Integration of arts and sciences using negative muon non-destructive analysis at J-PARC MUSE

Y. Miyake[1], M. Tampo[1], S. Takeshita[1], I. Umegaki[1], P. Strasser[1], S. Doiuchi[1], A. Hashimoto[1], K. Shimomura[1], T. Kutsuna[2], T. Saito[3], K. Kubo[4], and K. Ninomiya[5]

[1]High Energy Accelerator Research Organization (KEK), Institute of Material Structure Science (IMSS), Muon Science Laboratory (MSL)
[2]National Museum of Nature and Science,
[3]National Museum of Japanese History
[4]International Christian University
[5] Institute Radiation Sciences, Osaka University

E-mail: yasuhiro.miyake@kek.jp

Abstract. We have developed a non-destructive, position-selective, and quantitative elemental analysis method using negative muons at J-PARC MUSE. Our final goal is to establish a platform for integrating arts and science, where historical heritage can be analyzed non-destructively by combining quantum beam techniques utilizing muons, neutrons, or photons. At the symposium, the status of the negative muon non-destructive analysis held at J-PARC MUSE on old Japanese coins (Koban, Cho-gin), bell-shaped bronze vessels of the Yayoi period, and old horse harnesses will be introduced.

1. Introduction
Hold your palm up to the sky. Subatomic particles called muons fall from the sky on the palm at a frequency of 1 muon/second. These cosmic ray muons have a high penetrating power and are used to take images of mountains to help predict eruptions, search for hidden spaces in huge pyramids, and observe nuclear fuel debris melted down in nuclear reactor accidents. This paper introduces non-destructive elemental analysis using accelerator muons artificially produced at J-PARC. These muons are more than a few million times more intense than cosmic-ray muons and can be applied in archeology.

2. Pulsed muon source at J-PARC
We installed a superconducting decay/surface muon channel with a modest-acceptance (approximately 40 mSr) pion injector, the so-called D-line, at J-PARC in 2008. This channel allows us to extract the world's strongest pulsed muons. In 2015, we replaced the old superconducting magnet with a new superconducting magnet with a warm bore (inner bore radius of ϕ20 cm); therefore, no beam window was used to extract lower-momentum-intense negative muons; otherwise, they stopped at the beam windows. Consequently, negative muons from 2.6 MeV/c to 120 MeV/c are available at the D-line, corresponding to an implantation depth that can be controlled from 0.2 μm to 62 mm for carbon samples.

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3. Negative muon

Muons have no internal structure and belong to the second generation (standard model) of charged leptons; their mass is one-ninth that of protons, and they have antiparticle (positive muons: μ^+) and particle (negative muons: μ^-) forms. Because negative muons are 200 times heavier than electrons, their orbits are 200 times closer to the nucleus than those of electrons (Fig. 1).

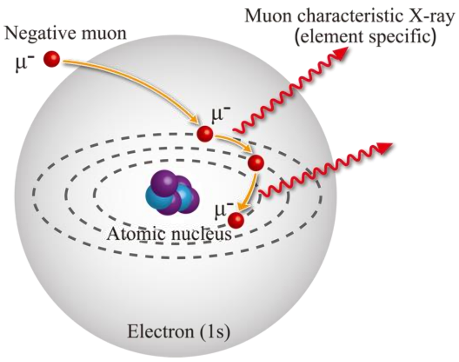

Figure 1. A negative muon is 200 times heavier than electron, and its orbits is 200 times closer to the nucleus than that of electrons

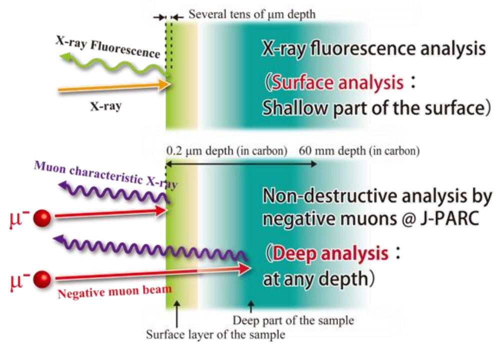

Figure 2. X-ray fluorescence analysis is limited to elemental analysis near the measured object's surface (10 – 100 μm depth) because X-rays have a low penetrating power. The negative muon non-destructive method at J-PARC MUSE allows us elemental analysis at any depth, from near the surface (~200 nm) to as deep as 60 mm (carbon)

4. Non-destructive analysis by negative muons at J-PARC MUSE

When a negative muon is injected into matter, it loses its kinetic energy and is captured by the Coulomb field of the nucleus, forming a muonic atom. A muonic atom is an atomic system with one negatively charged muon instead of an electron. As soon as the muonic atom is formed at a highly excited level, such as n~15, the captured muon immediately de-excites to lower atomic muon levels by emitting characteristic muonic X-rays with energies unique to each element of the matter. Because the muon mass is 207 times heavier than that of an electron, the muonic atom level is 207 times deeper than the corresponding electronic atom level. Thus, muonic X-rays are more difficult to capture than electronic X-rays. For copper, the muonic K_α X-ray energy is approximately 1500 keV, while that of the electronic K_α X-ray is only 8 keV. Electronic X-rays cannot pass through a 100-μm-thick Cu foil, whereas muonic X-rays can easily penetrate Cu plates with a thickness of several millimeters. In other words, the muonic X-ray method can analyze the interior of a bulk sample. Moreover, since the injection energy of the decay muons produced at J-PARC MUSE can be controlled by adjusting a bending magnet, muons can be stopped at any depth. Therefore, the muonic X-ray method can be considered a unique 3D bulk elemental analysis technique that combines a 2D imaging device or beam scanning, particularly for non-destructive archeological analysis, which would integrate the arts and science.

5. Application for the element analysis of historical heritage

The analysis of rare, sometimes unique, historical heritage must be performed in a non-deforming, non-destructive manner. Therefore, X-ray fluorescence analysis is often used. However, X-ray fluorescence analysis is limited to elemental analysis near the surface (10-100 μm depth) of the measured object owing to the low penetrating power of X-rays. On the other hand, the negative muon non-destructive method allows elemental analysis at any depth, ranging from near the surface (~200 nm) to as deep as 60 mm (carbon) (Figure 2) at J-PARC MUSE. Because of its high penetrating power and lower self-absorption, it can detect elements lighter than sodium, such as lithium and carbon, which are not sensible by conventional X-ray fluorescence elemental analysis. It is important to note that historical relics can be measured in air and vacuum without damaging valuable samples.We conduct three types of archaeological research categories (Fig. 3) at J-PARC MUSE based on the advantages of these unique features [1].

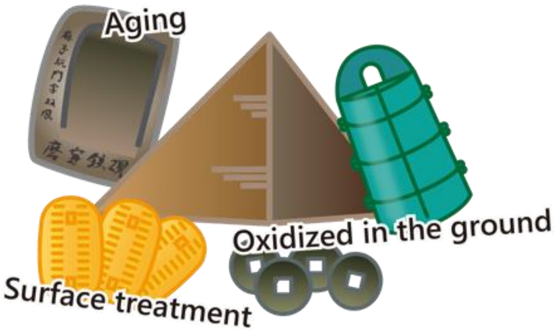

Figure 3. Three kind of research categories using negative muons have been conducted in the archeology at J-PARC MUSE.

5.1 Research investigating in the deep inside

The first category is research that investigates the deep inside of historical materials. A glass pharmaceutical bottle left by Ogata Koan, a treasure at Osaka University, had been in storage for so long that the glass lid had stuck and could not be removed. The negative muon non-destructive analysis method was chosen as the best way to identify the contents of the glass bottles without breaking them. The principle is straightforward: a beam is controlled to pass through the glass wall and stop only at the reagent, allowing non-destructive elemental analysis of the bottle's contents. We concluded that the medicine in the glass cap was mercury sulfate (Hg_2Cl_2) [2].

Another notable study in the first category is the chemical analysis of bronze objects. Bronze objects, such as bronze bells and mirrors, are often excavated from the ground, and sometimes tin is removed or rusted on the surface. In our experiments, the depth dependence of oxygen was examined. Then, by analyzing the depth at which the effects of oxygen (that is, rust) can be avoided, the chemical compositions of tin, lead, and copper at the time of manufacture can be determined, which was previously thought to be impossible. This analysis allows us to obtain archaeological indicators to determine the time, place, and raw materials used in producing these objects. There has been a theory that medieval bronze objects such as sutra case and bronze mirrors were made from northern Song coins that were in circulation at the time and that the people donated these coins. To verify this theory, experiments were conducted on a sutra case and a bronze mirror excavated from the ruins of the Dogaya Abandoned Temple in Shizuoka Prefecture without the influence of rust. The results showed that the compositional ratios of tin, lead, and copper in the excavated bronze artifacts differed significantly from those of the Song dynasty coins, indicating that the theory of the original Northern Song dynasty coins may not be valid [3]. Further verification, including isotopic analysis, is required in the future.

5.2. Research on the artificially surface-treated historical heritage

The second category is research involving artificial surface-treated historical heitage. Gold coins made of gold/silver alloys and silver coins made of silver/copper alloys, which were manufactured and circulated from the Warring States period to the end of the Edo period, have been studied through the creation of an "IRO AGE" technique that improves their appearance. The non-destructive analysis is a technique that can be used to determine the depth dependence of elemental composition from near the surface to a depth of several millimeters without destroying rare koban and cho-gin coins. By quantitatively clarifying the changes in color-finishing technology using non-destructive analysis methods, essential knowledge can be obtained to infer the political and economic conditions and sociocultural trends of the time [4]. Recently, research has begun to analyze the coins of the Ryukyu Dynasty and continental Japan for studying the spread of culture beyond Japan.

5.3. Research historical relics suffering from aging

The third category, which has just begun, is research on historical materials that have deteriorated or have been altered over time. Negative muon non-destructive analysis was used to determine whether a black deposit found on a stone product excavated from the Tawa-yama site in Matsue City, Japan, which is suspected to be an inkstone, was the first inkstone ever found in Japan. Although it was found that a carbon component had seeped into the stone to a depth of 10 μm, there is a cautious opinion that it was not ink but some other material [5]. Various alternative research methods have been used, and this debate continues.

6. Conclusion

The High Energy Accelerator Research Organization, Institute for Materials Structure Science (KEK-IMSS), has been developing a new non-destructive analysis method that has the potential to be used for research on historical heritage and other humanities materials. This method uses the world's most intense pulsed muon beam at MUSE, along with the beam's unique features. We have been developing new non-destructive analytical methods that have the potential to be used for research on historical and other humanities materials. The symposium has been held six times since 2019, bringing together researchers of cultural heritage materials who use quantum beams such as synchrotron radiation and neutrons to introduce archaeological research, related research, and analytical techniques. The symposium explores the possibilities of integrating humanities and science research. We hope that further integration of arts and sciences using quantum beams can extend to the rest of the world.

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