Induced radioactivity in accelerator materials and soil-shield samples

Francesco Paolo La Torre, Marco Silari
CERN European Organisation for Nuclear Research, Switzerland

Abstract

Several samples of materials used for accelerator components and shielding structures were irradiated in the high-energy stray radiation field of the H4IRRAD Facility, installed in one of the secondary beam lines (H4) from the SPS at CERN. After irradiation, the induced radioactivity of the samples was measured by gamma spectrometry at various cooling times up to 2 years, allowing identification of isotopes with a wide range of half-lives. The activation of soil-shield samples was also studied in detail. In particular, the mechanism and probability that the radioactivity produced in soil and ground water may transfer from the site of activation to the environment was investigated. Two techniques were used to quantify the amount of radioactivity leaching in the groundwater. Furthermore, the isotope production and their specific activities measured in the soil were simulated with the FLUKA Monte Carlo code.

Introduction

The operation of high-energy accelerators leads to nuclear activation of the surrounding material due to particle interactions in accelerator components, beam transfer line elements and shielding structures. A detailed estimation of the induced radioactivity is required in order to keep the impact on personnel and environment as low as reasonably achievable. A total of 30 material samples were collected from machine and shielding components manly used in the Large Hadron Collider (LHC) as well as in other CERN accelerator environments [1,2]. The samples were irradiated in a high-energy stray radiation field in a series of experiments recently performed at the H4IRRAD Facility at CERN.

The preliminary results on measured specific activities for seven samples are presented in this work. Conversion coefficients from unit lost beam power to induced specific activity at saturation for five metallic samples are shown. The activation of soil-shield samples was also studied in detail [3]. In particular, the mechanism and probability that the radioactivity produced in soil and ground water may transfer from the site of activation to the environment was investigated. Two techniques were used to quantify the amount of radioactivity leaching in the groundwater.

Material sample description

The AISI 304L steel is a general-purpose grade widely used for vacuum applications. At CERN it is mainly used in flanges and interconnections of the LHC vacuum vessel [4,5]. The AISI 316L steel is a molybdenum-containing grade. Due to its corrosion resistance, ductility and increased austenitic stability, this grade is used for specific application in the LHC interconnections. AISI 316LN is a nitrogen containing stainless steel. Both 316L and 316LN grades are widely used in the LHC main dipole cold masses (shell, insert, cover, plate, cold bore tubes, etc.) [4,5]. MUMETAL is a nickel-steel alloy with high magnetic properties, high magnetic permeability for low magnetic fields and small hysteresis effect. Vacuum chambers in the LHC injection and extraction septa were manufactured with MUMETAL [6]. The last sample is a Ni-Fe alloy called INVAR and used for the LHC helium transfer lines [7]. Table 1 shows the chemical composition of the metallic samples.

The soil samples were collected on the Prévessin site of CERN at a depth of 24 m. CERN is located in the Geneva basin, which is filled by sedimentary deposits called "molasse". The chemical analysis of the molasse rock was carried out by the EMPA laboratory in Dübendorf (Switzerland) via X-ray fluorescence spectrometry (WD-XRF). The results of the chemical analysis are shown in Table 2. The natural water content of the soil (moisture) was measured in CERN Environmental laboratory by drying a known amount of earth. The soil moisture content may be expressed by weight as the ratio of the mass of water present to the dry weight of the soil sample. To determine this ratio, the samples and container were weighed in the laboratory both before and after drying, the difference being the mass of water originally in the sample. The water content measured in the soil specimen was 5% by weight.

Table 1. Chemical composition in mass fraction (g/100 g) of metallic samples irradiated at H4IRRAD [2]

Element	Steel 304L	Steel 316L	Steel 316LN	INVAR	MuMetal
Density	8	7.99	8	8	8.75
C	0.03	0.03	0.03	/	0.011
Co	< 0.2	< 0.2	< 0.2	/	0.01
Cr	17 - 20	16 – 18.5	16 – 18.5	/	0.03
Cu	/	/	/	/	5.04
Fe	balance	balance	balance	64	13.8
Mg	/	/	/	/	0.015
Mn	2	2	2	/	0.57
Mo	/	2 – 2.5	2 - 3	/	4.02
N	/	0.05	0.14 – 0.2	/	/
Ni	10 – 12.5	11 - 14	12 - 14	36	76.4
P	< 0.045	< 0.03	< 0.045	/	0.002
S	< 0.03	< 0.01	< 0.015	/	/
Si	1	1	1	/	0.05

Table 2. Chemical composition of dried soil (density: ~1.4 g/cm^3)

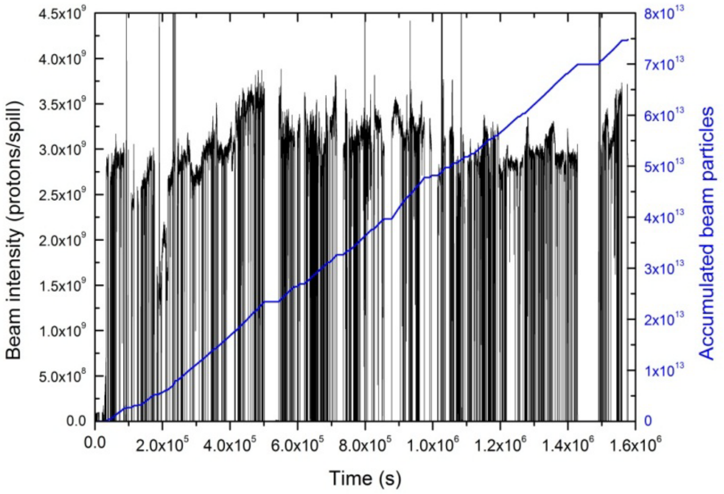
Element	O	Si	Ca	Al	C	Fe	Mg	K	Na	Ti
(g/100 g)	38.8*	24	16	6.8	5*	4	2	1.9	0.7	0.42
Element	Mn	Ba	P	Sr	Zn	Cr	Zr	Eu	Ni	S
(g/100 g)	0.11	0.06	0.06	0.05	0.03	0.02	0.02	0.01	0.01	0.01

*Extrapolated value, not quantifiable by XRF analysis.

Irradiation facility and sample activation

The activation experiment was carried out at the H4IRRAD Facility which is installed in one of the secondary beam lines (H4) from the SPS in the CERN North Area [8]. The samples were installed under the copper target (8 cm in diameter and 100 cm in length) struck by the SPS primary proton beam with momentum of 400 GeV/c and average intensity of about 3×10^9 protons per pulse (over a supercycle of about 45 seconds and an extraction length of ~5 seconds). An argon ionisation chamber (XION) placed in the H4 beam line just upstream of the copper target monitored the intensity of the primary beam. Figure 3 shows the beam intensity profile and the accumulated number of protons as a function of the irradiation time as recorded during the experiment: for the long-lived radionuclides of interest in the present study, the irradiation can be considered constant over the entire period (about 18 days).

Figure 1. Irradiation profile of the samples



The number of protons in each spill (left scale) is shown together with the accumulated total number of protons (right scale).

Gamma spectrometry results

Immediately after irradiation with ~7.5×10^{13}accumulated protons, the dose rate of the samples was of the order of a few mSv/h. Most of this radioactivity was due to very short half-life radioisotopes. Since the radioisotopes of interest to this study have medium or long half-life, the samples were allowed to decay for at least 10 days before counting. The activated samples were measured with a high sensitivity, low-background, high-purity germanium (HPGe) detector by Canberra. The data acquisition and analysis was carried out using Canberra's Genie-2000 spectrometry software and the PROcount-2000 counting procedure software. This is a comprehensive software package for data acquisition, display and analysis, which includes a set of advanced spectrum analysis algorithms providing a complete analysis of gamma ray spectra. Several gamma spectrometry analyses were performed for each sample at various cooling times up to 2 years.

The soil samples could not be directly counted for ^{3}H due to the low beta-particle endpoint energy (18 keV), which is absorbed in the sample. For this reason, the tritium activity in the soil was estimated via Monte Carlo calculations with the FLUKA code [9,10]. The tritium activity in the water was determined using a liquid scintillation counter (Packard TRI-CARB 3180TR/SL), measuring a mixture of 8 ml of activated water and 12 ml of so-called liquid scintillation cocktail (Packard Ultima Gold LLF). In the case of high precision measurements, distillation is usually recommended requiring well controlled conditions where other radionuclides present in the sample (e.g. ^{22}Na) may significantly increase the result for tritium. This was not needed in the present case, as the potential interference of other radionuclides in the tritium pulse-height window was negligible.

The results of the gamma spectrometry for all samples and of the tritium measurement for the irradiated water after 10 days of cooling time are shown in Table 3. Quoted errors include statistical and systematic uncertainties of the gamma spectrometry analysis and liquid scintillation analysis.

Table 3. Specific activity of material samples irradiated at H4IRRAD, after 10 days of cooling time

Nuclide	$t_{1/2}$	Specific Activity (Bq/g)						
		304L	316L	316LN	INVAR	MUMETAL	Soil	Water
^{3}H	12.32 y	/	/	/	/	/	14.1 ± 0.3*	28.9 ± 2.6
^{7}Be	53.1 d	30.4 ± 10.7	/	17.7 ± 5.8	87.8 ± 27	102 ± 25	425 ± 32	523 ± 48
^{22}Na	2.6 y	0.49 ± 0.08	0.34 ± 0.07	0.21 ± 0.04	0.47 ± 0.11	0.76 ± 0.15	12.5 ± 0.8	/
^{46}Sc	83.8 d	101 ± 9	77. ± 7.7	48.2 ± 3.7	75.7 ± 7.6	47.1 ± 4.2	3.02 ± 0.19	/
^{48}V	16 d	554 ± 97	497 ± 99	501 ± 100	524 ± 152	516 ± 139	9.33 ± 0.50	/
^{51}Cr	27.7 d	2048 ± 307	1619 ± 259	1004 ± 161	1744 ± 366	1176 ± 282	25.0 ± 2.3	/
^{52}Mn	5.59 d	273 ± 87	294 ± 91	534 ± 144	653 ± 287	396 ± 174	6.01 ± 0.43	/
^{54}Mn	312 d	287 ± 29	239 ± 24	138 ± 14	336 ± 34	151 ± 15	6.02 ± 0.42	/
^{56}Co	77.3 d	74.4 ± 6.0	74.1 ± 6	58.7 ± 4.1	323 ± 23	620 ± 43	0.40 ± 0.04	/
^{57}Co	272 d	76.4 ± 9.2	81.5 ± 9.8	64.0 ± 7.7	464 ± 56	746 ± 82	/	/
^{58}Co	70.9 d	270 ± 27	318 ± 32	256 ± 26	1451 ± 145	2639 ± 237	0.22 ± 0.05	/
^{59}Fe	44.5 d	/	2.93 ± 1.1	1.90 ± 0.66	9.67 ± 2.9	15.1 ± 3.8	/	/
^{60}Co	5.27 y	1.48 ± 0.15	1.40 ± 0.14	0.71 ± 0.07	3.88 ± 0.39	6.97 ± 0.63	/	/
^{75}Se	120 d	/	/	0.41 ± 0.18	/	1.94 ± 0.73	/	/
^{83}Ru	86.2 d	/	/	2.10 ± 0.59	/	6.60 ± 1.98	/	/
^{88}Y	107 d	1.03 ± 0.19	7.20 ± 0.72	6.30 ± 0.63	/	22.8 ± 2.0	/	/
^{88}Zr	83.4 d	0.64 ± 0.34	4.93 ± 0.79	4.03 ± 0.60	/	15.3 ± 2.29	/	/
^{95}Nb	115 d	/	13.0 ± 2.4	10.5 ± 2.4	/	41.2 ± 9.1	/	/
^{133}Sn	115 d	0.62 ± 0.33	/	/	/	14.5 ± 2.3	/	/

*From FLUKA simulations.

Conversion coefficients for metallic samples

The conversion coefficients obtained in this work should be regarded as an upper limit of the induced radioactivity that can be expected in materials irradiated by the secondary radiation generated by high-energy protons (beam losses) stopped in target-like objects (e.g. collimators). Since the samples were exposed very close (~ 1 cm) to the target, which is thick enough to develop most of the hadronic cascade, these coefficients should be considered conservative. In cases where the material exposed to the secondaries is at a larger distance and/or the component where the beam loss occurs is not so thick, the specific activity induced in the material will be lower.

The induced radioactivity in the materials nearby the beam loss evolves with time according to the well-known expression:

$$A(t) = A_S[1 - exp(-t_{irr}/\tau)]exp(-t_{cool}/\tau) \qquad (1)$$

where $A(t)$ is the activity at time $t = t_{irr} + t_{cool}$, A_S is the saturation activity, τ is the mean life of the radionuclide of interest, t_{irr} and t_{cool} are the irradiation time and the cooling time.

Let us assume a continuous loss of 1 W of beam power, on average, on a collimator. The value of 1 W is a fairly representative figure and the results can easily be scaled to a different power loss. Using Equation (1) one can calculate the specific activity at saturation A_S for the radionuclides of Table 3. Normalising A_S to 1 W of beam loss, we obtain the conversion coefficients from unit lost beam power to induced specific activity at saturation for the radionuclides produced in each material (Table 4).

Table 4. Conversion coefficients from unit lost beam power to induced specific activity at saturation

Nuclide	$t_{1/2}$	Conversion coefficient $A_S(Bq/g/W)$				
		304L	316L	316LN	INVAR	MUMETAL
^{7}Be	53.1 d	5.43E+04	/	3.16E+04	1.56E+05	1.81E+05
^{22}Na	2.6 y	1.23E+04	8.47E+03	5.20E+03	1.17E+04	1.90E+04
^{46}Sc	83.8 d	2.59E+05	1.98E+05	1.23E+05	1.94E+05	1.21E+05
^{48}V	16 d	5.30E+05	4.75E+05	4.80E+05	5.01E+05	4.94E+05
^{51}Cr	27.7 d	2.40E+06	1.90E+06	1.18E+06	2.04E+06	1.38E+06
^{52}Mn	5.59 d	3.71E+05	3.98E+05	7.25E+05	8.86E+05	5.38E+05
^{54}Mn	312 d	2.43E+06	2.03E+06	1.17E+06	2.84E+06	1.27E+06
^{56}Co	77.3 d	1.78E+05	1.78E+05	1.41E+05	7.73E+05	1.48E+06
^{57}Co	272 d	5.67E+05	6.05E+05	4.75E+05	3.44E+06	5.53E+06
^{58}Co	70.9 d	6.03E+05	7.10E+05	5.71E+05	3.24E+06	5.89E+06
^{59}Fe	44.5 d	/	4.60E+03	2.98E+03	1.52E+04	2.38E+04
^{60}Co	5.27 y	7.41E+04	7.00E+04	3.57E+04	1.95E+05	3.50E+05
^{75}Se	120 d	/	/	1.43E+03	/	6.78E+03
^{83}Ru	86.2 d	/	/	5.49E+03	/	1.73E+04
^{88}Y	107 d	3.25E+03	2.27E+04	1.99E+04	/	7.19E+04
^{88}Zr	83.4 d	1.64E+03	1.26E+04	1.03E+04	/	3.89E+04
^{95}Nb	115 d	/	1.74E+04	1.41E+04	/	5.53E+04
^{133}Sn	115 d	2.07E+03	/	/	/	4.87E+04

Leaching techniques for activated soil

Two leaching possibilities were investigated: water stagnation with irradiated soil (mixing system) and water percolation through the irradiated soil (flowing system). To measure the amount of radioactivity passed from the irradiated soil to the water in both systems, leach water samples were systematically measured by gamma spectrometry and by scintillation analysis.

In the mixing system, 100 g of the irradiated soil was placed in a graduated flask together with 1 litre of distilled (tritium free) water. After vigorous shaking to disperse the soil in the water, the mixture was stirred for 8 hours. To measure the radioactivity leached out, a 100 ml sample of the hazy water was filtered through a Millipore filter (0.45 µm). The gamma activity in the water was measured with a Germanium detector as discussed above while β emitters were measured with a liquid scintillation spectrometer after distillation. Activated soil and water were in contact for 4 months and the analyses on the leached water were repeated after 1, 2 and 4 months. The results for the mixing system are shown in Table 5.

In the flowing system, a sample of 100 g of irradiated soil was placed in a funnel with a Millipore filter (0.45 µm) connected to a graduated container. A glass separatory funnel was placed over the funnel and filled with 1 litre of distilled (tritium free) water. The glass stopcock allowed controlling the rate of addition of the water to 125 ml/hr. The gamma activity in the water was measured with a Germanium detector while β emitters were measured with a liquid scintillation spectrometer after distillation. The whole procedure was repeated after 1, 2 and 4 months. The results for the flowing system are shown in Table 6.

Table 5. Specific activity of ^{3}H and ^{22}Na measured in leached water after correction for the decay and the concentration for the mixing system

Mixing time	Specific Activity (Bq/l)	
	^{3}H $(t_{1/2} = 12.32\ \mathrm{y})$	^{22}Na $(t_{1/2} = 2.6\ \mathrm{y})$
8 hours	518 ± 31	96.7 ± 5.8
1 month	541 ± 32	131 ± 8
2 months	542 ± 32	136 ± 8
4 months	542 ± 33	144 ± 9

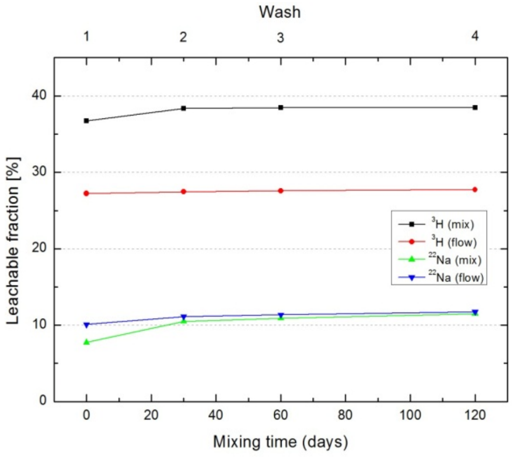
Table 6. Specific activity of ^{3}H and ^{22}Na measured in the leached water including decay correction for the flowing system

Wash	Specific Activity (Bq/l)	
	^{3}H $(t_{1/2} = 12.32\ \mathrm{y})$	^{22}Na $(t_{1/2} = 2.6\ \mathrm{y})$
1	384 ± 23	126 ± 6
2	3.23 ± 0.93	12.7 ± 0.9
3	1.67 ± 0.90	3.15 ± 0.22
4	2.05 ± 1.09	4.71 ± 0.54

Fraction of radioactivity leached out

The activity concentration of the leached water for both systems was compared with the radioactivity measured in the activated soil in order to estimate the fraction leached out. Figure 2 shows the ratio between the total activity measured in the leached water and the total activity measured in the activated soil for ^{3}H and ^{22}Na, the two radionuclides of major interest in this study. Since the liquid scintillation analysis cannot be performed on the soil, the tritium activity measured in the leached water was compared with the tritium estimated in the soil by the FLUKA simulations. Most of the radioactivity leached into the water just after 8 hours of mixing time (mixing system) or after the first wash (flowing system). In the first case, the longer the soil is mixed with the water, the more radioactivity leaches out. After two months of stirring, 39% of the ^{3}H and 11% of the ^{22}Na are leached by water. For the flowing system, the leachable fraction after one wash is 27% for ^{3}H and 10% for ^{22}Na. As from the second wash, the leachable fraction falls down to a fraction of per cent for both radioisotopes.

Figure 2. Cumulative fraction of ^{3}H and ^{22}Na activities extracted by water from the activated soil for both systems



Conclusions

Several metallic and soil samples were irradiated at the H4IRRAD test facility. Gamma spectrometry and beta scintillation analysis (leached water only) were performed to determine the radioactivity induced in the samples. Conversion coefficients from unit lost beam power to induced specific activity at saturation were calculated for each radionuclide produced in metallic samples. FLUKA simulations were carried out to estimate the concentration of ^{3}H in the soil. Two leaching procedures were used and compared to quantify the amount of radioactivity leached out of the soil into the water. The mixing system was able to remove up to 39% of ^{3}H and 12% of ^{22}Na from the irradiated soil.

Acknowledgements

The authors would like to thank A. Dziewa, F. Malacrida and A. El Gares for performing the gamma spectrometry and the liquid scintillation measurements. The authors are grateful to M. Calviani, M. Brugger and the H4IRRAD team for allowing the irradiation of the samples and for providing the FLUKA geometry of the facility.

References

[1] R. Froeschl, F.P. La Torre and N. Walter (2012), “The ActiWiz material composition catalogue”, CERN Technical Report, CERN-RP-2012-002-REPORTS-TN.

[2] R. Froeschl, FP. La Torre, S. Sgobba, C. Theis, H. Vinke, N. Walter (2012), “Radiological Hazard classification of material in CERN’s accelerators”, CERN Technical Report, CERN-DGS-2012-003-RP-IR.

[3] F.P. La Torre, M. Silari (2014), “Leaching of radionuclides from activated soil into groundwater”, CERN Technical Report, CERN-RP-2014-012-REPORTS-TN.

[4] S. Sgobba (2006), “Material for high vacuum technology: an overview”, CERN-TS/2006-004 (MME), EDMS 773904.

[5] S. Sgobba (2009), “Physics and measurements of magnetic materials”, CERN Accelerator School CAS 2009: Specialised Course on Magnets, Bruges, 16-25 June 2009.

[6] J.M. Jimenez, B. Henrist (2004), “Vacuum Chambers for the Circulating Beams in the LHC Injection and Extraction Septa”, Functional Specification, LHC-VCRS-ES-0001 rev. 1.0, EDMS 405734, (MUMETAL).

[7] S. Claudet et al. (2004), “Two 100 m Invar transfer Lines at CERN: design principles and operating experience for helium refrigeration”, CERN-AT-2004-021-ACR.

[8] B. Biskup, M. Brugger, M. Calviani, I. Efthymiopoulos, R. Kwee, J. Mekki, F.P. La Torre, E. Lebbos, P. Mala, G. Manessi, A. Nordt, F. Pozzi, K. Roeed, C. Severino, M. Silari, A. Thornton (2011), “Commissioning and Operation of the H4IRRAD Mixed-Field Test Area”, CERN Technical Note, CERN-ATS-Note-2011-121 PERF.

[9] A. Ferrari, P.R. Sala, A. Fasso, J. Ranft (2005), “FLUKA: A Multi-particle Transport Code”, CERN-2005-10, INFN/TC_05/11, SLAC-R-773.

[10] G. Battistoni, S. Muraro, P.R. Sala, F. Cerutti, A. Ferrari, S. Roesler, A. Fasso, J. Ranft (2007), “The FLUKA code: description and benchmarking”, *Proceedings of the Hadronic Shower Simulation Workshop 2006*, Fermilab 6-8 September 2006, M. Albrow, R. Raja Eds., AIP Conference Proceeding 896, 31-49.