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Results and outcomes in xenon mitigation under the European Union Council Decision V

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Introduction

The monitoring capabilities of the IMS noble gas systems may benefit from reductions of radioactive noble gas emissions. If these are achieved at nuclear facilities that have high radioxenon releases in normal operation, this could significantly enhance the CTBT verification capability. In this framework, the SCK•CEN was 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) exploration and selection of appropriate xenon adsorption materials, ii) study of operational conditions and trap design and iii) construction and testing of a mobile xenon trap. The project was performed in close collaboration with the Institute of RadioElements and was successfully completed by the end of 2015. The major results and outcomes of the project are shown.

Exploration and selection of appropriate xenon adsorption materials

In this phase of the project, different solid materials, with an emphasis on silver zeolites, that showed promising xenon adsorption properties in the literature were investigated. The different adsorbent materials that were investigated are summarized in Table 1.

Type of material	Name of Material	Supplier
Activated Carbon	RKJ 1	Cabot Norit Nederland B.V.
Activated Carbon	NuclearCarb 203C	Chemviron Carbon
Activated Carbon	Nusorb GXK	NUCON International Inc.
Carbon Molecular Sieve	MSC CT-350	Japan EnviroChemicals Ltd.
Silver Zeolite	Ag-ETS-10	Alberta Adsorbents Inc.
Silver Zeolite	Ag-Chabazite	Alberta Adsorbents Inc.
Silver Zeolite	Ag-Mordenite	Sigma Aldrich Co.

Table 1 – Investigated adsorbent materials

An existing experimental system was adapted and optimized to meet the requirements needed for this project. The system contains three main parts: the gas management system, the adsorption test bed and the detection and acquisition system. Stable xenon was used in the experimental system for safety reasons. The schematic drawing of the whole experimental system is shown in Figure 1.

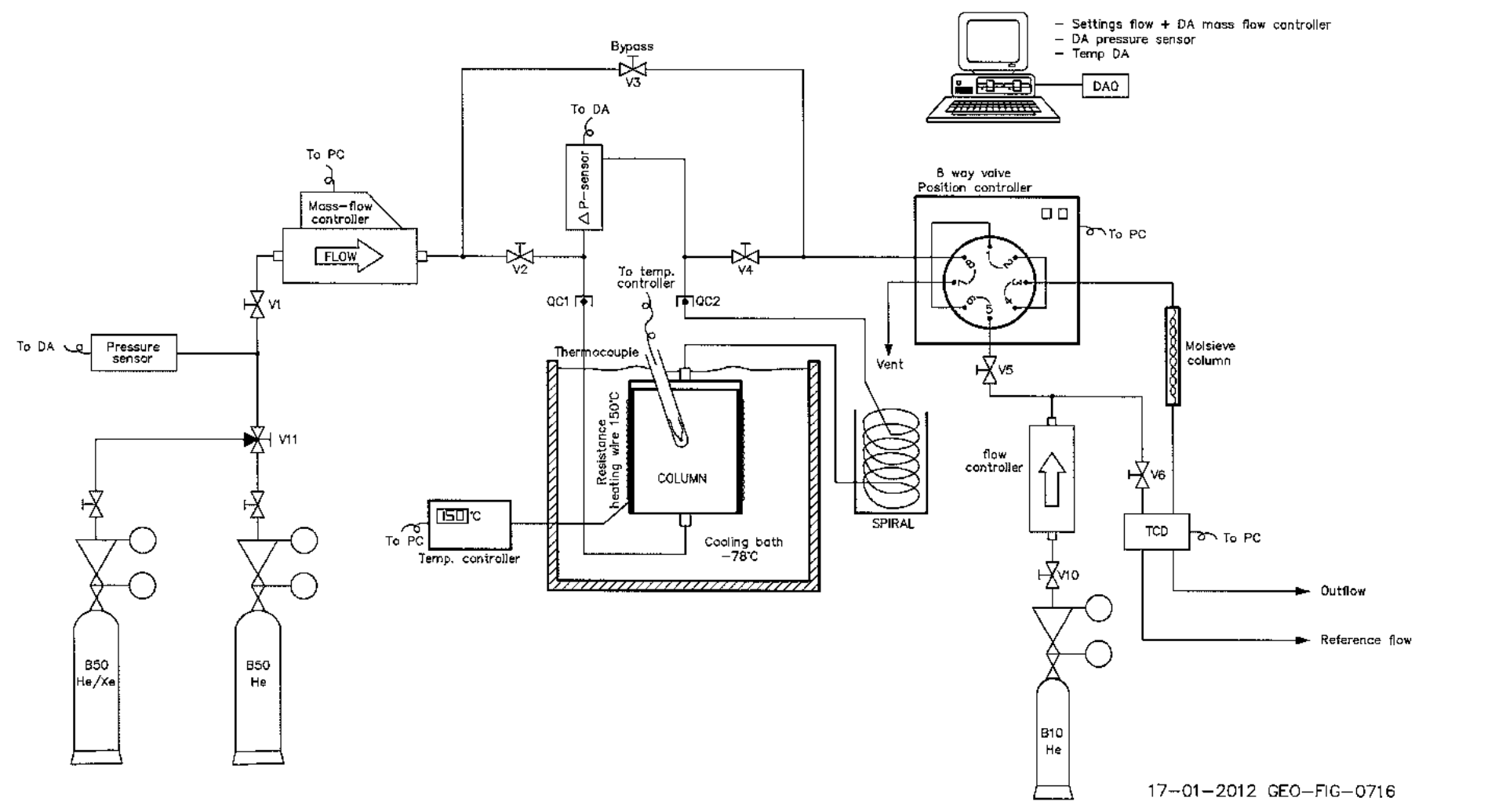

Figure 1 – Schema of the experimental set-up

The different adsorbent materials were compared under the same conditions as show in Table 2. These conditions are typical for the production step that was investigated at the radiopharmaceutical production facility. The xenon breakthrough curves obtained for the 4 most promising materials are shown in Figure 2. The best results were obtained with the Ag-ETS-10 silver doped zeolite (green curve). The measured adsorption capacity of the Ag-ETS-10 is 25 times higher than the best activated charcoal observed in our conditions.

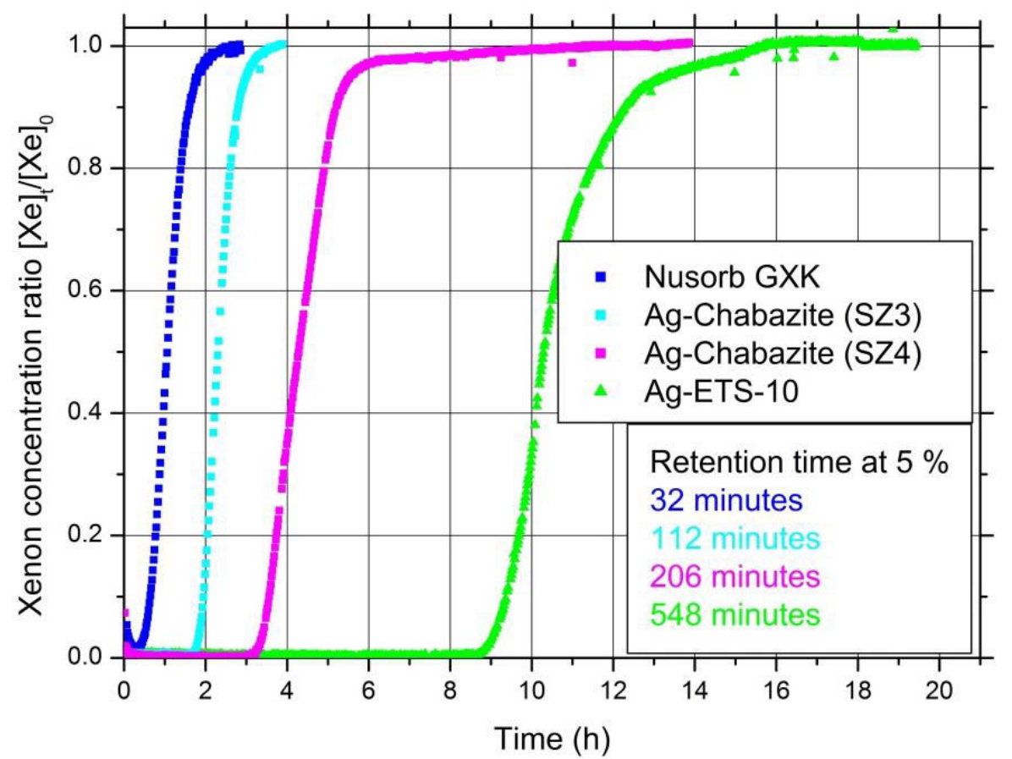

Figure 2 – Comparison of xenon breakthrough curves

Flow rate	400 cm³/min
Pressure	~ 1.8 bar
Temperature	~ 20°C
Adsorbent volume	~ 20 cm³
Xe/He	1000 ppm

Table 2 – Experimental conditions

References
[1] C. Gueibe, J. Camps and K. van der Meer, “Xenon mitigation project - Phase I: adsorption materials”, SCK•CEN,Mol, 2014.
[2] C. Gueibe, J. Camps, J. Rutten, K. van der Meer and J. Paridaens, “Xenon mitigation project - Phase II: adsorption materials”, SCK•CEN, Mol, 2015.
[3] SCK-CEN, “VISIPLAN,” [Online]. Available: http://www.visiplan.be/~/media/Files/Visiplan/Visiplan/visiplan-1.pdf. [Accessed 20 November 2014].
[4] C. Gueibe, J. Rutten, J. Camps and K. van der Meer, “Xenon mitigation project - Phase III: prototype construction and testing”, SCK•CEN, Mol, 2015.

Study of operational conditions and trap design

An advection-diffusion-adsorption model was developed in Comsol Multiphysics® to investigate different geometries, the volume required for the adsorption of xenon from the defined production process and different working processes for the prototype. The model was validated with all experiments performed in the various conditions investigated. For each adsorbent material the simulated breakthrough curves were fitted to the experimental ones to estimate the adsorption properties. An example of such a fit for the Ag-ETS-10 is shown in Figure 3.

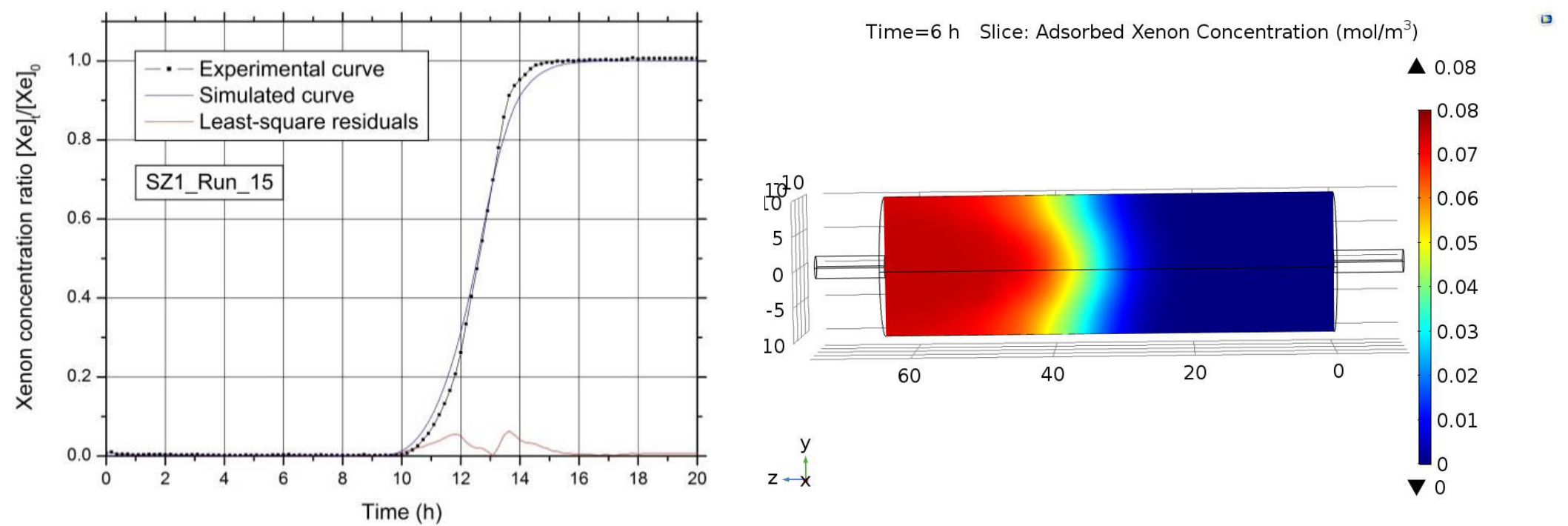

Figure 3 – Fit of an experimental breakthrough curve on the Ag-ETS-10 (left) and the simulated adsorbed xenon concentration after 6h (right)

The developed model was used to investigate the adsorption kinetic for different trap geometries under the same conditions. Resulting from this investigation, a new laboratory column was developed and was used to validate the model results as shown in Figure 4.

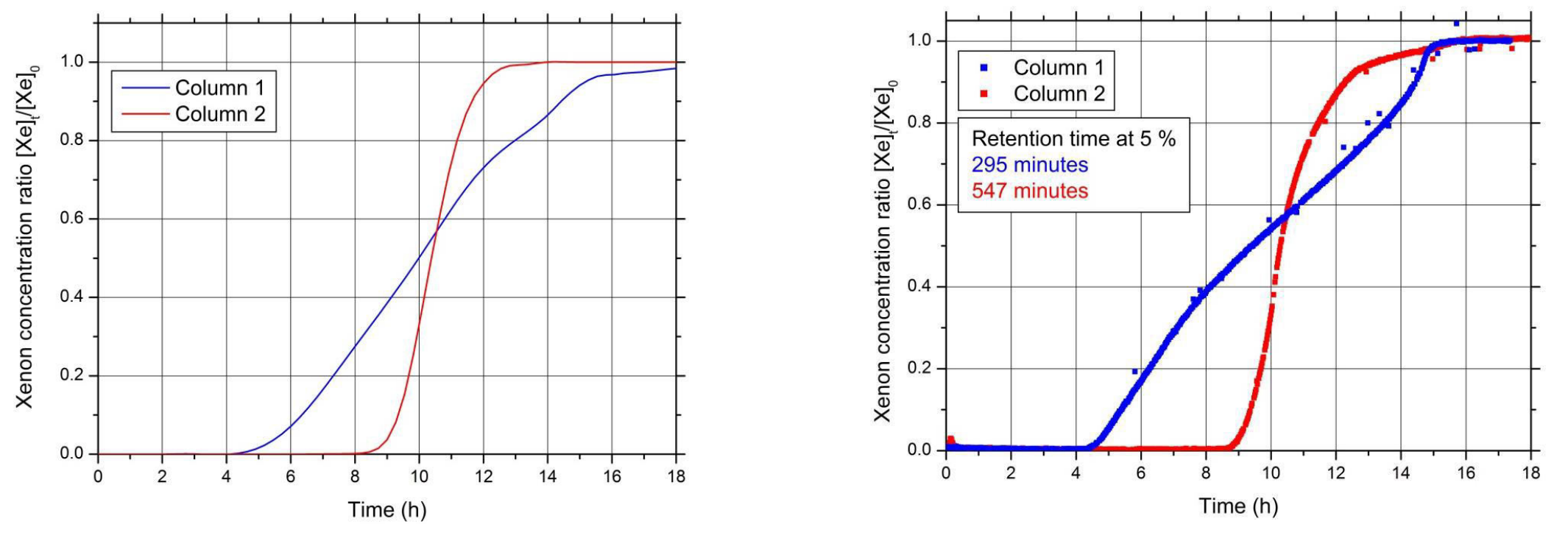

Figure 4 – Simulated adsorption kinetic under two different trap geometries (left) and experimental validation (right)

The durability of the Ag-ETS-10 with regard to regeneration cycles was investigated through 14 adsorption/regeneration cycles and showed only very small variations in adsorption capacity. In addition, the Ag-ETS-10 was irradiated in SCK•CEN’s gamma irradiation facility resulting in a 1 MGy absorbed dose and showed, based on the adsorption capacity, no degradation of the material.

The scientific design studies made it possible to optimize the working process, the trap geometry and the volume of adsorbent with regard to: xenon adsorption, heat dissipation and compactness of the system. In addition, the VISIPLAN software [4] was used to determine the shielding required for the xenon mitigation prototype in order to meet the dose constraints for workers around the system. The final technical design of the prototype is shown in Figure 5. The resulting prototype is very **compact** and **mobile**. In addition, the lead shielding can easily be removed. The prototype is also very **flexible** as the adsorption columns can be removed, used in series or in parallel.

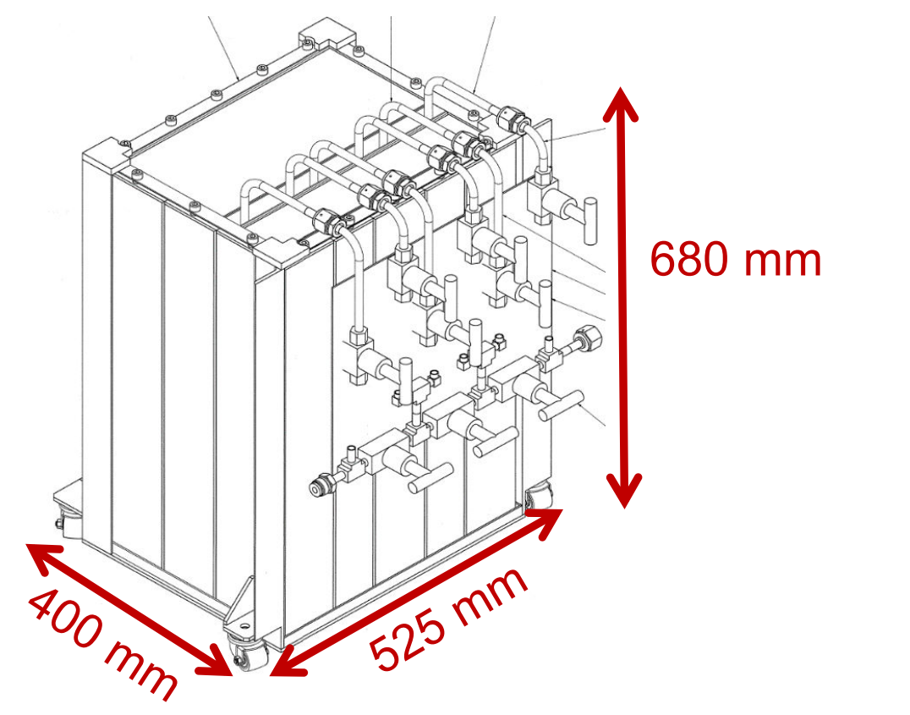

Figure 5 – Technical design of the prototype

Construction and testing of a mobile xenon trap

The prototype constructed based on the technical design is shown in Figure 6. The adsorption properties and regeneration of the Ag-ETS-10 in the adsorption columns of the prototype were experimentally tested at SCK•CEN with stable xenon. The experimental results behaved as expected.

The regeneration system used in the prototype is based on infrared lamps and is very efficient in regenerating the Ag-ETS-10. Finally, leak tests under helium were performed both in over and under pressure. At the end of the project, the prototype was donated to the Institute of RadioElements with a qualification file.

Figure 6 –Front (left), side (middle) and top (right) view of the xenon mitigation prototype

Conclusions

- Ag-ETS-10 concentrates xenon by a **factor 25** more efficiently than the best activated charcoal investigated in the defined conditions
- Exploration of irradiation (1 MGy) and temperature effects (up to 60°C) in Ag-ETS-10 showed no degradation of the Ag-ETS-10
- Simulations for: advection-diffusion-adsorption of xenon, heat build up due to decay and radiation protection
- Design studies for the optimization of the xenon mitigation system
- Xenon mitigation prototype tested (stable Xe) and delivered to IRE
- The experience gained in this project is also of relevance for xenon capture in IMS noble gas systems

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