BENCHMARK STUDIES OF INDUCED RADIOACTIVITY PRODUCED IN LHC MATERIALS, PART I: SPECIFIC ACTIVITIES

M. Brugger†, H. Khater‡, S. Mayer†, A. Prinz‡, S. Roesler†, L. Ulrici†, H. Vincke‡
†CERN, SC-RP, CH-1211 Geneva 23, Switzerland
‡ SLAC, M.S.48, 2575 Sand Hill Road, Menlo Park, CA 94025, U.S.A.

ABSTRACT

Samples of materials which will be used in the LHC machine for shielding and construction components were irradiated in the stray radiation field of the CERN-EU high-energy reference field facility. After irradiation, the specific activities induced in the various samples were analyzed with a high-precision gamma spectrometer at various cooling times, allowing identification of isotopes with a wide range of half-lives. Furthermore, the irradiation experiment was simulated in detail with the FLUKA Monte Carlo code. A comparison of measured and calculated specific activities shows good agreement, supporting the use of FLUKA for estimating the level of induced activity in the LHC.

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‡ SLAC, M.S.48, 2575 Sand Hill Road, Menlo Park, CA 94025, U.S.A.

INTRODUCTION

Accurate calculations for radionuclide inventories are typically performed with modern particle interaction and transport codes, in case of the Large Hadron Collider (LHC) mainly with the Monte Carlo code FLUKA [1, 2]. Predictions for the production of individual isotopes sometimes show significant deficiencies, when compared with experimental data, as they depend strongly on details of the experimental methods and the models implemented in the Monte Carlo code. Thus, benchmark experiments are of utmost importance in order to verify the accuracy and limits of applicability of these models. The present study combines activation and dose rate measurements, the latter being discussed in a separate paper [3]. This work is a continuation of the studies reported in Ref. [4]. It addresses a larger variety of materials and provides increased accuracy in both measurements and simulations. This paper includes the results for only a small selection of samples, more details of the experiment and the other results can be found in Ref. [5].

MEASUREMENTS AND SIMULATION

Samples

A total of 14 different materials (22 samples in total) used in the construction of the LHC were selected, ranging from various types of steel, copper, titanium, concrete and marble to light materials, such as carbon composites and boron nitride. Details of the materials

discussed in this paper, such as their elemental composition as well as their densities can be found in Ref. [5].

Irradiation Experiment

All samples were irradiated at the CERN-EU high-energy Reference Field (CERF) facility [6]. At this facility a pulsed, 120 GeV/*c* mixed hadron beam (1/3 protons, 2/3 positively charged pions) is aimed at a 50 cm long copper target creating a stray radiation field around the target that is similar to beam loss regions at high-energy accelerators (collimators, dumps, *etc.*). The samples were either laterally attached to the target or placed on a sample holder, located immediately downstream of the target and centred with its axis. Further details can be found in Ref. [5].

Gamma Spectrometry

The specific activities of the irradiated samples were measured at different cooling times ranging from about 20 minutes to two months. The gamma spectrometry measurements were performed with a high-sensitivity, low-background High-Purity Germanium (HPGe) detector by Canberra.

The samples were positioned on a custom made sample holder at different reproducible distances from the detector. The distance for each sample was chosen on the basis of its remanent dose rate and on the respective dead time of the measuring system. For each distance the efficiency of the detector was calculated using LABSOCS, a mathematical efficiency calibration software by Canberra taking into account geometrical effects and self-absorption in the samples.

The nuclide identification is based on standard or user-specific libraries. The latter were created for each sample material taking into account the chemical composition, the possible activation reaction channels as well as the cooling time. All results of the analyses were revised manually since, for some radionuclides (*e.g.,* in case of interference in the gamma energies of different nuclides), the semi-manual calculation of the specific activity turned out to be the most accurate method.

Monte Carlo Simulation

The specific activities of different radionuclides in the samples were calculated with FLUKA. The simulations were based on a detailed description of the experimental setup containing the copper target, the holder with the samples, the concrete enclosure of the beam-line shielding, as well as the accurate alignment of the beam axis with respect to the target axis. Furthermore, the elemental compositions of the samples were considered as obtained from the elemental analysis.

The full hadronic cascade was simulated in the target, in the samples and in the beamline enclosure. Neutrons were transported down to thermal energies; for all other hadrons a threshold of 1 keV was applied. Separate simulations were performed for proton and pion beams and their results were combined according to the actual beam composition. The total yield of all produced radionuclides was scored separately for all samples and specific activities were calculated taking into account the decay chains and build-up of isotopes, as well as the correct intensity profile of the respective irradiation experiment.

RESULTS

In the following the calculated and measured specific activities are compared for each material. All cooling times refer to the beginning of the respective gamma spectrometry measurement.

Many isotopes were detected at different cooling times and their specific activities were determined. However, for the final comparison with FLUKA predictions only one measurement result was selected for each nuclide based on the following criteria: smallest experimental uncertainty, lowest ratio between the measured specific activity and the respective "Minimal Detectable Activity" (MDA), as well as the cooling time as compared to the half-life. Isotopes predicted by the FLUKA simulations with an uncertainty larger than 20% are not listed.

Aluminium

Table 1 shows results for the isotopes detected in the aluminium sample. Many isotopes are relatively well reproduced by FLUKA. The light nuclide ^{7}Be is underestimated by FLUKA due to the fact that multi-fragmentation processes are not implemented in the code. Furthermore, ^{44}Sc and ^{46}Sc are overestimated by the simulation, possibly due to uncertainties in the assumed elemental composition (calcium content) or due to an overestimation of the respective production channels. Finally, in the case of ^{58}Co the (n,p) reaction on nickel most likely contributes a significant fraction and, thus, the higher FLUKA value might indicate an overestimation in the nickel content of the sample.

Table 1. Comparison of calculated and measured specific activities of isotopes in the aluminium sample for the following cooling times: (1) 1d 16h 55m, (2) 16d 8h 56m and (3) 51d 9h 47m. In addition, the ratios of the experimental results to the MDA (Exp/MDA) and the ratios of the calculated to the measured specific activities (FLUKA/Exp) are given. Note that the errors of the specific activities are quoted in percent whereas those of the ratios are given as absolute values.

Cooling time	Isotope	$t_{1/2}$	FLUKA (Bq/g) ± (%)	Experiment (Bq/g) ± (%)	Exp/MDA	FLUKA/Exp
3	^{7}Be	53.29d	0.287 ± 3.6	0.789 ± 12.6	20.4	0.36 ± 0.06
3	^{22}Na	2.60y	0.307 ± 1.2	0.365 ± 9.6	94.8	0.84 ± 0.09
1	^{24}Na	14.96h	33.0 ± 0.4	38.6 ± 3.6	821.3	0.85 ± 0.03
1	^{44}Sc	3.93h	0.508 ± 3.3	0.229 ± 23.6	2.1	2.22 ± 0.60
2	^{46}Sc	83.79d	0.039 ± 3.3	0.025 ± 15.7	5.2	1.57 ± 0.30
1	^{47}Sc	80.28h	0.161 ± 16.5	0.163 ± 10.6	6.0	0.99 ± 0.27
1	^{48}V	15.97d	0.185 ± 10.9	0.199 ± 7.4	5.0	0.93 ± 0.17
2	^{51}Cr	27.70d	0.224 ± 6.3	0.257 ± 16.8	4.8	0.87 ± 0.20
1	^{52}Mn	5.59d	0.531 ± 4.0	0.224 ± 5.6	8.3	2.37 ± 0.23
3	^{54}Mn	312.12d	0.078 ± 3.5	0.080 ± 11.4	17.7	0.97 ± 0.14
3	^{57}Co	271.79d	0.004 ± 17.6	0.004 ± 32.3	1.3	0.83 ± 0.42
2	^{58}Co	70.82d	0.035 ± 5.0	0.019 ± 21.6	3.7	1.82 ± 0.48

Stainless Steel

The results for stainless steel are given in Table 2. The specific activities of most of the heavier isotopes ($A > 43$) are remarkably well reproduced by FLUKA. Except for a few cases, the agreement of calculated and measured values is within the given uncertainties. The activities of light isotopes are underestimated as already observed for the aluminium sample. The nuclide ^{56}Mn, mainly produced by neutron capture on ^{55}Mn, is slightly overestimated, possibly due to uncertainties in the elemental composition. Finally, the two isotopes ^{56}Co and ^{57}Ni are products of spallation reactions on nickel. As the nickel content has also been determined with rather high accuracy and both the uncertainties of the calculated and

measured activities are rather small, inaccuracies in the simulation of the respective production channels remain as the only reason for the slight overestimation of the measured value by FLUKA.

Table 2. As in Table 1, here for the stainless steel sample. The cooling times are: (1) 22m, (2) 1d 6h 28m and (3) 17d 10h 39m.

Cooling time	**Isotope**	**$t_{1/2}$**	**FLUKA (Bq/g) ± (%)**	**Experiment (Bq/g) ± (%)**	**Exp/MDA**	**FLUKA/Exp**
3	^{7}Be	53.29d	0.02 ± 9.3	0.205 ± 24.3	1.9	0.10 ± 0.03
2	^{24}Na	14.96h	0.142 ± 4.3	0.513 ± 4.3	65.2	0.28 ± 0.02
1	^{42}K	12.36h	4.73 ± 4.3	5.98 ± 23.9	3.4	0.79 ± 0.22
2	^{43}K	22.30h	0.678 ± 4.1	1.08 ± 4.6	19.0	0.63 ± 0.05
1	^{38}Cl	37.24m	3.5 ± 5.3	6.9 ± 11.5	11.6	0.51 ± 0.09
1	^{39}Cl	55.60m	1.39 ± 13.3	2.24 ± 14.4	3.5	0.62 ± 0.17
1	^{m34}Cl	32.00m	3.34 ±5.4	2.67 ± 17.3	6.7	1.25 ± 0.28
2	^{47}Ca	4.54d	0.042 ± 18.9	0.098 ± 25.1	2.5	0.42 ± 0.19
1	^{43}Sc	3.89h	15.3 ± 3.1	12.9 ± 17.9	6.5	1.19 ± 0.25
2	^{44}Sc	3.93h	9.54 ± 1.0	13.8 ± 4.8	131.4	0.69 ± 0.04
2	^{m44}Sc	58.60h	8.93 ± 1.0	6.51 ± 7.1	126.4	1.37 ± 0.11
3	^{46}Sc	83.79d	0.734 ± 0.8	0.873 ± 8.3	63.3	0.84 ± 0.08
2	^{47}Sc	80.28h	6.37 ± 1.5	6.57 ± 8.2	128.1	0.97 ± 0.09
2	^{48}Sc	43.67h	1.98 ± 3.2	1.57 ± 5.2	68.9	1.27 ± 0.11
2	^{48}V	15.97d	13.1 ± 0.7	8.97 ± 3.1	411.5	1.46 ± 0.06
2	^{48}Cr	21.56h	0.633 ± 4.8	0.584 ± 6.7	10.8	1.08 ± 0.12
1	^{49}Cr	42.30m	27.2 ± 3.1	23.4 ± 10.2	12.1	1.16 ± 0.15
3	^{51}Cr	27.70d	19.1 ± 0.4	15.1 ± 12.5	102.0	1.26 ± 0.16
1	^{52}Mn	5.59d	1.5 ± 1.5	1.4 ± 13.0	5.1	1.10 ± 0.16
1	^{m52}Mn	21.10m	27.0 ± 1.6	23.3 ± 8.8	54.9	1.16 ± 0.12
3	^{54}Mn	312.12d	3.02 ± 0.3	2.85 ± 10.1	195.2	1.06 ± 0.11
1	^{56}Mn	2.58h	74.3 ± 6.4	53.8 ± 6.8	98.5	1.38 ± 0.18
1	^{52}Fe	8.28h	0.79 ± 7.3	0.72 ± 39.1	1.1	1.10 ± 0.51
2	^{55}Co	17.53h	1.16 ± 3.1	1.04 ± 4.6	19.3	1.11 ± 0.09
3	^{56}Co	77.27d	0.69 ± 1.4	0.49 ± 7.6	48.8	1.42 ± 0.13
3	^{57}Co	271.79d	0.546 ± 0.9	0.463 ± 10.7	24.6	1.18 ± 0.14
2	^{58}Co	70.82d	2.06 ± 0.4	2.21 ± 5.9	45.8	0.93 ± 0.06
2	^{57}Ni	35.60h	5.2 ± 2.0	3.5 ± 4.5	103.5	1.48 ± 0.10

Copper

The specific activities together with the ratios of calculated and measured values are given in Table 3. For many isotopes the simulated and measured activities agree within about 30%. The underestimation by FLUKA of the production of low-mass isotopes, as observed for most metallic samples, is at least partially due to deficiencies in the description of fragmentation in FLUKA. Furthermore, activities of individual meta-stable states cannot be predicted by FLUKA and, thus, equal sharing of the calculated activity among the different states is assumed. This might lead to discrepancies between calculated and measured activities as in case of $^{m44}Sc/^{44}Sc$ and $^{m52}Mn/^{52}Mn$. For some isotopes an error in the treatment of the parent-daughter correction in the gamma spectrometry analysis led to so far unresolved problems and could explain the significant overestimation by the gamma spectrometry measurement as in the case of ^{28}Mg , the daughter of ^{28}Al. In the case of ^{65}Ni and ^{64}Cu a careful investigation of production cross sections calculated with FLUKA which are also available in the literature might give further insight into the reasons for the disagreement. The underestimation of ^{65}Zn could be caused by the gamma spectrometry analysis and is presently under study.

Table 3. As in Table 1, here for the copper sample. The cooling times are: (1) 34m, (2) 1h 7m and (3) 48d 3h 21m.

Cooling time	Isotope	$t_{1/2}$	FLUKA (Bq/g) ± (%)	Experiment (Bq/g) ± (%)	Exp/MDA	FLUKA/Exp
3	^{7}Be	53.29d	0.06 ± 4.5	1.29 ± 12.6	11.9	0.05 ± 0.01
3	^{22}Na	2.60y	0.02 ± 2.5	0.029 ± 14.3	5.6	0.66 ± 0.11
2	^{24}Na	14.96h	3.94 ± 2	14.8 ± 8.5	121.3	0.27 ± 0.03
2	^{28}Mg	20.91h	0.16 ± 13.9	1.89 ± 14.6	5.5	0.09 ± 0.03
1	^{38}K	7.64m	0.21 ± 2.7	4.59 ± 34.5	4.0	0.05 ± 0.02
1	^{42}K	12.36h	12.7 ± 1.7	21.6 ± 15.3	6.8	0.59 ± 0.10
2	^{43}K	22.30h	4.19 ± 2.5	6.38 ± 11.1	11.4	0.66 ± 0.09
2	^{43}Sc	3.89h	15.9 ± 1.4	24.6 ± 24.1	9.2	0.65 ± 0.17
2	^{44}Sc	3.93h	52.7 ± 0.7	45.4 ± 9.5	88.0	1.16 ± 0.12
3	^{46}Sc	83.79d	0.77 ± 0.7	0.865 ± 8.3	128.0	0.89 ± 0.08
2	^{47}Sc	80.28h	10.2 ± 1.4	11.0 ± 14.2	6.1	0.93 ± 0.14
2	^{48}Sc	43.67h	3.64 ± 2.9	3.16 ± 12.8	18.2	1.15 ± 0.18
2	^{m44}Sc	58.60h	23.6 ± 0.7	18.4 ± 13.2	27.8	1.28 ± 0.18
3	^{48}V	15.97d	1.84 ± 0.6	1.12 ± 7.8	186.0	1.65 ± 0.14
1	^{49}Cr	42.30m	20.4 ± 1.2	15.0 ± 24.9	1.2	1.36 ± 0.35
3	^{51}Cr	27.70d	4.64 ± 0.5	3.55 ± 12.7	38.7	1.31 ± 0.17
1	^{52}Mn	5.59d	14.5 ± 0.8	18.3 ± 5.5	24.9	0.79 ± 0.05
1	^{m52}Mn	21.10m	17.8 ± 0.8	9.2 ± 33.3	7.9	1.94 ± 0.66
3	^{54}Mn	312.12d	1.33 ± 0.5	1.13 ± 10.2	97.4	1.18 ± 0.13
1	^{56}Mn	2.58h	21.7 ± 1.3	27.7 ± 5.8	20.1	0.78 ± 0.05
3	^{59}Fe	44.50d	0.39 ± 1.8	0.558 ± 10.4	42.9	0.70 ± 0.08
2	^{55}Co	17.53h	6.34 ± 2.3	7.41 ± 10.2	16.1	0.86 ± 0.11
3	^{56}Co	77.27d	1.4 ± 0.9	1.2 ± 7.2	127.0	1.16 ± 0.09
3	^{57}Co	271.79d	1.6 ± 0.5	1.8 ± 9.9	92.6	0.92 ± 0.10
3	^{58}Co	70.82d	5.79 ± 0.3	6.51 ± 10.2	533.6	0.89 ± 0.09
3	^{60}Co	5.27y	0.14 ± 0.4	0.172 ± 8.5	9.1	0.80 ± 0.07
1	^{61}Co	99.00m	44.0 ± 0.9	52.7 ± 12.3	4.2	0.84 ± 0.11
2	^{57}Ni	35.60h	4.14 ± 2.7	4.78 ± 12.1	15.8	0.86 ± 0.13
2	^{65}Ni	2.52h	5.38 ± 2.4	3.46 ± 19.3	3.5	1.55 ± 0.34
1	^{60}Cu	23.70m	13.9 ± 1.2	16.4 ± 8.7	12.2	0.85 ± 0.08
1	^{61}Cu	3.33h	173.0 ± 0.4	165.0 ± 27.2	13.2	1.05 ± 0.29
2	^{64}Cu	12.70h	336.0 ± 0.7	595.0 ± 13.2	15.1	0.56 ± 0.08
2	^{62}Zn	9.19h	6.86 ± 2.4	5.66 ± 19.9	4.4	1.21 ± 0.27
3	^{65}Zn	244.26d	0.071 ± 2.5	0.117 ± 12.0	8.6	0.64 ± 0.09

Titanium

The results for the specific activities of the titanium sample are summarised in Table 4. Again, relatively good agreement is found for most isotopes. The activity of ^{43}Sc seems to be overestimated by FLUKA. However, the experimental uncertainty is rather large such that the calculated value is still within the calculated and measured uncertainties. As in the case of copper, the assumption of equal sharing of the activity between the different states of an isotope might explain the discrepancies in the case of ^{m44}Sc/^{44}Sc. Finally, the so far unresolved problem in the parent-daughter correction most probably led to the significant overestimation of the specific activity of ^{28}Mg by the gamma spectrometry measurement.

Table 4. As in Table 1, here for the titanium sample. The cooling times are: (1) 2h 49m, (2) 4d 1h 30m and (3) 20d 4h 3m.

Cooling time	Isotope	$t_{1/2}$	FLUKA (Bq/g) ± (%)	Experiment (Bq/g) ± (%)	Exp/MDA	FLUKA/Exp
2	^{22}Na	2.60y	0.061 ± 6.8	0.056 ± 10.7	3.9	1.08 ± 0.19
1	^{24}Na	14.96h	15.1 ± 2.6	25.1 ± 3.6	546.8	0.60 ± 0.04
1	^{28}Mg	20.91h	0.524 ± 13.6	2.35 ± 5.5	13.0	0.22 ± 0.04
1	^{42}K	12.36h	41.5 ± 1.8	46.9 ± 5.2	134.0	0.89 ± 0.06
1	^{43}K	22.30h	16.2 ± 2.6	20.4 ± 3.7	124.4	0.79 ± 0.05
2	^{47}Ca	4.54d	0.42 ± 9.7	0.58 ± 15.7	43.3	0.73 ± 0.18
1	^{43}Sc	3.89h	31.5 ± 1.9	19.6 ± 56.7	21.3	1.61 ± 0.94
1	^{44}Sc	3.93h	118. ± 1	97.6 ± 4.2	503.1	1.21 ± 0.06
2	^{m44}Sc	58.60h	16.8 ± 1.1	7.61 ± 5.5	272.8	2.20 ± 0.14
3	^{46}Sc	83.79d	4.86 ± 0.7	5.82 ± 8.2	559.6	0.84 ± 0.07
2	^{47}Sc	80.28h	52.7 ± 1	61.6 ± 8.2	1422.6	0.86 ± 0.08
2	^{48}Sc	43.67h	5.23 ± 2.1	4.79 ± 3.7	281.8	1.09 ± 0.06
2	^{48}V	15.97d	2.73 ± 2.3	2.16 ± 6.1	213.9	1.27 ± 0.11
3	^{51}Cr	27.70d	0.078 ± 9	0.094 ± 36.3	1.4	0.82 ± 0.37

Concrete

Table 5 shows results for the isotopes detected in the concrete sample. Again, good agreement exists for calculated and measured activities of many isotopes. Uncertainties in the elemental composition are expected to be the main reason for discrepancies, such as ^{47}Ca (sensitive to the calcium content of the concrete) or ^{48}V and ^{52}Mn (produced in reactions with trace elements).

Table 5. As in Table 1, here for the concrete sample. The cooling times are: (1) 11h 41m, (2) 12d 6h 40m and (3) 55d 2h 31m.

Cooling time	Isotope	$t_{1/2}$	FLUKA (Bq/g) ± (%)	Experiment (Bq/g) ± (%)	Exp/MDA	FLUKA/Exp
3	^{7}Be	53.29d	2.63 ± 1.0	2.95 ±11.9	263.4	0.89 ± 0.11
3	^{22}Na	2.60y	0.060 ± 1.4	0.061 ± 9.9	101.5	0.98 ± 0.11
1	^{42}K	12.36h	1.34 ± 8.8	1.03 ± 6.1	20.3	1.30 ± 0.19
1	^{43}K	22.30h	1.58 ± 3.7	1.52 ± 3.4	157.7	1.04 ± 0.07
1	^{47}Ca	4.54d	0.239 ± 6.8	0.343 ± 14.5	29.6	0.70 ± 0.15
1	^{44}Sc	3.93h	0.304 ± 5.2	0.315 ± 6.3	12.0	0.97 ± 0.11
1	^{m44}Sc	58.60h	0.242 ± 5.7	0.127 ± 9.1	15.5	1.91 ± 0.28
1	^{47}Sc	80.28h	0.296 ± 6.4	0.325 ± 8.3	35.0	0.91 ± 0.13
2	^{48}V	15.97d	0.086 ± 7.7	0.045 ± 8.8	36.4	1.90 ± 0.31
2	^{51}Cr	27.70d	0.111 ± 5.0	0.085 ± 15.8	4.8	1.30 ± 0.27
1	^{52}Mn	5.59d	0.19 ± 7.2	0.11 ± 4.1	15.1	1.74 ± 0.20
3	^{54}Mn	312.12d	0.016 ± 5.5	0.015 ±11.9	9.5	1.06 ± 0.18
2	^{56}Co	77.27d	0.0024 ± 19.8	0.003 ± 21.8	2.2	0.80 ± 0.33

CONCLUSION

Samples of different materials typically used at accelerators were irradiated in the stray radiation field of the CERF facility. The specific activities of produced radioactive isotopes were measured at different cooling times after the irradiation and results were compared to predictions from detailed FLUKA simulations.

For most of the identified isotopes good agreement of calculated and measured specific activities was found. As expected, discrepancies were observed for intermediate and small-mass isotopes (as compared to the mass of the sample material) which can most likely be attributed to deficiencies in the FLUKA simulation models.

Furthermore, in a few cases disagreements between the simulation and the measurement results are assumed to be caused by trace elements in the composition not identified by the elemental analysis. In addition, cross sections for specific isotopes as calculated by FLUKA are the subject of further investigations.

It can be concluded that isotope production in high-energy radiation fields is well described by FLUKA and reliable predictions for isotope inventories, *e.g.*, at the LHC, could be achieved. In addition, it can be shown that quantities based on isotope production, such as remanent dose rates, show an equally good agreement between measured and calculated values [3].

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