Experimental study of concrete activation in MCNP safety simulations for neutron sources

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Abstract

Radiation shielding properties, characteristics and simulation of concretes are well-studied issues, unlike the dose consequences of their activation by incident radiations. A detailed work in this field was carried out for the European Spallation Source (ESS) ERIC on samples of three types of concrete: the newly developed PE-B4C-concrete, its Reference concrete – from which it was developed – and a general concrete produced by Skanska company. Neutron irradiation experiments were performed in the Budapest Research Reactor. PE-B4C-concrete showed significantly smaller total activity concentration than the Reference concrete. Then composition measurements were performed with different analytical techniques (Prompt Gamma Activation Analysis, Neutron Activation Analysis, energy dispersive X-ray fluorescence analysis), and on their basis, detailed MCNP concrete material cards were generated and validated by the reproduction of measured data with the combination of MCNPX and Cinder90 codes. Finally, as an application of the created recommended material

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cards, a realistic irradiation scenario of the ESS bunker was simulated. Decay gamma dose rates of concrete activation were found to be 29-72% higher with the use of the measurement-based input compositions, compared to the generic ones, highlighting the importance of correct determination of trace elements in materials in relation to safety planning.

Keywords: PE-B4C-concrete, neutron activation, Monte Carlo, MCNP, neutron shielding

1. Introduction

The European Spallation Source (ESS) ERIC [1] – which is now under construction in Lund, Sweden – will be the brightest neutron source of the world [2]. Due to the high neutron fluxes with wide variety of energies in the bunker, the different neutron guides and the experimental caves, an extensive and detailed safety planning is essential for all the components of the facility exposed to radiation. Moreover, special focus should be paid on the newly developed materials for applying in radiation protection. One of these is the so-called ‘PE-B4C-concrete’ [3]. This concrete has been developed at ESS from an ordinary concrete, – in the following referred to as ‘Reference concrete’, – by replacing a certain part of the pebble-content with B\(_4\)C grains and polyethylene beads, giving in total \(~10\) w\% of the new mixture (0.76% and 10.2%, respectively).

In the PE-B4C-concrete the hydrogen content of the added polyethylene slows down the neutrons more effectively than the water content of other concretes. These lower energy neutrons are then effectively absorbed by the natural \(^{10}\)B content of B\(_4\)C. The shielding properties of this concrete [4] and other concretes are well-investigated [5; 6] and simulated with different Monte Carlo codes [7]. The the effects of activation products on nuclear safety is also known, although less extensively studied [8; 10]. Most of the concrete compositions that are currently available for simulation purposes are limited to the major components of the concrete (i.e. the nominal composition given by the manufacturer). This indeed determines the shielding properties, but trace elements are expected to
be the dominant sources of neutron induced radiation, because of their significant neutron activation [11]. The published studies in shielding material activation mostly focus on long-lived radioisotopes for radioactive waste management considerations, highlighting the importance primarily of cobalt and europium content of concretes [12]. However, the neutron activated materials may have short-term effects as well in terms of occupational exposure. As an example, maintenance of neutron scattering instruments and beamlines usually performed with a few days of cooling after closing the beam, when the dose consequences of decay gamma radiation of the activated shielding and other instrument components should decrease below the pertinent constraint.

The two-fold aim of the current study is to determine the neutron activation properties of the newly developed PE-B4C-concrete in comparison with other common shielding concretes and to facilitate more realistic simulation of concrete activation in Monte Carlo modelling.

For this purpose, three types of concrete samples were studied: the above introduced PE-B4C-concrete and its Reference concrete, and a general concrete produced by Skanska company [13] – referred to as ‘Skanska concrete’ in the followings –, that is currently used in ESS. Neutron irradiation experiments were performed in the vertical irradiation channels of the Budapest Research Reactor (BRR) [14] with two different neutron spectra, determining the activation properties of all these concretes. In order to provide detailed data for realistic activation calculations, the initial composition of concrete samples were measured with different analytical techniques: Prompt Gamma Activation Analysis (PGAA) [15] was used along with Neutron Activation Analysis (NAA) [16] in the BRR as a first approach. The combination of these analytical methods are practically suitable for the whole periodic table. However, the high cost and and difficulties with implementation do not make it possible to apply neutron activation techniques for continuous quality assurance measurements during construction. To examine a feasible alternative, widely used energy dispersive X-ray fluorescence (EDXRF) [17] measurements were also performed. The impact of the initial concrete composition on activation simulations was also studied.
via reproducing the performed neutron irradiation measurements with various
measured and nominal concrete compositions jointly using MCNPX [18] with
Cinder90 [19], similarly to MCNPX tests in MEDINA facility [20]. The ef-
fact of concrete activation on dose consequences of decay gamma radiation was
also demonstrated by the simulation of a realistic maintenance scenario in the
ESS bunker. With this study, the importance of trace elements in safety cal-
culations is highlighted, and a generally applicable method is presented for the
development of simplified but realistic input compositions for neutron activation
simulation purposes.

2. Methodology

Three types of concrete samples: PE-B4C-concrete, its Reference concrete
and Skanska concrete were examined in the current study. The samples were
received as grist. Elemental compositions of the samples were measured with
NAA, PGAA and EDXRF analytical techniques. During the NAA experi-
ments, activation properties of the samples were also determined. Based on
these experiments, simplified but realistic concrete compositions were deter-
moved and validated for activation simulation purposes, specifically in ‘material
card’ format for MCNP simulations. Finally, an ESS bunker case study was
performed to demonstrate the applicability and importance of the developed
material cards in safety planning. The main focus of the current study is on
the decay gamma emission of the irradiated shielding concretes, and its poten-
tial dose consequences in terms of occupational exposure. For this reason, –
considering the high self-attenuation properties of the concrete for alpha- and
beta-radiation, – only the contribution of gamma-emitting activation products
of the concretes are discussed in the following.

2.1. NAA measurements

For the NAA analysis ~0.1 g samples of each concrete were filled to high-
purity quartz ampoules, measured by a Mettler-Toledo XPE 26 microbalance
having <0.7 µg reproducibility. Two sets of samples were prepared and encapsulated in 10 cm long aluminum tubes and irradiated in the water-filled, vertical irradiation channels of the BRR [14]. The first set was irradiated in a rotated, well-thermalized irradiation channel (referred to later on as ‘Thermalized channel’) located in the beryllium reflector around the reactor core. The second set was placed into an irradiation channel in the reactor core, with higher fast/thermal neutron flux ratio (referred to in the followings as ‘Fast channel’). The irradiation time was 2 hours for both cases. Neutron flux parameters during the irradiation were measured with Bare Triple-Monitor method [21] and are presented in Table 1.

Table 1: Measured neutron fluxes in the vertical irradiation channels of BRR [14]. (The uncertainties are 15-20%.)

| Flux Type            | Thermalized channel | Fast channel |
|----------------------|---------------------|--------------|
| Thermal flux [cm²/s] | 2.00E+13            | 5.00E+13     |
| Epithermal flux [cm²/s] | 4.30E+11           | 3.80E+12     |
| Fast flux [cm²/s]    | 1.30E+12            | 4.70E+13     |

The activities of the irradiated samples were determined by measuring their gamma response with a Canberra HPGe detector (p-type detector, 36% relative efficiency, 1.75 keV/1332.5 keV resolution connected to Ortec 502 MCA-2) in a low background measurement chamber. The first measurements were performed after 4 days of cooling due to the high initial activity of the samples. Samples were measured 5 times in a 3-week long time period, to determine their decay characteristics. The measurement time increased from 10 minutes to 2 hours in this period. Gamma spectra were evaluated with Hypermet PC [16] and KayZero for Windows 3.06 programs [22] resulting elemental compositions.

The measured gamma spectra were not only used for elemental composition analysis, but also to characterise the activation properties of the concrete samples, especially comparing the activation and decay profiles of the PE-B4C-concrete with that of the Reference concrete. In addition, in order to facilitate the comparison of the measured results with simulated activity concentrations too, – as Cinder90 output tends to give an overflowed, hardly searchable
inventory—‘key isotopes’ were identified in the measured spectra, giving the majority of the total activity in all the measured and extrapolated activities. All parent elements of these key isotopes were also identified, and were in the focus of the composition measurements.

2.2. PGAA measurements

Prompt gamma activation analysis was performed on all three types of concrete samples at the PGAA Experimental Station of the BRR [15]. In the experiment 6 g samples of each concrete was irradiated with a 20 × 20 mm collimated cold neutron beam. The emitted gamma-rays were detected with HPGe detector surrounded by a bismuth germanate (BGO) scintillator detector. Signals were processed and evaluated with a Canberra AIM 556A multichannel analyser and Hypermet-PC gamma software [23]. Element identification was performed with the ProSpeRo program, utilising the prompt gamma analysis libraries from Molnar et al. [24]. In this PGAA station 16 elements could be measured: H, C, Na, Al, Si, S, K, Ca, Ti, Mn, Fe, B, Cl, V, Sm and Gd.

2.3. EDXRF measurements

In addition to the above described methods, as a relatively fast and low-cost technique, energy-dispersive X-ray fluorescence (EDXRF) [17] spectroscopy was also carried out on the concrete samples for elemental analysis in the Nuclear Science and Instrumentation Laboratory of the IAEA [25].

To increase the homogeneity of the grist samples, they were ground with a ball grinder using tungsten carbide (WC) balls and mortars. The material used for grinding was considered during evaluation of elemental concentrations. After the addition of 0.25 g wax to each sample, they were pressed to 2.5 g pellets. Three samples were prepared from all of the concretes, and all samples were measured with an Epsilon 5 (PANalytical, The Netherlands) triaxial polarising EDXRF device [26]. The instrument is equipped with a 100-kV X-ray tube, a series of secondary targets and a HPGe detector with high efficiency for hard X-rays. Each sample was measured under ten different conditions using
secondary targets suitable for different groups of elements. Concentrations were calculated using the built-in software based on all ten spectra for each sample. Calibration curves generated from measurements of several geological certified reference materials were used for the concrete samples. Using several secondary targets is advantageous since it allows a full elemental analysis of the samples for elements from Na to U, considering major elements as present in the common oxide forms \[27\]. The condition using a 100-keV excitation and a Barkla polarising secondary target makes it possible to quantify lanthanide elements based on their K lines. EDXRF in general has a high dynamic range covering major to trace elements.

The composition of the so-called dark matrix was considered from the nominal composition of the samples, given by the manufacturer. Preliminary results of PE-B4C and Reference concretes have already been published elsewhere \[28\].

2.4. Simulations of concrete activation measurements

MCNPX neutron activation simulations were performed in order to study the impact of the concrete composition on the produced activity. For this purpose, the neutron irradiation experiments (see Section \[2.1\]) were reproduced via simulations. The constructed geometry consisted of the sample and the aluminium sample holder (see Figure 1) thus placed in a \(20 \times 20 \times 20\) cm cube, representing a homogeneous volumetric neutron source.

![Irradiation geometry in MCNPX simulations with blue aluminium sample holder, purple sample and white void in between.](image)

The experiments in both the Fast and the Thermalized channels were reproduced in simulations, with their typical neutron spectra which are presented in
Figure 2. The default spectral distributions of the channels were adjusted to the current measurements by scaling them with the measured flux parameters presented in Table 1.

Simulations were performed with different initial compositions (i.e. ‘material cards’) for all three concretes: i) the nominal composition, having similar level of details as the currently available MCNP concrete material cards, e.g. the Los Alamos (MCNP) material card [11], ii) the composition obtained from EDXRF and iii) the composition based on the NAA and PGAA measurements. The results of the PGAA and the NAA methods were considered in combination, as they are practically complementary methods. The measurement-based compositions involved all elements indicated in the nominal composition, extended with the parent elements of the key radioisotopes. Nominal values were applied for non-detectable bulk elements.

Irradiation conditions were reproduced using MCNPX [18] and Cinder90 [19] codes. Activity concentrations were calculated for the end of the respective irradiation time (t₀) and also for the times of the activity measurements. Considering radioactive waste production, the activity concentrations of the samples
were determined for 1 year- and 5 years of cooling as well. The simulated activity concentrations were compared to the measured ones for the whole described time period, where the measured activities were extrapolated to \( t_0 \) and 1 y and 5 y cooling times. While the focus of the current study is on the gamma-emitting isotopes, Cinder provides a detailed but indiscriminate inventory. For the fast and easy comparison of the measured and simulated data, the total simulated activity concentrations were approximated as the sum of the activities of the key isotopes, described in Section 2.1.

On the basis of the measured compositions and the activity simulations, combined input compositions were constructed for the PE-B4C and the Reference concrete that sufficiently reproduced the measured activities, and therefore recommended for further activation simulations for e.g. radiation safety planning. These compositions consist of two kinds of elements: major elements indicated in the nominal compositions and the parent elements of the key isotopes. For each element a measured value was preferred if it was available, otherwise the nominal one was used. As (in this unique case) the EDXRF analyses were performed with the most number of samples, the EDXRF produced values were favoured as default against other measured results, considering the better statistics. However, NAA produced values were considered for elements in very low amount \( 10^{-4} \text{ w\%} \), as in this case NAA provided lower detection limit than EDXRF giving presumably more precise values. In addition, when the different methods provided conflicting results – mostly due to the general inhomogeneity of the samples –, the more conservative values were selected. Finally, as the oxygen content varies significantly in the concrete, it was chosen to assume that the mass fraction unaccounted for by the above, consisted of oxygen. The material compositions created this way, i.e. the iv) recommended compositions were validated via the above described MCNPX simulations.

2.5. Dosimmetrical simulations in the ESS bunker

A set of simulations was also performed in order to evaluate the impact of realistic concrete composition from dosimetric aspects. Decay gamma dose
rate coming from the irradiated concrete shielding was determined with nominal and measurement-based recommended compositions. The West Hall of the ESS’s bunker was selected as a test case, as described in [29]. For this purpose, a detailed MCNP model of the bunker complex was developed, including the penetration through the monolith and guide segment corresponding to the BIFROST instrument, as shown in Figure 3. The ESS bunker is planned to be built from two types of concrete: a heavy concrete for the radial walls and the roof, and a normal concrete for the axial wall. For the similarity with the real case of the ESS design, the Reference concrete was applied in the model in the axial walls. The neutron activation process and generated decay gamma dose rates were modelled for only this single wall, eliminating the additional decay gamma background originating from the heavy concrete walls.

Figure 3: Overview of the MCNP model of the bunker complex. 1-9: 50 cm radius spheres along the neutron guide of the BIFROST instrument, marked areas for numerical comparison of decay gamma dose rates.

The simulations were performed with the combination of MCNPX and Cinder90 code in a realistic operational scenario of the ESS: 10 years of operation on average power (0.616 × 5 MW) and 6 month operation in full power (5 MW) followed by three days of cooling, representing e.g. a maintenance period. For
the MCNP transport the source was constituted by 2 GeV protons impinging on the tungsten target. In turn, neutrons of different energies and directions were generated in the collisions. Then the neutron flux and isotope production rates were determined for the axial wall, divided into 36 segments considering the effect of self-shielding. Cinder90 code was used to determine the production of radioisotopes, providing the corresponding Gamma source description with the usage of Gamma Script. This source was applied again in a second set of MCNP simulation, to determine the decay gamma dose rates – originating from the axial wall made of the Reference concrete – for the whole West Hall of the ESS bunker. In addition, the dose rates were recorded in 9 spherical areas – shown is Figure 3 – for numerical comparison.

3. Results and Discussion

3.1. Concrete activity measurements

The activities of the three studied concrete samples were determined after irradiation in the Fast and Thermalized vertical irradiation channels of the BRR, with the main goal of comparing their total activity in a 2-week long follow-up period. The total activities of the samples were calculated as the sum of the activities of all the determined gamma-emitting radioisotopes in their respective measured spectra. The measured decay curves of the three concretes are presented in Figure 4 for both irradiation channels.

It is shown that, in accordance with the expectations, the measured total activity of the newly developed PE-B4C-concrete is consistently lower than that of the Reference concrete for the whole studied time period in both sets of measurements, and significantly lower during the first week after irradiation. PE-B4C-concrete produced ~53-54% and 66-80% lower activity during the first week of cooling for the Thermalized and Fast channels, respectively. For this reason, PE-B4C-concrete is proven to be more advantageous in terms of neutron induced decay gamma emission, especially for a few days of cooling, i.e. in case of maintenance. Comparing the decay profiles of the PE-B4C- and Reference
Figure 4: Measured decay profiles of the three concrete samples after irradiation in the Thermalized (4a) and Fast channels (4b). The statistical uncertainties are too small to be discernible. The measured points are connected for better visibility.

Concrete with that of the Skanska concrete, it is also revealed that the reduction of the neutron-induced activity of the Skanska concrete, compared to the Reference concrete, is in the same range that of the PE-B4C-concrete: \( \sim 50-76\% \) and 76-80\% during the first week of cooling for the Thermalized and Fast channels, respectively. This indicates that the remanent activity of a concrete can be significantly reduced with the application of polyethylene and boron-carbide, although, the variation of activity production due to the different concrete types can already be within the same range, highlighting the importance of the initial composition.
In addition, in order to provide guidelines for simplified but realistic Monte Carlo activation simulations, and to enlighten the comparison of measured and simulated data, a set of 15 key isotopes was determined, giving the majority (minimum 83%) of the total activity for all the measured data in all samples and both irradiation channels. Both short- and long-lived isotopes are selected, giving a sufficient estimation of the total activity considering the potential occupational exposure after the end of irradiation, and for radioactive waste management after a typical 5-year cooling. The key radioisotopes and their typical activity concentrations in the studied time period – extrapolated to 5 years of cooling – are presented in Figure 5. Among the 15 isotopes, $^{24}$Na and $^{187}$W were found to be dominant in the first few days of cooling, while $^{60}$Co and $^{152}$Eu are giving the majority of the total activity in long-term, in accordance with the results of similar studies [12; 30]. Activity simulations are expected to include these key isotopes as neutron activation products.

![Figure 5: Decay curves of the 15 key radioisotopes of PE-B4C-concrete after irradiation in the Fast channel. Extrapolated from measured data.](image)

For this purpose, potential parent elements of the isotopes – shown in Figure 5 were identified. Most of them are produced via (n,γ) reaction, with a few exceptions. Beside (n,γ) reaction, $^{46}$Sc can also be a product of fast neutron (n,p) reaction of the 8% natural $^{46}$Ti content of titanium. Similarly, $^{54}$Mn is produced as a fast neutron reaction product of $^{54}$Fe, and $^{24}$Na also can be pro-
duced via a fast reaction from $^{27}$Al, in addition to the $(n,\gamma)$ reaction of $^{23}$Na. The parent element of $^{233}$Pa is thorium, as it is the decay product of short-lived $^{233}$Th, produced by the $(n,\gamma)$ activation of natural thorium. In total 14 parent elements are determined and recommended to be involved in the initial concrete compositions.

3.2. Composition measurements

In order to provide detailed input data for Monte Carlo neutron activation simulations, the composition of the three concretes were determined via PGAA, NAA and EDXRF analytical measurements. The results of all samples are presented in Tables A1 – A3. It is revealed that in accordance with the expectations, the combination of PGAA and NAA measurements provided detailed and reliable data of the composition. PGAA mostly provided the bulk element content, while NAA the trace elements. It must be noted, that the NAA method may be less accurate compared to EDXRF in case of those parent elements which were activated to the same radioisotopes in different reaction routes.

EDXRF measurements were also performed, providing input data both on bulk and trace elements beyond Na. Energy-dispersive X-ray fluorescence spectroscopy measurements show good agreement with the results of activation analysis for most of the elements. However, the parent elements of a few key isotopes were not measurable: Sm and Yb were under detection limit of EDXRF, of 2 and 20 ppm, respectively. Furthermore, the two most important elements generating long-lived isotopes were not detected either: Eu concentration found to be under the detection limit of 1 ppm and Co was not measurable due to severe spectral overlapping with the high iron concentration of the samples. For this reason, although EDXRF is an easy and relatively cheap technique for the quality assurance of concretes, it may not be adequate for characterizing shielding materials which will be posed to high neutron flux. The use of wavelength-dispersive XRF (WDXRF) might overcome the difficulty with cobalt due to its high spectral resolution.
3.3. Monte Carlo modelling of sample activation

The 2 hours irradiation of the samples in the Thermalized and Fast channels of the BRR were reproduced via MCNPX simulations applied in combination with Cinder90 analytical activation code. The measured and simulated activity concentrations (extrapolated to 1 y and 5 y of cooling in the case of the measured data) are presented in Figures 6 – 8 for all three concretes. It is revealed that the simulations performed only with the nominal concrete composition significantly underestimated the measured activities. The simulations gave only 0.1–28% and 10–41% of the measured activity for PE-B4C-concrete, and 0.2–20% and 12–28% for the Reference concrete in the Thermalized and Fast channels, respectively. However, it is shown that the discrepancies between the simulated and measured results are remarkably reduced with the application of either of the measurement-based composition. For all the concretes the first measured activity is consequently underestimated with all initial composition, understandably as the contribution of the key isotopes here is the lowest. Except of this data point, for the Thermalized channel (Figures 6a, 7a and 8a) the discrepancies are in average within 40% based on either analytical method for most of the studied time period. In the case of EDXRF measurement based simulations, the activity after 5 y cooling is underestimated with one order of magnitude, as it was shown that the main contributors to the activity at this time are the \( ^{60}\text{Co} \) and \( ^{152}\text{Eu} \), and none of their parent elements were measurable by the EDXRF method. The results of the Fast channel (Figures 6b, 7b and 8b) are more varied, most likely due to the presence of threshold reactions and the resonance integral region, making the simulations particularly sensitive to the uncertainties of the irradiating neutron spectra. However, the activity simulations with the measured compositions are considerably more realistic for this case too, resulting at least 2 times higher activities than with the nominal composition. In essence, the activity increase due to the trace elements is comparable with the typical two-threefold safety reserve typically imposed on Monte Carlo dose simulations.
Figure 6: Measured decay profiles of PE-B4C-concrete and simulated decay profiles with different initial compositions after irradiation in the Thermalized (6a) and Fast channels (6b). The statistical uncertainties are too small to be discernible. The measured points are connected for better visibility.

Figure 7: Measured decay profiles of Reference concrete and simulated decay profiles with different initial compositions after irradiation in the Thermalized (7a) and Fast channels (7b). The statistical uncertainties are too small to be discernible. The measured points are connected for better visibility.

Revealing the relevance of the initial concrete composition in the neutron activation simulations, simplified but realistic input compositions (material cards) were prepared for the PE-B4C- and Reference concretes from the combination of all the measured initial compositions focusing on the nominal bulk composition and the parental elements of the key isotopes. The derived compositions, which are recommended for activation simulations are presented as ready-to-use MCNP material cards in Table 2.
Table 2: MCNP style compositions (material cards) for PE-B4C and Reference concretes, recommended for neutron activation simulations.

| PE-B4C concrete composition | Reference concrete composition |
|-----------------------------|--------------------------------|
| by weight fraction          | by weight fraction             |
| Density [g/cm³] = 1.97      | Density [g/cm³] = 2.33         |
| 1001 -1.2633E-02            | 1001 -6.0087E-03              |
| 5000 -2.0000E-03            | 5000 -2.4900E-05              |
| 6000 -5.4583E-02            | 8016 -4.8801E-01              |
| 8016 -4.6618E-01            | 11000 -1.9676E-02             |
| 11000 -1.2910E-02           | 12000 -9.5302E-03             |
| 12000 -9.8257E-04           | 13000 -6.6560E-02             |
| 13000 -5.5120E-02           | 14000 -3.0098E-01             |
| 14000 -2.7037E-01           | 15000 -2.8035E-03             |
| 16000 -2.3669E-03           | 17000 -3.0200E-05             |
| 17000 -1.3000E-04           | 19000 -2.1899E-02             |
| 19000 -1.9467E-02           | 20000 -6.6338E-02             |
| 20000 -8.5427E-02           | 21000 -3.9660E-06             |
| 21000 -3.4450E-06           | 22000 -1.7600E-03             |
| 22000 -1.5900E-03           | 24000 -6.5710E-05             |
| 24000 -9.4300E-05           | 26000 -1.5500E-02             |
| 26000 -1.5870E-02           | 27000 -5.5230E-06             |
| 27000 -5.7810E-06           | 30000 -1.0680E-04             |
| 30000 -1.0006E-04           | 32000 -8.6160E-05             |
| 32000 -6.8500E-05           | 37000 -5.9570E-04             |
| 37000 -6.8500E-05           | 40000 -7.6160E-06             |
| 55000 -1.9830E-06           | 50000 -2.3716E-05             |
| 57000 -1.9966E-05           | 60000 -2.3716E-05             |
| 62000 -1.5670E-06           | 63000 -6.9960E-07             |
| 63000 -5.8510E-07           | 74000 -5.9570E-04             |
| 70000 -1.0260E-06           | 90000 -7.6160E-06             |
| 74000 -7.5460E-05           |                        |
| 90000 -4.3010E-06           |                        |
Irradiation simulations were performed with the ‘recommended composition’ of the PE-B4C- and the Reference concretes as well, for the whole studied time period. In addition, the isotopic activities were determined for the t₀ – end of irradiation time – and compared to that of extrapolated from measured data for the same time. As an example, results for the PE-B4C-concrete are presented and compared in Table 3. It was found that the activities agree well, within 15% for most isotopes. High, >50% differences found for the ¹⁴⁰La in the Thermalized irradiation channel, and for ⁵⁴Mn and ¹⁵⁷Yb in the Fast channel. The case of the La can be attributed to the fact that the La content was found to be very inhomogeneous among the samples, while the Yb and Mn both have a significant resonance integral in the high energy end of the applied neutron spectrum, being particularly sensitive for spectral uncertainties. These differences highlight the importance of extensive sampling for such inhomogeneous materials, like concretes.

Table 3: Comparison of simulation- and measurement-based extrapolated isotopic activity concentrations at t₀ in PE-B4C-concrete.

| Isotope | Thermalized channel | Fast channel |
|---------|---------------------|--------------|
|         | Activity concentration [Bq/g] | Difference [%] | Activity concentration [Bq/g] | Difference [%] |
| Na-24   | 2.65E+08            | 6.23E+07     | 3.53E+08            | 9.15E+08     | -17 |
| W-187   | 4.51E+06            | 3.96E+06     | -12                 | 1.80E+07     | -16 |
| La-140  | 3.90E+05            | 5.90E+05     | 13.3                | 1.46E+06     | -9  |
| Sm-153  | 4.13E+05            | 3.00E+05     | -12                 | 1.92E+06     | -25 |
| Yb-175  | 3.46E+04            | 2.40E+04     | -31                 | 1.08E+05     | -54 |
| Rb-86   | 1.28E+04            | 1.50E+04     | 17                  | 7.08E+04     | -1  |
| Pa-233  | 4.45E+03            | 3.88E+03     | -13                 | 2.24E+04     | -29 |
| Cr-51   | 3.16E+04            | 3.55E+04     | 12                  | 7.60E+04     | 0   |
| Fe-59   | 1.75E+04            | 1.63E+04     | -7                  | 4.47E+04     | -16 |
| Sc-46   | 1.71E+04            | 1.94E+04     | 13                  | 4.61E+04     | -11 |
| Zn-65   | 1.85E+03            | 1.87E+03     | 1                   | 4.83E+03     | -1  |
| Mn-54   | 2.99E+02            | 2.89E+02     | 7                   | 8.53E+03     | 49  |
| Cs-134  | 6.06E+02            | 5.97E+02     | -1                  | 2.69E+03     | -7  |
| Co-60   | 1.35E+03            | 1.41E+03     | 3                   | 3.76E+03     | -8  |
| Eu-152  | 1.60E+03            | 1.67E+03     | 5                   | 4.35E+03     | -18 |

The total activity concentrations – from the 15 key isotopes – were also determined with the recommended concrete compositions for both concretes for the studied time interval, and are presented in Figures 6 and 7. The simulations with the recommended compositions gave agreement with the measured activities in the Thermalized channels: agreeing within 38% and 49% in the whole
time period for the PE-B4C- and the Reference concrete, and within 16% and 23% without the consistently underestimated 4 d data points. In the Fast channel the recommended composition-based simulated activities are more similar to the purely measurement-based ones, fortifying that high energy simulations may be more sensitive to other parameters, e.g. the uncertainties of the irradiating spectrum.

For the Skanska concrete no specific ‘recommended composition’ was developed as it was not considered as shielding material in the region of interest of the current study, although, in Figure 8 it is shown that the activation analysis based composition can be recommended for simulations as well.

3.4. Decay gamma dose rate simulations in the ESS bunker

The power of the developed realistic ‘recommended compositions’ were studied not only for activation, but in terms of dosimetric simulations as well, for a single radial wall in the West Hall of the ESS bunker. Pure decay gamma dose rates were determined in the Hall, generated by the activated isotopes of the wall in a typical maintenance scenario with 3 days of cooling after the beam shut down. The simulated dose maps for both compositions are shown in Figure 9 and the dose rates are numerically compared in nine locations along the BIFROST instrument, as presented in Table 4.

![Figure 9](image_url)

Figure 9: Simulated dose rate maps \( \frac{d\mu Sv}{d \text{h}} \) from a the segmented axial wall made of Reference concrete. Simulations were performed with nominal (9a) and recommended (9b) initial concrete compositions.
In Figure 9 it is demonstrated that the dose rates significantly increase in the close proximity of the first quarter of the axial wall, and the area with $>1 \mu \text{Sv h}^{-1}$ gamma dose rate (yellow) is also increased, due to the trace elements in one single wall of the bunker.

As the gamma dose rate is quite inhomogeneous in the bunker, the increase of dose rate in the selected areas also varies between 29–72%, exceeding 50% in five of nine positions sampled for numerical comparison, as described in Figure 3. It was revealed, that the presence or lack of trace elements in these simulations by itself can multiply the simulated activities and doses, depleting the typical conservative allowance of Monte Carlo safety simulations. Herewith the need for more realistic material compositions for activation and safety simulations is confirmed.

Table 4: Simulated dose rates $[\mu \text{Sv h}^{-1}]$ in the West Hall of the ESS bunker. Locations are indicated in Fig. 3.

| Nominal composition | Recommended composition |
|---------------------|-------------------------|
| Dose rate           | Unc. [%]                | Dose rate | Unc. [%] | Ratio [%] |
| 1 1.89E+00          | 7 2.73E+00              | 6         | 45       |
| 2 1.79E+00          | 8 2.37E+00              | 6         | 33       |
| 3 9.37E-01          | 9 1.32E+00              | 7         | 41       |
| 4 4.99E-01          | 11 6.45E-01             | 7         | 29       |
| 5 2.52E-01          | 9 4.02E-01              | 7         | 59       |
| 6 1.87E-01          | 9 3.01E-01              | 7         | 61       |
| 7 1.24E-01          | 10 2.14E-01             | 8         | 72       |
| 8 7.75E-02          | 12 1.33E-01             | 8         | 72       |
| 9 6.07E-02          | 27 1.04E-01             | 17        | 71       |

4. Conclusions

A comprehensive study was carried out on activation properties of neutron shielding concretes. Neutron activation of three different concretes that were considered to be used at ESS: PE-B4C-concrete, its Reference concrete and a Skanska concrete were measured in the Thermalized and Fast channels of the BRR. Following the decrease of activity of the irradiated concrete samples in a 2 weeks period, it was found that the overall activity concentration of the PE-B4C-concrete is significantly lower than that of the Reference concrete: 54%
and 66% lower during the first week of cooling after being irradiated in the Thermalized and Fast channels, respectively. With this it is proven that the PE-B4C-concrete is more advantageous than the Reference concrete in terms of activation, especially during the first week of cooling, i.e. in a potential maintenance period.

In order to take into account the activation products more realistically in Monte Carlo simulations, the composition of all three concretes were determined with three analytical methods (NAA, PGAA, EDXRF). On the basis of the performed neutron activation measurements, the 15 most important gamma-emitting radioisotopes – giving together a minimum of 83% of the total measured activity for the whole studied time range – were identified, allowing a simple comparison of the measured and simulated activities. This simplification can be used in activation related safety considerations to evaluate both short- and long-term effects, not only in ESS, but in other facilities concerned with neutron irradiation (e.g. in nuclear power plants) as well.

Activation simulations of the three concretes were performed with MCNP and Cinder90 codes for all the measured and nominal compositions (similar to currently available general material cards). It was shown that the simulations with the nominal composition underestimate the measured activities with one order of magnitude, highlighting the importance of trace elements in terms of activation. On the basis of the measured composition MCNP material cards were developed and validated for the PE-B4C- and Reference concretes that are recommended for activation calculations reproducing the measured data mostly within 23% difference in the Thermalized channel.

The importance of accurate material cards was also demonstrated in terms of radiation safety: decay gamma dose rates were determined for a single concrete wall of the ESS West Hall with realistic irradiation scenario for the Reference concrete. It was found that after 3 days of cooling i.e. in a maintenance period, the decay gamma dose rates simulated with the recommended, measurement-based material card resulted 29-72% higher dose rates than with the nominal composition which does not contain trace elements.
To sum up, a detailed, comprehensive study was performed on the neutron activation properties of neutron shielding concretes and the measurement-based methodology was presented for the construction of detailed material cards for more realistic activation simulations.

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Appendix
Table A1: Measured elemental composition of PE-B4C-concrete with PGAA, EDXRF and NAA analytical methods.

| Element | Nominal [w%] | PGAA Unc. [%] | EDXRF Unc. [%] | NAA Unc. [%] |
|---------|--------------|---------------|----------------|--------------|
| H       | 2.31         | 1.26          | -              | -            |
| C       | 8.99         | 5.46          | 11.9           | -            |
| Na      | 0.62         | 1.11          | 4.2            | 1.29         |
| Al      | 2.35         | 3.44          | 3.7            | 5.51         |
| Si      | 28.6         | 21.5          | 3.3            | 27.0         |
| S       | 0.28         | 0.23          | 5.5            | 0.24         |
| K       | 1.26         | 1.39          | 3.7            | 1.95         |
| Ca      | 8.10         | 6.50          | 3.9            | 8.54         |
| Fe      | 0.84         | 1.12          | 1.6            | 1.47         |
| B       | 0.6          | 0.2           | 20             | -            |
| O       | 45.8         | 56.4          | 4              | -            |
| Mg      | 1960         | -             | 983            | 30           |
| Ti      | 521          | 1030          | 1.7            | 1590         |
| P       | 260          | -             | -              | -            |
| Cl      | 36           | 156           | 2.7            | 130          |
| V       | -            | 77            | 6              | 57.6         |
| Sm      | -            | 1.5           | 6              | -            |
| Gd      | -            | 1.6           | 10             | -            |
| Mn      | -            | -             | 230            | 4.3          |
| Sc      | -            | -             | 12             | 29           |
| Cr      | -            | -             | 82.1           | 1.6          |
| Co      | -            | -             | -              | 5.78         |
| Ni      | -            | -             | 11.7           | 6.7          |
| Cu      | -            | -             | 43.5           | 1.5          |
| Zn      | -            | -             | 100            | 0.9          |
| Ga      | -            | -             | 7.84           | 5.5          |
| Ge      | -            | -             | 4.87           | 5.8          |
| As      | -            | -             | 2.92           | 4.9          |
| Rb      | -            | -             | 59.4           | 0.7          |
| Sr      | -            | -             | 311            | 0.4          |
| Y       | -            | -             | 11.6           | 1.7          |
| Zr      | -            | -             | 91.4           | 0.4          |
| Nb      | -            | -             | 5.36           | 2.5          |
| Mo      | -            | -             | 3.03           | 4.3          |
| In      | -            | -             | 1.70           | 11.4         |
| Sn      | -            | -             | 2.81           | 1.7          |
| Sb      | -            | -             | 1.59           | 14.4         |
| Cs      | -            | -             | 2.58           | 11.7         |
| Ba      | -            | -             | 509            | 0.2          |
| La      | -            | -             | 20.0           | 1.4          |
| Ce      | -            | -             | 41.16          | 1.3          |
| Pr      | -            | -             | 4.84           | 9.5          |
| Nd      | -            | -             | 19.6           | 1.1          |
| W       | -            | -             | -              | 75.5         |
| Pb      | -            | -             | 18.3           | 4.7          |
| Th      | -            | -             | 3.85           | 7.8          |
| U       | -            | -             | 2.70           | 13.6         |
| Es      | -            | -             | -              | -            |
| Hf      | -            | -             | -              | -            |
| Tb      | -            | -             | -              | 0.30         |
| Yb      | -            | -             | -              | 1.03         |

[ppm weight mass ratio]
Table A2: Measured elemental composition of Reference concrete with PGAA, EDXRF and NAA analytical methods.

| Element | Nominal | PGAA Unc. [%] | EDXRF Unc. [%] | NAA Unc. [%] |
|---------|---------|---------------|----------------|--------------|
| H       | 0.72    | 0.60          | 1.6            | -            |
| Na      | 1.06    | 2.00          | 2.4            | 1.97         | 10            |
| Al      | 3.70    | 5.29          | 2.2            | 6.66         | 0.5           |
| Si      | 32.7    | 31.3          | 0.8            | 30.1         | 0.1           |
| S       | 0.24    | 0.28          | 3.8            | -            | -             |
| K       | 2.12    | 2.13          | 2.2            | 2.19         | 0.1           |
| Ca      | 7.12    | 7.08          | 2.8            | 6.63         | 0.1           |
| Ti      | 0.09    | 0.16          | 2.5            | 0.18         | 0.6           |
| Mn      | -       | 0.03          | 3.5            | 0.02         | 4.3           |
| Fe      | 1.16    | 1.36          | 2.6            | 1.34         | 0.1           |
| O       | 50.8    | 50.2          | 4              | -            | -             |

[ppm weight mass ratio]

| Element | Nominal | PGAA Unc. [%] | EDXRF Unc. [%] | NAA Unc. [%] |
|---------|---------|---------------|----------------|--------------|
| B       | -       | 24.9          | 1.2            | -            |
| Cl      | 30      | 169           | 2.9            | -            |
| V       | -       | 70.0          | 7.0            | 55.5         | 0.4           |
| Sm      | -       | 1.97          | 1.8            | -            | 2.38         | 4.1 |
| Gd      | -       | 2.20          | 6.0            | -            | -            |
| Mg      | 2370    | -             | -              | 9530         | 31           |
| Sc      | -       | -             | -              | 14.3         | 14.9         | 3.97 | 3.6 |
| Cr      | -       | -             | -              | 44.3         | 0.7          | 65.7 | 4.2 |
| Co      | -       | -             | -              | -            | -            | 5.52 | 4.4 |
| Ni      | -       | -             | -              | 6.40         | -            | -    |
| Cu      | -       | -             | -              | 22.4         | 2.1          | -    |
| Zn      | -       | -             | -              | 87.7         | 0.5          | 107  | 6.0 |
| Ga      | -       | -             | -              | 10.9         | 5.7          | -    |
| Ge      | -       | -             | -              | 3.20         | 7.1          | -    |
| As      | -       | -             | -              | -            | -            |
| Rb      | -       | -             | -              | 76.0         | 1.0          | 86.2 | 6.4 |
| Sr      | -       | -             | -              | 380          | 0.4          | -    |
| Y       | -       | -             | -              | 12.4         | 3.4          | -    |
| Zr      | -       | -             | -              | 114          | 0.7          | -    |
| Nb      | -       | -             | -              | 6.20         | 1.0          | -    |
| Mo      | -       | -             | -              | 2.18         | 3.9          | -    |
| In      | -       | -             | -              | 1.28         | 5.1          | -    |
| Sn      | -       | -             | -              | 2.81         | 10.8         | -    |
| Sb      | -       | -             | -              | 1.25         | 12.4         | 1.25 | 7.9 |
| Cs      | -       | -             | -              | 3.95         | 1.6          | 2.59 | 7.8 |
| Ba      | -       | -             | -              | 665          | 0.2          | 641  | 4.7 |
| La      | -       | -             | -              | 23.7         | 1.2          | 21.44 | 3.9 |
| Ce      | -       | -             | -              | 37.0         | 21.3         | 45.75 | 4.0 |
| Pr      | -       | -             | -              | 5.49         | 10.0         | -    |
| Nd      | -       | -             | -              | 18.6         | 23.1         | 20.32 | 8.0 |
| W       | -       | -             | -              | -            | -            | 596  | 3.6 |
| Pb      | -       | -             | -              | 19.4         | 1.9          | -    |
| Th      | -       | -             | -              | 4.71         | 4.9          | 7.62 | 4.0 |
| U       | -       | -             | -              | 3.39         | 4.6          | -    |
| Eu      | -       | -             | -              | -            | -            | 0.70 | 4.3 |
| Hf      | -       | -             | -              | -            | -            | 2.98 | 4.5 |
| Th      | -       | -             | -              | -            | 0.37         | 10.3 |
| Yb      | -       | -             | -              | -            | -            |
| P       | 454     | -             | -              | -            | -            |
| Ta      | -       | -             | -              | -            | -            | 0.62 | 7.4 |
Table A3: Measured elemental composition of Skanska concrete with PGAA, EDXRF and NAA analytical methods.

| Element | Nominal [w%] | PGAA Unc. [%] | EDXRF Unc. [%] | NAA Unc. [%] |
|---------|--------------|---------------|----------------|--------------|
| H       | 0.35         | 2.1           | -              | -            |
| C       | 0.55         | 57.1          | -              | -            |
| Na      | 0.55         | 2.7           | 0.5            | 46           | 0.56         | 3.8         |
| Al      | 2.59         | 2.6           | 4.9            | 15.7         | -            |
| Si      | 34.1         | 1.3           | 29.9           | 4.3          | -            |
| S       | 0.31         | 3.2           | 0.258          | 2.5          | -            |
| K       | 1.28         | 2.6           | 1.48           | 0.6          | -            |
| Ca      | 8.36         | 3.0           | 9.21           | 1.1          | -            |
| Ti      | 0.16         | 3.1           | 0.187          | 2.7          | -            |
| Mn      | 0.05         | 3.2           | 0.045          | 2.2          | -            |
| Fe      | 1.61         | 3.2           | 1.64           | 2.4          | 2.10         | 3.7         |
| O       | 50.46        | 3             | -              | -            |

| Element | Nominal [ppm weight mass ratio] | PGAA Unc. [%] | EDXRF Unc. [%] | NAA Unc. [%] |
|---------|---------------------------------|---------------|----------------|--------------|
| B       | 25.2                            | 1.3           | -              | -            |
| Cl      | 151                             | 1.8           | 140            | 7.1          | -            |
| V       | -                               | -             | 64             | 11.7         | -            |
| Sm      | 1.7                             | 1.9           | -              | 2.25         | 4.0          |
| Gd      | 2.2                             | 6.0           | -              | -            |
| Mg      | -                               | -             | 6360           | 15.7         | -            |
| Sc      | -                               | -             | -              | 4.75         | 3.6          |
| Cr      | -                               | -             | 63.6           | 3.5          | 110          | 3.9         |
| Co      | -                               | -             | -              | 7.69         | 4.1          |
| Ni      | -                               | -             | 9.3            | 6.3          | -            |
| Cu      | -                               | -             | 17.3           | 4.9          | -            |
| Zn      | -                               | -             | 53.4           | 3.4          | 65.7         | 8.0         |
| Ga      | -                               | -             | 4.0            | 6.5          | -            |
| Ge      | -                               | -             | 1.7            | 13.8         | -            |
| As      | -                               | -             | 2.0            | 4.9          | -            |
| Rb      | -                               | -             | 54.6           | 1.1          | 65.9         | 7.8         |
| Sr      | -                               | -             | 171            | 0.7          | -            |
| Y       | -                               | -             | 15.1           | 2.7          | -            |
| Zr      | -                               | -             | 145            | 0.3          | -            |
| Nb      | -                               | -             | 4.9            | 2.3          | -            |
| Mo      | -                               | -             | 3.0            | 5.6          | -            |
| In      | -                               | -             | 1.1            | 9.4          | -            |
| Sn      | -                               | -             | 1.5            | 18.0         | -            |
| Sb      | -                               | -             | 1.3            | 15.3         | 1.33         | 11.3        |
| Cs      | -                               | -             | 1.9            | 11.3         | 1.10         | 10.9        |
| Ba      | -                               | -             | 387            | 0.4          | 405          | 5.9         |
| La      | -                               | -             | 16.1           | 2.7          | 14.6         | 3.9         |
| Ce      | -                               | -             | 28.5           | 2.1          | 38.9         | 4.2         |
| Pr      | -                               | -             | 4.9            | 11.3         | -            |
| Nd      | -                               | -             | 16.7           | 5.6          | 17.29        | 9.1         |
| W       | -                               | -             | -              | 175          | 3.8          |
| Pb      | -                               | -             | 12.6           | 4.7          | -            |
| Th      | -                               | -             | 3.0            | 7.7          | 6.10         | 4.1         |
| U       | -                               | -             | -              | -            |
| Eu      | -                               | -             | -              | 0.62         | 4.5          |
| Hf      | -                               | -             | -              | 4.70         | 5.9          |
| Tb      | -                               | -             | -              | 0.43         | 9.4          |
| Yb      | -                               | -             | -              | 1.69         | 5.4          |
| P       | -                               | -             | -              | -            |
| Ta      | -                               | -             | -              | 0.40         | 10           |
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