QUANTOM® – Optimization of the online neutron flux measurement system

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Abstract—**For the final disposal of radioactive waste, the waste packages have to meet the acceptance requirements defined by national licensing and supervisory authorities. Non-destructive methods are very much preferred over destructive methods for the qualification or re-qualification. Existing non-destructive methods as integral or segmented gamma scanning or neutron counting only determine the isotope specific activity but do not allow quantifying other non-radioactive hazardous substances. These should have been documented during creation, conditioning, and packaging of the waste. But especially for legacy waste, this documentation is often poor or even missing. This gap is to be filled by the QUANTOM® measurement device that will determine the mass fraction of elements within a 200-l-drum using the Prompt- and Delayed-Gamma-Neutron-Activation-Analysis. In order to obtain a spatially resolved characterization, it will employ a segmented scanning approach. For the determination of the absolute mass fractions, the neutron flux inside the drum has to be known accurately. As the waste itself will alter the neutron distribution and flux, it is not possible to calculate the latter a priori from the gamma measurement. Hence the neutron flux has to be measured simultaneously with the gamma radiation. In this presentation, we will introduce the system for measuring the thermal neutron flux surrounding the waste drum from which the flux within the waste package has to be reconstructed. We performed a simulation study to score several possible detector placements for an improved reconstruction performance. We will show the outcome of these calculations and present the final design of the detector arrangement.**

Index Terms—**Prompt Gamma Activation Analysis, PGAA, Radioactive Waste disposal, Waste qualification**

I. INTRODUCTION

FOR the safe intermediate storage and final disposal of radioactive waste, waste packages have to meet the waste acceptance requirements defined by national licensing and supervisory authorities. In Germany the federal company for radioactive waste disposal BGE (Bundesgesellschaft für Endlagerung) has established waste acceptance requirements for the geological repository Konrad and is responsible for the qualification of radioactive waste packages.

For the qualification or re-qualification, non-destructive methods are very much preferred over destructive methods, as the latter lead to additional exposures of operating personnel, increase of the waste volume, increase of cost, and are very time consuming. Existing non-destructive methods as integral or segmented gamma scanning or neutron counting only determine the isotope specific activity but do not allow quantifying other non-radioactive hazardous substances. These should have been documented during creation, conditioning, and packaging of the waste. But especially for legacy waste, this documentation is often poor or even missing. This makes it difficult to prove that the waste does comply with the acceptance criteria. Thus there is an urgent need for the non-destructive determination of the elemental and chemical composition of the content of waste drums.

This gap is to be filled by the QUANTOM® measurement device. This device will measure the mass fraction of many elements within a 200-l-drum using the Prompt- and Delayed-Gamma-Neutron-Activation-Analysis (P&DGNAA). These mass fractions can be used to check the plausibility of a declared material vector. In order to obtain a spatially resolved characterization, QUANTOM® uses a segmented scanning approach. A comprehensive description of this system and its measurement geometry is given in the paper of A. Havenith [1]. For the scope of this paper, it is sufficient to know, that the device consists of a large graphite moderator/reflector with a central cavity which holds a 200-l-drum. A DD-Neutron generator is used as the source of 2.45 MeV neutrons. Two collimated HPGe-detectors record the gamma signals resulting from neutron captures within the drum. The collimation allows a segmented scanning of the drum in four axial layers with 12 radial segments each, resulting in a total of 48 segments. To determine the gamma spectrum for each of the 48 segments, 24 different orientations of a single drum have to be measured.

The gamma signal is proportional to the amount of material present and also to the neutron flux within the drum. To calculate accurate absolute mass fractions from the gamma signal the spatial distribution of the neutron flux within the waste drum needs to be known to normalize the signal. As the waste itself will alter the neutron distribution and flux by moderation and absorption, it is not possible to calculate the latter a priori from the gamma measurement. Hence the neutron flux has to be determined simultaneously

with the gamma radiation. Due to the non-destructive nature of the measurement, the flux inside the drum is not directly accessible and only the flux surrounding the drum can be measured. Based on the measured neutron flux outside of the drum and the determined mass fractions of the drum contents the neutron flux and distribution within the drum will be calculated by a deterministic transport code for neutrons assuming a diffusion approximation model. This newly determined flux within the drum can then be used to obtain an improved estimate for the mass fractions. This iteratively process is repeated until the convergence is reached.

II. THE NEUTRON MEASUREMENT SUBSYSTEM

As outlined above, for the normalization of the gamma signal the spatially and energy resolved neutron flux within the drum has to be known. Furthermore, only the flux outside of the drum is accessible for a direct measurement.

The largest contribution to the signal of the HPGe-detector is from the segment directly in front of the detector. Therefore the flux within this segment is the most important to know. But even with a collimator, there will always be some crosstalk from adjacent segments. This contribution will increase with rising gamma-energy, as the attenuation of the collimator and the matrix will decrease. Furthermore, segments just on the opposite side of the detector can contribute as well, especially if the matrix material is only weakly absorbing for gammas. This consideration makes clear, that the gamma signal is determined by multiple segments with different degrees of contribution. Therefore, also the neutron flux has to be determined for more than only those two segments in front of the HPGe-detectors. Considering only the adjacent segments this will result in at least 10 neutron count rates that need to be measured.

A. Selection of the detection technology

The thermal neutron flux at several possible measurement positions was determined by AiNT by a MCNP simulation to be in the range of 2×10^5 n/cm²/s to 7×10^5 n/cm²/s. In order to obtain count rates that do not result in extensive dead times, a sensitivity for thermal neutrons in the order of 0.1 cps/nv or less is desirable. Several detector technologies have been taken into account. Fission Chambers offer very small sensitivities and would be well suited. They are a proven technology for the measurement of the thermal and fast neutron flux. But they are also quite expensive and the use of in the order of 10 to 20 fission chambers would have been cost prohibitive for the QUANTOM® project. Another technique with intrinsic low sensitivity is ^{6}Li-doped glass fibers. Due to their low mass, this technology would have resulted in very little additional material in the measurement cavity and thus no increase in the background signal. Readout with position sensitive photomultiplier tubes (PMT) or multi-channel silicon photomultipliers (SiPM) might result in large channel counts with relatively low effort. But no commercially available system could be identified and the development within the QUANTOM® project was deemed too risky. The third technology that was considered is the Microstructured Semiconductor Neutron Detector from RDT Radiation Detection Technologies [2]. It uses a neutron capturing film that is deposited on a 3D-structured silicon pn junction. While it has a small form factor and is a low power, low voltage solid state device, the development of a large count readout electronic was also considered not in reach within the QUANTOM® project.

The last technology that was considered is conventional ^{3}He-tubes. These are usually designed for high neutron sensitivities in the order of 10 to 100 cps/nv, much too large for the predicted neutron flux. The lowest available sensitivity we found was a 6.6 mm x 30 mm large tube with a ^{3}He partial pressure of 10 bar and a sensitivity of 0.6 cps/nv. The manufacturer suggested reducing the ^{3}He-partial pressure to 0.5 bar which results in a simulated sensitivity of roughly 0.05 cps/nv, right in the envisioned range. This detector was then selected for the neutron measurement subsystem for QUANTOM®.

B. Detection Electronics

After the selection of ^{3}He-proportional tubes as the detection technology, a suitable frontend electronic has to be selected. As a channel count in the order of 10 to 30 was discussed, a setup with a large number of discrete preamplifiers/discriminators and counter modules was deemed too costly. Thus a system with a high integration and a large channel count was requested. One such system that is usually used as front end electronics for instrumentation at large scale neutron research facilities, e.g. for neutron powder diffractometers, could be identified. This system is manufactured by Mesytec and consists of Preamplifier/Discriminator modules and a central aggregation unit. One Preamplifier/Discriminator module can handle up to 16 ^{3}He-tubes, digitize the signals and send the multiplexed data for all channels via a high speed proprietary digital link to the aggregation unit. It is available as a NIM module or as a stand-alone variant. The aggregation unit can combine the signals of up to 8 Preamplifier/Discriminator modules and send the data via an Ethernet connection to an analysis computer. The data for each neutron signal contains the channel and module number, a timestamp with 100 ns resolution and the pulse height.

With this electronic and a custom made software, not only count rates can be measured, but also pulse height spectra and multi channel scaling data. The latter may perspectively be used to determine the neutron die away time with the pulsed mode of the neutron generator.

III. PLACEMENT OF THE NEUTRON DETECTORS

With the selection of the ^{3}He-tubes as detection technology and the Mesytec system as the front-end electronic, two important decisions remain to be met. The first one is how many ^{3}He-tubes should be used and the second one is where these tubes should be located. To answer these questions based on an objective quantity and not pure intuition, a simulation study on the influence of the number and positions of ^{3}He-tubes on the flux reconstruction has been carried out.

The idea of this study is, to calculate for all possible detector placements the count rate at the detector positions and simultaneously the flux within each segment of the waste drum for a (large) number of possible drum contents. With a simple flux reconstruction model, the flux within
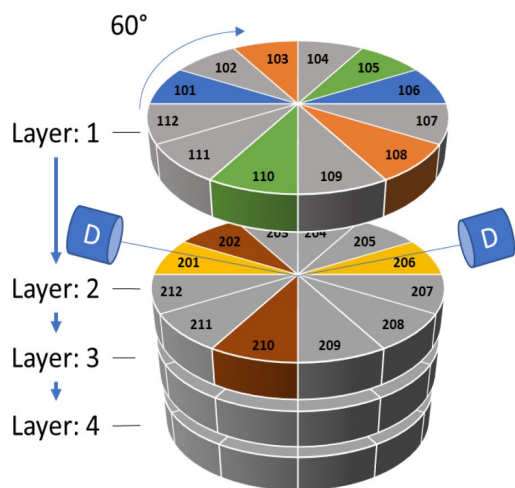
Fig. 1. Sketch of segmentation of the drum and the position of the HPGe-detectors. The drum is virtually divided in four horizontal layers of an approximate height of 20 cm. Each layer is subdivided into 12 segments. The two segments in front of the HPGe-detectors of each layer are measured simultaneously, as indicated by the color coding. The three digit segment code consists of the layer number and the serially numbered segment code. In this sketch, the neutron generator is located in the back.

each segment can be determined from the count rates and compared to the simulated flux. The detector placement can then be scored by the agreement between these two fluxes. In a first step, some constraints on the detector placements have been defined:

1) All detectors are located as close as possible to the drum, within a PTFE layer serving as the boundary of the measurement cavity. Otherwise the detectors would have to be placed inside the graphite moderator, which would make the fabrication of the graphite blocks much more difficult.

2) The detectors should be located on a grid, with 24 radial positions and vertical spacing of one half of a measurement layer. Up to three detector layers below and above the measurement plane will be considered. This results in a finite number of 24×7=168 possible detector positions.

3) The underlying symmetry of the QUANTOM® system should be preserved. There is one real mirror symmetry concerning the left and right half of the system and one approximate mirror-symmetry within the horizontal measurement plane (up and down).

4) No neutron detector should be placed directly in front of the HPGe detectors, as this presumably results in an increased gamma background from neutron captures in the ³He-tube material directly in the view of the HPGe.

These constraints result in a very large, but finite number of possible detector configurations. When looking at the segment faces, neutron detectors are either placed on the center, in the corners or on the edges.

A. Setup of the MCNP simulation

The neutron simulation were performed by the Monte-Carlo program MCNPX version 2.7.0 [3]. A detailed geometric model of the QUANTOM® system was provided by AiNT. At all 168 possible detector positions, we defined a cylindrical volume considerably larger than the envisioned

³He-tube in order to obtain a good statistic with a modest number of MCNP histories. The neutron detector signal was simulated by a F4 volume flux tally, weighted by the neutron capture cross section of ³He. The material within the probe volume is not changed from the original one, so no distortion of the neutron flux is introduced by this approach. Especially no neutrons are actually captured and removed from the simulation history. This approach was verified for several drum contents by comparing to the result obtained from a calculation with a detailed model of the selected ³He-tube. Likewise, the neutron flux within the drum segments was weighted by the radiative capture cross section of ¹H, as a prototypic (n,γ)-isotope. This value is proportional to the generated gamma source term and allows to include the complete neutron spectrum within a single number and to renounce on an arbitrary threshold for "thermal neutrons". As the average neutron flux within the drum has a very large thermal contribution, where basically all isotopes show a 1/v behavior of the cross section, the qualitative result does not depend strongly on the actual used isotope for flux weighting, except for the absolute value.

Four different materials have been used as the drum contents. Air is used as an inert empty space, polyethylene as a very strong moderating material, sodium chloride as an absorbing material and concrete as a typical matrix material. Except for air, 10%, 33% and 100% of the nominal density have been considered for the actual density. All segments of the drum were randomly filled by one of these materials. The drum was positioned, such that the second layer from the top was in front of the detector. With this setup, over 6000 different drum configurations have been simulated, each with 10^7 histories. This resulted in statistical uncertainties of 1-2% for the neutron count rates and approx. 5‰ for the weighted neutron fluxes.

To assess the flux reconstruction performance of a specific detector placement, a very simple linear model was used here: the flux within each segment is simply expressed as a linear combination of the count rates from all detector positions. Suppose on has a candidate detector placement with n detectors, F denoted the 48×N segment fluxes and C the n×N count rates, where N is the number of calculated drum configurations (N~6000), then a 48×n matrix M is searched, such that:

$$F = M \times C \qquad (1)$$

This equation can be easily solved in a least square sense by a singular-value decomposition. The mean root square of the difference of the simulated to the predicted flux is then an indicator, how good the flux in each drum segment could be reconstructed by a particular detector arrangement. Finally the results for each segment are weighted by the contribution of each segment to the total gamma signal. These weighting factors were determined for a drum filled with concrete of 33% nominal density and a gamma energy of 2223 keV (capture on hydrogen). This represents a situation, where the relatively high gamma energy results in only little absorption within the drum and a reduced collimator efficiency and ultimately in an increased cross-talk of the segments. This calculation yields a contribution
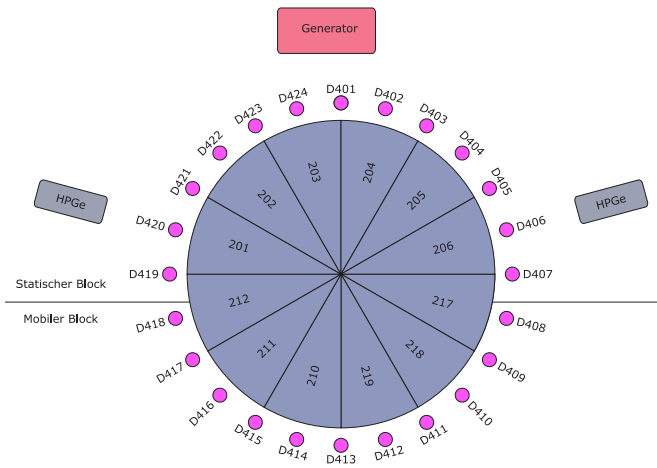
Fig. 2. Sketch of the measurement plane of the QUANTOM® device. Neutrons are produced in the neutron generator on top and generate gammas via (n,γ) reactions. These are recorded by the two HPGe detectors. All possible neutron detector positions within this plane are indicated by magenta circles. Not shown are all internal details like moderator, collimator and radiation shielding.

of roughly 25 % for the segment directly in front of the detector and 9 % each for the horizontally adjacent segments. Alternative scores could weight all segments equally or consider only two segments in front of the two HPGe-detectors. With this framework, one has a tool to compare the performance of different detector arrangements. The most important advantage of this approach is that after the time consuming MCNP simulation of the ~6000 different drum configurations, the investigation of different detector arrangements only consists of selecting the right count rates for the matrix C, calculating the matrix M and from that the score. In total 17 different configurations have been investigated in detail in an internal report for the QUANTOM® consortium, from which one has been selected for the final design. Due to space restrictions, only four different ones will be presented and discussed here.

IV. RESULTS

For the description of the detector configurations, the following nomenclature is used: Segments and detector positions are described by a 3 digit code. The first digit describes the vertical layer, 1-4 for the drum segments and 1-7 for detector positions. The last two digits enumerate the tangential position, 01 to 12 for drum segments and 01-24 for detector positions. To distinguish drum segments from detector positions, the latter is prefixed by a "D". In the actual simulation the drum is positioned in such a way, that layer 2 is in the measurement plane and the segments 201 and 206 are in front of the HPGe-detectors. The detector positions are stationary with respect to the QUANTOM® device, with the layer 4 coinciding with the measurement plane and the detector position D401 pointing towards the neutron generator. Fig. 2 shows a measurement plane with segment numbers and detector positions. Results are presented for four different detector placements, which are described in the following sections:

A. Configurations

For all configurations a short description and a sketch of the projection on a cylindrical surface is given. In these sketches from Fig. 3 to Fig. 6, the two segments highlighted in red are those directly in front of the HPGe-detectors. Blue segments denote the additionally contributing segments within the measurement plane and above and below the red segments. Detector positions at the left and right side are the same due to the circular projection.

1) All detector positions

The obvious test is using all possible detector positions for the reconstruction of the neutron flux. While this is not a feasible configuration due to the vast number of detectors, it gives the best reconstruction performance and shows the limit of this method. Fig. 3 shows a projection of the detector positions on the circumference of the drum.
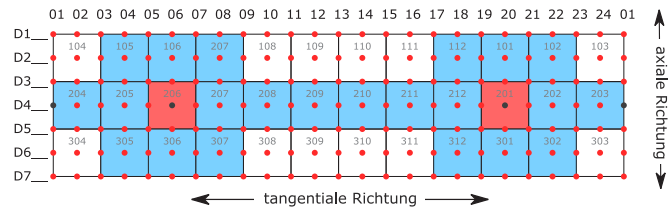

Fig. 3. Sketch of all detector position, which is also the first calculated arrangement. The results are given in Table I. The two segments highlighted in red are those in front of the HPGe-detectors. The blue segments are those within the measurement plane and above and below the red segments. Detector positions at the left and right side are the same.

2) Detectors on the centers of 24 segments

For this configuration, detectors are positioned on the segment centers of the twelve segments within the measurement plane and six each above and below this plane. In contradiction to requirement 4) detectors are placed directly in front of the HPGe-detectors. This configuration is shown in Fig. 4.
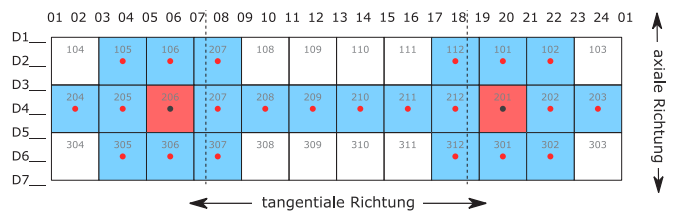

Fig. 4. Sketch of configuration 2). Detectors are positioned in the segment centers within the measurement plane and above and below the segments adjacent to the segment in front of the HPGe-detectors.

3) Constraint placement

This placement is like configuration 2), but the detectors in front of the HPGe-detectors are removed.
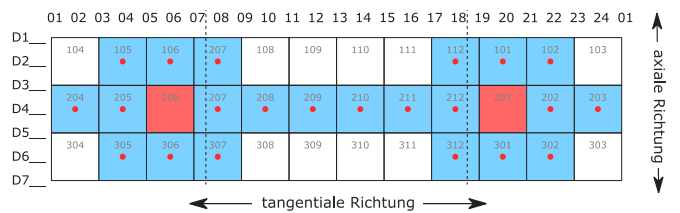

Fig. 5. Sketch of configuration 3). Detectors are positioned in the segment centers within the measurement plane and above and below the segments adjacent to the segment in front of the HPGe-detectors. The detectors in front of the HPGe-detectors are removed.

4) Final placement

For the final placement, some additional detectors compared to configuration 3) have been added, especially to

aid in the reconstruction of the most important segments in front of the HPGe-detector.
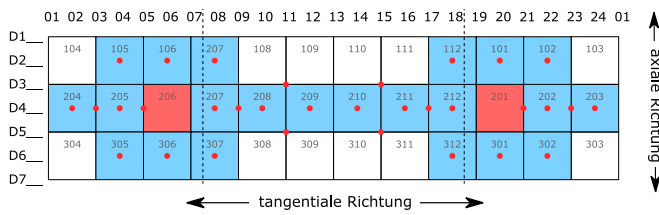

Fig. 6. Sketch of configuration 4). Detectors are positioned in the segment centers within the measurement plane and above and below the segments adjacent to the segment in front of the HPGe-detectors. For better reconstruction of the flux in the most important segments in front of the HPGE-detector additional detectors were added in the neighboring segments.

B. Scores

Table I shows the results of the four configurations. In addition to the weighted score, the non-weighted score and the score for only the two red segments, the total number of detectors and the number in the front and back block of the moderator are given.

As can be seen, the configuration with all detectors results clearly in the best score (lowest value). Removing the detectors in front of the two red segments from 2) to 3) strongly increases the value of the score, which indicates a much worse reconstruction capability. This is especially apparent for the S_{HPGe} score.

The final configuration(N° 4) results in a very good score, especially when compared to the "naïve" configuration 2).

V. Conclusions

In this work, we present the approach taken to optimize the design of the neutron measurement subsystem for the QUANTOM® device. The neutron detectors in the system have to be small, robust and of low detection efficiency for thermal neutrons. Therefore small ^{3}He-tubes have been selected and modified for lower efficiency. A simplified model of the measurement system was developed and used in a Monte-Carlo-Calculation for simulating various sets of possible detector positions around the measured waste drum. A single score value was defined to compare the quality of different sets. Taking into account also budget considerations and spatial requirements in the system an optimal design with 32 detectors was chosen. With this system, we will be able to record simultaneously the neutron count rates at a total of 32 positions surrounding the drum. These count rates will be a crucial input to the procedure that calculates the spatially resolved neutron flux inside of the waste drum. This is needed to normalize the PGNAA signal for the determination of accurate mass fractions for non-radioactive hazardous substances in radioactive waste drums.

VI. References

[1] A. Havenith *et al.,* "QUANTOM® - Non-destructive scanning of radioactive waste packages for material characterization," *EPJ Web of Conferences*, vol. 2019.

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TABLE I
RESULTS FOR DIFFERENT DETECTOR ARRANGEMENTS

No.	N	N_F	N_B	S_w	S_{all}	S_{HPGe}
1	168	91	77	1.68	1.68	1.53
2	24	14	10	3.69	3.81	2.13
3	22	12	10	4.77	4.56	8.20
4	32	16	16	2.01	2.05	1.30

Results for the four described detector configurations. The number of total detectors is N, the numbers in the front and back part N_F and N_B, respectively. The scores S_w and S_{all} denote those with and without weighting, respectively. S_{HPGe} is the score that only considers the two segments in front of the HPGe detectors.

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