

# Development of cosmic-ray muon spin rotation radiography to investigate chemical and physical states of steels in large-scale architecture

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We propose a cosmic-ray muon spin rotation radiography to investigate physical and chemical status of steels in large-scale architectures based on the experimental results obtained for Fe and concrete by using cosmic-ray and accelerator muons. Spin polarized positive muons contained in the cosmic-rays were stopped in the Fe plates provide a characteristic spin rotation signal of decay positrons. Signals of decay electrons from negative muons stopped in Fe are separated from those of decay positrons, by nature of their different lifetimes in the samples. After subtraction of decay electrons, we have successfully extracted muon spin rotation with  $47.2 \pm 0.7$  MHz in frequency and  $8.7 \pm 4.6 \,\mu\text{s}^{-1}$  in relaxation rate. These values are consistent with those obtained from reference experiment using intensive muons provided by an accelerator, verifying validity of analysis. This method is quite promising to investigate steels at near-surface (20 cm thick from the surface).

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# 1. Introduction

In an earthquake zone called the Circum-Pan-Pacific Earthquake Belt, large-scale architectures such as buildings, bridges, highway roads, dams are exposed to a danger of big earthquakes. By 2023, 43 % of bridges (~171000) and 34% of tunnels (~3000) on highways in Japan exceed their durable lifetime of 50 years after construction [1]. They have been exposed to severe natural environment and hard wear long years. In these architectures, aging effects such as fatigue, damage and corrosion are worried to cause serious deterioration and accidents by these earthquakes. Under these situations, it is urgently required for national and world-wide safety precautions to develop a nondestructive inspection method to explore inner status of architectures.

In architectures constructed by the iron reinforced concrete (RC) method, a sign of corrosion appears typically as cracks of concrete surface by aging effects on steels. For further enforcement, the pre-stressed concrete (PC) method has been developed in the middle of 19th century, and has been extensively used in last several decades. In the PC method, a sheath covering steels restrain progress of corrosion. However in PC, a sign of corrosion such as cracks of concrete is hard to be observed until when steels inside are seriously deformed or broken; i.e. architectures made by PC method shows few precursor before serious destruction.

For inspection of the inner structure of the large-scale architectures, a use of cosmic-ray muon is quite effective to observe large scale objectives and landforms [2]. The structure of the Second Pyramid of Giza was confirmed to have no hidden chambers by measuring directional variation of cosmic-ray muons incident to the detectors installed in the Pyramid [3]. It took, however, a quarter of century to get started a real application of cosmic-ray muon radiography by using near-horizontal cosmic-ray muons whose intensity is two orders of magnitude lower than vertical ones [4]. By measuring the positional distribution of the intensity attenuation of the penetrating cosmic-rays, the inner-structure of the world's largest blast furnaces of the Nippon Steel Corporation have been studied during a time of its full operation [5], based upon the experiences of radiography of the volcanic mountains [4, 6]. Investigations of the inner status of the damaged Fukushima nuclear reactors are in progress by using multiple-scattering method of cosmic-ray muons [7, 8] along with the transmission radiography [9]. However, it is hard to detect chemical as well as physical properties of steels by transmission or multiple scattering methods. The use of polarized cosmic-ray muons was proposed for examination of the physical and chemical status of steel rods in reinforced concrete in which a characteristic spin rotation pattern was observed in the time evolution of decay positron emitted from the cosmic-ray muons stopped in the iron plates by the test experiment [10].

In order to verify the potential application of the cosmic-ray muons for nondestructive materials analysis in large-scale architectures, we performed a series of reference experiments in various Fe and steel rods, and a concrete block by using intensive decay muons produced from accelerator. Based on detailed analysis of the cosmic-ray muon measurements with reference to those, we propose a new method of cosmic-ray muon spin rotation radiography.

### 2. Cosmic-ray muon spin rotation radiography

### 2.1. Nature of cosmic-ray muons and decay electrons/positrons

Cosmic-ray muons at the sea level of the earth are known to have the following properties; a characteristic Zenith-angle dependent energy spectrum with common natures of high energy

(GeV to TeV) and low intensity  $(1/(cm)^2/min in vertical direction)$  [11], a mixed charge state (60%  $\mu^+$  and 40%  $\mu^-$ ) and a partial spin polarization (-0.33 for  $\mu^+$  [12]). Because of high energy nature, a stopping distribution of the cosmic-ray muon in concrete extends from cm to a few km in depth.

After the cosmic-ray muons stopped in material,  $\mu^+$  and  $\mu^-$  takes a different history [3]. The  $\mu^-$  is a heavy electron-like elementary particle interacting with surrounding atoms and molecules electromagnetically. Once it is stopped inside the material, a small atomic state called muonic atom is formed around the nucleus, where a radius is approximately  $270/Z \times 10^{-13}$  cm for light nuclei. Since the ground state of the muonic atom reached in sub-ns after  $\mu^-$  injection has a radius close to that of the nucleus for heavy nuclei, a weak interaction between nucleons and muon becomes effective so that the negative muon becomes captured to nucleus through the elementary process of  $\mu^- + p \rightarrow n + \nu_\mu$ , competing with the free decay at the orbit of the muonic atom of  $\mu^- \rightarrow e^- + \nu_\mu + \nu_e$ . Thus, mean lifetime of  $\mu^-$  takes a Z-dependent change.

On the other hand, the  $\mu^+$  takes a slowing-down and stops at the microscopic interstitial site at the material crystal. During the slowing-down, spin polarization of  $\mu^+$  is negligibly small. After stopping, spin polarized  $\mu^+$  causes  $\mu^+SR$  signal by feeling a characteristic microscopic magnetic field at the interstitial site [6].

Most of decay e<sup>-</sup> and e<sup>+</sup> from muons stopped deep inside of objectives are absorbed because of their low initial energy below 50 MeV. Decay e<sup>+</sup> and e<sup>-</sup> emitted from muons stopped near surface of objectives within about 20 cm can be coming out to the detectors placed in front of the objectives. Thus, physical and chemical status of near-surface steels can be selectively investigated. From view point of mechanical strength of pre-stressed concrete, these outermost steels near surface play essentially important role supporting total stresses predominantly.

# 2.2. Principle of the cosmic-ray muon spin rotation radiography

By detecting a signal of the decay  $e^-$  and  $e^+$  whose energies are up to 50 MeV from various selected position of the stopping muons near surface of the objective, one can obtain a radiographic imaging information such as a spatial distribution of microscopic magnetism from the  $\mu^+$  spin rotation ( $\mu^+$ SR) signal and an element distribution from the amplitude distributions of components with different  $\mu^-$  lifetime due to different capture rates of nucleus [13].

Time evaluation of mixed decay  $e^+/e^-$  signal N(t) is detected with reference to the time of stopping of the incoming cosmic-ray muons for an area of each stopping position.

N(t) can be written as follows,

$$N(t) = N_0 \left[ \alpha e^{-t/\tau(0)} \left\{ 1 + \sum_i A_i e^{-\lambda(i)t} \cos(2\pi \nu_i t + \phi_i) \right\} + (1 - \alpha) \sum_j P_j e^{-t/\tau(j)} + C \right]$$
(1)

where  $\alpha$ : a fraction of positive muon in the cosmic-ray,  $\tau(0)$ :  $\mu^+$  lifetime,  $A_i$ : decay positron asymmetry parameter in the *i*th material including  $\mu^+$  polarization,  $v_i$ :  $\mu^+$  spin precession frequency due to internal field of the *i*th material,  $\lambda(i)$ : relaxation rate of the  $\mu^+$  spin precession due to the *i*th internal field,  $P_j$ : fraction of the  $\mu^-$  stopping in the *j*th material and  $\tau(j)$ :  $\mu^-$  mean lifetime in the *j*th material, respectively.

Fig. 1 shows a proposed set-up for the muon spin rotation radiography [10]. The stopping position can be obtained by the direction of the incoming cosmic-ray muons determined by a telescope of more than two arrays of the segmented counters [6]. Tracking hodoscope of muon passage is placed in front of the decay  $e^+/e^-$  counter. The hodoscope determines muon stopping position in steels and concrete separately by using different  $\mu^-$  mean lifetimes. It enables us to focus on  $\mu^+SR$  in steels.

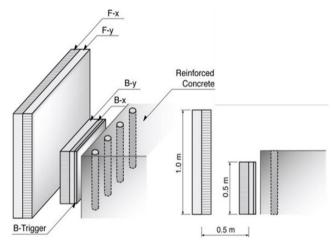


Fig. 1 Proposed set-up for the cosmic-ray muon spin rotation radiography using decay e<sup>+</sup>/e<sup>-</sup> from the stopped cosmic-ray muon. Muon beam tracking hodoscope is placed in front of decay e<sup>+</sup>/e<sup>-</sup> to determine muon stopping position [10].

Suggested procedure is as follows; a) unit segmentation of the objective volume of muon stopping by radiography counters by detecting muon incoming direction and decay  $e^+/e^-$ ; b) selection of the iron containing volume units by lifetime analysis for the decay of the stopped  $\mu^-$  and c) spin rotation analysis of that iron containing volume unit.

# 2.3 Test experiment

As described in the previous report [10], time evolution of the decay e<sup>+</sup> and e<sup>-</sup> from the cosmic-ray muons stopping in Fe plate of regular 99.3 % in purity (2 cm thick) and in concrete block of commercial use (6 cm thick) was measured separately, by using the test set-up and logics

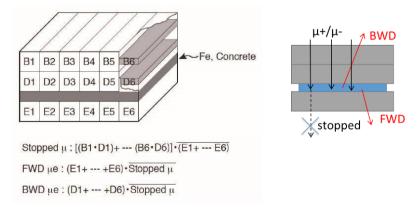


Fig. 2 Counter geometry and logics of decay  $e^+/e^-$  detection from stopped muons in the test experiment. Inside each counter box, a plastic scintillator with the size of 3 cm thick, 10 cm wide and 100 cm long is located [10].

shown in Fig. 2. Each counter is a plastic scintillator with a size of 10 cm width 100 cm long and 3 cm thick. There, the data for the FWD (BWD) direction where decay e<sup>+</sup>/e<sup>-</sup> were detected along (opposite to) the direction of incoming vertical cosmic-ray muons were presented.

The time evolution of detected e<sup>+</sup>/e<sup>-</sup> signals in FWD geometry is shown in Fig. 3 (a) for Fe plate (red) and concrete block (blue). It is clearly seen the difference of lifetime between Fe and concrete due to nuclear capture in heavy elements, and precession signal with about 50 MHz which is typically observe in  $\mu^+$ SR experiment in pure Fe [14]. Preliminary analysis was reported in ref. 6 by using Eq. 1. By detailed analysis in this study,  $\mu^+$  and  $\mu^-$  signals in Fe were successfully separated to 66% and 34% respectively. We corrected distortion that is commonly seen in time spectra before 100 ns and slowly relaxing background before extraction of  $\mu^+$ SR signal in Fe. Fig. 3 (b) shows the asymmetry spectra of  $\mu^+$  spin rotation in Fe, obtained by dividing  $\mu^+$ SR signal in Fig. 3(a) by natural decay term. Solid line shows fitting function by using the following equation,

$$A(t) = A_2 e^{-\lambda_2 t} \cos(2\pi \nu_{Fe} t + \phi) + A_1 e^{-\lambda_1 t} + B$$
 (2)

where  $A_2$ ,  $\lambda_2$ ,  $\nu_{Fe}$  and  $\phi$  is amplitude, relaxation rate, precession frequency and initial phase of precessing component and  $A_1$  and  $\lambda_1$  is amplitude and relaxation of relaxing component, B is constant background respectively. Precession frequency  $\nu_{Fe}$  is  $47.2 \pm 0.7$  MHz and relaxation rate  $\lambda_2$  is  $8.7 \pm 4.6 \,\mu\text{s}^{-1}$ . The relaxation of precessing signal is one order of magnitude larger than that observed for highly purified single crystalline Fe with 99.995 wt% purity in which precession frequency is 50MHz and relaxation rate is 0.80  $\mu\text{s}^{-1}$  [14].

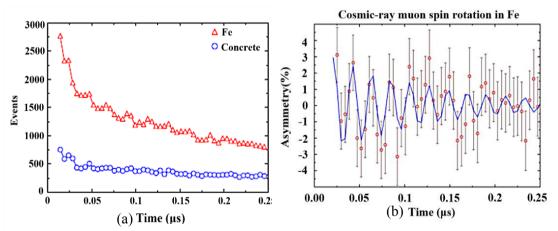


Fig. 3 Time evolution of cosmic-ray muon spin rotation in Fe (46 days of measurement time) and concrete (20 days): (a) observed zero-field spin rotation time spectrum, (b) asymmetry spectrum of  $\mu^+SR$  signal extracted from spectrum (a) for Fe plates. Solid line is fitting by Eq. 2.

# 3. Positive and negative µSR experiment by accelerator muons

As a reference for the cosmic-ray muon experiment, a series of  $\mu$ SR experiments have been conducted for the Fe plate and concrete block by using positive and negative muons at the General Purpose Decay-Channel Spectrometer (GPD) beam line in the Paul Scherrer Institut (PSI).

## 3.1. Negative muon experiment in Fe and concrete

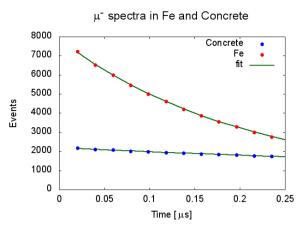


Fig. 4 Time evolution of decay e<sup>-</sup> detected by BWD geometry in Fe plate with 99.3% in purity (6.4h) and in concrete (3.2h). Solid lines are fitting by using mean lifetimes described in the text.

Fig. 4 shows the negative muon spectra observed for a piece cut from the Fe plate for cosmic-ray muon experiment (red) and a piece of concrete (blue). A life of  $\mu^-$  was confirmed as 206.5  $\pm$  0.3 ns in Fe. In concrete blocks, on the other hand, three component with different lives are decomposed from the spectra; i.e.  $\tau_i$ : 1737  $\pm$  97 ns, 787  $\pm$  532 ns, and 326  $\pm$  76 ns, corresponding to nominal values of 1795.4  $\pm$  2.0 ns in oxygen, 756.0  $\pm$  1.0 ns in silicon and 864.0  $\pm$  1.0 ns in aluminum, and 332.7  $\pm$  1.5 ns in calcium, respectively [15]. These results will be utilized to analyze cosmic-ray muon data in concrete which consist of both negative and positive muons.

# 3.2. Positive muon experiment in Fe and steels

Fig. 5 shows the asymmetry spectrum observed in the same Fe sample with that in Fig. 4. Precession frequency and relaxation rate is  $47.42 \pm 0.30$  MHz and  $6.0 \pm 0.1 \mu s^{-1}$  respectively. These values are coincident well with those extracted by detailed analysis of the cosmic-ray muon data within the error, supporting the validity of the experimental data and analysis method in cosmic-ray muon experiment.

As shown in Fig. 6, precessions with about 50 MHz in frequency were also confirmed in various steel bars of RC (a, c), and PC (b, d), while relaxation rates are an order of magnitude larger than Fe in this experiment. It is remarkable that RC steels manufactured by different companies shows similar relaxation, while relaxation is much faster in PC than that in RC even manufactured by the same company. These results imply that  $\mu^+SR$  is more sensitive to residual

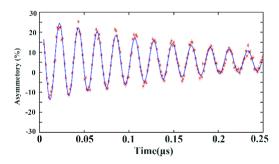


Fig. 5 Time evolution of BWD/FWD asymmetry observed by  $\mu^+SR$  in Fe with 99.3% in purity. Solid line is fitting by eq. 2.

tension applied in the process than composition. At the same time, feasibility of cosmic-ray muon spin rotation method is strongly supported for practical application to investigate chemical states of steels bars in RC and PC. Details with respect of effect of impurity, residual tension and defects will be discussed elsewhere.

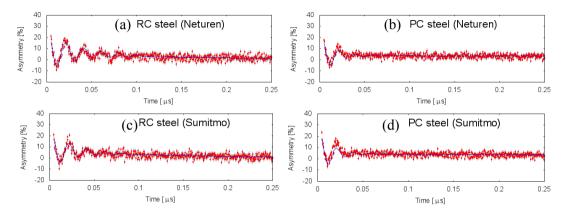


Fig. 6 Time evolution of BWD/FWD asymmetry observed by  $\mu^+SR$  in various steel bars commercially available and used in large-scale architectures. Solid lines are fitting by eq. 2

# 4. Conclusion

We demonstrated potential application of the cosmic-ray muon spin rotation method to nondestructive investigation of the large-scale architecture consisting of steels. Parameters of precession frequency and relaxation rate determined by the analysis of the cosmic-ray  $\mu^+$  spin rotation for Fe were precisely confirmed by accelerator muon experiment, showing validity of analysis method.

These results support feasibility of the muon spin rotation radiography proposed by Nagamine et al for imaging physical and chemical status of steel bars in PC and RC to increase efficiency as well as position sensitivity[10].

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