NON-DESTRUCTIVE ANALYSIS OF BULK CONCRETE SAMPLES USING PROMPT GAMMA NEUTRON ACTIVATION ANALYSIS: PRELIMINARY RESULTS

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Abstract - A prompt-gamma neutron activation analysis (PGNAA) system for non-destructive elemental analysis of bulk concrete samples is evaluated. The system incorporates a Pu-Be radionuclide neutron source within a graphite neutron collimator assembly. Prompt-gamma rays produced by neutron capture reactions in concrete elements were detected by a HPGe semiconductor detector. A C25/30 concrete sample of 15x 15 x 15 cm^3 was analysed. Preliminary results of silicon and calcium determination in concrete by analysis of the 4.934 and 6.420 MeV gamma ray lines, respectively, are presented. Moreover, the determination of chlorine in concrete by measurement of the 6.111 MeV gamma ray line is discussed. Gamma-ray self-attenuation in the sample was derived by Monte Carlo simulations using the MCNP code. The results of this study demonstrated the feasibility of developing an "in situ" PGNAA technique facility for non-destructive elemental analysis of bulk concrete.

Keywords: Prompt gamma neutron activation, non-destructive analysis, bulk concrete samples.

1 Introduction

Prompt-gamma neutron activation analysis (PGNAA) is an established nuclear technique that has found important applications in analytical investigations of bulk materials. Collimated neutron beams from radionuclide neutron sources, accelerators or reactor facilities are used to irradiate the sample. Prompt-gamma rays produced by neutron capture reactions within the sample elements are detected by appropriate gamma ray spectrometry systems. The fundamentals of the PGNAA technique and its applications have been reviewed recently [1].

A PGNAA facility based on a radionuclide neutron source was developed at the Institute of Nuclear Technology and Radiation Protection (INT-RP), NCSR "Demokritos" [2]. The design aim was to determine total body nitrogen in small experimental animals [3, 4]. In the present work the PGNAA technique was modified to perform non-destructive elemental analysis of bulk concrete samples. Preliminary results of silicon and calcium determination in concrete by analysis of the 4.934 and 6.420 MeV gamma ray lines, respectively, are presented. Moreover, the determination of chlorine in concrete by measurement of the 6.111 MeV gamma ray line is discussed.

The results of this work demonstrated the feasibility of developing of a relatively low cost, non-destructive, *in situ* method for elemental analysis of bulk concrete.

2 Method

2.1 System description

A schematic diagram of the PGNAA facility is given in Figure 1. The facility uses an isotopic Pu-Be neutron sources within an optimized neutron collimator – reflector assembly and a combination of high efficiency NaI(Tl) and high energy resolution HPGe gamma ray detectors. The technical characteristics of the system are shown in Table 1.

2.2Sample

A C25/30 Portland cement based concrete sample was analysed. The sample was prepared by mixing the concrete ingredients and casting it in rectangular cubical moulds of dimensions 15x 15 x 15 cm^3. The composition of concrete in the pre-mix phase is shown in Table 2. The bulk density of the sample was 2.3 g/cm^3.

2.3Monte Carlo simulations

Monte Carlo simulations using MCNP-4C2 code [5] and the ENDF/B-VI cross section data set were performed in order to derive the gamma ray detector efficiency for the bulk concrete sample. The model included the source, collimator, gamma ray detectors, shielding and the

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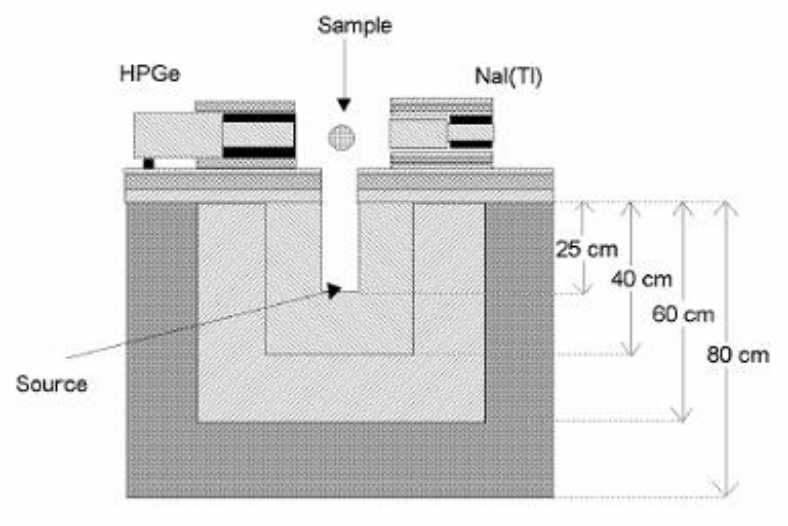

Figure 1 : PGNAA schematic diagram

Table 1: Technical characteristics of the PGNAA system

Neutron Source	^{239}Pu-Be 185 GBq
Source reflector material	Graphite
Collimator	Selection between graphite (C), polyethylene (CH_2) and borated polyethylene (CH_2-B)
Source to Sample Distance	25 – 50 cm
Sample to Detector Distance	10 – 40 cm
Beam dimensions (at the sample)	20×40 cm^2 (max)
Detector 1	Na((Tl) 10 cm in diameter × 10 cm in length
Detector 2	HPGe 10% or 85%
Data acquisition	Gamma-fast (Eurisis Measures) PC-card
Biological Shielding	Boron-lead-doped wax (min thickness 50 cm)

Table 2: Premix phase composition of the concrete sample

Material	Wt. (%)
Portland cement	8.2
Fine aggregate	28.7
Coarse aggregate	56.4
Water	6.7

concrete sample. The developed model enabled prediction of voluminous source gamma ray detector efficiency for the characteristic prompt-gamma rays of the elements of interest.

3 Results and Discussion

3.1 Prompt-gamma ray spectrum of concrete

The prompt-gamma ray spectrum of the concrete sample is shown in Figure 2. The spectrum was obtained by a 10% in relative efficiency HPGe semiconductor detector after irradiation time of 7200 s. Silicon, calcium and chlorine in concrete resulted in peaks at 4.934, 6.420 and 6.111 MeV, respectively, that could be well discriminated above the complex background spectrum. Prominent features of the PGNAA background spectrum are peaks at 0.48 MeV from $^{10}B^*$, at 2.223 MeV due to thermal neutron capture in hydrogen, at 2.61 MeV due to inelastic neutron scattering in lead and at 4.43 MeV from $^{12}C^*$ as well as their single and double escape peaks. The Doppler broadened peak at 4.43 MeV is attributed to the Pu-Be source itself. Moreover, the spectral lines at 596 keV and 692 keV are attributed to thermal neutron capture and inelastic neutron scattering events in the Ge crystal, respectively. The Ge lines can be used in order to directly evaluate the detector shielding efficiency to the source neutrons.

3.2 Si to Ca ratio determination

The silicon to calcium concentration ratio in concrete ([Si]/[Ca]) was given by equation (1)

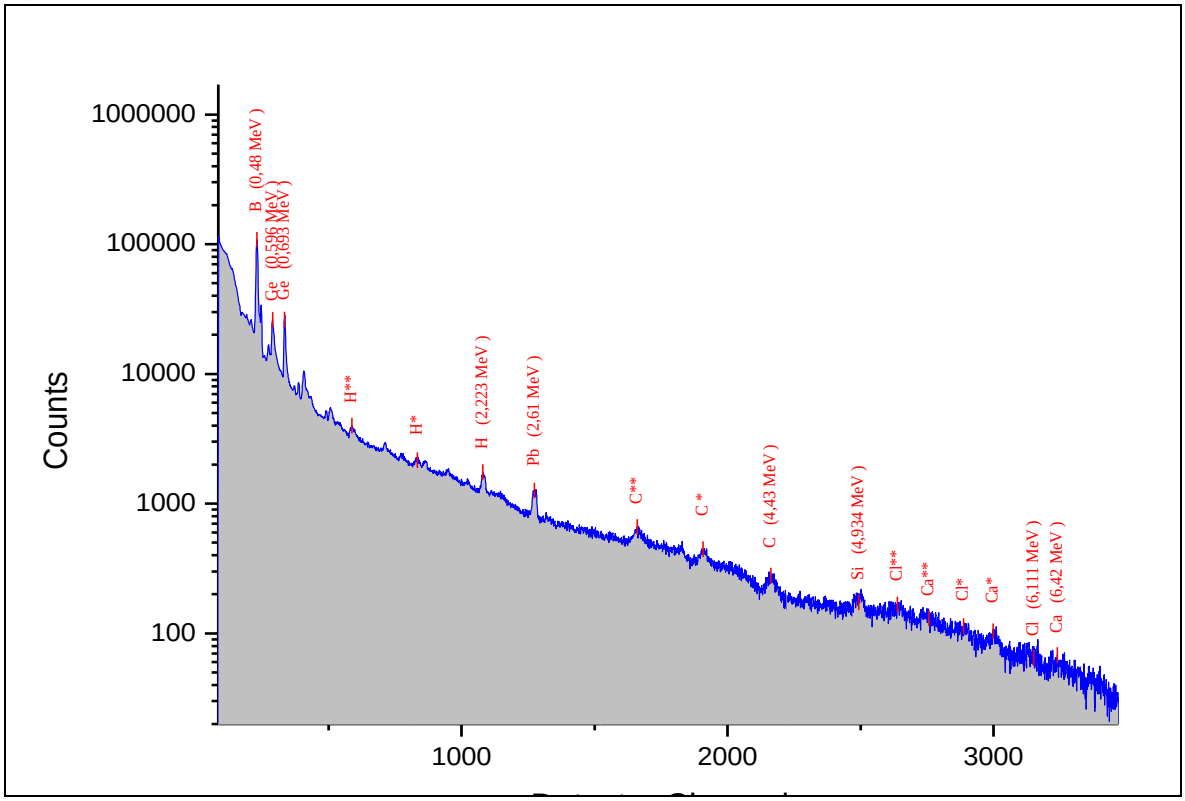

Figure 2 : PGNAA spectrum of concret, in the photon energy range 0 – 7 MeV

$$\frac{[Si]}{[Ca]} = \frac{N_{\gamma,Si}\,\sigma_{Ca}\,\varepsilon_{\gamma,Ca}\,I_{\gamma,Ca}}{N_{\gamma,Ca}\,\sigma_{Si}\,\varepsilon_{\gamma,Si}\,I_{\gamma,Si}} \qquad (1)$$

where $N_{\gamma,Si}$ and $N_{\gamma,Ca}$ are the net counts, σ_{Si} and σ_{Ca} the thermal neutron capture reaction cross-section, $\varepsilon_{\gamma,Si}$ and $\varepsilon_{\gamma,Ca}$ the gamma ray detector efficiency for the bulk sample at 4.934 and 6.420 MeV, $I_{\gamma,Si}$ and $I_{\gamma,Ca}$ the gamma ray branching ratio for Si and Ca, respectively. The silicon and calcium gamma ray detection efficiencies for the bulk sample were calculated using the MCNP code. The measured gamma ray energies, their branching ratios, neutron cross-sections, net peak areas obtained and calculated detector efficiencies are shown in Table 3.

Table 4 shows a comparison of the [Si]/[Ca] ratio determined using the PGNAA technique and that derived by wet chemical analysis of the concrete constituents materials in the pre-mixing phase. A good agreement between the two methods is observed. The large uncertainty in the PGNAA result is due to the low counting statistics obtained. It is stressed that in this preliminary study a HPGe detector of only 10% efficiency was utilized. Nevertheless, in future studies a significantly larger HPGe detector of 85% efficiency will be used. Thus, a reduction in the uncertainty of the PGNAA measurement will be achieved, i.e. by a factor of 3.

Table 3: Silicon and calcium gamma ray energy, branching ratio, neutron cross section, detector efficiency and net peak counts obtained

Element	Gamma ray energy (MeV)	Branching ratio (%)	Neutron cross section (b)	Peak area (cps)	Detector efficiency $\times 10^{-5}$ (%)
Si	4.934	70.55	0.16	1403 ± 295	3.87 ± 0.20
Ca	6.420	28.09	0.43	444 ± 229	2.47 ± 0.16

Table 4: Comparison of Ca/Si ratio obtained by PGNAA and from mixture ingredients

Method	[Si]/[Ca] (w/w)
Chemical analysis	1.9 ± 0.3
PGNAA	2.2 ± 1.2

4 General Discussion and Conclusions

In this work a PGNAA method for evaluation of concrete composition was discussed. The method allows non-destructive determination of calcium, silicon and chlorine concentration in bulk concrete samples. Knowledge of the [Si]/[Ca] ratio is considered to be a key parameter for durability studies of concrete structures, since it may provide an index of cement proportion in concrete [6, 7]. Moreover, determination of chlorine in concrete is important, since chloride salts play a dominant role in initiating reinforcement steel corrosion in concrete [8].

The major advantages of the PGNAA technique over traditional concrete analysis techniques are:

- non-destructiveness
- possibility for *in situ* performance
- minimum sample preparation requirement
- absence of sample size reduction and homogenization procedures
- minimum probability of sample contamination or element lose during the analysis procedure
- determination of Ca, Si, Cl (Al and H), simultaneously

On the other hand, wet chemical analysis of concrete samples is destructive, it cannot be performed in situ, it requires a great effort to achieve representative sub-sampling and moreover particular care is required to minimize contamination probability during sample preparation and analysis.

The results of this study will assist on the design of a PGNAA system based on isotopic neutron sources allowing non-destructive field tests of concrete structures.

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