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A Cost-Effective and Environmentally Friendly Gaseous Waste Management

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Abstract- Dealing with radionuclides includes trapping the vapors and storing it for decay. However, these methods can involve considerable expense in infrastructure, manpower and monitoring. Legislation requires the presence of air filtration systems with detectors to monitor and control the release of radionuclides into the air in nuclear medicine centers. We describe a method for the treatment of gaseous waste that is economically feasible and environmentally friendly. This method complies with legislation and has the advantage of using the already existing resources in the radiopharmaceutical production facility.

Keywords: Environmental Economics; Waste Management; Radioactive Waste
1. INTRODUCTION

During radiochemical processing in nuclear medicine facilities, a substantial part of the positron-emitting radiopharmaceuticals is released into the environment (Mukherjee 2002; Giardina et al. 2015). Council Directive 2013/59/Euratom requires the proper monitoring and evaluation of radioactive airborne material released in the environment and reporting of results to the competent authority in order to avoid the accidental release of high levels of radionuclides during the production of radiopharmaceuticals. In particular, Article 35 states that all Member States shall ensure that an adequate program is in place to monitor the level of radioactivity in the environment. In addition, Article 36 stipulates that Members shall communicate the results of such monitoring to the Commission.

Traditional methods of treating gaseous wastes with above-legal toxicity levels include waste gas release incorporating buffer tanks (Oehninger and Weinreich 1994), automated waste gas compression systems (Pascali et al. 1996) and compression systems (Calandrino et al. 2007). If the level of toxicity is higher than allowed by law, toxic radionuclides are diverted to a place where they will decay.1

Although most commercially available hot cells and automated synthesis modules have capture devices, such as activated carbon filters, chemical adsorbents and liquid nitrogen traps (Calandrino et al. 2007), not all the radioactive gases and aerosols created are effectively trapped. Therefore, these gas compression storage systems need additional protection; otherwise, the by-products generated in the process are dumped directly into the atmosphere. We present a method to obviate these problems as well as being effective for the treatment of these radioactive gases and residues, for a biomedical research center, focusing on conventional nuclear medicine and positron emission tomography (PET).

2. LEGISLATION

The regulatory system. At the international level, there is an obligation to establish a regulatory mechanism. In some countries, a Regulatory Authority has been created consisting of a single entity specifically dedicated to radiological protection.

In Portugal, the respective competencies are assigned to a set of existing entities, of which: the Directorate General of Health (DGS); Regional Health Administrations (ARS); Technological and Nuclear Institute (ITN); the Portuguese Environment Agency (APA); Directorate General of Energy and Geology (DGE); the Regional Directorates of the Economy (DRE). The specific competences of each entity, with regard to radiological protection, are defined in Decree-Law no. 165/2002. Later, Decree-Law no. 139/2005, as an independent supervisor of the entire system, created the Independent Commission for Radiological Protection and Nuclear Safety. This Commission, composed of elements directly appointed by the Prime Minister, has among its powers the capacity to propose amendments and recommendations to the other authorities involved. Despite the existence of a large number of radiological installations for medical and industrial purposes, the only nuclear installation in Portugal for research and teaching purposes, is the Portuguese Research Reactor (RPI) operated by the Technological and Nuclear Institute.
Radiation protection activities in the Directorate General for Health are assigned to the Environmental and Occupational Health Division of the Directorate for Health Promotion and Protection Services. However, the Guidelines for the Management of Radioactive Waste in Portugal suggest that it is technically acceptable that the effluent discharge contains small volumes of radioactive material in a controlled manner in certain situations, as the most reasonable option in the case of very short-lived waste (VSLW) in situations duly provided for by law or authorized by the authority (Sampaio and Fonseca 2015, p. 8).

3. PRODUCTION OF RADIONUCLIDES

For the production, research and development of radiopharmaceuticals, the center has two radiopharmaceutical laboratories and a cyclotron, used in both diagnostic and therapy. In diagnostic, it produces radioactive isotopes typically used in PET treatments. Housed in the bunker, the Cyclone 18/9™ Cyclotron from IBA accelerates protons of up to 18 MeV and deuterium up to 9 MeV to produce radionuclides. These are transported to the radiopharmacy laboratory, where they are used in the synthesis of radiopharmaceuticals. Table 1 shows the levels of present and future nuclide production.

<table>
<thead>
<tr>
<th>Nuclides</th>
<th>Present</th>
<th>Future</th>
<th>Duration</th>
<th>Activity (UNITS)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Production Frequency (times a week)</td>
<td>(in minutes)</td>
<td>Min.</td>
<td>max</td>
</tr>
<tr>
<td>18F</td>
<td>10-15</td>
<td>Slight decrease</td>
<td>60-120</td>
<td>250</td>
</tr>
<tr>
<td>11C</td>
<td>10-12</td>
<td>same</td>
<td>20-30</td>
<td>37</td>
</tr>
<tr>
<td>68Ga</td>
<td>2</td>
<td>10</td>
<td>30-60</td>
<td>3.7</td>
</tr>
<tr>
<td>64Cu</td>
<td>1</td>
<td>2</td>
<td>480-720</td>
<td>2</td>
</tr>
</tbody>
</table>

The main radioisotope currently produced is 18F in the synthesis of 18F-Fluorodeoxyglucose (FDG), but other radioisotopes such as 11C, 68Ga and 64Cu are also produced. Table 2 shows the characteristics of the nuclides.

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Half-life (minutes)</th>
<th>Decay mode</th>
</tr>
</thead>
<tbody>
<tr>
<td>18F</td>
<td>109.8</td>
<td>β⁺</td>
</tr>
<tr>
<td>11C</td>
<td>20.4</td>
<td>SF</td>
</tr>
<tr>
<td>68Ga</td>
<td>68</td>
<td>β⁺</td>
</tr>
<tr>
<td>64Cu</td>
<td>762</td>
<td>β⁺, β⁻</td>
</tr>
</tbody>
</table>

Fluoride (F-18) is one of the most popular short-lived positron emission radioisotopes ($T1/2 = 109.8$ min) (Wilson et al. 2008). Its use as a radioactive marker in combination with noninvasive and metabolic PET imaging techniques is of great importance in medical imaging (Cai et al. 2008). In fact, FDG is the most successful and widely used
clinical imaging PET marker in oncology (Hoh 2007; Yu 2006). Thus, PET radioisotope production is well established and is routinely monitored for the generation of volatile radioactive gaseous and aerosol by-products, whose storage and disposal are challenging in financial and environmental terms (Pascali et al. 1996).

Carbon-11, whose half-life is 20.334 min, is the most stable artificial radioactive isotope and is often used in the radioactive labeling of molecules in PET. Its decay occurs mainly due to the emission of positrons.

Gallium-68 is a positron-emitting isotope with a half-life of 68 min, which is generated from germanium-68 for use in diagnostic tomography. It is normally attached as a label to a carrier molecule.

Finally, copper-64 has a half-life of 762 min and is a positron emitter, making it a viable radionuclide for PET imaging. Thus, it can be used in cancer radiotherapy, medical research and diagnostic practice (Blower 2015). Its characteristics allow simultaneous monitoring of the distribution of drugs and bio-kinetics.

Irradiation during 18FDG production is typically < 2 h, with varying downtime between the production cycles. For long-lived nuclides ($T_{1/2} > 90$ days), the waiting time between production cycles (typically < 3 days) is insignificant compared to half-life, and sample activity is relatively constant (less than 3%). For very short-lived nuclides ($T_{1/2} < 15$ min), saturation activity is easily achieved within a single production line, but the nuclide also decays very rapidly.

The radionuclides produced at the center are between these two limits (long- and very short-lived nuclides). In this case, a part of the product decays between the production cycles, accumulates during the next cycle and then decays further in a repetition cycle. Thus, the actual time needed to achieve saturation varies with the half-life and time intervals between production cycles.

In Portugal, dose limits are regulated by Decree-Law no. 22/2008 of 17 November and are in conformity with the requirements of Council Directive 96/29/Euratom of 13 May, of which the effective dose limit for exposed workers is set at 100 mSv averaged over a period of five consecutive years, provided that it does not exceed a maximum effective dose of 50 mSv in each year.

Notwithstanding this limit, the following are also fixed: The equivalent dose limit for the lens was set at 150 mSv per year but current regulations has reduced this limit to 20 mSv per year with significant impact on the monitoring and dose implications to the worker; the equivalent dose limit for the skin is set at 500 mSv per year; the equivalent dose limit for the purposes is set at 500 mSv per year. The effective dose limit for the public is set at 1 mSv per year and may be exceeded in a given year, if the average dose for five consecutive years does not exceed 1 mSv per year.

Radiosynthesis equipment. Preventive maintenance and operational use of the Cyclone 18/9™ Cyclotron from IBA used for the production of nuclides produce long-lived activated waste that must be classified by radionuclide and pre-disposal activity as LLRW (low-level radioactive waste). The cyclotron can generate large volumes of waste from the production of various positron emitters; however, full spectrometric gamma analysis of all wastes is expensive, and excessive waste handling is contrary to the ALARA concept (Breuning 2010).
It is the center’s responsibility and obligation to minimize gaseous radioactive waste using the most feasible means. Therefore, our goal is not to discard, but to safely retain as much radioactive waste as possible, which is created during any isotope production.

In each of the two radiopharmacy laboratories, there are two armored COMERER™ cells that have their protection mechanisms. Indeed, the cells possess outer surface and internal work area of seamless stainless steel with rounded edges for easy decontamination. In addition, the safety locking system prevents its opening in the presence of greater activity within the limits predetermined by the user. Other characteristics of these cells include extraction of activated carbon filter and configurable ventilation system with pressure control under the cell and warning in case of pressure failure. The purity of the lead responsible for the radioprotection characteristics of these cells is 99.97%. Moreover, since the synthesis and fraction systems installed in armored cells are automated, exposure to radiation is minimized. As the cyclotron is not self-protected, it is placed in a bunker with walls made of two meters of concrete and a door equipped with an automatic control system for opening and closing. Before it is activated, people are evacuated from the sealed area.

The bunker door fits into the walls and has safety mechanisms such as an audible alarm when it opens/closes slowly, stopping its movement when the sensors detect any impediment and cease immediately displacement in case of emergency.

Since the synthesis and fraction systems installed in armored cells are automated, exposure to radiation is minimized. The cyclotron also has a safety key that needs to be placed in the control room in the ON position prior to the production of radionuclides. During cyclotron operation, neutron and gamma radiation levels are measured outside the bunker. The area dose rate monitoring system is located within the cyclotron.

4. COST-EFFECTIVE ANALYSIS OF GASEOUS WASTE MANAGEMENT

Radiation is mainly the gamma resulting from the annihilation of positrons. Background radiation is measured when the cyclotron is not in operation. To this end, there is a neutron flux monitoring system located in the chamber that accesses the bunker: in the technical corridor (behind the bunker) and on the ground floor of the waiting room (because it is situated above the bunker).

Figure 1 shows the blueprint of the center. The synthesis of FDG is produced in the radiopharmacy laboratory 1, whereas radiopharmacy laboratory 2, which is dedicated to research, receives the radioactive atoms of $^{11}$C, $^{13}$N and $^{15}$O produced in the cyclotron. The technical corridor links the cyclotron bunker (1) to the radiopharmacy laboratories (2 and 3).
Instead of being released into the atmosphere, the radionuclides with a level of toxicity above that allowed by law are transported through the technical corridor (4), from the radiopharmacy laboratories (2) and (3) to the cyclotron bunker (1).

Figure 2 shows the interior of the bunker where the cyclotron is located.
The insertion of the radioactivity is done in the northwest corner of the bunker along the ground. The air inside the bunker is exhausted through the ventilation system in the center of the ceiling. The principle of the radioactive management is the simple dispersion in the cyclotron bunker, which is particularly large (58.02 m²) when compared to other cyclotron bunkers.

There are a number of problems related to protecting humans and the environment from exposure to radiation. First of all, and according to Wagner (2003), environmental law faces the major problem of lack of scientific information to assess the impact of industrial activities on public health and the environment. In the absence of such research, a number of authors (for example, Mukherjee 2002; Giardina et al. 2015; Infantino 2015; Ferdous et al. 2017) have focused on measuring the radiation level of their medical facilities and comparing it with the legal permitted levels. In addition, in Portugal, although there are more than 100 laws, regulations and decrees governing nuclear activities, the legal framework is incomplete (NEA 2011). Many PET plants 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 tanks armored lead. However, these devices take up a lot of space (Won et al. 1997) and the costs of disposal have risen substantially as disposal facilities have developed. As a result, and as part of cost-reduction efforts, some countries show a trend to minimize the production of radioactive wastes (Rau et al. 2000). At the same time, less expensive solutions have been sought for disposal of VLLW.

These solutions are basically two: buffer tanks or long pipes for delaying the expulsion of gases (Tochon-Danguy et al. 1994; Pascali et al. 1996) and gas capture bags with electronic feedback that are integrated with the cyclotron safety system (Schweiger 2011; Stimson et al. 2016). Table 3 shows a comparison of costs between three methods: gas compression systems, gas capture bags and our method.
<table>
<thead>
<tr>
<th>Gas compression systems</th>
<th>Gas capture bags</th>
<th>Our method</th>
</tr>
</thead>
<tbody>
<tr>
<td>&gt; USD $ 100,000</td>
<td>USD $20,000</td>
<td>USD $ 0</td>
</tr>
</tbody>
</table>

Source: Own elaboration based on Stimson et al., 2016.

Our new simple technique allows treating radioactive gases and aerosols generated during production without the need for additional space and it is less expensive than the other solutions. Thus, our method is practical, effective, reliable and cost-effective.

**Limitations.** It may be noted that this method of waste disposal undermines the long-term profitability of cyclotron production. However, so far this argument has not been proven.

### 5. CONCLUSION

Compliance of levels of toxicity with legislation is checked on the battery monitor so that it never exceeds legal limits. Air dispersion immediately decreases the level of toxicity to legally accepted levels. The bunker is especially large (58.02 m²); therefore, future research would be checking if the dispersion of gaseous wastes in a bunker of smaller dimensions would immediately reduce activity to acceptable levels of toxicity, and to calculate the minimum closed area that allows the immediate dispersion of toxicity to levels below the legal limit. This method complies with the legislation and has the advantage of using the existing resources in the Nuclear Medicine Center. Therefore, this method is reliable and cheaper than traditional methods.

**References**


