Development of elemental analysis by muonic X-ray measurement in J-PARC

K Ninomiya¹, T Nagatomo², K M Kubo³, P Strasser³, N Kawamura³, K Shimomura³, Y Miyake³, T Saito⁴ and W Higemoto¹
¹ Japan Atomic Energy Agency, Tokai, Naka, Ibaraki, 319-1195, Japan
² International Christian University, Mitaka, Tokyo, 181-8585, Japan
³ High Energy Accelerator Research Organization, Tsukuba, Ibaraki, 305-0801, Japan
⁴ National Museum of Japanese History, Sakura, Chiba, 285-8502, Japan

E-mail: ninomiya.kazuhiko@jaea.go.jp

Abstract. Muon irradiation and muonic X-ray detection can be applied to non-destructive elemental analysis. In this study, in order to develop the elemental analysis by muonic X-ray measurement we constructed a new X-ray measuring system in J-PARC muon facility. We performed muon irradiation for Tempo-koban (Japanese old coin) for test experiment of elemental analysis. Muonic X-rays originating from muon transition in muonic silver and gold atoms were identified. The contents of Tempo-koban (Au:61%) was determined by muonic X-ray intensities.

1. Introduction

When a free negative muon stopped in matter, the muon was captured in the Coulomb field of a nucleus and a muonic atom is formed. The captured muon deexcites to the 1s state through Auger electron and muonic characteristic X-ray emissions. The energies of muonic X-rays are higher than these of electronic X-rays because of a large mass of a negative muon (105.7 MeV/c², 207 times larger than that of electron). Such high energy muonic X-rays (0.1-6 MeV) are emitted from bulk samples without photon self-absorption, and the source elements of these muonic X-rays are easily identified using a semiconductor detector. The depth of muon implantation in sample can be tuned by adjusting the incident muon momentum. In addition, the samples are hardly activated by muon irradiations. These are very important advantages for elemental analysis compared with other methods (e.g. X-ray fluorescence and prompt gamma-ray analysis by neutron irradiation). It is expected that muonic X-ray spectroscopy can be applied to non-destructive elemental analysis [1, 2].

In spite of such advantages, this technique has not been used widely. There are few fundamental studies for development of this technique because this method is hardly known in the field of analytical sciences. In our present knowledge, we are not able to determine the elemental contents of sample material quantitatively only from muonic X-ray intensity because of the molecular effects on the formation process of muonic atoms. Muonic X-ray intensities and muon atomic capture ratios depends on the muon-capturing molecule. Although some muon capture models were studied [4-6], the molecular effects on the formation process have still been unclear. Therefore, it is essential to know the relation between muonic X-ray intensities and material contents from muonic X-ray spectra for the standard samples that components have already known. In this study, to develop elemental
analysis by muonic X-ray measurement and to investigate the molecular effects on the muonic atom formation, we constructed a new X-ray measuring system in J-PARC.

2. Experimental

The system for muonic X-ray measurement was constructed at D2-area in Muon Facility (Muon Science Establishment; MUSE) in Materials and Life Sciences Facility (MLF), J-PARC (see Figure 1). This system consisted of radiation shields, beam collimator with 8.5 cm diameter, two germanium detectors and MCA in CAMAC system. The primary proton beam intensity during the present work was about 20 kW. We selected the momentum of negative muon as 35 MeV/c. For muonic X-ray measurement, photo events that correlated with muon pulse signals were detected by germanium detectors. We optimized experimental conditions such as detector position, arrangement of radiation shields, the beam transport optics and muon stopping region in zinc plate with 0.5 mm thickness and 10 x 10 cm$^2$. Muon irradiation for Tempo-koban (provided from National Museum of Japanese History, old Japanese coin produced in the middle of the 19th century) was also performed about 4 hours for test experiment of elemental analysis. The sample dimension of the Tempo-koban was about 0.5 mm thickness and 6.5 x 4 cm$^2$ with an oval shape. The range of incident muon in the Tempo-koban was estimated as ~0.3 mm. The detection efficiencies of germanium detectors were determined from EGS-5 calculations [7] in which parameters were adjusted to reproduce experimental efficiencies of $^{137}$Cs and $^{60}$Co standard radiation sources.

3. Results and discussion

3.1. Muon irradiation for zinc plate

Figure 2 shows a typical muonic X-ray spectrum by muon irradiation for zinc plate. Muonic K, L, M and N X-ray lines were clearly found. Muonic X-ray intensities per muon capture were estimated from cascade calculation [8]. In this calculation, we gave the well accepted initial parameter as follows; the initial principal quantum number is equal to 14 with statistical initial angular momentum distribution and L and M electrons are never ionized completely by muon-electron and electron-electron Auger processes during muonic cascade. In this way, we estimated that 15 muons per pulse
were stopped in the zinc plate from muonic 2-1 X-ray intensity. The consistent values were obtained from analysis of 5-4, 4-3 and 3-2 muonic X-ray intensities.

![Zinc X-ray spectrum](image1)

**Figure 2.** Muonic X-ray spectrum for zinc plate. The muon irradiation time was 5500 s with 20 kW primary proton power (total incident muon: $2 \times 10^6$). The numbers in each bracket show a change of principal quantum numbers by muonic X-ray transition.

3.2. **Muon irradiation for Tempo-koban**

Muonic silver and gold X-rays were identified by muon irradiation for the Tempo-koban (see Figure 3). This result shows the Tempo-koban is silver-gold alloy. We analyzed the number of stopped muons per muon pulse by the same method of zinc experiment from muonic silver 4-3 and muonic gold 5-4 X-ray lines. These muonic X-ray lines had good statistics and well separated from other peaks. The muon capture ratio in gold atom is estimated as 0.59 ± 0.06.

![Tempo-koban X-ray spectrum](image2)

**Figure 3.** Muonic X-ray spectrum for Tempo-koban (measuring time: 14000 s).
The atomic muon capture ratio ($A(Z_1/Z_2)$) for silver-gold alloy is essential to determine the contents of the Tempo-koban by muonic X-ray intensities. However, the accurate atomic capture ratio has not been reported. We estimated the atomic capture ratio for silver-gold alloy as $A(Au/Ag) = 1.68$ from the Z-law by Fermi and Teller ($A(Z_1/Z_2) = Z_1/Z_2$ [4]). From the muon atomic capture ratio and the number of captured muons in silver and gold atoms, the ratio of atoms in Tempo-koban was determined as $I(Ag/Au) = 1.17$, $Au = 61 \pm 6 \%$ ($Ag = 39 \%$) in elemental mass ratio. This result does not include the systematic errors originating from muonic X-ray intensity per captured muon (cascade calculation) and atomic capture ratio for silver-gold alloy (muon capture model).

Our result is consistent with the result for well known contents of Tempo-koban ($Au: 57\%$) and for the same Tempo-koban that was analyzed from prompt gamma-ray analysis by neutron irradiation in J-PARC NOBORU ($Au: 54 \pm 1\%$) [9].

4. Conclusion

We developed a muonic X-ray measuring system for elemental analysis at MUSE in MLF, J-PARC. Low background muonic X-ray spectra were successfully obtained by extracting correlation photo events with muon beam pulse, and the contents of the Tempo-koban were determined from muonic X-ray intensities without destruction.

In the next step, we are planning to perform muon irradiation for silver-gold alloys that are already known their contents. From these studies, atomic capture ratio and muonic X-ray intensity pattern will be understood. As a result, the systematic errors will be decrease and more accurate elemental analysis will be available. We are also planning to investigate muon capture phenomena for various materials and establish the muon capture model to determine muonic X-ray intensities per captured muon and atomic capture ratios for sample materials without muon irradiation for standard samples.

5. References

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