{SiO}_2\)) based glass beads using high energy alpha particle alpha beams. The method utilises the set of \(^{16}\text{O}(\alpha, x)^{18}\text{F}\) reactions, exploiting the high concentration of natural oxygen in the glass as well as the relatively high cross-sections for the formation of the positron emitter \(^{18}\text{F}\). The competing \((\alpha, x)\) reactions on natural silicon, oxygen, and other contaminant species within the target matrix, have been shown to yield no long-lived radioisotopes instead producing short-lived (maximum half-life 2.5 minutes) or stable activation products. The activation is expected to yield \(^{18}\text{F}\) of relatively high radioisotopic purity. Advantageously, \(^{18}\text{F}\) has a longer half-life (110 minutes), higher positron branching ratio (96.73\%) without any associated gamma emissions, and shorter positron range compared to \(^{68}\text{Ga}\) implying longer experimental timescales at higher spatial resolution.

2 The \(\text{SiO}_2(\alpha, x)\) reactions

The alpha particle induced reactions occurring with both naturally occurring silicon (92.22\% \(^{28}\text{Si}\), 4.69\% \(^{29}\text{Si}\), and 3.09\% \(^{30}\text{Si}\)) and naturally occurring oxygen (99.76\% \(^{16}\text{O}\), 0.04\% \(^{17}\text{O}\), and 0.21\% \(^{18}\text{O}\)) result in elastic and inelastic scattering and predominantly short-lived or stable activation products. The only significant long lived activation product is \(^{18}\text{F}\) produced by competing alpha particle induced reactions on \(^{16}\text{O}\) within the target [9]. The reactions, in order of significance given by relative total, cross-section are direct production by \(^{16}\text{O}(\alpha, np)^{19}\text{F}\), \(^{16}\text{O}(\alpha, d)^{18}\text{F}\), and indirect production by \(^{16}\text{O}(\alpha, 2n)^{18}\text{Ne} \rightarrow ^{18}\text{F}\) which proceeds by \(\beta^-\) decay of the \(^{18}\text{Ne}\). The cumulative excitation function of the \(^{16}\text{O}(\alpha, x)^{18}\text{F}\) reaction peaks at about 35 MeV, with a threshold at 18 MeV beam energy. Figure 1 shows the microscopic cross-sections for the three significant \(^{18}\text{F}\) producing reactions, their sum, and the total cross-section for alpha particles of energy in the range up to 100 MeV, taken from TALYS calculations given in the TENDL database [10].

3 Target systems

Glass spheres (molar composition roughly 61\% \(\text{O}\), 23\% \(\text{Si}\), 10\% \(\text{Na}\), 3\% \(\text{Ca}\), 2\% \(\text{Al}\), 2\% \(\text{B}\), 1\% \(\text{Mg}\)) of diameters 5.0 to 10 mm were used as targets, detailed composition can be found in references [8] and [9]. A target holder and capsule system were designed to deliver \(\alpha\)-particles accelerated by the iThemba LABS separated sector cyclotron (SSC) to the targets and to maintain integrity by providing circulating cooling water. A standard target holder used for the batch production of radionuclides at iThemba LABS using the horizontal-beam target station 1 (“Elephant”) [11] was modified for this purpose. The entrance window thickness of the target holder and the wall thickness of the target capsule were reduced. The target capsule was placed in the “high energy slot” inside the holder. Normally, in the case of radionuclide production, the capsule serves to protect the target material from direct contact with the cooling water. When activating glass beads, direct contact of the target material with the cooling water is essential to reduce the effects of beam heating. Deformation by swelling and/or melting potentially affect the representation and non-invasiveness of the tracer particle when performing PEPT. The target capsule was therefore modified by drilling a matrix of small holes through the front and side walls through which cooling water can freely flow. A further advantage of this approach is that the coolant (10 bar, 30 l/min flow rate) serves to agitate the target spheres in situ, where rotation allows beam entrance from different directions relative to the sphere centre, effectively isotropically irradiating the targets. Depending on the beam energy and target attenuation this approach enables the entire surface of the target sphere to be irradiated, rather than just the forward-facing surface as when using a fixed target.

Due to dead layers upstream of the target (including entrance windows, cooling layers, etc.) a significantly higher beam energy is required to reach the glass surfaces with the desired energy. The target holder and capsules were designed for a primary \(\alpha\)-particle beam
energy of 100 MeV. The beam passes two 25 µm thick Havar (ICRU-470 / UNS R30004) foils separated by a 10 mm helium cooling layer (maintained at 1.2 bar), a further 4.0 mm air gap, and aluminium entrance window of 0.5 mm to enter the target station, as shown in figure 2. For a 100 MeV α-particle beam extracted from the SSC, the beam energy impinging on the target surface is around 80 – 70 MeV determined by Monte Carlo calculation. The typical linear range of these alpha particles within the target material is around 3 mm, with energy above the reaction threshold within the first 2 mm of penetration. For small diameter targets (< 4 mm), the radioisotope activation is expected to be volumetric, with larger targets expected to activate to a surface layer of uniform depth, provided they are rotated under the coolant flow.

Fig. 2. Target stack, including dead layers.

4 Method

Multiple target spheres are placed within a single target capsule as shown in figure 3 and activated with the beam defocussed and swept in a circular pattern to increase the beam-strike area. Practically, this approach leads to a redundancy in activated targets and reduces the heating load per sphere, but not all receive the same amount of accumulated charge. A distribution of radioactivity (and heating damage) is then expected, from which a suitable candidate can be selected for the PEPT measurement.

Fig. 3. Target capsule, perforated containment, and open capsule holding SiO₂ target spheres.

Several bombardments were performed with 100 MeV α-particles delivered by the separated sector cyclotron (SSC) of iThemba LABS. Helium gas was ionised in the ECR ion source and accelerated to 8 MeV for injection into the SSC, where the beam is further accelerated to 100 MeV. The extracted beam was directed to the target station through a set of dedicated beamlines and a k = 100 switching magnet, which limits the maximum beam energy delivered to the target in practice. Approximately two-hours bombardment at an average beam current of 0.8 µA was sufficient to deliver 2.0 µA of integrated charge, at which point the bombardment was terminated. A short cooling period (20 minutes – 1 hour) following end-of-beam allowed any short-lived radioisotopes to decay, and the target holder and capsule were recovered from the target station using robotic manipulation. The target spheres were unloaded from the capsule in a hot cell and transferred to the PEPT laboratory for activation product characterisation and activity measurements.

5 Results

Characterisation followed multiple simultaneous measurements using a wide range of detector techniques and extended over multiple half-lives of the produced species [9]. A calibrated high purity germanium (HPGe) spectrometer (energy resolution 1.2% at 511 keV) was used to determine the gamma radiation spectrum emitted by the targets. The most intense peak was observed at 510.5 ± 2.6 keV with associated sum peaks and Compton continua, corresponding to the positron annihilations expected from the decay of proton rich species. Based on the HPGe measurements, and assuming the yield of positron activity due to predominantly ¹⁸F, a Capintec CRC25-R 4π ionization chamber calibrated to ¹⁸F was used to measure the absolute activity of each sample over many hours, allowing the decay curve to be reconstructed and projected back to the time at end-of-beam. Ionisation chamber results from four independent bombardments, and 24 samples total, are shown in figures 4 and 5.

Fig. 4. Measured frequency distribution histogram of target activities, the point shows the mean for all samples.

Three target spheres had no appreciable activity above background, implying the beam did not impinge upon them with sufficient energy for activation. Three
of the samples had measured activities above (75 MBq) (2.0 mCi), with the maximum activity around 110 MBq (3 mCi). The mean activity was 30 MBq (850 µCi) measured for all 24 samples. Time series gamma-ray spectroscopy using a calibrated sodium iodide (NaI) spectrometer enabled half-life measurements for each of the primary photopeaks, adding a high degree of confidence in their likely origin. Singles counting windowed around the 511 keV photopeak (350 – 850 keV) using 34 independent detector modules of the HR++ positron camera, and simultaneous prompt and delayed time coincidence measurements (12 ns coincidence gate) were used to calculate the absolute activity for each of the samples, confirming the ionisation chamber measurements [9]. The consolidated activity data for all samples are shown in figure 5 as a function of the squared radius of the target spheres, where the linear trend shows that expected from activation of the surface layer implying that the targets did undergo some rotation within the beam.

![Graph showing activity vs. squared radius for target size.](image)

**Fig. 5.** Mean activities per target size.

Low intensity photopeaks measured with the HPGe spectrometer at energies 372.3 ± 1.1 keV and 1367.1 ± 1.7 keV were observed. By combining these measurements with the time series decay data per peak, these were attributed to activation of target composition admixtures. The positron emitter $^{24}$Sc (511 keV (annihilation), 176.2% and 372.9 keV, 22.5%) was produced by alpha particle interactions with the 3% calcium admixture. The neutron rich $^{24}$Na (14.98 hour half-life, principle gamma transitions at 1368.6 keV 99.99% and 2754.0 keV 99.85%) was produced by thermal and fast neutron interactions with Na, Mg, and Al, within the glass targets, present in ~1% amounts. For PEPT, the additional positron yield from $^{41}$Sc is beneficial, extending the possible experimental timescale. $^{24}$Na is not a positron emitter; however, annihilation photons are observed from this isotope due to the high energy photon emissions undergoing pair production. These are a likely source of background in PEPT, although the intensity is insignificant compared to the yield from positron emitting species. If these activation channels were to be used to produce $^{18}$F for alternative applications (e.g. medicine), the observed contaminants can be controlled by modifying the purity of the target. Table 1 combines all measurement analyses and presents the yield of observed activation products projected back to the EOB.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Decay mode</th>
<th>Half-life, hours</th>
<th>Relative activity, %</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{18}$F</td>
<td>β⁻ (96.73%)</td>
<td>1.8</td>
<td>$\sim$ 95%</td>
</tr>
<tr>
<td>$^{41}$Sc</td>
<td>β⁻ (88.1%)</td>
<td>3.9</td>
<td>&lt; 5%</td>
</tr>
<tr>
<td>$^{24}$Na</td>
<td>β⁻ (99.99%)</td>
<td>14.9</td>
<td>&lt; 5%</td>
</tr>
</tbody>
</table>

### 6 Concluding remarks

The $^{16}$O(a, x)$^{18}$F reactions using 100 MeV alpha particle beams at nominal intensity 0.8 $\mu$A have been investigated in producing $^{18}$F within 5.0 – 10 mm diameter glass targets of naturally occurring isotopic composition. The observed purity of $^{18}$F was consistently 95% in all experiments, with absolute activity ranging up to 110 MBq (3 mCi). Future work will aim to optimise the beam energy to maximise activation yield and control effects of beam heating, and will explore alternative target materials and reactions. These results offer potential new production mechanisms for $^{18}$F and complement the existing portfolio of tracer particles used in positron emission particle tracking experiments.

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### References