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Radioxenon Project – Task II/2

Design Studies

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1 Introduction

The preparatory commission of the Comprehensive nuclear-Test Ban Treaty Organization (CTBTO) has set-up a wide variety of high precision sensors designed to detect possible violations of the treaty. These sensors are divided in four technologies: seismic, infrasound, hydroacoustic and radionuclide identification. Together they constitute the International Monitoring System (IMS) (CTBTO, 2017). Half of the radionuclide stations contain noble gas detectors. The noble gases are very important for verification purposes since they have a very low interaction with their surroundings, which makes it possible to detect them over long distances and even from underground nuclear tests. Moreover, the radioxenons that would give a significant signature from a nuclear test are: Xe-131m, Xe-133, Xe-133m and Xe-135, all of them having a relatively short half-life which results in no memory effect in the atmosphere. During a nuclear test they are produced in high quantities, which make them a key component in order to validate that a seismic, infrasound or hydroacoustic detection is of nuclear testing nature.

However, it has become clear in the previous decade that there is a worldwide radioxenon background coming from various civilian sources. The main sources of radioxenon releases to the atmosphere were determined to be coming from a very limited number of Radiopharmaceutical Production Facilities (RPF's), which are dispersed around the globe (Saey, 2009). This radioxenon background makes it very difficult to interpret the measurements obtained by the international noble gas monitoring system, which is operated by the CTBTO. A reduction of this global radioxenon background would significantly enhance the verification capability of the CTBTO. Moreover, it is expected that the radioxenon background will increase and become more complex in the near future because of the new startup of radiopharmaceutical production facilities in the world. An acceptable upper limit of 5 GBq/day for radioxenon releases from RPF's was proposed by *T. Bowyer et al.* This upper limit is based on the perspective of minimal impact on monitoring stations and it could possibly be achievable for large RPF's (Bowyer, et al., 2013).

In this framework, SCK•CEN has been contracted by the CTBTO under the EU Council Decision V to design a mobile system for the reduction of radioxenon emissions from radiopharmaceutical production facilities. The project was subdivided in three phases: i) investigation and selection of xenon adsorption materials (Gueibe, et al., 2014), ii) study of operational conditions and trap design (Gueibe, et al., 2015a) and iii) construction and testing of a mobile trap (Gueibe, et al., 2015b). This project was performed in partnership with the Institute for RadioElements (IRE) in Fleurus, Belgium. The project was successfully completed by the end of 2015. The major result of this project was the development and design of a retention system prototype that fits into the process line of the IRE. This first prototype demonstrated promising results and was delivered to the IRE at the end of the EU CD V.

In order to pursue ongoing efforts in mitigating the radioxenon emissions from RPFs and as a follow up to the outcomes of the EU CD V related work, further developments and testing of the retention system prototype were needed. In this regard, SCK•CEN and IRE were contracted in 2016 by the CTBTO under the EU Council Decision VI to: i) analyze the scale-up and the long term behavior of the prototype at IRE, ii) perform design studies at additional facilities and iii) investigate further the stack releases and stack monitoring at IRE. This report concerns the design studies at additional facilities.

The second task (*i.e.* design studies at additional facilities) is performed for a maximum of three facilities taking into account their specific operational features and conditions. The first step of the second task was the selection of the facilities, which was performed in two stages: i) request for letters of interest containing the necessary information to demonstrate that the requirements to participate in the study are met and ii) a dedicated web conference with each facility selected from the received letters of interest. The results of this first step in the second task of the project are fully described in (Gueibe, et al., 2017a) and (Gueibe, et al., 2017b). Resulting from this first step, three facilities, namely: Nordion (Canada), ANSTO (Australia) and PT INUKI (Indonesia), were selected to perform the foreseen design studies.

The design studies that were performed for these facilities are discussed in the current report. The work performed for each facility is discussed in a separate section (*i.e.* Nordion in Section 2, ANSTO in Section 3 and PT INUKI in Section 4). Finally, the conclusions of the work performed within this task are drawn in Section 5.

2 Nordion

Nordion is a global health science organization that provides market-leading products used for the prevention, diagnosis and treatment of disease. Nordion is a leading provider of gamma technologies and medical isotopes that benefit the lives of millions of people in 45 countries around the world. Nordion's products are used daily by pharmaceutical and biotechnology companies, medical-device manufacturers, hospitals, clinics and research laboratories (Nordion, 2017). Nordion's medical isotopes are used in the diagnosis and treatment of disease, including applications for cardiology, neurology, oncology and research (Nordion, 2017).

In February 2015, Nordion announced a new isotope supply agreement with U.S.-based Missouri University Research Reactor Center (MURR) and General Atomics. This agreement will replace its supply when the Chalk River facility closes in 2018. A new technology for Mo-99 production, General Atomics' patented Selective Gaseous Extraction (SGE) technology, will be used. The targets, which will incorporate LEU, will be irradiated at MURR where the SGE will be performed. The extracted material will then be shipped to Nordion for processing (Critch, et al., 2015).

As described in the previous report, the initial information collected through the questionnaire, email exchanges and the web conference, confirmed that the intended design study was technically feasible for the facility (Gueibe, et al., 2017b). A preliminary investigation for the design study was performed based on the information received. A technical visit of the facility was carried out in June 2017 to gather a more detailed insight on the production processes and the radioxenon releases they possibly induce. In addition, meetings were organized with Nordion's experts to discuss the preliminary investigation and collect their feedback on this preliminary investigation. Resulting from the technical visit, it was agreed that two different design studies would be performed with working conditions defined by Nordion. The first design study was dedicated to a specific production process, which was linked to the major radioxenon release of the production facility. The second design study was performed to provide Nordion with some design options for other, smaller, radioxenon releases. In addition to these two design studies, a feasibility study for a xenon mitigation system working on the whole production ventilation was performed. During the technical visit, the reduction of yet another type of small radioxenon release was discussed. Resulting from these discussions, it became clear that some of these smaller radioxenon releases could be reduced by enhancing the radioiodine trapping system to avoid the subsequent radioxenon releases. This will be further investigated by Nordion, as this was not part of the current project.

During the development of the design studies, intermediate results were presented to Nordion's experts to collect their feedback. The feedback received from Nordion's experts was used to refine the design studies to meet their specific needs.

Regarding the major radioxenon release, the dimensions of a radioxenon mitigation system (working with the Ag-ETS-10 silver zeolite or with the NUCON NUSORB® GXK activated carbon) for the specific production process were provided to Nordion. In addition, the detailed working process of the proposed radioxenon mitigation system was shared with Nordion. Finally, the heating up of the Ag-ETS-10, due to the decay of radioxenon, was investigated. The detailed calculations, analysis and discussions were shared with Nordion as well.

Resulting from the second design study, the dimensions of a radioxenon mitigation system (working with the NUCON NUSORB® GXK activated carbon) for different generic working conditions were provided to Nordion. The reduction in the dimensions of the system by using the Ag-ETS-10 silver zeolite instead of the previously studied activated carbon were detailed as well. The detailed calculations, analysis and discussions were shared with Nordion.

The results, as well as the detailed calculations, of the feasibility study for a xenon mitigation system, working on the whole ventilation of the production facility, were provided to Nordion.

The major outcome of this investigation is that such a system would not be efficient and requires very large dimensions to be able to reduce significantly the radioxenon releases. This highlighted the need to work as closely as possible to the radioxenon release source (to avoid dilution in the gas stream and to have a better control on the ventilation conditions).

3 ANSTO

ANSTO Health, a division of the Australian Nuclear Science and Technology Organisation (ANSTO), currently produces around 2 milion doses of potentially life-saving medical isotopes per year. These medicines are used to diagnose and treat a wide variety of conditions including cancers, heart disease and skeletal injuries in patients in Australia and around the globe (ANSTO, 2017).

ANSTO Health produces Mo-99 from Low Enriched Uranium (LEU) targets in ANSTO's LEU fueled research reactor OPAL. ANSTO Health currently exports Mo-99 to countries including the USA, China, Japan and South Korea, and has the capacity to double its exports (ANSTO, 2017).

There is a lack of reliability in global Mo-99/Tc-99m supply due to the closure of aging infrastructures across the globe. ANSTO is establishing a new production plant that will meet 25-30% of future global demand for Mo-99, and is building a waste treatment facility which will utilize ANSTO-developed technologies to treat Mo-99 waste (ANSTO, 2017).

As described in the previous report, the initial information collected through the questionnaire, email exchanges and the web conference confirmed that the intended design study was technically feasible for the facility (Gueibe, et al., 2017b).

It has to be noted that the design study was performed for the existing facility and not for the new production plant that is currently being built. This is due to the fact that the new production plant will have a dedicated radioxenon mitigation system that has been designed, based on ANSTO's experience with radioxenon releases in the existing production plant. This system consists of a combination of decay tanks and activated carbon filters (specifically designed for trapping noble gases). As a result of the dedicated radioxenon mitigation system, the radioxenon emissions from the new production facility are expected to be lower than the existing facility. However, this will only be ascertained when the facility will be operational. The purpose of the design study, which was agreed with ANSTO's experts, is to develop a xenon trapping column for testing purposes, based on the Ag-ETS-10 material, that could replace a currently used activated carbon column while decreasing the underlying xenon emissions. The aim for ANSTO is to have a proof of concept for the efficient and reliable xenon trapping with the Ag-ETS-10 material in their operational conditions, which is outside the scope of the current project. If the Ag-ETS-10 column, designed in the current project, proves to be efficient and reliable for xenon capture, the Ag-ETS-10 material could be used in the new facility (if needed) to reduce further the xenon releases.

A preliminary investigation for the design study was performed based on the information received. A technical visit of the facility was carried out in September 2017 to gather a more detailed insight on the production processes and the radioxenon releases they may induce. In addition, meetings were organized with ANSTO's experts to discuss the preliminary investigation and collect their feedback on this preliminary investigation. Resulting from the technical visit, it was agreed to further develop the design study for the intended xenon trapping testcolumn, based on the Ag-ETS-10 material.

During the development of the design study, intermediate results were presented to ANSTO's experts to collect their feedback. The feedback received from ANSTO's experts was used to refine the design study to meet their specific needs.

The detailed calculations, analysis and discussions of the design study were shared with ANSTO. The required dimensions of the Ag-ETS-10 testing column that would yield the same retention time as the currently applied activated carbon filters were provided to ANSTO as well, together with the working process of the proposed testing column.

4 PT INUKI

PT Industri Nuklir Indonesia (Persero) or PT INUKI (Persero) previously known as PT Batan Teknologi (Persero) is the only State-Owned Enterprise in Indonesia engaged in nuclear technology-based industries. PT INUKI focuses on the businesses of radioisotope production, radiopharmaceutical production, nuclear fuel fabrication, nuclear power operation and maintenance, and other nuclear engineering services. Radioisotope productions include Mo-99, I-131, P-32, Zn-65 pip tag (for radioactive marker sub), and Ir-192 sealed source. Radiopharmaceutical products include Generator Mo-99/Tc-99m, sodium I-131 (oral, injection and capsule), and sodium I-131-Hippuran (PNNL, 2016).

As described in the previous report, the initial information collected through the questionnaire, email exchanges and the web conference confirmed that the intended design study was technically feasible for the facility (Gueibe, et al., 2017b).

A preliminary investigation for the design study was performed, based on the information received. A technical visit of the facility was carried out in May 2017 to gather a more detailed insight on the production processes and the radioxenon releases they possibly induce. In addition, meetings were organized with PT INUKI's experts to discuss the preliminary investigation and collect their feedback on this preliminary investigation. Resulting from the technical visit and the following email exchanges with PT INUKI's experts, it was decided to perform two generic design studies. These generic design studies were developed to provide some design options for PT INUKI, which could be used by PT INUKI once they will have clearly identified the radioxenon releases in their production process. The identification of the radioxenon releases could not be done in the framework of the current project as the radioxenon stack monitoring data could not be made available in due time. However, it is clear that two different types of radioxenon release can occur in the production process. The first type of radioxenon release is related to prompt releases resulting from specific production steps. The second type of radioxenon release is resulting from the decay of iodine that is precipitated on surfaces or captured in specific filters. This second type of radioxenon release is resulting in an ongoing, continuous radioxenon release. According to the previous remarks, one design study for each kind of release has been developed.

The design study for prompt radioxenon releases was developed with two different options, *i.e.* a radioxenon mitigation system that would work for a specific period of time on the ventilation of the respective hot cell and a radioxenon mitigation system that would work for a specific period of time on an isolated ventilation pathway in the hot cell. The second option allows to better control the ventilation conditions and to avoid the dilution of the radioxenon before being treated by the system. This is resulting in a much smaller radioxenon mitigation system. However, this would require larger adaptations to the current production process. The dimensions of the adsorption columns and the working process of the proposed radioxenon mitigation system for both options were provided to PT INUKI together with the detailed calculations, analysis and discussions for each proposed system.

The second design study is focusing on radioxenon releases resulting from the decay of radioiodine trapped in filters. For this design study, a hot cell integrated container box is proposed to store all radioiodine containing waste, which can then be ventilated in well-controlled conditions. In such a design, the outgoing gases of the container box will be passing through an activated carbon filter for iodine capture and then through a dedicated xenon mitigation system before being released to the atmosphere. The dimensions of the adsorption columns and the working process of the proposed radioxenon mitigation system were provided to PT INUKI. The detailed calculations, analysis and discussions were shared with PT INUKI as well.

5 Conclusions

Based on the initial selection process, three facilities, namely: Nordion (Canada), ANSTO (Australia) and PT INUKI (Indonesia), were selected to perform the design studies intended in the Radioxenon Project.

Based on the initial information collected through the questionnaire, email exchanges and the web conference, a preliminary investigation for a radioxenon mitigation system was performed for each facility. Each preliminary investigation was used to feed the discussions that were held during the technical visit of the respective facility. The feedback and additional information collected during the technical visit was used to develop the agreed design stud(y)(ies) for the respective facility.

During the development of the design studies, intermediate results were discussed with the respective facility to meet as well as possible the specific needs of the respective facility.

The respective detailed calculations, analysis and discussions were provided to the facility. These results can be used by the facility to develop a radioxenon mitigation system or to improve their current radioxenon mitigation means.

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