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Simulation of the Determination of NaCl Concentration in Concrete samples by the Neutron induced Prompt Gamma-ray Method

Hyeon-Soo Kim

Department of Physics, Kyonggi University, Suwon 442-760, Korea

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A prompt gamma-ray neutron activation (PGNA) system was simulated by the Monte Carlo N-Particle transport code (MCNP-4A) to estimate the level at which the scattered photon fluence rate, the absolute efficiency of the HPGe-detector, the volume of the concrete sample and the $^{35}Cl(n,\gamma)$ reaction rate in this sample contribute to the count rate in the NaCl concentration measurement. The n-γ fluence rates at the ST-2 beam tube exit of the HANARO reactor were used as input data, and the GAMMA-X type HPGe detector was modeled to tally 1.1649 MeV γ-rays emitted from the $^{35}Cl(n,\gamma)$ reaction in the concrete sample. For three cylindrical concrete samples of 13.8, 46.8 and 157.1 cm^3 volumes, respectively, the relations between the NaCl weight fractions of 0.1, 1, 2 and 5 % in each of the concrete samples and the 1.1649 MeV pulses created in the HPGe detector model were studied. As a result, it was found that the count rate at the same NaCl concentration nearly depends on the volume of the samples in a simulated condition of the same NaCl concentration samples, and that the linearities of the NaCl concentration calibration curves were reasonable in the narrow range of the NaCl weight fraction.

Key words : MCNP, Simulation, PGNA, NaCl concentration

1. Introduction

Chlorine ions lead to corrosion in steel concrete buildings, the corrosion being capable of causing cracks in the steel and concrete, resulting in the collapse of the building. Thus, the NaCl concentration must be regulated by an weight fraction of NaCl less than 0.1 % in dried sea sand[1]. The conventional method of verifying the NaCl concentration, however, employs destructive methods. A prompt gamma-ray neutron activation analysis (PGNA) can analyze the NaCl concentration promptly without destroying the concrete sample[2].

In this study, PGNA was simulated by the Monte Carlo N-Particle transport code (MCNP-4A) to estimate the level which the absolute efficiency of the HPGe-detector, the volume of the concrete sample, and the $^{35}Cl(n,\gamma)$ reaction rate in this sample contribute to the count rate in the NaCl concentration measurement[3]. For these estimations, calibration curves of NaCl weight fractions depending on the cylindrical concrete sample volumes, were determined. For this simulation, the neutron and gamma-ray fluence rates at the ST-2 horizontal beam tube exit of the HANARO research reactor, which were estimated by reactor designers, were used as input source radiation data, and the GAMMA-X type HPGe detector was modeled to tally the 1.1649 MeV γ-rays emitted from the $^{35}Cl(n,\gamma)^{36}Cl$ reaction in the concrete sample. To estimate the uncertainty of this result and to verify the suitability of the geometric data of the HPGe-detector inputted for the MCNP simulation, the relative efficiency of our detector model was calculated for the same experimental conditions as those carried out by the EG&G ORTEC manufacturer.

Corresponding Author : Hyeon-Soo Kim, Department of Physics, Kyonggi University, Suwon 442-760, Korea
Phone : +82-31-249-9623
E-mail : kimsa3377@hanmail.net

2. Simulation

2.1. The PGNA System

he model of the PGNA system used and the schematic diagram of the GAMMA-X type HPGe detector modeled for the MCNP calculation are shown in Fig. 1 and 2, respectively. The actual beam size of the neutron and gamma-rays which emerge from the ST-2 beam tube exit is 7 cm × 14 cm in a rectangular shape. However, we simulated the radiation as a mono-directional source emitted from a circular surface with a diameter of 11.2 cm in the direction of the positive x-axis normal to the surface. The distance from the source to the center of the concrete sample and the distance from the center of the sample to the endcap of the HPGe detector model were chosen to be 35 and 25 cm, respectively.

2.2. Source Radiation and Sample Preparation

The photon and neutron fluence rates at the ST-2 beam tube exit of the HANARO research reactor are listed in Tables 1 and 2, respectively.

In Tables. 1 and 2, n indicates the number of neutrons (or photons) per cm^3 and v is their velocity (cm/s). The weight fractions of the elements for 0.1, 1, 2 and 5 % NaCl in cylindrical concrete samples are given in Table 3. The dimensions of the samples are shown in Table 4.

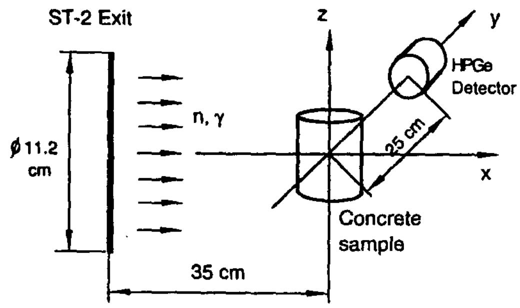

Fig. 1. Model of the PGNA system at the exit of the ST-2 horizontal beam tube of the HANARO reactor.

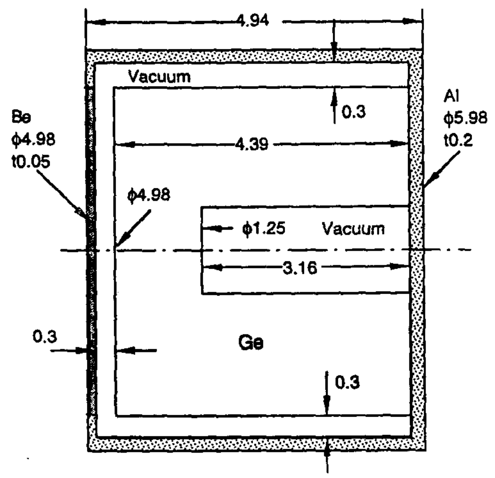

Fig. 2. Cross-sectional view of the HPGe detector for the MCNP simulation (Dimensions are in cm.).

Table 1. Gamma-ray fluence rate Φ at the ST-2 beam tube exit

Energy E (MeV)	Fluence rate (nv)
1.00×10^{-2}	Low energy boundary
1.00×10^{-1}	8.611×10^{9}
5.10×10^{-1}	1.624×10^{10}
6.00×10^{-1}	1.186×10^{9}
1.33	3.536×10^{9}
3.00	1.127×10^{9}
7.50	2.473×10^{8}
14.0	4.866×10^{8}

Table 2. Neutron fluence rate Φ at the ST-2 beam tube exit

Energy E (eV)	Fluence rate (nv)
1.000×10^{-5}	Low energy boundary
4.140×10^{-1}	3.249×10^{10}
3.727×10^{1}	2.582×10^{9}
2.035×10^{3}	2.273×10^{9}
4.087×10^{4}	1.695×10^{9}
1.500×10^{5}	5.121×10^{8}
4.505×10^{5}	2.841×10^{8}
1.003×10^{6}	1.575×10^{8}
2.019×10^{6}	1.738×10^{8}
6.703×10^{6}	2.710×10^{8}
1.733×10^{7}	3.801×10^{6}

Table 3. Weight fractions of the elements in cylindrical concrete samples (ρ= 2.3 g/cm^3). Cl, Mg, Si, K, Ca and Fe are naturally occurring elements

Element	NaCl concentration			
	0.1 %	1 %	2 %	5 %
^{1}H	0.0102	0.0101	0.0100	0.0097
^{16}O	0.5371	0.5321	0.5269	0.5106
^{23}Na	0.0003	0.0030	0.0060	0.0156
Cl	0.0007	0.0070	0.0140	0.0344
Mg	0.0020	0.0020	0.0020	0.0019
^{27}Al	0.0345	0.0342	0.0339	0.0328
Si	0.3421	0.3391	0.3356	0.3254
K	0.0132	0.0131	0.0129	0.0126
Ca	0.0447	0.0443	0.0438	0.0425
Fe	0.0142	0.0141	0.0139	0.0135
^{12}C	0.0010	0.0010	0.0010	0.0010

2.3. Counts of the 1.1649 MeV γ-rays

The relation between the ^{35}Cl(n,γ) reaction rates induced in the sample and the tally rates of the 1.1649 MeV photon pulse to be created in the HPGe detector is as follows:

$$R_\sigma \cdot \Phi \cdot \sigma_{n\gamma} \cdot N \cdot V \cdot I \cdot \varepsilon = C(0) \quad (1)$$

according to

R_σ : atomic fraction weighted cross-section ratio of ^{35}Cl(n,γ) to ^{37}Cl(n,γ) reaction = 0.75× ($\sigma_{n\gamma}$ for ^{35}Cl) / {0.75×($\sigma_{n\gamma}$ for ^{35}Cl) +0.25×($\sigma_{n\gamma}$ for ^{37}Cl (n,γ)};

Φ : neutron fluence rate through the concrete sample [cm^{-2}s^{-1}];

$\sigma_{n\gamma}$: sum cross-section of the ^{35}Cl(n,γ) and ^{37}Cl(n,γ) reactions [cm^2];

N : sum atomic number density of ^{35}Cl and ^{37}Cl in the concrete samples [cm^{-3}];

V : volume of the concrete sample [cm^3];

I : emission rate of the 1.1649 MeV γ-rays per ^{35}Cl(n,γ) reaction;

ε : absolute efficiency of the 1.1649 MeV γ-rays to be tallied by the HPGe detector model;

Table 4. Dimension of the cylindrical concrete samples

Model	R(cm)*	H(cm)**	V(cm^3)***
Vol-1	1.11	3.56	13.8
Vol-2	1.67	5.34	46.8
Vol-3	2.5	8	157.1

* R : radius of the samples
** H : height of the samples
*** V : volume of the samples

C(0) : tally rate of the 1.1649 MeV photon pulses created in the HPGe detector model

Because MCNP-4A uses the Cl(n,γ) cross-section, R_σ is introduced to acquire the ^{35}Cl(n,γ) reaction rate from a 75 % atomic fraction of ^{35}Cl in naturally occurring Cl. Each ^{35}Cl emits 1.1649 MeV γ-rays from the ^{35}Cl(n,γ) reaction with an emission probability of 27.7 %. The values of $\sigma_{n\gamma}$ for ^{35}Cl and ^{37}Cl can be obtained from reference data. Neutron fluence rates, Φ[cm^{-2} s^{-1}], averaged over the concrete samples and the Cl(n,γ) reaction rates, $\Phi\sigma_{n\gamma}N$[cm^{-2} s^{-1}], in the samples, were calculated by an f4:n tally and ENDF/B reaction type 102 in an fm14 tally of MCNP-4A, respectively.

The atomic number density of ^{35}Cl in the concrete sample can be deduced from the weight fraction of Cl in Table 3. The volumes of the concrete samples are shown in Table 4, and the ratios of height to diameter of the samples are approximately 1.6. To determine the absolute efficiency of the HPGe detector model, an f8:p tally of MCNP-4A was used.

The meaning of the MCNP-4A tallies mentioned above are as follows:

F:4 n = neutron fluence averaged over volume [neutrons/cm^2];

fm 14 (102) = ENDF/B reaction type for a (n,γ)reaction;

f8: p = energy distribution of pulses created in a detector by incident photon beams [pulses]

The tally rates C(0) of the HPGe detector model for different NaCl concentrations in the concrete samples and for different volumes of the samples were calculated from equation (1). Because the tally rate of the 1.1649 MeV photon pulses created in the HPGe detector are proportional to the $^{35}Cl(n,\gamma)$ reactions in the concrete sample, the NaCl concentrations can be deduced from the tally rates. Finally the calibration curves depending on the NaCl concentrations and the volume sizes of the concrete samples were determined from the tally rates C(0).

3. Results and Discussion

3.1. Scattered Photon Contribution

The scattered photon contribution from the concrete sample to the HPGe detector was calculated for the PGNA system model in Fig. 1. The gamma-ray fluence rate spectrum in Table 1 was used as input source radiation data in this simulation, and the result is shown in Fig. 3. From this result, it is clear that it is not necessary to shield the scattered photons for the 1.1649 MeV γ-ray measurement with the HPGe detector model.

3.2. $^{35}Cl(n,\gamma)$ Reaction Rate in the Concrete Sample

In equation (1), $\Phi \sigma_{n\gamma} N$ indicates the $Cl(n,\gamma)$ reaction rate in the concrete sample. The neutron fluence rates Φ and the $Cl(n,\gamma)$ reaction rates in the concrete samples were calculated by MCNP-4A for the PGNA arrangement model at the ST-2 beam tube exit. The neutron fluence rates are shown in Fig. 4. In Fig. 4, the differences between the neutron fluence rate spectra in various concrete sample volumes were neglected. The $^{35}Cl(n,\gamma)$ reaction rates in the samples were derived by correcting the $Cl(n,\gamma)$ reaction rates with an atomic fraction weighted cross-section ratio R_σ, and are shown in Fig. 5. The values of R_σ used in this calculation were 0.996701, 0.996206 and 0.785169 for the thermal range (E_n < 1eV), the intermediate range (1 eV < E_n < 10 keV), and the high-energy range (10 keV < E_n < 15 keV), respectively[4]. Fig.5 shows that the $^{35}Cl(n,\gamma)$ reaction rates were calculated to be at almost the same level in the same NaCl concentration samples, Vol-1, Vol-2, Vol-3. Therefore, it is shown that these differences in the $^{35}Cl(n,\gamma)$ reaction rates have little influence on the difference of the count rate in the above samples.

The NaCl concentration calibration curves were derived from equation (1) under the assumption that the tally rates of the 1.1649 MeV γ-rays emitted from the $^{35}Cl(n,\gamma)$ reaction are proportional to the concentrations of NaCl in the concrete samples, and this result is shown in Fig. 6. The linearity between the count(= tally) rates and the NaCl concentrations for each concrete sample volume was satisfactory. The absolute efficiency ε of the HPGe detector in equation (1) was calculated under this simulation condition with MCNP-4A, and the results are shown in Table 5. According to the increase of radius in the sample, the absolute efficiencies around each sample

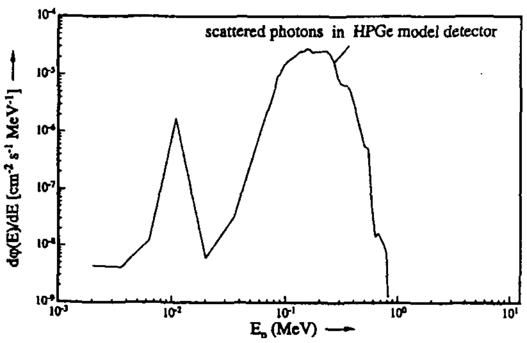

Fig. 3. Scattered photon fluence rate in the HPGe detector for the PGNA model at the ST-2 beam tube exit.

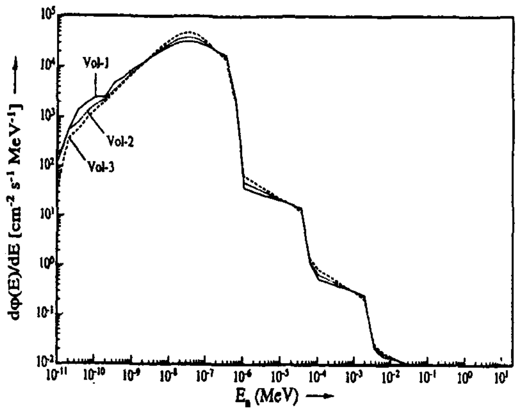

Fig. 4. Neutron fluence rates Φ in three different volumes of concrete samples for the PGNA model at the ST-2 beam tube exit. The weight fraction of NaCl in these concrete samples is 0.1 %.

decreased slightly owing to the attenuation effect of the γ-rays. Therefore, this result, shown in Table 5 and Fig. 6, show that the difference of its values contribute slightly to the difference of count rate in the same NaCl concentration. Because the absolute efficiencies(Table 5) of the HPGe detector in the same volume and different NaCl concentration samples were not the same, this result wrongly contributed to the linearity of the NaCl concentration calibration curve(Fig. 6).

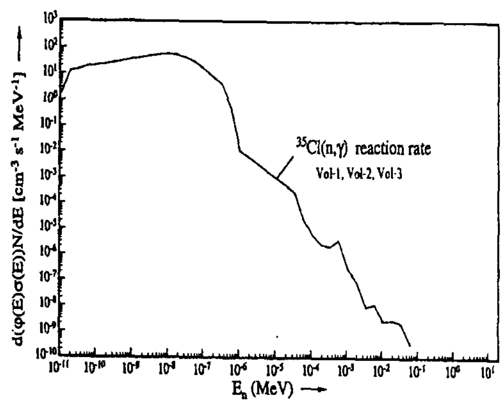

Fig. 5. $^{35}Cl(n,\gamma)$ reaction rates in three different volumes of concrete samples (Vol-1, Vol-2, Vol-3).

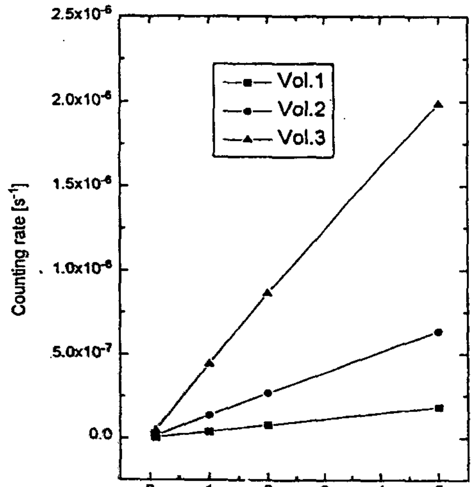

Fig. 6. NaCl concentration calibration curves for three different volumes of concrete samples for the PGNA arrangement at the ST-2 beam exit of the HANARO reactor.

To verify the suitability of the geometric data of the HPGe-detector inputted for the MCNP simulation, the relative efficiency of our detector model was calculated for the same experimental conditions as used by the EG&G ORTEC manufacturer[5]. So, we simulated the condition that 1.33 MeV γ-ray point source is set at 25 cm from the center of the front surface of the endcap on a line perpendicular to the endcap face. The absolute efficiency of the HPGe detector model under this simulation condition was estimated to be 1.849×10^{-4} with a relative error of 5.2 %. The relative efficiency of our detector model was calculated from the absolute efficiency by equation (2):

$$\varepsilon_r = \frac{\varepsilon}{1.2\times10^{-3}} \times 100\,[\%] \qquad (2)$$

according to

ε : the absolute efficiency of the HPGe detector model for the 1.33 MeV γ-ray point source under the same simulation conditions as the experimental conditions of EG&G ORTEC;

ε_r : the relative efficiency of the detector model under the same simulation conditions as EG&G ORTEC's [%];

Table 5. Efficiency ε of the HPGe detector (Gamma-X) for PGNA arrangement at the ST-2 beam tube exit

Concentration sample	Concentration* of NaCl (%)	Efficiency (%)
Vol-1	0.1	2.52846E-02
	1	2.52846E-02
	2	2.52846E-02
	5	2.44500E-02
Vol-2	0.1	2.44804E-02
	1	2.43759E-02
	2	2.43688E-02
	5	2.43668E-02
Vol-3	0.1	2.19658E-02
	1	2.19918E-02
	2	2.20079E-02
	5	2.19473E-02

* Weight fraction of NaCl in the concrete sample

1.2×10^{-3} : the efficiency of a 3-in.×3-in. NaI(Tl) scintillation detector under the experimental conditions of EG&G ORTEC, and this value is known as a constant.

The relative efficiency calculated of the HPGe detector under the same conditions as those the EG&G ORTEC manufacturer, and the value from the certificate data offered by EG&G ORTEC, was 15.4 % and 15% respectively, with 0.8 % of absolute statistical uncertainty.

4. Conclusions

The scattered photon fluence rate and the absolute efficiencies in the HPGe detector for PGNA, the $^{35}Cl(n,\gamma)$ reaction rates and the NaCl concentration calibration curves for concrete samples of 13.8, 46.8 and 157.1 cm^3 volumes in which the NaCl weight fractions were 0.1, 1, 2 and 5 % for each volume, were calculated by the MCNP-4A Monte Carlo code. To verify the suitability of the data of the HPGe detector inputted for the MCNP simulation, its relative efficiency was calculated. For these calculations, the PGNA simulation method, was suggested at the ST-2 Beam tube exit of HANARO, was suggested. The GAMMA-X type HPGe detector was modeled to tally the 1.1649 MeV γ-rays emitted from the $^{35}Cl(n,\gamma)$ reactions in the concrete samples and the samples were simulated to be irradiated by n-γ mixed radiations at the ST-2 beam tube exit of the HANARO research reactor.

In the above simulation results, it was found that the count rate of the same NaCl concentration samples nearly depends on the volume of these samples in this simulated condition of the same NaCl concentration samples, and the linearities of the NaCl concentration calibration curves were reasonable within the narrow range of the NaCl weight fraction. The relative efficiency of our detector model was calculated to be 15.4 %, with 0.8 % of absolute statistical uncertainty. Therefore, the calculated relative efficiency agreed with the certified relative efficiency within the range of statistical uncertainty. From this comparison of the above two relative efficiencies, the suitability of the geometric data of the HPGe-detector input for the MCNP simulation was verified, and also these results showed that the NaCl concentration calibration curves obtained by this simulation were valid.

Through this research, it was found that if the optimum volume and geometric shape of the sample is selected by using this PGNA simulation method, the NaCl concentration in concrete samples can be optimally and reasonably well obtained by this method in measuring the NaCl concentration using its calibration curve. This curve must be obtained within the narrow range of the NaCl weight fraction and one of the applications of the HANARO research reactor is suggested in order to verify the NaCl concentration in concrete constructions through a non-destructive and prompt method.

References

1) Kim, W. S. and H. S. Kim, 1996, Study on Prospective Projects of the Division of Chemistry and Radiation, KRISS-95-161-IR, Korea research Institute of Standards and Science, 30-41pp.
2) Matsumoto, T. and O. Aizawa, 1990, Prompt Gamma-ray Neutron Activation Analysis of Boron-10 in Biological Materials, Appl. Radiat. Isot., 41(9), 897.
3) Briesmeister, J. F.(Ed.), 1993, MCNP(4A)-A general Monte carlo code for neutron and photon transport Version 4A, Los Alamos National Lab., 1-164pp.
4) Chang, J. H., 1995, Table of Nuclides, KAERI/GP-106/95, Korea Atomic Energy Research Institute, 75-76pp.
5) EG&G ORTEC, 1987, Solid-state Photon Detector Operation Manual of Gamma-X plus Series, EG&G ORTEC Model No. GMX-1585-plus, USA.