

A Study on the Non-Destructive Detection of Salt in Concrete Using Neutron-Captured Prompt-Gamma Rays at RANS^{*)}

Yasuo WAKABAYASHI¹⁾, Yuichi YOSHIMURA^{1,2)}, Maki MIZUTA¹⁾, Yoshimasa IKEDA¹⁾, Takao HASHIGUCHI¹⁾, Tomohiro KOBAYASHI¹⁾, Atsushi TAKETANI¹⁾, Shinzo YANAGIMACHI¹⁾, Makoto GOTO¹⁾, Hideyuki SUNAGA¹⁾, Yujiro IKEDA^{1,3)} and Yoshie OTAKE¹⁾

¹⁾RIKEN, RIKEN Advanced Photonics, 2-1 Hirosawa, Wako-shi, Saitama 351-0198, Japan

²⁾Tokyo Institute of Technology, Laboratory for Advanced Nuclear Energy, 2-12-1 Ookayama, Meguro-ku, Tokyo 152-8550, Japan

³⁾J-PARC Center, Japan Atomic Energy Agency, 2-4 Shirakata, Tokai-mura, Naka-gun, Ibaraki 319-1195, Japan

(Received 31 July 2017 / Accepted 31 March 2018)

We are studying the non-destructive detection of salt gradients in concrete using a neutron-captured prompt-gamma-ray analysis at the RIKEN Accelerator-driven compact Neutron Source (RANS). The neutrons at RANS are generated via a 7-MeV proton and 300- μm -thick beryllium target and are moderated via a polyethylene moderator. Samples, e.g., mortar samples with varying salt contents and part of an existing salt-damaged bridge, were irradiated by neutrons; then, the gamma rays following the neutron capture reaction were detected by germanium detectors placed near the samples. From the results, we obtained the detection sensitivity of the chloride content in a medium.

© 2018 The Japan Society of Plasma Science and Nuclear Fusion Research

Keywords: neutron capture, prompt gamma ray, compact neutron source, salt damage, non-destructive diagnosis

DOI: 10.1585/pfr.13.2404052

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/m^3 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

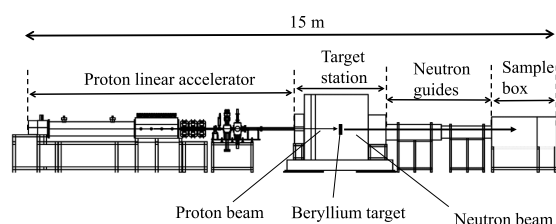


Fig. 1 Schematic structure of RANS.

author's e-mail: y.wakabayashi@riken.jp

^{*)} This article is based on the presentation at the Conference on Laser Energy Science / Laser and Accelerator Neutron Sources and Applications 2017.

Table 1 Summary of samples used in this study.

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

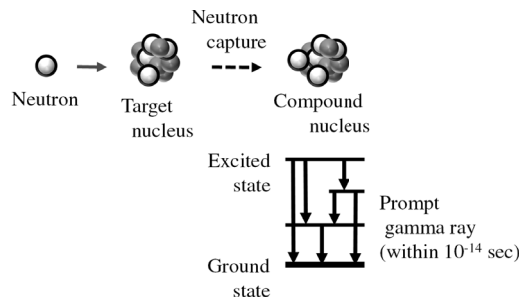


Fig. 2 Diagram of a neutron-captured prompt gamma ray.

target station and is bombarded by the 7-MeV protons, and then neutrons are generated via the $^9\text{Be} + p$ 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 $1 \times 10^4 \text{ n/cm}^2/\text{sec}/100 \mu\text{A}$.

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 $^{35}\text{Cl}(n, \gamma)^{36}\text{Cl}$ 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 $[\text{C}_2\text{H}_3\text{Cl}_2]_n$. 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_2O_3 , Fe_2O_3 , and H_2O . The salt densities in the cement pastes were controlled to be 0 kg/m³, 10 kg/m³, and 20 kg/m³ (= mg/cm³). The chloride

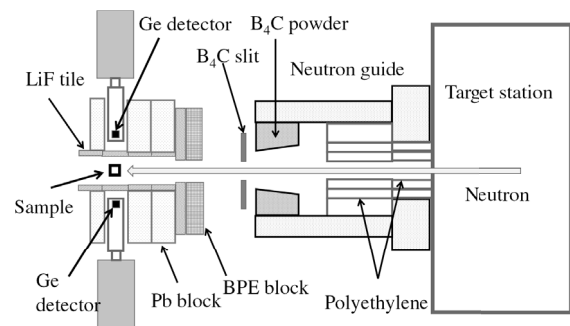


Fig. 3 Layout of the experimental setup.

ion concentrations in the mortar samples were controlled to be 0 kg/m³, 0.3 kg/m³, 0.5 kg/m³, 1 kg/m³, 3 kg/m³, and 5 kg/m³. 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 B₄C 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 $^{35}\text{Cl}(n, \gamma)^{36}\text{Cl}$ 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/m³ and 10 kg/m³. 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 as an imitation of a concrete object. The varying chloride ion densities in the mortar samples were 0 kg/m³, 0.3 kg/m³, 0.5 kg/m³, 1 kg/m³, 3 kg/m³, and 5 kg/m³. Figure 5 shows part of the obtained spectra of the measured mortar samples. The gamma-ray energies of 786+788 keV, 1165 keV, and 1951 keV from the $^{35}\text{Cl}(n, \gamma)^{36}\text{Cl}$

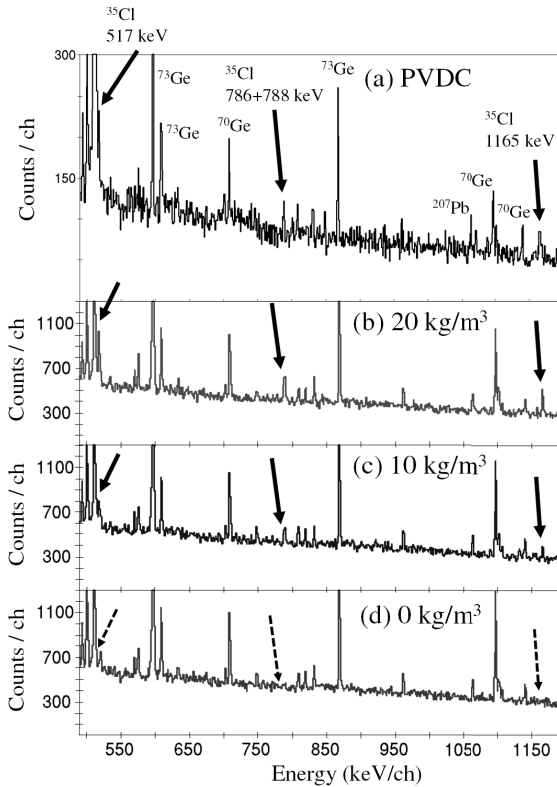


Fig. 4 Gamma ray spectra measured for (a) the PVDC film and (b - d) the cement pastes.

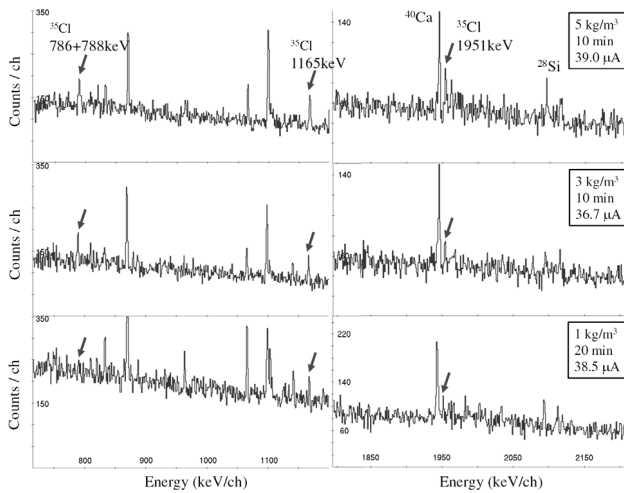


Fig. 5 Gamma ray spectra for the measured mortar samples with chloride ion concentrations of 5 kg/m³, 3 kg/m³, and 1 kg/m³.

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

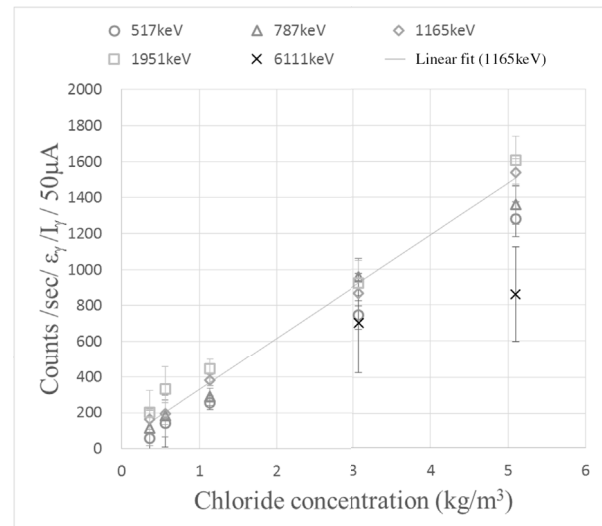


Fig. 6 Relationship between the chloride ion concentration of the mortar samples and the count rate of each gamma ray.

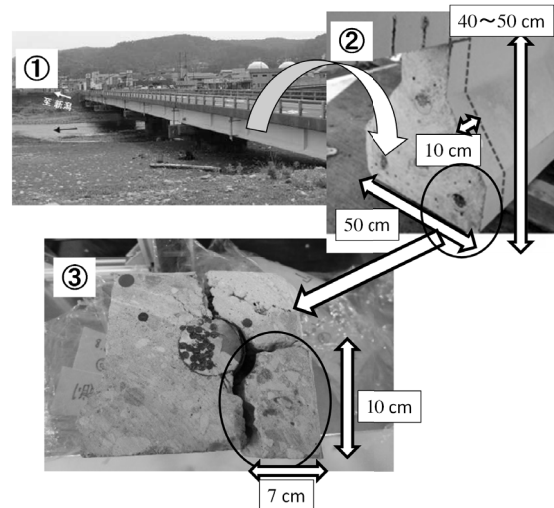


Fig. 7 Pictures of an existing salt-damaged bridge sample.

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 ³⁵Cl (n, γ) reaction in the bridge sample is 0.72(11) cps

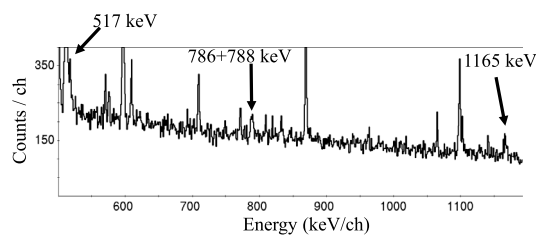


Fig. 8 Gamma ray spectrum obtained by measuring an existing salt-damaged bridge sample.

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.

Acknowledgments

This work was (partially) supported by Council for Science, Technology and Innovation (CSTI), Cross-ministerial Strategic Innovation Promotion Program (SIP), “Infrastructure maintenance, renovation and management” (Funding agency: JST) and was (partially) supported by Photon and Quantum Basic Research Coordinated Development Program from the Ministry of Education, Culture, Sport, Science and Technology, Japan.

- [1] Y. Yamagata *et al.*, J. Radioanal Nucl. Chem. **305**, 787 (2015).
- [2] Y. Otake (a partial auth.), M. Uesaka, H. Kobayashi, “Compact Neutron Sources for Energy and Security” Reviews of Accelerator-Science and Technology ‘Accelerator Applications in Energy and Security’ Vol. **08**, pp.196-198 (2015).
- [3] Y. Ikeda *et al.*, Nucl. Instrum. Methods A **833**, 61 (2016).