



An effective technique for the storage of short lived radioactive gaseous waste

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ABSTRACT

An effective technique is described to deal with volatile, short lived radioactive waste generated as a result of the routinely produced positron emission tomography (PET) radiopharmaceutical 2-deoxy-2-[¹⁸F]fluoro-D-glucose (FDG). All radioactive gases and aerosols created during the synthesis are collected and stored safely in commercially available TEDLAR gas sampling bags. Once these collected PET by-products decay, the TEDLAR gas bags can be easily emptied and reused. This improved technique is effective, safe, reliable and economical.

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1. Introduction

[¹⁸F] Fluorine (F-18) is one of the most popular short lived ($t_{1/2} = 109.8$ min) positron emitting radioisotopes (Wilson et al., 2008), which is usually cyclotronically generated in liquid form by means of a ¹⁸O(p,n)¹⁸F nuclear reaction. Its major application as a label for radioactive tracers used in combination with the non invasive, metabolic PET imaging technique is of high importance (Cai et al., 2008). In recent years, one particular F-18 labelled radiopharmaceutical, FDG, has even been elevated to the most successful and widely used clinical imaging PET tracer in oncology, neurology and cardiology (Hoh, 2007; Yu, 2006). Besides being well established, the production of the PET radioisotope as well as the labelled product are routinely accompanied with the generation of volatile radioactive gaseous and aerosolic by-products, e.g. [¹⁸F]HF, [¹³N]NH₃, whose retention, storage and disposal has been shown to be financially and environmentally challenging (Pascali et al., 1996; Calandrino et al., 2007). Although nowadays most commercially available hot cells and automated synthesis modules are conceptualised and facilitated with trapping devices, such as activated charcoal filters, chemical absorbents and liquid nitrogen traps (Calandrino et al., 2007; Hot Cell Manufacturer COMECER, personal communication, 2009), still, not all created radioactive gases and aerosols are effectively immobilised. Hence worldwide many PET Centres attempt to deposit radioactive waste using expensive, large gas compression storage systems that need additional shielding or the generated by-products are simply discharged directly into the atmosphere.

The disposal of radioactive waste gases in PET centres around Scotland is regulated by the Scottish Environment Protection Agency (SEPA) by means of the Radioactive Substances Act (1993). Medical cyclotron facilities such as the John Mallard Scottish PET Centre in Aberdeen have to comply strictly with daily and annual gaseous disposal limits as agreed and authorised by SEPA. However, prior to the disposal it is the authorisation holder's responsibility and obligation to minimise radioactive gaseous waste using the best practicable means. Therefore our permanent aim is not to discharge but to safely retain as much radioactive waste possible, which is created during any PET isotope and PET tracer production.

2. Materials and methods

2.1. Radiation Protection

In August 2010, SEPA authorised a daily disposal limit of 74 GBq as well as an annual disposal limit of 500 GBq for any radionuclide (¹⁸F, ¹¹C, ¹³N, ¹⁵O) generated as a product or by-product of cyclotron operation taken together.

All components of the radiation stack monitoring and data management system (CMS-PET) for continuous radioactive gas detection in the air extract system were purchased from and installed by Lab Impex Systems. The ventilation outlets of the hot cells were connected to the stack. The average stack flow rate was 2300 m³/h. The level of background radiation detected by the stack detector was on an average 76 kBq/m³. TEDLAR gas sampling bags were purchased from GRACE/ALLTECH Associates. TEDLAR gas sampling bags (volume: 10 l and 40 l) were connected

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via PARFLEX or PTFE tubing and SWAGELOK fittings. On an average five FDG syntheses could be carried out before they had to be emptied. The contents of all TEDLAR bags were measured and checked using a contamination monitor (Mini-Monitor—Series 900 Thermo Fisher) and only then were deflated every Monday morning before FDG production. To avoid any malfunction, as part of our FDG preventive maintenance plan, the gas bags were replaced every 3 months.

2.2. [^{18}F]Fluoride production

[^{18}F]Fluoride was prepared by proton bombardment of 97% enriched [^{18}O]H $_2$ O (Isotrade GmbH, Germany) by the $^{18}\text{O}(p,n)^{18}\text{F}$ nuclear reaction. The silver target (1.1 ml) was pressurised to 600 psi and irradiated with 11 MeV protons produced by the CTI/SIEMENS RDS-111 cyclotron at the John Mallard Scottish PET Centre in Aberdeen. Irradiations of 50 min with a beam current of 27 μA were typically used. At the end of bombardment (EOB) the target was unloaded within 5 min using argon gas. The 10 l Tedlar gas bag was filled up to 20%. On average a maximum surface dose rate of 5000 $\mu\text{Sv/h}$ was measured after the end of the unloading process. Unloading the target without the use of this 10 l gas bag would discharge on average 1.6 GBq.

2.3. FDG production

The John Mallard Scottish PET Centre is currently producing FDG on average once a day, three times a week. On average, 34 GBq of [^{18}F]fluoride were transferred with argon gas to the remote controlled FDG synthesis module (GE Healthcare TRACERlab Fx_{FDG} synthesiser) that was contained in a lead shielded mini hot cell (Von Gahlen). The FDG synthesis was carried out according to Hamacher et al. (1986) via nucleophilic substitution followed by acid hydrolysis. Chromafix, Chromabond purification cartridges and pharmaceutical grade FDG production reagent kits (K-320 TF) were purchased from ABX GmbH, Germany. FDG production took ca. 35 min and yielded on average 17 GBq of FDG at end of synthesis (EOS). The 40 l Tedlar gas bag was filled up to 20%. On average a maximum surface dose rate of 1000 $\mu\text{Sv/h}$

was measured at EOS. Without the use of this 40 l gas bag a radioactive discharge of on average 0.5 GBq would be detected during every FDG synthesis.

3. Results

For the [^{18}F]fluoride production the cyclotron target is loaded with 97% enriched [^{18}O]H $_2$ O, pressurised and then bombarded. Although mainly [^{18}F]fluoride is being formed during this process also ^{13}N is created (Guillaume et al., 1991) via the $^{16}\text{O}(p,\alpha)^{13}\text{N}$ reaction. After EOB the target unload process starts. For this the initial target pressure is vented and subsequently the aqueous [^{18}F]HF bolus is transferred via argon gas to the target collect vial of the FDG synthesis module (Fig. 1). It is during this high pressure unloading process where radioactive waste gases and aerosols, mainly in the form of [^{13}N]NH $_3$ and [^{18}F]HF, would be discharged via the vent of the target delivery vial into the hot cell. Therefore many PET Centres simply discharge the waste through the ventilation system into the atmosphere or are equipped with devices such as gas compression systems to collect, transfer and store the waste in additional lead shielded tanks. However these devices are not only very expensive but also consume a lot of space. We on the other hand use a practical, effective, reliable and economical alternative. We connected a small, 10 l TEDLAR gas sampling bag (Fig. 2) to the vent of the target delivery vial and positioned it on top of the synthesis module. Consequently, during the unload process of the target, the radiation monitoring system did not register any elevated radiation levels in the stack (Fig. 3). Hence any potential radioactive discharge is safely deposited in the bag inside the lead shielded hot cell.

After the target is unloaded the FDG production programme can be started. As a matter of fact the FDG synthesis contains two reaction steps during which volatile radioactive substances are created. Both steps, the azeotropic distillation of the water/acetonitrile solution and the nucleophilic substitution generate mainly [^{18}F]CH $_3\text{F}$ and [^{18}F]fluoride as suggested by Calandrino et al. (2007). During both steps, volatile radioactive contaminants are supposed to be trapped by a liquid nitrogen trap that is usually installed between the vacuum pump and the reaction vessel (Fig. 1).

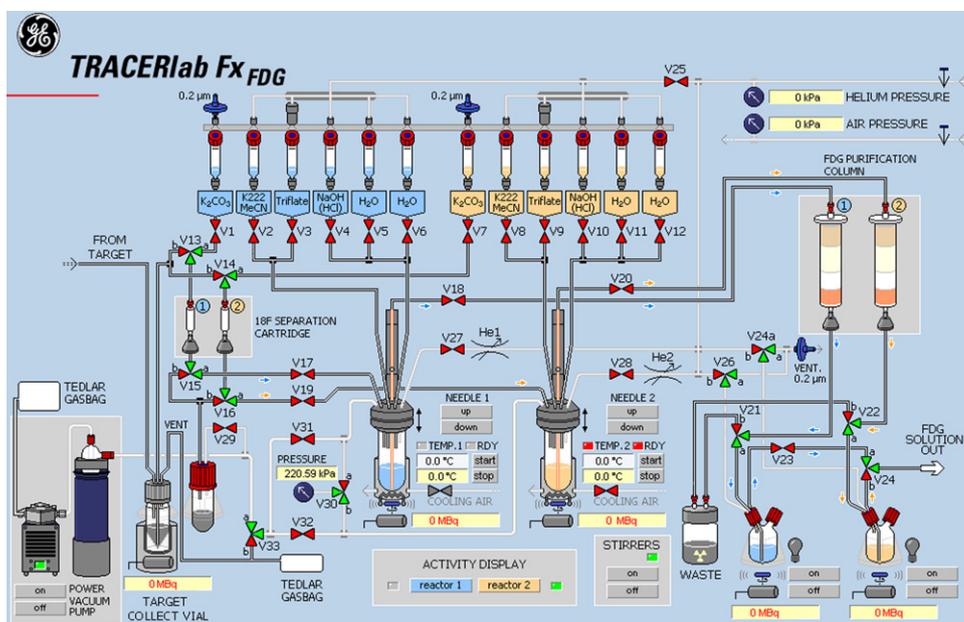


Fig. 1. Graphical representation of the GE Fx_{FDG} synthesis module. The 10 l TEDLAR gas sampling bag is connected to the vent of the target collect vial with Paraflex tubing while the 40 l TEDLAR gas bag is connected to the exhaust of the vacuum pump via PTFE tubing.

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