THE SMART EFFLUENT MONITORING SYSTEM AT THE AUSTRALIAN NATIONAL MEDICAL CYCLOTRON

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The continuous monitoring and quantitative assessment of radioactive effluent release through the exhaust stack of a medical cyclotron facility are mandatory duties imposed by the relevant regulatory authority. At the Australian National Medical Cyclotron large activities of short lived positron emitting gaseous PET-Radiopharmaceuticals like $^{11}CO_2$, $^{13}NH_3$, and ^{18}FDG in aqueous form as well as ^{123}I in the form of $Na^{123}I$ are routinely produced. For the selective detection of the positron emitting gases and vapours containing ^{123}I, a stand alone stack monitoring system has been implemented. Two types of sensitive, fast response radiation detectors for the selective detection of ^{18}FDG and ^{123}I have been developed at our laboratory. This real time stack monitoring system is capable of recording the stack discharges at interval of 1 minute and to actuating acoustic and visual alarms, if the permissible levels of radioactivity release are exceeded.

1. Introduction

The Australian National Medical Cyclotron (NMC) produces large activities of short lived positron-emitting radioisotopes particularly, $^{11}CO_2$, $^{13}NH_3$, and ^{18}FDG (Fluoro-deoxyglucose) in aqueous form for the National PET Centre in Royal Prince Alfred Hospital, situated 320 meters north east of the cyclotron complex. During chemical processing of the PET radiopharmaceuticals in the automatic chemistry cell, a small fraction (usually < 1% of the final yield) of the volatile radioactive product passes through the ventilation system. However, in the event of system failure a much higher effluent release in the order of several GBq may occur[1]. Similarly, during radiochemical processing and dispensing a small fraction of ^{123}I is released into the atmosphere.

It is obligatory for the NMC Health Physics Section to provide the state Environmental Protection Authority (EPA) with the instantaneous activity concentrations (Bqm^{-3}) of the stack release for every single PET or ^{123}Iodine production run. As explained in 3.1, we have found conventional stack effluent monitors unsuitable to serve this purpose. Innovative fast response positron and iodine detectors have therefore been developed for real time detection of short, instantaneous bursts of stack discharge. In this paper, some important features of the smart effluent monitoring facility of the NMC is presented (Figure 1).

2. Principle of Operation

2.1 Hot Cell Ventilation

After the proton bombardment of the PET target (PT) in the cyclotron vault, the resulting PET radioisotopes are transferred through a teflon capillary tube to an auto-chemistry cell (AC) for further radiochemical processing. Following completion of the processing the PET product (collected in a sterile bottle) is carried to the dispensing cell (DS), sealed in sterile vials, capped and delivered to the National PET Centre via a pneumatic rabbit-transfer system (RS). At present, about 5 batches ^{18}FDG (30 GBq per batch) and 3 batches of $^{13}NH_3$ (20 GBq per batch) are produced per week. A ventilation duct (VD) connects the automatic chemistry and dispensing cells and the rabbit transport station to the main stack via the HEPA (High Efficiency Particulate Air) filter banks (HF1 and HF2) and exhaust fans (EF1 and EF2). The residual radioactivity trapped in the HEPA filter is continuously monitored by the gamma monitor (GMH) connected to the Health Physics Data Acquisition Syatem[2].

The ^{123}I is produced by bombardment of highly enriched (99.99%) ^{124}Xe gas-target (IT) with 31 MeV protons in the beam room. The enriched ^{124}Xe at 1 atm. is initially pumped from the storage tank (ST) into the target (IT). The intensity of the energetic proton beam is optimised on the Faraday Cup (FC) and steered into the gas target by the switching magnet (SM) and the magnetic quadrupole lens (QPL). The reaction products are processed in a computer-controlled ^{123}I production cell (IPC). The radioactive product transfer from target to processing cell is monitored by the gamma monitor (GMT) connected to the Health Physics Data Acquisition System[2]. At present about 2 batches of ^{123}I (120 GBq per batch) are produced per week. The containment of the iodine processing cell and the storage tank is connected to the main stack thorough the ventilation duct (VD) via the HEPA (High Efficiency Particulate Air) (HF0) and SIAM (Specific Iodine Absorption Module) (SM1and SM2) filter banks and exhaust fans (EF3 and EF4). The schematic diagram of the ventilation system is shown in Figure 1.

2.2 Stack Monitoring

The exhaust air is extracted from the central region of the main stack at the rate of 0.06 m^3min^{-1} by the suction pump (SP) connected to a flow meter (FM), a flow control valve (FCV) and a critical orifice to ensure a constant flow. The effluent sample is passed through the smart positron (PD) and ^{123}I detectors (ID) linked in cascade, and finally returned to the stack. The radiation detectors are individually connected to two Single Channel Analysers (SCA1 and SCA2).The analog output (chart recorder) terminals of the SCAs are connected to

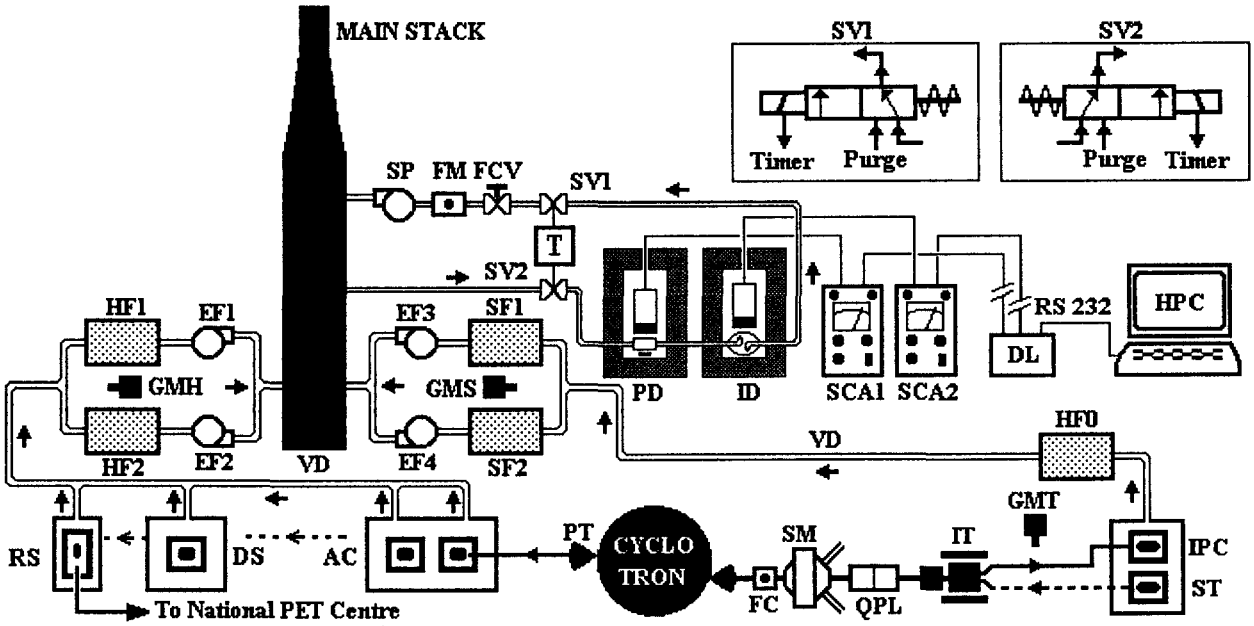

Figure 1: Schematic diagram of the NMC Stack monitoring facility showing the cyclotron targets, hot cells and filter ventilation system, flow control solenoid valves and the real time radiation monitors with the data acquisition system. The diagram is explained in the text.

a locally manufactured industrial standard datalogger (DL), interfaced to the central health physics computer (HPC) through a RS232 serial port[2]. The stack exhaust is sampled every one minute and the relevant data is stored in the hard disc of a personal computer for statistical analysis. The NMC stack monitoring system is capable of actuating visual and audible alarms, when the activity concentrations of the released gases exceed the recommended value, thus allowing the responsible person to take prompt actions to prevent the stack release.

3. Materials and Method

3.1 The Smart Effluent Detectors

Conventional stack detectors usually assay the radioactivity of effluent releases trapped in an active-charcoal cartridge therefore, are incapable to response to any sudden burst of radioactivity release. A gradual degradation of the detection efficiency due to poisoning of the charcoal cartridge during its operating life also causes a serious problem.

The effluent detectors developed at the NMC have excluded the use of such charcoal filters. The principle of the positron detector (PD) is as follows: A small spiral made of thin teflon tube (1mm internal diameter, net volume: 3ml) has been pressed between two 5cm diameter 1.5mm thick aluminium discs. The spiral is connected to the effluent suction line as shown in Figure 1. The positrons are partially attenuated in the walls of the teflon tube annihilate in the aluminium discs, hence producing 511 keV photons. The 1.5 mm thick aluminium disc is adequate to stop all positrons[3] emitted by the commonly used PET isotopes produced at the Australian National Medical Cyclotron.

The ^{123}I detector (ID) consists of a 250 ml glass flask housing two oppositely facing nozzles creating a vigorously turbulent air flow in the flask and thereby producing a homogeneous distribution of the effluent in the flask volume. The ^{123}I detector is connected in cascade with the positron detector (PD) and the flow control valve (FCV). Two 5cm diameter NaI(Tl) scintillators (Model: 44-12, Manufacturer: Ludlum Measurements, Inc, Texas USA) are placed at immediate surface contact with the detectors and connected to two single channel analysers (SCA1 and SCA2) (Model: 2200, Manufacturer: Ludlum Measurements, Inc, Texas USA). The energy discrimination windows of the SCAs are set for 511 ± 25 keV (for positron detector PD) and 160 ± 25 keV (for iodine detector ID). Both stack detectors are placed in two separate lead castles of 10 cm wall thickness as shown in Figure 1.

3.2 The Calibration Procedure

Prior to installation, the stack detectors (PD and ID) were calibrated with known activities of ^{18}FDG and ^{123}I. About 100kBq of ^{18}FDG in aqueous form was received from the Quality Control laboratory of the NMC. The sample was diluted and carefully injected into the teflon spiral of a net internal volume of 3ml and thoroughly sealed. The activity of ^{18}FDG in the chamber was determined within a ± 10% accuracy using a Radionuclide Calibrator (Model: CRC 712M, Manufacturer: Capintec Incorporated, Pittsburgh, USA) and placed in the lead castle. The 5cm diameter NaI (Tl) scintillator was placed on the detector and the analog voltage output

from the SCA1 recorded by the datalogger[4] for 24 hours (ie. 13 half lives of ^{18}F ie. 110 min). The activity concentration [MBq.m^{-3}] of ^{18}FDG in the teflon spiral was calculated from the initially injected activity and is shown with the corresponding output voltages from SCA1 in Figure 2a.

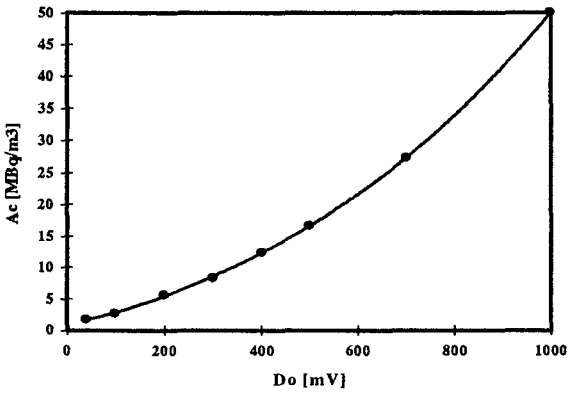

Figure 2a: Calibration curve of the positron detector showing the output voltage Do from the SCA1 with the corresponding activity concentration Ac in the detector volume (3ml). The data points are fitted with the 4th order polynomial: $y = 1.16\times10^{-11}x^4 - 1.33\times10^{-8}x^3 + 3.49\times10^{-5}x^2 + 1.59\times10^{-2}x + 9.19\times10^{-1}$.

Similarly, 5kBq of ^{123}I in the form of aqueous solution of $Na^{123}I$ was received from the QC laboratory and a piece of sponge been soaked with the solution. A 250ml glass flask (same volume as the detector chamber ID) was filled with the sponge, thoroughly sealed to prevent loss of activity due to evaporation. Evidently, this configuration (ie. a uniform distribution of 158 keV photon emitting radioactive source throughout the flask volume) embodied a fairly perfect emulation of the real stack monitoring situation. The flask was placed in the lead castle, the 5cm diameter NaI (Tl) scintillator is placed on the flask and the analog voltage output from the SCA2 has been recorded with the datalogger[4] for 7 days (ie. 12.5 half lives of ^{123}I, ie. 13.4 h). The activity concentrations [MBq.m^{-3}] of ^{123}I in the flask are calculated from the initially injected activity in the sponge and are shown with the corresponding output voltages from SCA2 in Figure 2b.

3.3 Effluent Release Assessment

The activity concentration a_t of the effluent in the detector volume at the time t is given as:

$$a_t = k(v_t) \quad (1)$$

where, k and v_t are the calibration polynomial and the output voltage at the instant t respectively.

The average activity concentration during the period of a single production run is given as:

$$A = \Sigma a_t/\Delta t \quad (2)$$

where, Σa_t and Δt are sum of the activity concentrations detected and the total duration (min) of the stack sampling process respectively.

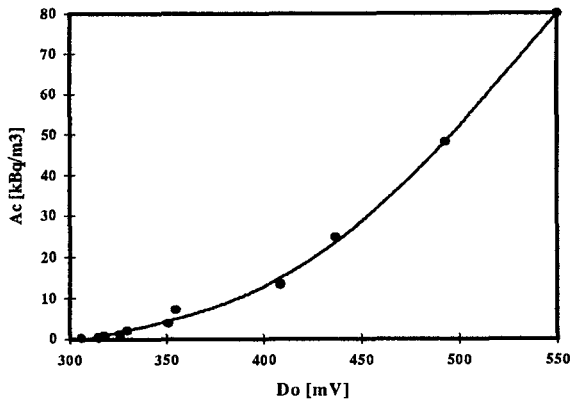

Figure 2b: Calibration curve of the 123-Iodine detector showing the output voltage Do from the SCA2 with the corresponding activity concentration Ac in the detector volume (250ml). The data points are fitted with the 4th order polynomial: $y = -2.40\times10^{-8}x^4 + 4.28\times10^{-5}x^3 - 2.60\times10^{-2}x^2 + 6.92\times10^{0}x - 6.80\times10^{2}$.

4. Results

4.1 Activity Concentration of ^{18}FDG

The activity concentrations of a typical ^{18}FDG production run is shown in Figure 3a. The instantaneous detector output voltages have been modified in real time to the corresponding activity concentrations by the datalogger, using the calibration polynomial (equation 1) shown in Figure 2a. The results were stored in the Excel V5 spread sheet format. The electrical noise and the background signals have been subtracted from the raw data by using a spread sheet macro program. The result indicate the occurrence of a maximum activity concentration of 2.25 MBqm^{-3} at around 7.30hr, which was later verified by the PET radiopharmacist. The average activity concentration value during the period (7.30 hr - 8.30 hr) of the PET production run was calculated using the spread sheet arithmetic (equation 2) and found to be 0.02 MBqm^{-3}. The relevant data acquisition software has been developed at our laboratory and it constitutes a vital part of the present health physics surveillance program[2].

4.2 Activity Concentration of ^{123}I

The results of a typical ^{123}I release is shown in the Figure 3b. The activity concentrations have been derived from the analog voltage outputs of the SCA2 recorded by the datalogger, the methodology as described earlier. The first peak P1 (2.5×10^{-3} MBqm^{-3}) in the figure corresponds to the ^{123}I release occurred during the transfer of residual ^{124}Xe gas from the iodine target (IT) to the storage tank (ST) and the second peak P2 (2.4×10^{-3} MBqm^{-3}) represents the ^{123}I release during rinsing of the target (IT) and radiochemical processing in the iodine processing cell (IPC) as shown in Figure 1. The mean activity concentration of ^{123}I during 24 hour period

has been calculated as 2.46×10^{-4} MBqm^{-3} from the area under the peaks P1 and P2 as shown in Figure 3b.

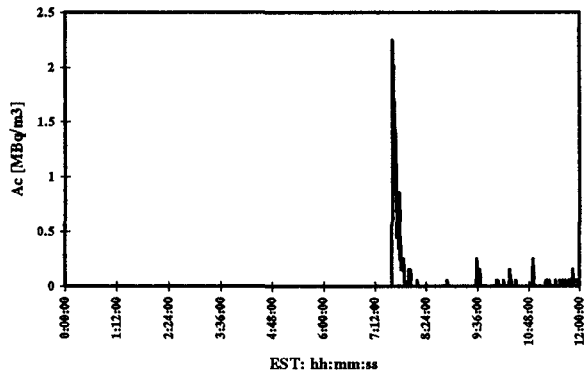

Figure 3a: The results of a typical real time stack discharge analysis during the production of ^{18}FDG at the NMC. The back ground noise has been subtracted from the raw data. The instantaneous characteristics of the effluent release is distinctly visible in this figure.

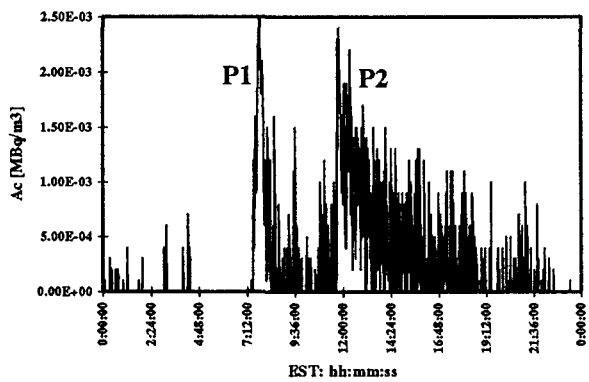

Figure 3b: The results of a typical real time stack discharge analysis during the production of ^{123}I at the NMC. The back ground noise have been subtracted from the raw data. The instantaneous characteristics of the effluent release is distinctly visible in this figure.

5. Discussion

A long term monitoring of the meteorological data and atmospheric tracer test[5] has revealed a stack to boundary dilution factor of the order of 10^{-3} at various critical receptor locations (for members of the public) in the vicinity of the NMC. Therefore, the actual activity concentrations (averaged over one routine production run per day) at those locations would be 20 Bqm^{-3} (for ^{18}FDG) and 0.25 Bqm^{-3} (for ^{123}I). These releases constitute 1% and 2.5% of the maximum permissible limits for ^{18}FDG and ^{123}I respectively. The maximum permissible activity concentrations of ^{18}F and ^{123}I in air for the members of the public[6,7] endorsed by the EPA are 2×10^{3} and 10^{1} Bqm^{-3} respectively.

The novelty of these smart effluent monitors is the absence of the charcoal trapping filter thereby manifesting the following advantages: a) short residence time of the effluent in the detector volume making the detector response independent of air flow rate, b) fast response time making these detectors suitable for instantaneous release assessments, c) long detector life due to the absence of the filter poisoning problem and d) low level activity detection capability.

At present, the dynamic detection range of these stack detectors are restricted by the degree and the span of the calibration polynomials. To overcome this problem wide range digital radiation detection instruments will be used in the near future. The residence times of the effluent flowing at a rate of 0.06m^{3}s^{-1} through the 3ml (positron) and 250ml (iodine) detector volumes are 3ms and 250ms respectively. The response time of the NaI scintillation detectors and the associated electronics are evidently much longer than these residence times. Therefore, the real time activity, computed by the data-logger using the static calibration factors will under-estimate the actual stack release.

To rectify this problem, the installation of two 2-way solenoid valves (SV1 and SV2) in the effluent suction line has been planned (Figure 1). A timer (T) set at 10s duty cycle will actuate the solenoid valves, thereby confining the effluent in both detectors (PD and ID) for 5 seconds in every 10 second duty cycle. The calibration factors estimated with static sources could therefore be used with minor duty cycle correction to assess the activity of the stack release.

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