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Preparation for activation measurements of concrete and PE-B4C-concrete to be applied for shielding at the European Spallation Source

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Abstract. To improve the effect of the concrete below 10 MeV where iron has resonances in the cross section, a new concrete have been developed. The PE-B4C-concrete utilizes hydrogen containing PE to thermalize the neutron and boron for in situ absorption. It is of utmost importance that the activation of the shielding material itself is well-understood, since it is planned to be used at the ESS. The first steps in this direction are shown the present study, in which concrete as well as reference aluminium samples are subject to XRF measurements to precisely determine the element content. This is compared to data sheets from the vender, and simulations are carried out to predict the sample activity.

The samples are planned for insertion into the the Budapest Research Reactor, followed be activity and spectral measurements.

1. Introduction

When the European Spallation Source (ESS) reaches its design configuration, protons of 2 GeV will impact a tungsten target at a rate of about $1.5 \times 10^{16} s^{-1}$, corresponding to 5 MW of proton beam power. Neutrons are generated by spallation processes in the target and will be released in substantial numbers with energies reaching up to the incident proton energy. The task of shielding instruments and personnel from high energy neutrons escaping the target-moderator-reflector system as well as from gamma photons will be undertaken by a combination of mainly steel and concrete. To improve the neutron absorption effect of concrete below 10 MeV where iron has resonances in the cross section, a new concrete has been developed. The PE-B4C-concrete utilizes hydrogen containing polyethylene to thermalize the neutron and boron for in situ absorption. It is of utmost importance that the activation of the shielding material itself is well-understood, both considering short-term effects on personnel during the operation phase and long-term effects on the decommissioning of the ESS facility.





Figure 1: Simulated (a) and real (b) aluminum sample holder tubes.

2. Methodology of material testing

A study is carried out using samples of PE-B4C-concrete as well reference samples of regular concrete and aluminum, to establish the methodology. First, the elemental concentrations are determined using X-ray fluorescence techniques and the results are compared to the nominal composition of the material. Samples of the materials are planned for insertion in the Budapest Research Reactor [1] where they will be subject to irradiation in both thermal and fast neutron fluence for a duration ranging from a few minutes to hours. After irradiation the activities of the samples will be measured and compared to the corresponding measurements on standard concrete as a function of cooling time. In addition the decay gamma spectrum will be measured. Due to an unforeseen long-widened repair of the reactor, the samples have not yet been activated therefore only results from XRF measurements and Monte Carlo activation simulations are available at present.

3. MCNP activation simulation with measured and nominal sample compositions

Monte Carlo simulation [2] was performed under conditions identical to the planned irradiation experiment at the Budapest Research Reactor. The samples will be irradiated in a “fast” and a “thermal” vertical channel, in aluminium sample holders (see Fig. 1(b)). The neutron flux spectra of the two irradiation channels of the Budapest Research Reactor are well-characterized from Monte Carlo simulations of reactor core (see Fig. 2). Using these spectra as source distributions, a simplified model consisting of only the source (uniform volume source) surrounding an aluminum sample holder containing the sample is prepared as shown in Fig. 1(a). The resulting spectra incident on the sample are input into CINDER’90[3] activation calculation program, thus time-dependent radionuclide activities are mapped, and in the future compared to the measurements. The measured element compositions and predicted activity is shown in Table 1 and Table 2.

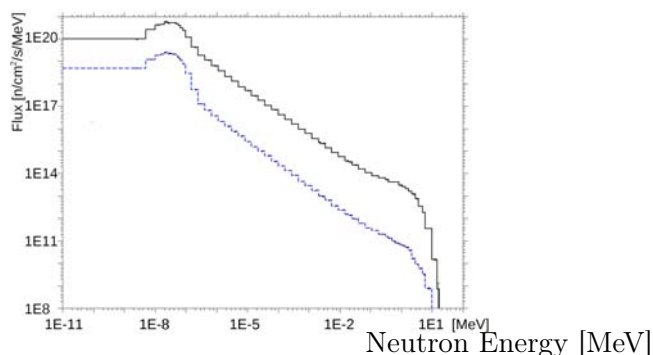


Figure 2: Simulated neutron flux at irradiation channels.

4. XRF-measurement on metal and concrete samples

Element-analysis was achieved with X-ray fluorescence (XRF) spectroscopy on metal and concrete samples. Three metal samples, $5 \times 5 \times 5 \text{ mm}^3$ cubes of aluminum, copper and steel

were measured with a handheld XRF device (Niton X3Lt GOLDD+, Thermo Scientific). In this paper we only report on the aluminium data while the steel and copper samples give similar results.

Two concrete samples, an average reference concrete and the PE-B4C-concrete, which contains 0.76 wt% B₄C and 10.2 wt% polyethylene were also studied with a polarizing XRF device (Epsilon5, PANalytical). The samples were received as grist. For sample preparation, both samples were ground again with ball grinder in tungsten carbide (WC) mortar, to increase the homogeneity (see Fig. 4(left)). Then pressed pellets of 2.5 g were prepared from both concrete samples, with the usage of 0.25 g wax (see Fig. 4(right)).



Figure 3: Metal samples (aluminium, stainless steel and copper) and handheld XRF device.



Figure 4: Grained concrete and pressed concrete pellets for XRF analysis.

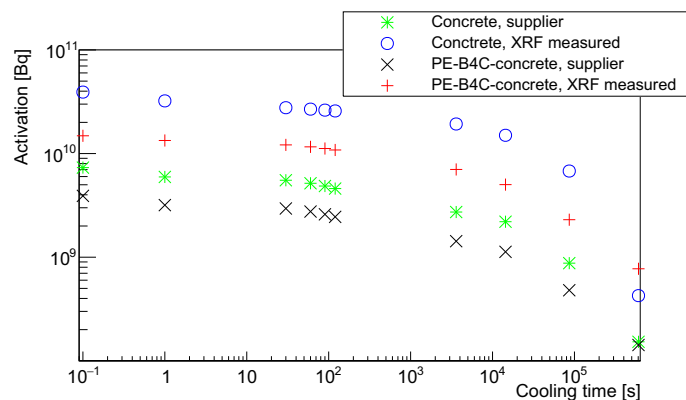


Figure 5: Simulated activity of PE-B4C-concrete and reference concrete with the XRF-measured and the nominal composition (1 week irradiation 1 week cooling).

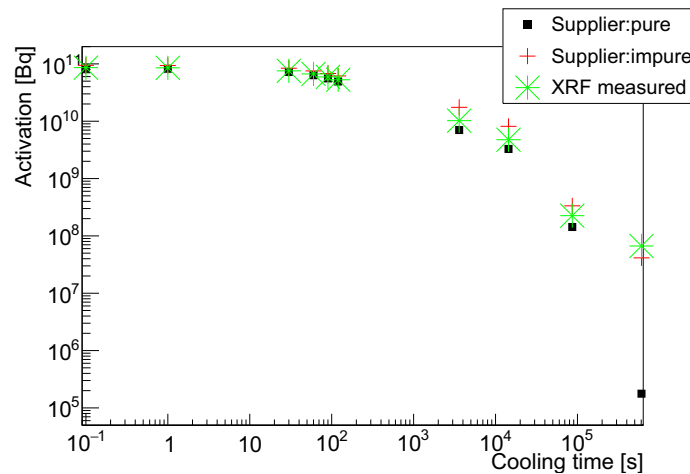


Figure 6: Simulated activity of aluminium sample with the XRF-measured and the nominal composition (1 week irradiation 1 week cooling).

5. Conclusions

The MCNP simulations show that PE-B4C-concrete have a much lower activation than normal concrete with both for the reference composition and for the XRF data. This is expected since the PE is 10.2 wt% which is about 20 vol% of the concrete and that is replacing the same volume of granite. Since PE does not activate significantly and we removed granite that activates. Also by adding PE and B increase the moderator power of the concrete and hence the neutron spectrum is driven towards lower energies, where the B absorption cross section increases (and B does not activate).

What is also clear that neither the reference values nor the XRF data give the full information of the activation. For this reason it is very important to perform the actual irradiation experiment to understand the long term effect of activation of the concrete and therefore a facility as ESS.

References

- [1] www.bnc.hu
- [2] Waters L. S. and others. The MCNPX Monte Carlo radiation transport code. *AIP Conf.Proc.* **896**, 81-90 (2007)
- [3] W. B. Wilson, S. T. Cowell, T. R. England, A. C. Hayes & P. Moller, A Manual for CINDER'90 Version 07.4 Codes and Data, LA-UR-07-8412 (December 2007, Version 07.4.2 updated March 2008).

Table 1: Composition of aluminium sample. Reference composition that is the standard of the supplier and measured composition with handheld XRF. (LOD: Lower Detection Limit)

Element	Reference material 6082-T6		Handheld XRF		
	Min	Max	Average	St Dev	LOD
wt%					
C					
Mg	0.600	1.200	0.952	0.511	0.650
Al	95.200	98.300	97.032	0.570	0.518
Si	0.700	1.300	1.024	0.072	0.087
P			< LOD	-	0.003
S			< LOD	-	0.003
Ti		0.100	< LOD	-	0.007
V			0.010	0.004	0.008
Cr		0.250	0.438	0.290	0.014
Mn	0.400	1.000	0.591	0.100	0.030
Fe		0.500	0.220	0.036	0.016
Co			< LOD	-	0.010
Ni			< LOD	-	0.011
Cu		0.100	< LOD	-	0.005
Zn		0.200	< LOD	-	0.003
Se			< LOD	-	0.003
Zr			< LOD	-	0.003
Nb			< LOD	-	0.003
Mo			< LOD	-	0.003
Ru			< LOD	-	0.003
Pd			< LOD	-	0.003
Ag			< LOD	-	0.005
Cd			< LOD	-	0.004
Sn			< LOD	-	0.003
Sb			< LOD	-	0.004
W			< LOD	-	0.004
Au			< LOD	-	0.003
Pb			< LOD	-	0.003
Bi			< LOD	-	0.003

Table 2: Composition of concrete samples. Reference composition that is best theoretical estimation of the composition taking into account supplier data and average compositions, and measured composition with Polarized XRF Epsilon5. (LOD: Lower Detection Limit)

*Tungsten presence from grounding in tungsten-carbide mortar.

Element	Concrete			PE-P4C-Concrete			Lower Detection Limit
	Reference material	Polarized XRF Average	Epsilon5 St Dev	Reference material	Polarized XRF Average	Epsilon5 St Dev	
Matrix wt%							
PE+B ₄ C				11.200	11.025	1.533	
Major elements wt%							
Na	1.060	1.968	0.197	0.617	1.291	0.197	0.281
Mg	0.237	0.953	0.295	0.196	0.983	0.295	0.116
Al	3.700	6.656	0.036	2.350	5.512	0.036	0.250
Si	32.700	30.098	0.003	28.600	27.037	0.003	0.223
P	0.045	< LOD	-	0.026	0.000	0.000	0.050
S	0.236	0.169	0.015	0.278	0.237	0.015	0.009
K	2.120	2.190	0.001	1.260	1.947	0.001	0.002
Ca	7.120	6.634	0.000	8.100	8.543	0.000	0.002
Fe	1.160	1.343	0.001	1.160	1.467	0.001	0.001
Trace elements ppm							
Cl	30.0	< LOD	-	35.5	130.0	10.0	30.0
Sc		14.3	2.1		11.8	3.4	6.8
Ti	910.0	1760.0	10.0	520.0	1590.0	10.0	10.0
V		55.5	0.2		57.6	7.1	7.1
Cr		44.3	0.3		82.2	1.3	2.3
Mn		230.0	0.0		230.0	10.0	10.0
Co		32.2	2.2		61.1	1.5	1.2
Ni		6.4	0.7		11.7	0.8	1.2
Cu		22.4	0.5		43.5	0.6	0.7
Zn		87.7	0.5		100.1	0.9	0.7
Ga		10.9	0.6		7.8	0.4	0.8
Ge		3.2	0.2		4.9	0.3	1.3
As		< LOD	-		2.9	0.1	1.3
Se		< LOD	-		0.0	0.0	1.3
Rb		76.0	0.7		59.4	0.4	0.4
Sr		380.9	1.3		311.7	1.2	0.4
Y		12.3	0.4		11.6	0.2	0.7
Zr		114.1	0.8		91.4	0.4	0.7
Nb		6.2	0.1		5.4	0.1	0.4
Mo		2.2	0.1		3.0	0.1	0.4
Ag		< LOD	-		< LOD	-	0.4
Cd		< LOD	-		< LOD	-	0.4
In		1.3	0.1		1.7	0.2	0.4
Sn		2.8	0.3		2.8	0.0	0.5
Sb		1.2	0.2		1.6	0.2	0.5
Cs		3.9	0.1		2.6	0.3	0.4
Ba		665.1	1.6		508.5	1.1	1.1
La		23.7	0.3		20.0	0.3	1.3
Ce		37.0	7.9		41.2	0.5	1.5
Pr		5.5	0.5		4.8	0.5	2.6
Nd		18.6	4.3		19.5	0.2	1.8
W*		111.4	0.4		164.8	0.5	0.5
Pb		19.4	0.4		18.3	0.9	0.9
Th		4.7	0.2		3.8	0.3	0.9
U		3.4	0.2		2.7	0.4	0.8