Characterization of Solid Building Structures with NaI Gamma Spectroscopy

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ASTRACT

This paper presents an in-situ gamma spectroscopy measurement setup, which utilizes a NaI detector for clearance measurements of concrete building structures. As such an apparatus can be operated at room temperature, large and costly supporting accessories are not required. This is a major improvement in comparison to existing approaches that work with semiconductor technology, e.g., high pure germanium detectors. The method under discussion allows to create versatile and handy measurement systems, which lower cost and time efforts, required for characterization measurements during the disassembly of nuclear power plants, considerably.

This novel characterization method has been developed jointly by E.ON and the University Rostock to foster the dismantling activities of E.ON nuclear power plants. The regulatory acceptance for this method has been granted for the facility Nuclear Power Plant Isar (KKI) in July 2013. This paper details the method under discussion and how an acceptance has been reached, according to applicable legislation. Furthermore, a comparison with state of the art characterization methods plus experiences from the practical application of the method will be shown.

During the decommissioning process of a typical nuclear power plant, roughly 75.000 m² of concrete structures need to be processed. This task starts with the radiological assessment. Once categorized, the further treatment and disposal of the structures under investigation will be chosen in accordance to the applicable national laws and regulations. Therefore, a characterization method, which is efficient in time and material demands, as well as reliable and accurate at the same time is needed. Meeting this requirement is a major contributor to cost and time efficient dismantling activities.

Manual contamination monitors are established as common procedure to assess surface contamination. However, while being reliable and accurate, their application tends to emerge as time consuming task that requires a high amount of manually executed labor for larger structures like room walls or concrete slabs.

As alternative, characterization methods based on in-situ gamma spectroscopy are considered as a more efficient method to determine the contamination of large and plain structures. To enable the generation of the required impulse height spectra, required for this technique, two different detectors types exist: The nuclide specific verification can be performed with semiconductor technology or alternatively through utilization of a scintillation counter method. While both technologies enable operators to assess larger areas than contamination monitors with each single measurement, the underlying technologies come with different advantages, disadvantages and restrictions, which make them more or less fitting for different application scenarios.

Utilizing In-Situ Gamma Spectroscopy for the Characterization of Building Structures

Using semiconductors for in-situ gamma spectroscopy is a widely established method, in use within disassembly projects of nuclear facilities since more than 20 years. It is majorly used for release measurements of floors, walls, complete rooms and other plain surfaces. Also, release measurements of barrels, filled with different materials, constitute an established scenario, where gamma spectroscopy comes to use. Setups for these fields of application typically utilize a semiconductor-based detector, made of high purity germanium, to transform the photons.

The major advantage of the semiconductor technology is its superior radio-nuclide energy resolution in comparison to scintillation counter methods, as depicted in Figure 1a for the radio nuclide Co-60. The major disadvantage on the other hand, besides the relatively small response capability for photons, is the occurrence of dark currents, triggered by thermal influences. To suppress these dark currents, the detector needs to be held on a very low temperature level constantly. Hence, germanium detector based setups will only operate accurately at temperatures around 77 Kelvin. Cooling them with liquid Nitrogen or other advanced cooling technologies is required. Due to these reasons, germanium detector setups are both, bulky and cost intensive for characterization activities [3]. A setup using this measurement method has been used for example in the German Nuclear Power Plant Stade (KKS) for release measurements. However, using the same measurement setup to assess the contamination of building structures to determine required decontamination steps is not ideal, as it would be way to expensive.

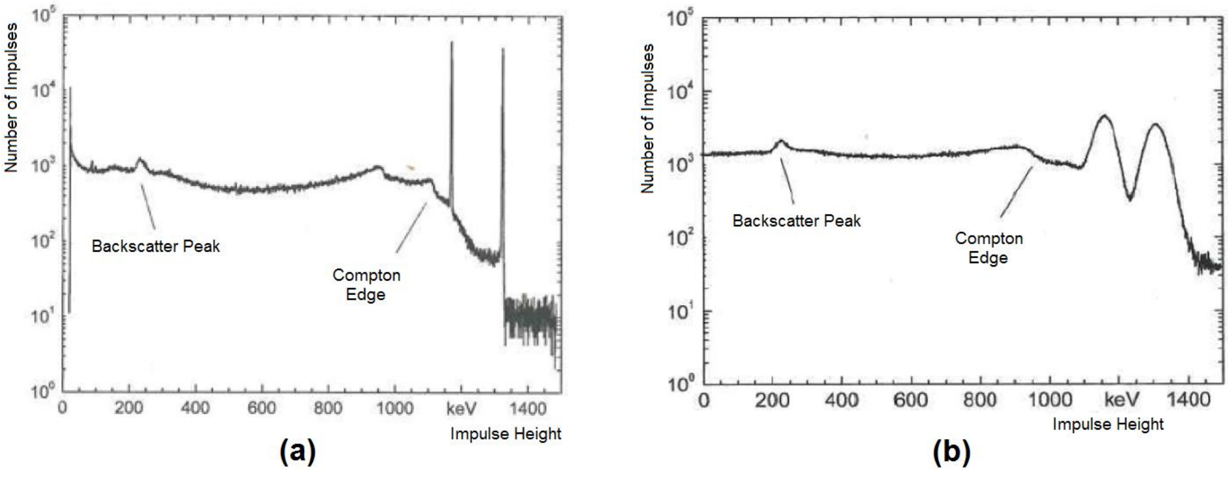


Figure 1: *Pulse height spectra of the gamma radiation of Co-60; (a): Measurement with coaxial semiconductor detector - high pure germanium, diameter: 62mm, length: 75mm; (b): Measurement with NaI crystal, diameter: 50.8mm, length: 50.8mm*

As alternative, a measurement setup employing scintillation counter technology can be used. A first and most obvious advantage in comparison to semiconductor based methods is that no cooling of the detector is required. Additionally, this method comes with a significantly higher detection probability. This allows for the utilization of a considerably smaller crystal and consequently also a smaller shielding to reach the same efficiency as with semiconductor technology. Hence, our aim was making this technology applicable for orientation measurements on large mineral concrete structures to determine required decontamination activities. For this attempt, we relied on a NaI detector based setup. Figure 1b shows the according impulse spectra for Co-60, as measureable with this detector type.

Major drawback of this approach is its inferior energy resolution capability in comparison to a gamma spectroscopy based setup, using a germanium detector. The relatively large peak width of our method might result in scenarios, where peaks in close proximity to each other might not be resolved as two different peaks. The severity of the adverse influence, peak widening will have to the results that can be achieved, depends mainly on the nuclides to detect. Considering the German Nuclear Power Plant Stade as example, no serious influence is expected with its key nuclide Cs-137. In other environments, where Co-60 constitutes the key nuclide, measurements will be more prone to be exposed to this adverse effect. The reason is that one of the two peaks of Co-60, specifically the one with the lower energy level, lies already close to the compton continuum of K-40. Here, peak widening might result in resolution problems. Additionally, it is to mention that an adverse influence of the background noise through naturally occurring radionuclides of the concrete structure under inspection might occur. The gravity of this effect will depend on the actual individual energy situation and the activity of the object under inspection.

Clearance of Radioactive Substances According to German Regulations

The current state of the art knows two different strategies to release buildings from controlled nuclear areas: First option is to break the structures up into smaller pieces and to measure them, second option is to assess the structures in their original state. Obviously, the later one of the two strategies lies in the focus of this paper. The following chapter addresses how this can be achieved in accordance to the existing rules and regulations in effect for a nuclear facility placed on German territory.

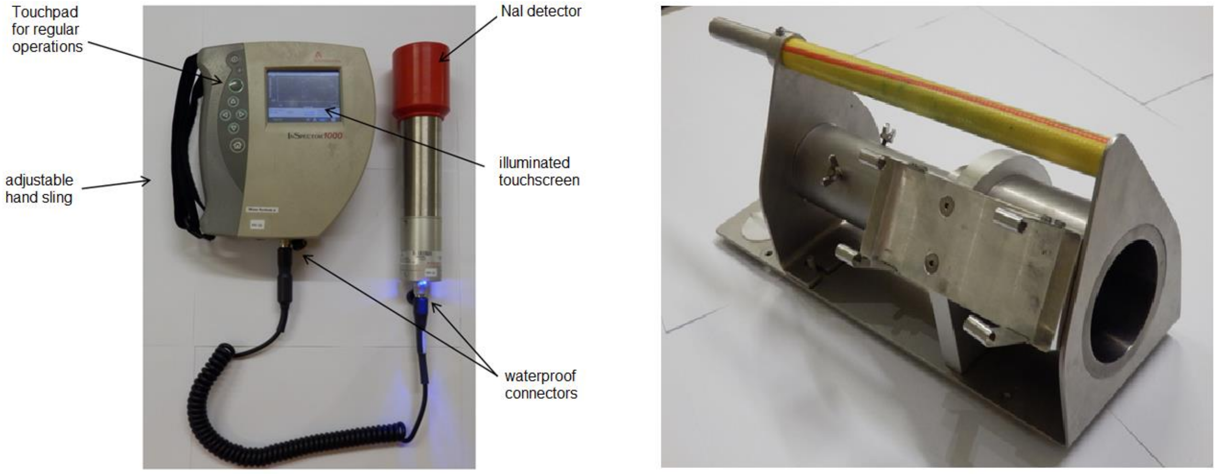
For nuclear power plants in Germany, the clearance of radioactive substances is regulated by the German Nuclear Power Act (Deutsches Atomgesetz, AtG) and the German Radiation Protection Order (Strahlenschutzverordnung, StrlSchV). According to §3 StrlSchV, the clearance of radioactive substances in accordance to §29 StrlSchV is considered as an administrative measure to release substances, parts of facilities, soil, tools, etc., which may be activated or contaminated due to their utilization during the use of nuclear substances or ionizing radiation, from the ruling scope of the German Nuclear Law. The consequence is that objects that have been cleared or released are no longer considered as radioactive. Hence, waste, which is generated during the disassembly of a nuclear power plant, will be in the scope of German waste disposal laws, as soon as the clearance according §29 StrlSchV has been performed, but no longer in the scope of nuclear legislation.

To release an object according to §29 Para. 2 StrlSchV, it needs to be secured that no single person will be exposed to an effective dose higher than 10 Microsievert. To ensure this, the StrlSchV defines values of permitted activities per radio nuclide (Appendix III) and additional boundary constraints (Appendix IV). If an object confirms to these permitted maximum activities and the constraints, it can be released according to §29 StrlSchV. If more than one radio nuclide is present, the sum formula according to Appendix IV Part A StrlSchV, as disclosed as Formula 1, needs to be applied. It basically defines that the sum of the ratios between the specific activity (C_i) and the according permitted activity for this radio nuclide according to StrlSchV (R_i) needs to be built, taking into account all present radio nuclides (i). This sum has to be smaller than 1, to allow for a release of this object from nuclear legislation [6].

$$\sum \frac{C_i}{R_i} < 1 \tag{1}$$

To release building structures for demolition, some conditions need to be considered: In general, the measurement shall be performed on the building, before it is disassembled. Both, penetrated activity and activity on the surface, need to be projected to the surface of the structure under investigation to determine its total activity as relevant for the release according to §29 StrlSchV. Hence, the state of the object to measure has major influence on the selection of a fitting measurement method. An area of up to 1 m² can be used as area to build an average from. Deviations from these conditions are allowed if accepted by the according administrative authority. After the structure has been released from the nuclear legislation, its demolition waste does not require to undergo any special treatment before it is disposed. However, the part of the legislation that builds the foundation of the method discussed, makes it mandatory that the structures that have been released will be destroyed and shredded into fist-sized pieces.

The method discussed in this paper aims specifically on scenarios as described above, i.e., release of large building structures and objects with plain concrete surfaces for demolition. Due to the fact that radio nuclides we intend to detect with this method need to have a relatively high maximum permitted activity, the method fits not for all environments. In the environments, where we worked successfully with this setup, Cs-137 or Co-60 had been the key nuclides. Both of them have a sufficient high maximum permitted activity according to the StrlSchV (Co-60: 3E4 Bq / m², Cs-137: 1E5 Bq / m²).



(a) (b)

Figure 2: Measurement system as used in the KKI; (a): InSpector™ 1000, 2“ x 2“temperature stabilized IPROS-2 NaI detector; (b): Tungsten – collimator, 1cm thickness

The Employed Measurement Setup, Utilizing a NaI Detector

This chapter will provide a detailed overview of the measurement setup, as it comes to use in the German Nuclear Power Plant Isar. It bases on the initial setup as disclosed in [4] and has been further refined and improved.

As measuring device, the InSpector™ 1000 from Canberra® Industries come to use, see Figure 2a. The system allows to connect different probes to identify different radio nuclides with the help of spectrometric analysis [1]. This versatility allows to utilize the instrument for a multitude of applications, e.g., the detection of radiation sources with simultaneous nuclide identification or measurement of dose rates just shy of the background level. The complete system is relatively small and light which makes it handy and therefore fitting for the intended measurement setup.

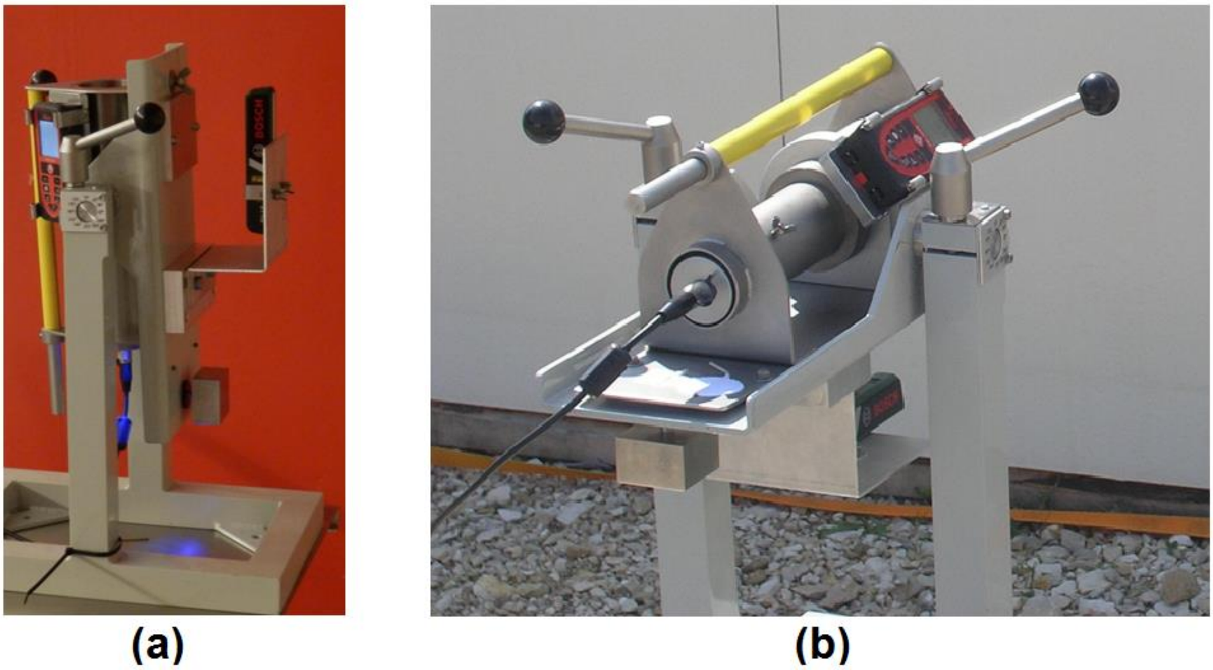


Figure 3: Measurement system as used in the KKI, probe inserted into the collimator, complete with handling frame, laser range finder and laser pointer

A characterized NaI detector is utilized as probe in this measurement setup. As detailed before, contrary to a semiconductor detector based system, this probe can be plugged to the InSpector™ 1000 and used without any cooling system, as the measurement system roots on the scintillation counter technology. The probe comes with an integrated NaI crystal in the dimension of 2 by 2 inches. Prior to its application in this setup, it had to be characterized by Canberra®, to allow the utilization of a calibration software.

The development of such a characterization is based on a Monte Carlo model. The process consists of three steps: First, a Monte Carlo N particle (MCNP) model for the detector under characterization needs to be developed and validated. Second, a large number of data regarding the effectiveness and discrimination behavior of the validated MCNP detector model to punctual radiation sources, distributed around the detector, need to be generated. Finally, a characterization needs to be derived from the data gathered and the validated characterization model. The result is a detector-parameter-file, which can be utilized by the ISOCS calibration software [5].

The utilized detector needs to be shielded against radiation from behind and from the side. This will define the viewing direction of the measurement system [2]. This aim will be reached by the use of a collimator, as depicted in Figure 2b. The opening angle of the collimator determines the distance between the detector and the surface to inspect. This topic will be treated in more detail in the chapter, which details the qualification measurements.

To foster efficiency during the operation of the measurement system, it has been completed with a handling frame, a laser range finder and a laser pointer to aim for the target, see Figure 3. The frame with all mounted devices has a total weight of 16kg and a dimensioning of 51cm x 26cm x 46cm. On top comes the weight and bulk of the InSpector™ 1000 (1kg) and an off the shelf Laptop for operation, which places the complete system on a scale below 20kg.

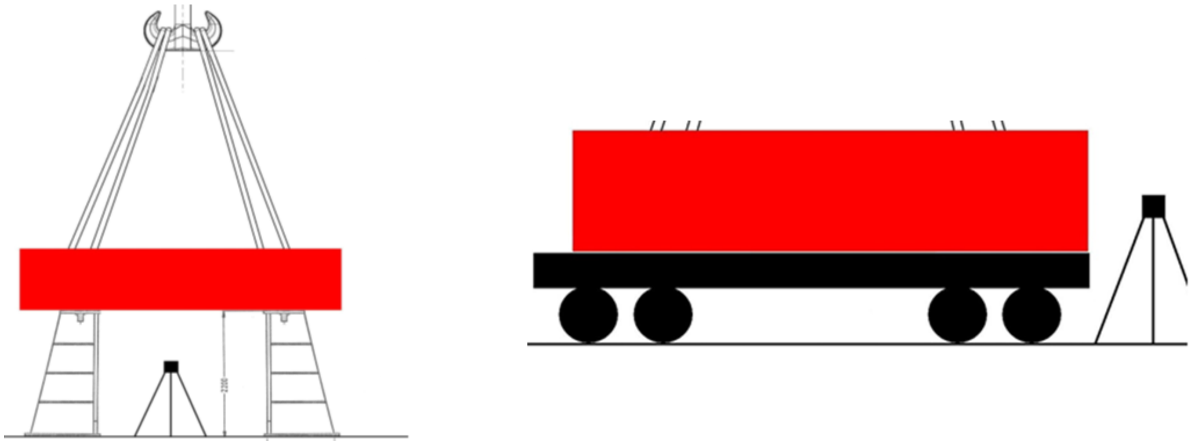


Figure 4: Measurement setups as used in the KKI, schematic display of feasible positioning for the detector

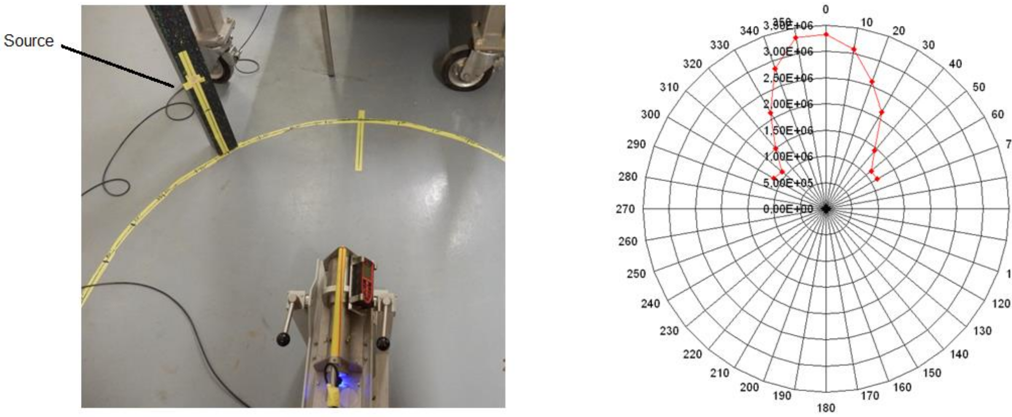
As it can be seen in Figure 4, the small scale of the complete measuring system in combination with negligible infrastructural requirements – not even electric power is required, as the system runs on battery power for 8 hours – allows a multitude of versatile measurement scenarios.

Measurement Series for Qualification and Commissioning of the Setup

To allow the utilization of novel methods to assess the activity of objects within a German nuclear power plant, like the method under discussion in this paper, a formal qualification act needs to be performed. For this purpose, the nuclear facility, which intends to use a new method, files a formal request to its responsible ministry. This request contains a detailed description of the measurement method, the purpose it shall be utilized for and a proposal for a commissioning program. The ministry reviews the request, may formulate additional conditions, restrictions and test procedures, and finally agrees to a proposal eventually. Consecutively, the ministry supervises the execution of the commissioning procedure and accepts the new measurement method. This chapter describes the measurement series which had to be performed by the KKI as part of the commissioning program, prior to the acceptance of this new method for clearance measurements according to §29 StrlSchV.

The aim of the first measurement series is to determine angular dependence on a circular arc for a measurement setup with attached collimator. This series shall verify the efficiency of the collimator employed as absorbing device and will also provide an optical representation of the angular dependence of the setup regarding yield and measurement errors. As can be seen in Figure 5a, the measurement setup will be installed statically, while the activity source is moved around the center of the setup on a circular arc. As Figure 5b discloses, it has been shown that, under the constraint of a constant calibration, the measured activity for the same sample diminishes with its increasing angular displacement out of the detector axis. This proves the effectiveness of the collimator, incorporated into the setup.

The second measurement series was devised to determine the opening angle of the collimator. In order to use the measurement method for clearance measurements, it needs to be assured that the relative detection efficiency on the outermost point of the area under inspection is at least 30%. Hence, the outermost point of angular displacement, where the detection efficiency lies still above 30%, has been determined with a measurement series, as shown in Figure 6a. For the setup under inspection, this collimator opening angle lies at 74°.

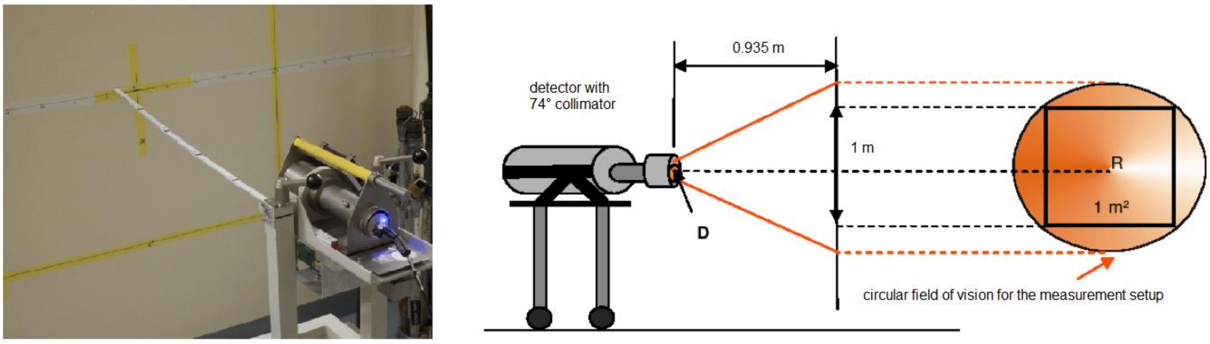


(a) **(b)**

Figure 5: Determination of the angular dependency of the measurement setup with collimator; (a): measurement setup; (b): activity calculation for Co-60, according to DIN ISO 11929

As depicted in Figure 6b, the ideal distance of the detector to the area to measure can be derived from the opening angle of the collimator: The measuring area is sized at 1m², which means 1m x 1m. The diagonal of a 1m x 1m square is 141cm long, which will be the diameter of the circular field of vision, the detector needs to have at the point of measurement. This translates to an ideal distance of 93.5 cm between the end of the collimator and the object under measurement.

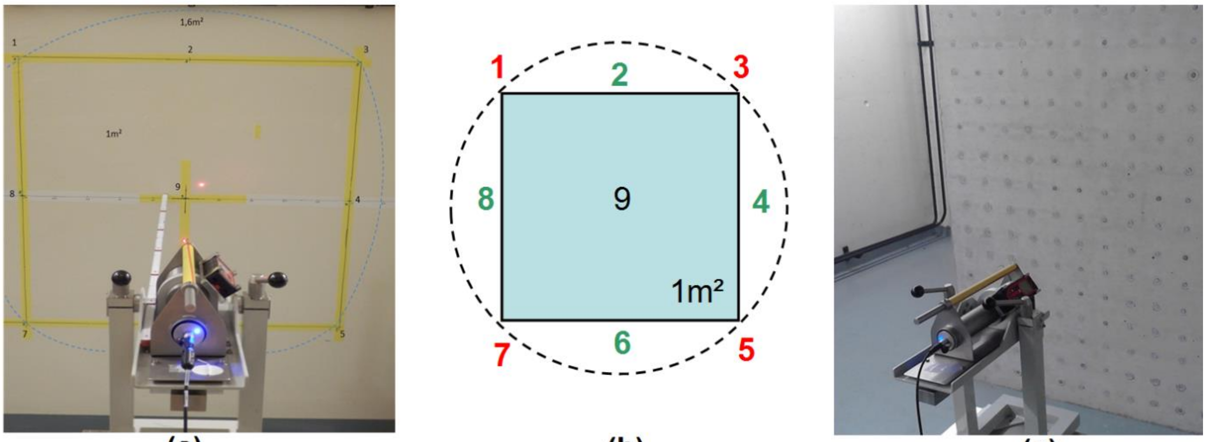
The third measurement series consists of rectangular measurements, see Figure 7a. The aim is to calculate the potential error occurring, if the activity is not distributed evenly on the area under investigation. This is considered as hot spot activity. Therefore, different sources are placed at the spots 1 to 9 of the measurement range, as shown in Figure 7b. Afterwards, measurements with the real calibration geometry for this specific case and with the homogeneous geometry as it is intended for the measurements under real conditions are performed.



(a) **(b)**

Figure 6: Determination of the opening angle of the collimator; (a): measurement setup; (b): schematic view

The trial proved that even with the activity sources placed on the outermost points of the measurement area (1, 3, 5, and 7) and the homogenous geometry as it is intended for real measurements applied, the activity value determined by the measurement system lies above the real activity value present. Hence, the homogeneous geometry can also be utilized if an adverse hot spot activation might be present on the object to asses.



(a) (b) (c)

Figure 7: *Rectangular measurement to calculate potential flaws if hot spot activity is present (a): measurement setup; (b): schematic view; (c): validation measurement - practical measurement at the K-risk phantom wall*

The forth measurement series consists of zero effect measurements to determine the detection limit of the measurement setup. Within this series, it needs to be proven that the detection limit lies significant below the permitted activity as defined in Appendix III of the StrlSchV. As the detection limit (LOD) is dependent on the measurement duration, this series serves also to determine the required measurement time. The measurement series proved that a measurement duration of 300 seconds is sufficient to use this method in the KKI. With this duration, the detection limit for Co-60, the lead nuclide within this environment, lies at 1.1E4 Bq. This is less than half the activity, as permitted by the StrlSchV, i.e., 3E4 Bq.

The final measurement series serves as validation of the method under inspection. Therefore, the operator had do perform measurements with the system on a wall with several different contamination scenarios. During this measurements, the operator was not aware of the real contamination setup. The test was passed, when contamination scenarios had been classified correctly as able or unable for clearance according to §29 StrlSchV by the operator.

After all measurement series had been performed, the method was finally commissioned and is now actively used in the Nuclear Power Plant Isar. However, some additional constraints have been determined by German authorities: The measurements need to be performed in a grid, where the single measurement overlap each other by 10cm. Although a homogeneous surface contamination is expected at the objects, where the method comes to use, a conservative calibration geometry has to be used, which assumes activity 1 cm below the concrete surface with a homogeneous distribution.

During the calendar years 2013 and 2014, the KKI performed clearance measurements with this method on a large scale. In total, over 1.600 tons of concrete structures with a total surface area of more than 3.200 m² have been successfully cleared during these two years, see Figure 8. Permanent control measurements and regular inspections by German authorities have proven that the reached results are accurate and reliable.

Conclusion and Outlook

Within this paper, a fully qualified measurement system for in-situ gamma spectroscopy, utilizing a NaI detector, has been presented. Compared with established measurement systems, based on semiconductor technologies, the novel method represents a major improvement in terms of weight, bulkiness, and handling complexity.

Figure 8: Application of the measurement method in the German Nuclear Power Plant Isar (KKI)
(a): measurement setup; (b): material after clearance measurement

Under the right conditions and within the right application area, as the ones considered within this paper, the presented method proved its superiority in terms of costs, time consumption and personnel requirements not only on a theoretical level, but also under real world conditions within a nuclear power plant.

For the future, we intend to participate in regular comparison measurements, as the one arranged by the German University Regensburg on a bi-annual basis. This would provide us with more comparison possibilities of the method introduced with concurring approaches. As a result, we expect to identify additional scenarios, where our method can support nuclear facilities in their regular nuclear protection activities or during dismantling projects.

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