A Study on the Non-Destructive Detection of Salt in Concrete Using Neutron-Captured Prompt-Gamma Rays at RANS∗

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1. Introduction

A large amount of public and private infrastructure is made of concrete. These structures are degraded by various factors, e.g., carbonation, chloride attacks, and frost attacks. Of these degradation factors, chloride attacks on concrete structures are critical because Japan is surrounded by seas and has many mountainous areas. Therefore, concrete structures are influenced by salt supplies such as airborne salt, seawater, and deicing agents. Chloride ions (Cl−) in salt (NaCl) passing through concrete promote steel corrosion in the concrete structures. Corrosion starts when the chloride ion concentration near the steel bars exceeds a marginal chloride ion concentration of 1.2 - 2.5 kg/m³ in accordance with the “Standard Specification [Design]”. If a chloride attack proceeds, eventually, a serious accident such as a collapse of a bridge will occur.

To prevent such serious accidents, it is important to know the chloride ion concentration distribution inside the concrete. However, measuring the chloride ion concentration distribution using existing methods requires taking sampling cores from structures and pre-processing them prior to measurement. In addition, it is not possible to obtain information concerning the time-dependent change at the same sampling position with such core sampling methods. Therefore, we propose an on-site non-destructive diagnostic technique using a neutron-captured prompt-gamma ray analysis (NPGA) combined with a movable compact neutron source. We are developing the proposed method at the RIKEN Accelerator-driven compact Neutron Source (RANS) [1–3].

2. Experimental Outline

2.1 RIKEN Accelerator-driven compact Neutron Source, RANS

Figure 1 shows the schematic structure of RANS, which consists of four parts: a proton linear accelerator, a target station, neutron guides, and a sample box. Protons are accelerated to energies of 7 MeV. The proton beam is pulsed, and the maximum proton current is 100 µA. The frequency and pulse width can be changed from 20 Hz to 200 Hz and from 10 µs to 150 µs, respectively. A 300-µm-thick beryllium (Be) target is placed at the center of the
Table 1 Summary of samples used in this study.

| Sample name | Sample size          | Condition            | Amount of $^{35}$Cl |
|-------------|----------------------|----------------------|---------------------|
| PVDC film   | $30 \times 30$ mm ($t = 100\mu$m) |                      | 50 mg               |
| Cement paste| $76 \times 57$ mm ($t = 50$ mm)     | NaCl concentration : 0, 10, 20 kg/m$^3$ | 0, 575, 1150 mg     |
| Mortar      | $40 \times 40$ mm ($t = 40$ mm)     | Cl concentration : 0, 0.3, 0.5, 1, 3, 5 kg/m$^3$ | 0, 15, 24, 48, 145, 242 mg |

Fig. 2 Diagram of a neutron-captured prompt gamma ray.

target station and is bombarded by the 7-MeV protons, and then neutrons are generated via the $^9$Be + p reaction. The neutrons are moderated via a 40-mm-thick polyethylene moderator. The moderated neutrons are extracted through two neutron guides and injected onto a sample in a sample box. The thermal neutron flux 5 m from the Be target is estimated to be approximately $1 \times 10^4$ n/cm$^2$/sec/100 $\mu$A.

2.2 Neutron-captured prompt-gamma-ray analysis, NPGA

NPGA uses the prompt-gamma rays emitted from an excited nucleus after a target nucleus captures a neutron, as shown in Fig. 2. Because each nucleus has specific gamma-ray energies and intensities, we can identify what elements exist in an object. For example, in the $^{35}$Cl (n, $\gamma$) $^{36}$Cl reaction, which is used in this study, primarily gamma rays of 517 keV, 786 keV, 788 keV, 1165 keV, 1951 keV, and 6110 keV are emitted from the excited states of $^{36}$Cl. Because the cross section of the thermal neutron capture reaction of chlorine is 10 times or more larger than the other elements in concrete, it should be relatively easy to detect in a concrete object even if the content of chlorine is small.

2.3 Samples and experimental setup

Polyvinylidene chloride (PVDC) films, cement paste samples, and mortar samples were tested. The PVDC has a chemical formula of $[C_3H_3Cl_2]_n$. The cement pastes were made from cement and water, and the mortar samples were made from cement, water, and sand. Both types of samples consist of CaO, SiO$_2$, Al$_2$O$_3$, Fe$_2$O$_3$, and H$_2$O. The salt densities in the cement pastes were controlled to be 0 kg/m$^3$, 0.3 kg/m$^3$, 0.5 kg/m$^3$, 1 kg/m$^3$, 3 kg/m$^3$, and 5 kg/m$^3$. Other properties of the samples, such as the size and condition, are summarized in Table 1.

Figure 3 shows the setup of the measurement for the NPGA at RANS. To detect gamma rays, two germanium (Ge) detectors were used. The neutron beam size was restricted to 50 mm × 50 mm by sintered a B$_4$C slit. The Ge detectors were shielded by Pb blocks, LiF tiles, and boron containing polyethylene (BPE) blocks.

3. Experimental Results

3.1 Detection sensitivity of chlorine

A PVDC film was used to confirm whether the NPGA was feasible at RANS by observing gamma rays with energies of 517 keV, 786 keV, 788 keV, and 1165 keV from the $^{35}$Cl (n, $\gamma$) $^{36}$Cl reaction. First, these energy peaks were identified, as shown by the solid arrows in Fig. 4 (a). The gamma rays were also detected from the bulk cement samples. As shown in Figs. 4 (b) and 4 (c), the peaks from $^{35}$Cl were observed for the samples with salt concentrations of 20 kg/m$^3$ and 10 kg/m$^3$. No peaks were observed in the spectrum of the sample without salt, as shown by the dotted arrows in Fig. 4 (d).

To determine the detection sensitivity of chlorine, we measured the mortar samples with varying chloride ion concentrations in the mortar samples were controlled to be 0 kg/m$^3$, 0.3 kg/m$^3$, 0.5 kg/m$^3$, 1 kg/m$^3$, 3 kg/m$^3$, and 5 kg/m$^3$. Other peaks were observed in the spectrum of the sample without salt, as shown by the dotted arrows in Fig. 4 (d).

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To determine the detection sensitivity of chlorine, we measured the mortar samples with varying chloride ion concentrations in an imitation of a concrete object. The varying chloride ion densities in the mortar samples were 0 kg/m$^3$, 0.3 kg/m$^3$, 0.5 kg/m$^3$, 1 kg/m$^3$, 3 kg/m$^3$, and 5 kg/m$^3$. Other peaks were observed in the spectrum of the sample without salt, as shown by the dotted arrows in Fig. 4 (d).

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γ) reaction are indicated with arrows. The chloride ion concentrations, measuring times, and average proton beam currents are given in the rectangular insets. The count rates of the peak areas at each chloride ion concentration are shown in Fig. 6 with statistical error bars. The results indicate that we can detect gamma rays for chloride ion concentrations of up to 0.3 kg/m³ excluding 6111 keV under the conditions at RANS. Because the detection sensitivity for the chloride ion concentration is sufficiently lower than the marginal chloride ion concentration for causing corrosion, approximately 1.2 - 2.5 kg/m³, non-destructive diagnoses should be feasible.

### 3.2 Estimating chlorine in an existing salt-damaged bridge sample

As a trial of the NPGA on an actual concrete structure, a brick taken from an existing salt-damaged bridge was analyzed. Pictures of the sample are shown in Fig. 7. The measured γ-ray spectrum is shown in Fig. 8. Via comparisons with the cement paste samples, the salt content in the bridge sample was estimated to be between 10 kg/m³ and 20 kg/m³ because the count rate of 1165 keV from the $^{35}$Cl(n, γ) reaction in the bridge sample is 0.72(11) cps.
while the equivalent count rates for the cement pastes with chloride ion concentrations of 10 kg/m$^3$ and 20 kg/m$^3$ are 0.55(7) cps and 0.98(7) cps, respectively. However, all the neutron and gamma-ray processes in the bulk samples need to be corrected due to the different volumes of the cement samples and the bridge sample.

4. Summary

We are studying a non-destructive diagnosis of the salt concentration distribution in concrete objects and have proposed a method using the NPGA and a compact neutron source. As a first feasibility study, we performed salt detection using the NPGA at RANS. Measuring mortar samples with varying chloride ion concentration, we observed a minimum chlorine detection sensitivity of 0.3 kg/m$^3$. This value was sufficiently lower than the marginal chloride ion concentration for causing corrosion in concrete structures. In addition, an existing salt-damaged bridge sample was tested. The salt content in this bridge sample was estimated to be between 10 kg/m$^3$ and 20 kg/m$^3$, even though it is necessary to correct all the neutron and gamma-ray processes in the bulk samples.

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