Elemental Analysis with Negative Muons

M. Kenya KUBO
International Christianity University,
Mitaka, Tokyo, Japan

(KEK-IMSS/J-PARC) M. Tampo, K. Hamada, N. Kawamura, P. Strasser, K. Shimomura, S. Doiuchi, T. Nagatomo, I. Umegaki, S. Takeshita, Y. Miyake, K. Nishiyama (KEK) G. Yoshida (JAEA) T. U. Ito, W. Higemoto, T. Osawa, S. Sakamoto , K. Ninomiya (Osaka U.) K. Ninomiya, G. Yoshida, M. Inagaki, K. Terada, M. Uesugi,T. Kudo, A. Shinohara, A. Sato, D. Tomono, Y. Kawashima, A. Nambu (Hokkaido U.) S. Tachibana (Kyoto U.) A. Tsuchiyama, M. Inagaki (TMU) M. Ebihara (U. Tokyo) T. Takahashi, S. Takeda, S. Nagasawa (JAXA) S. Watanabe, M. Katsuragawa (Tohoku U.) T. Nakamura, D. Wada (Nat. Mus. Jap. History) T. Saito (Toyota Central R&D Lab) I. Umegaki, H. Nozaki, Y. Higuchi, J. Sugiyama, (Hiroshima U.) K. Ninomiya

2024.10.17 The 4th J-PARC Symposium 2024

Outline

Introduction

- Non-destructive, depth-selective, multi-elemental analysis
- Negative muon and muonic atom
- Elemental analysis method with muonic X-ray
- Muon induced nuclear reaction and gamma-ray

Experimential

- J-PARC Muon facility (MUSE)
- Experimental setup

Examples

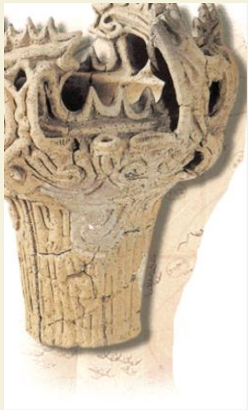
- Chinese bronze coin and mirror, Japanese gold coin
- Extra-terrestrial sample
- Li-ion battery
- Japanese sword (lifetime method)

Summary

Elemental Analysis of Relics, Ancient Arts, Handiworks etc.

Elemental composition and distribution reflect

- Techniques
- Raw materials and trade
- Transition of techniques

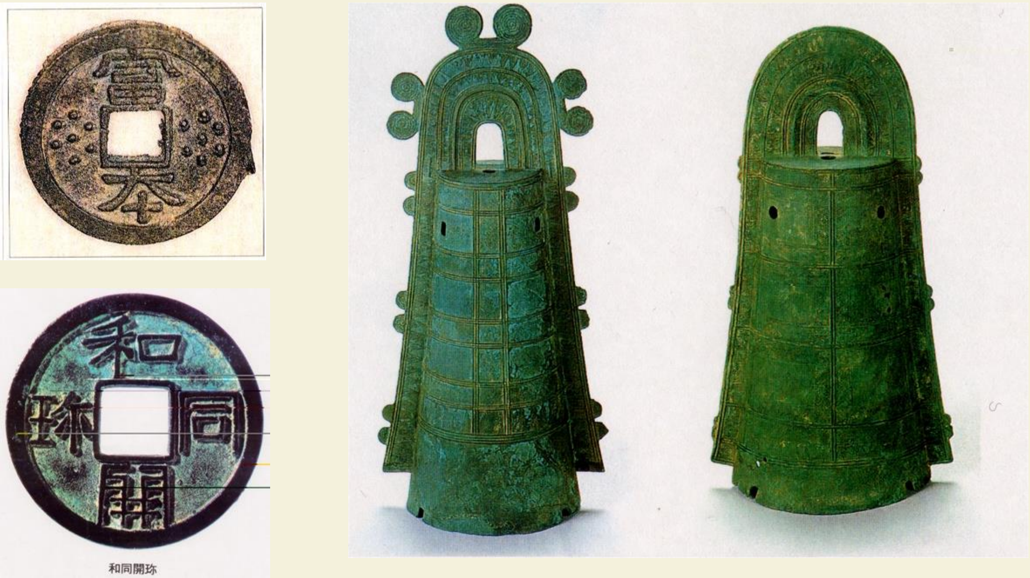


Sampling may damage materials.

Analysis is not practical.

Necessity of depth-selective, non-destructive, multi-elemental analysis

Rusted or modified surface has different composition from the inside.



Bronze coins, bells

Plated statue

Typical elemental analysis methods

ICP-AES, ICP-MS

highly sensitive, quantitative,
needs sample destruction

XPS, PIXE

position sensitive(2D), material surface analysis,
poorly quantitative for light elements

Neutron activation analysis (NAA and PGA)

isotope analysis, bulk analysis, very high sensitivity for some nuclides, no depth-profiling ability

SIMS

microanalysis, destructive

Required conditions

- ☆Non-destructive
- ☆Multi-elemental
- ☆Depth-selective

Muonic X-ray method meets these conditions.

What is muon?

Elementary particle

- Charge -*e*
- Spin：1/2
- Lifetime：2.2 μs

$$\mu^- \rightarrow e^- + \bar{\nu}_e + \nu_\mu$$

- No strong interaction
- Mass: 106 MeV/c^2

~207 m_e、~1/9 m_p

Muons in material

μ^+ : light proton

μ^- : heavy electron

Leptons

e	μ	τ
ν_e	ν_μ	ν_τ

Muon production

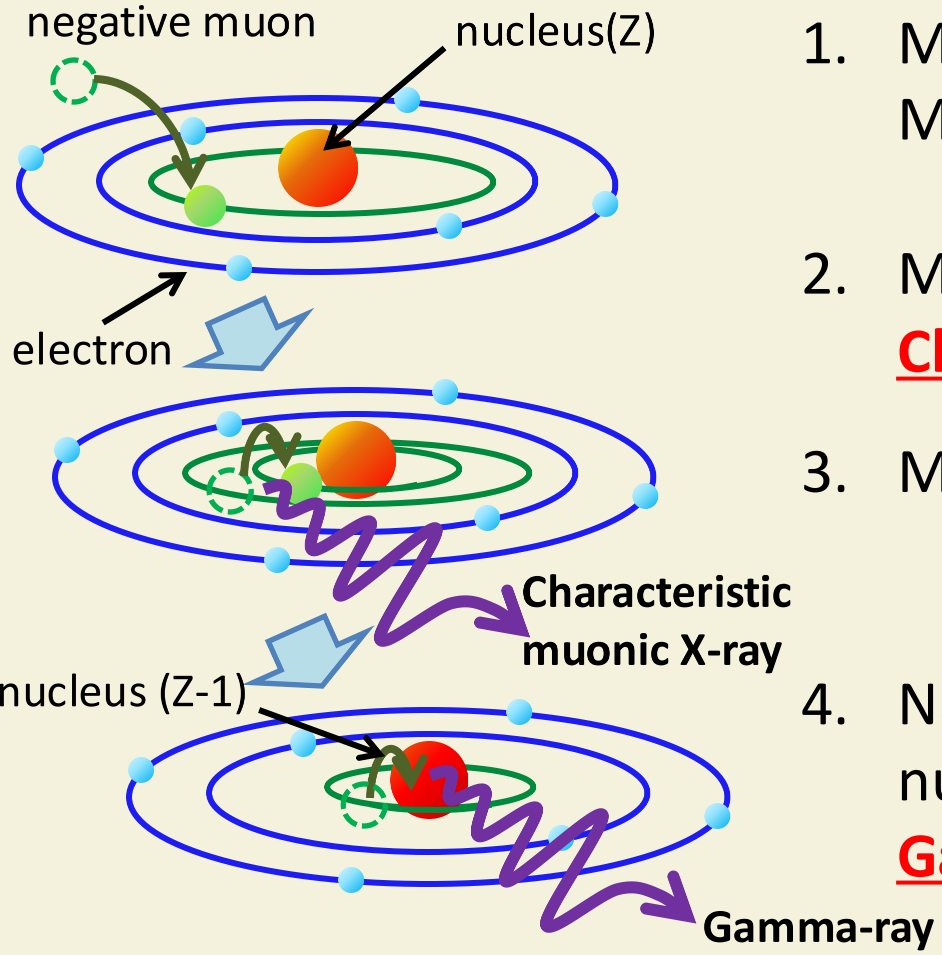
$$\pi^- \underset{26\text{ ns}}{\rightarrow} \mu^- + \overline{\nu_\mu}$$

π^{+-} are produced by

p^+ (≥ 280 MeV) + nucleus

Negative muon and muonic atom

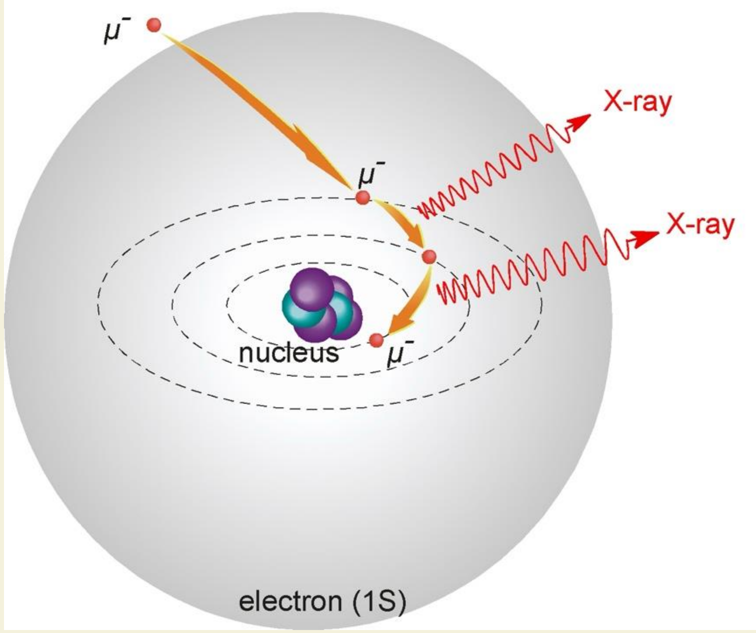
Muonic atom formation and following processes



1. Muonic atom formation
   Muon capture in atomic muonic orbital
2. Muon cascading process
   <u>**Characteristic muonic X-ray emission**</u>
3. Muon in muonic 1s state (50-20000 ns)
4. Natural decay or muon capture in the nucleus
   <u>**Gamma-ray emission**</u>

What is muonic X-ray?

Characteristic X-ray emitted during de-excitation process of muonic atom



$$E_n = -\frac{Z^2 m e^4}{8 n^2 \varepsilon_o^2 h^2}$$

$$r_n = -\frac{4\pi\varepsilon_o n^2 \hbar^2}{Z m e^2}$$

$$\frac{m_\mu}{m_e} \approx 207 \approx \frac{E_\mu}{E_e} \approx \frac{r_e}{r_\mu}$$

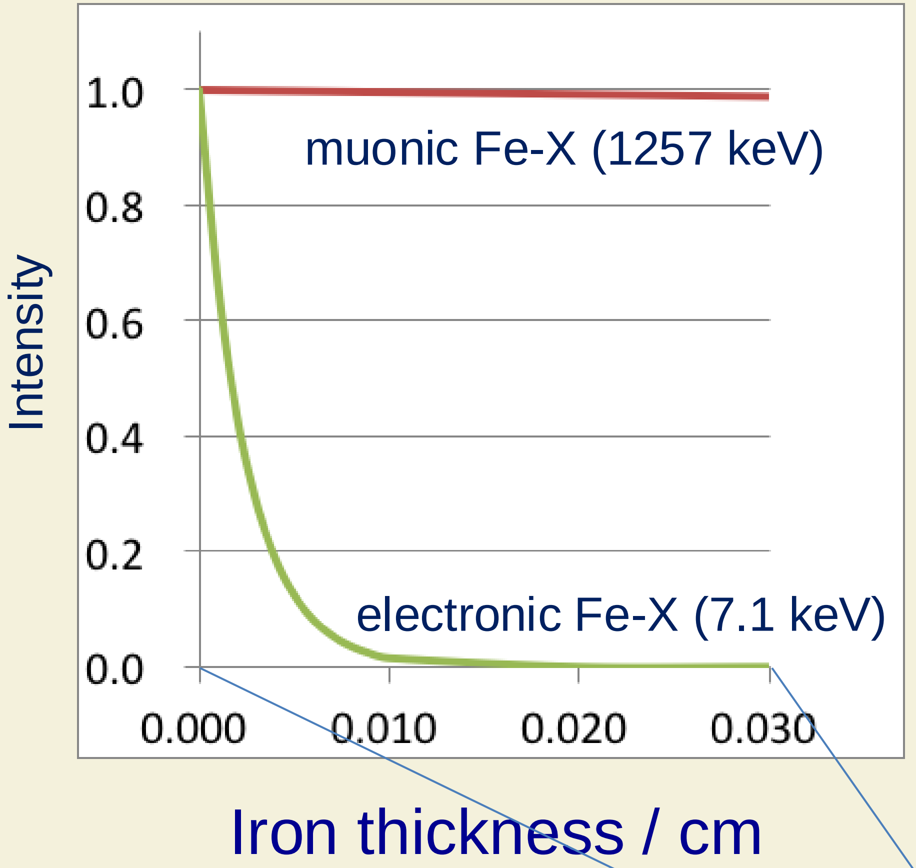
Bohr model

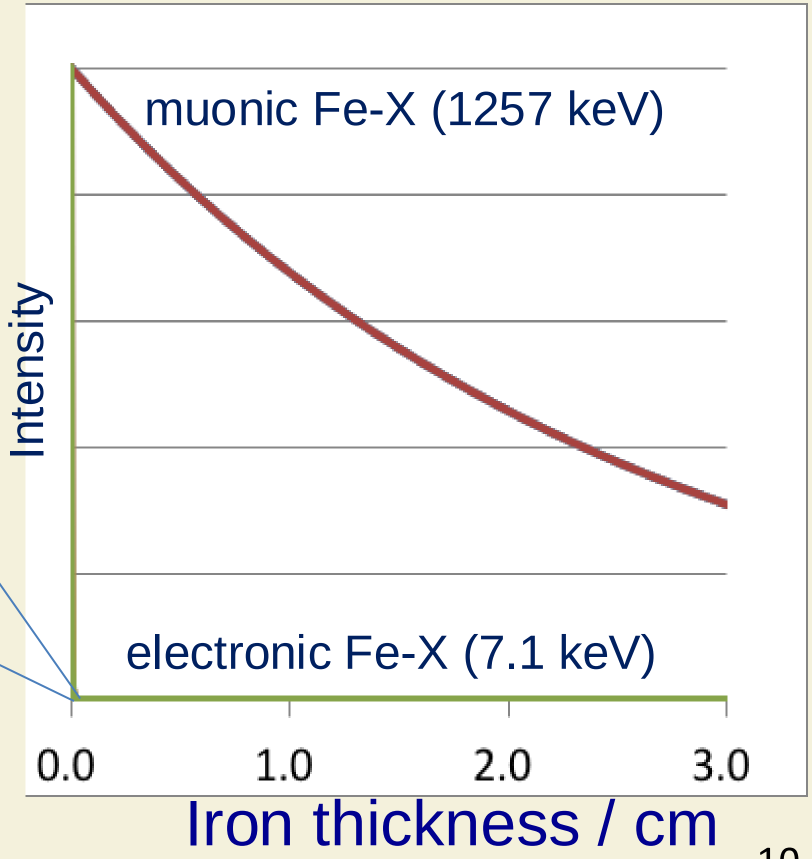
Muonic X-ray Energy（keV）

Element	Kα	Kβ	Lα	Lβ
C	76	89	14	19
O	134	158	25	34
Al	347	422	66	89
Fe	1256	1704	264	357
Cu	1513	2126	307	444
Pb	5966	8466	938	1372

~200 times larger than the corresponding electronic X-ray

Electronic and Muonic X-ray attenuation in Iron



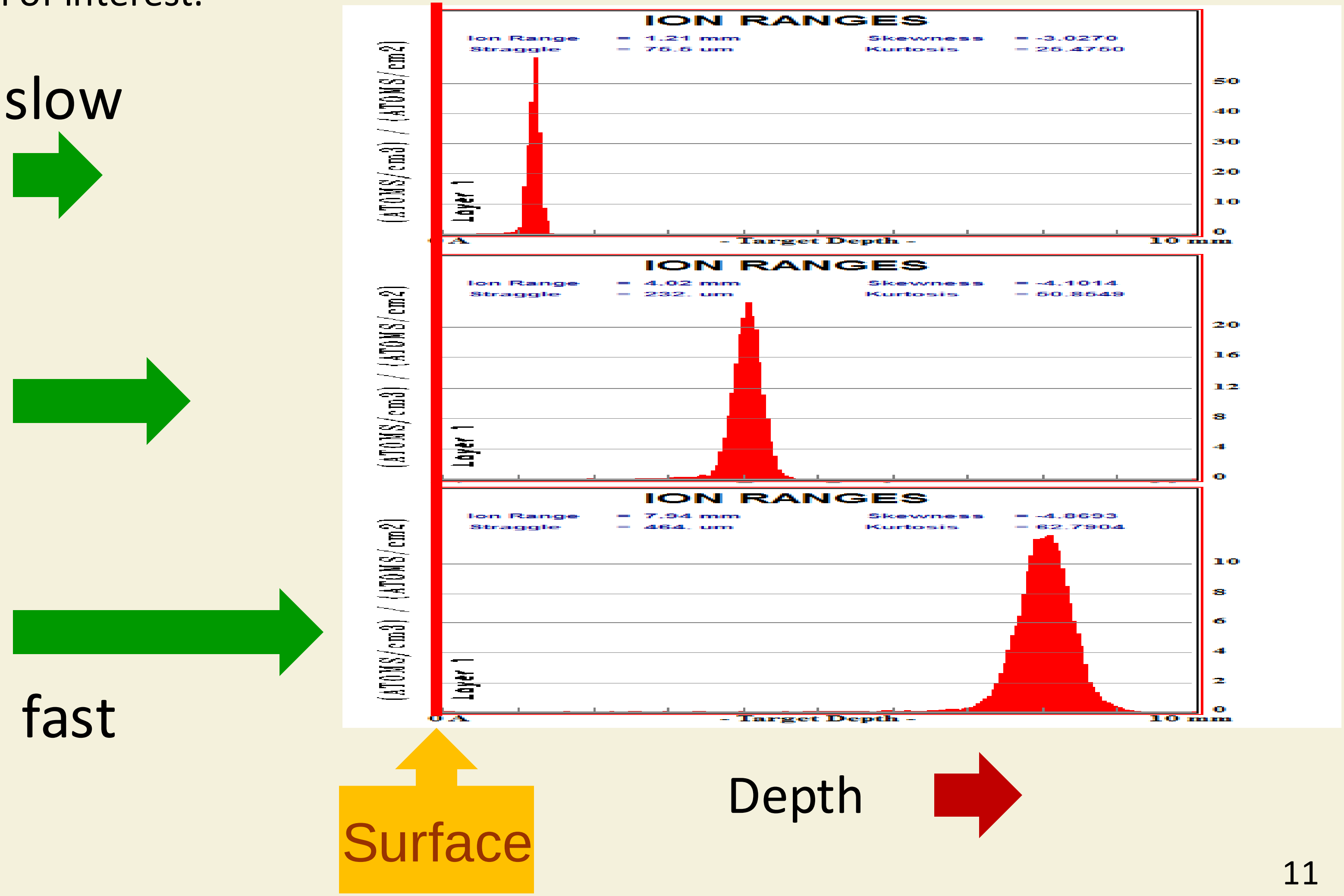


Muonic X-ray penetrates

a few cm thick Iron.

Stopping depth of muons

Muons with a variety of momentum are produced at the same time. We choose and use muons of proper momentum depending on the depth of interest.



Elemental analysis with Negative Muons

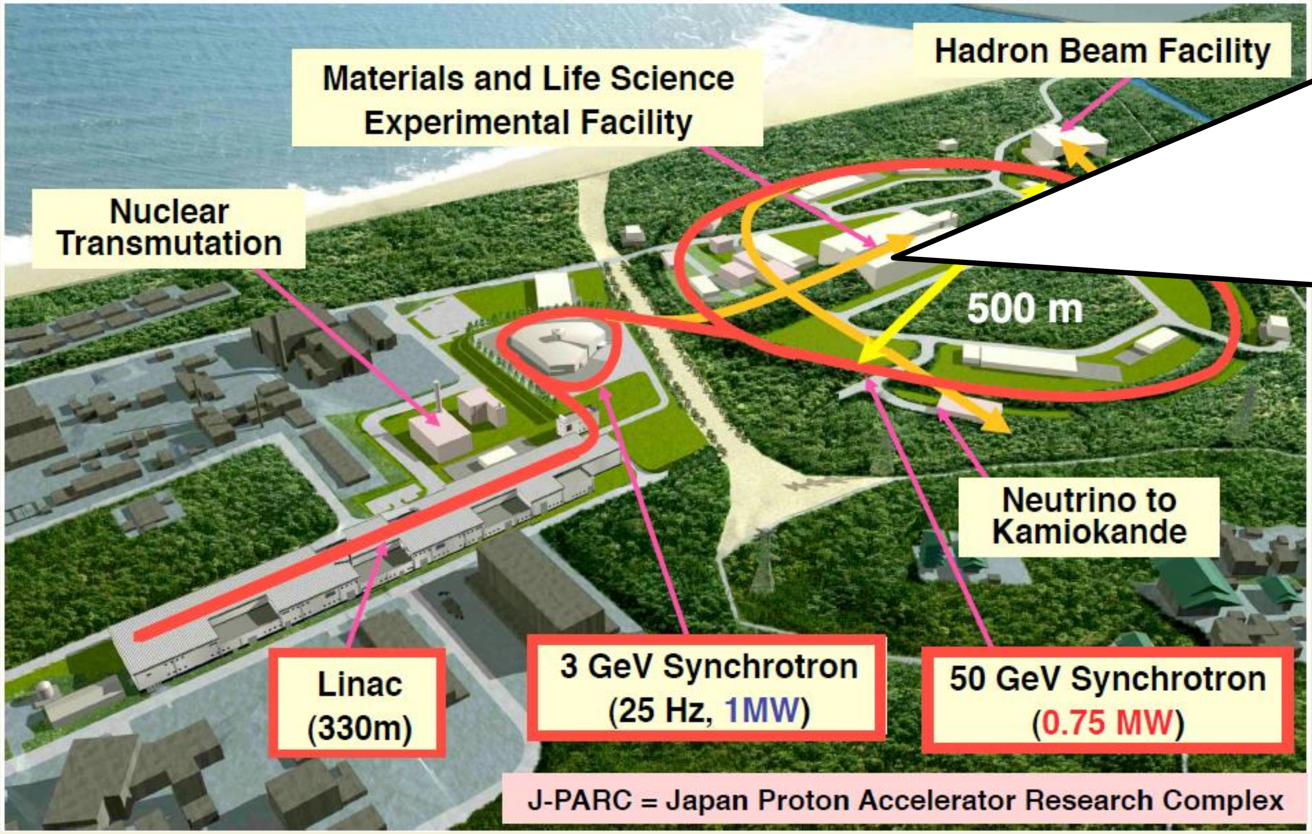
Formation of muonic atom.
Emission of characteristic muonic X-ray with specific energy to the element. **More than one muonic-Xray by one muon**.

- Applicable to every element **Multi-elemental, simultaneous**
  no need of previous knowledge
- 10 keV to 10 MeV
  Observable from outside of sample **Deep inside**
  No need of vacuum -> applicable to **Huge/Porous/Bio samples**
- No chemical process
  damageless **Nondestructive**
- Depth selective by changing incident energy, easy beam scan
  Site selective, 3D mapping

< Capture probability: proportional to *Z* with small chemical effects >

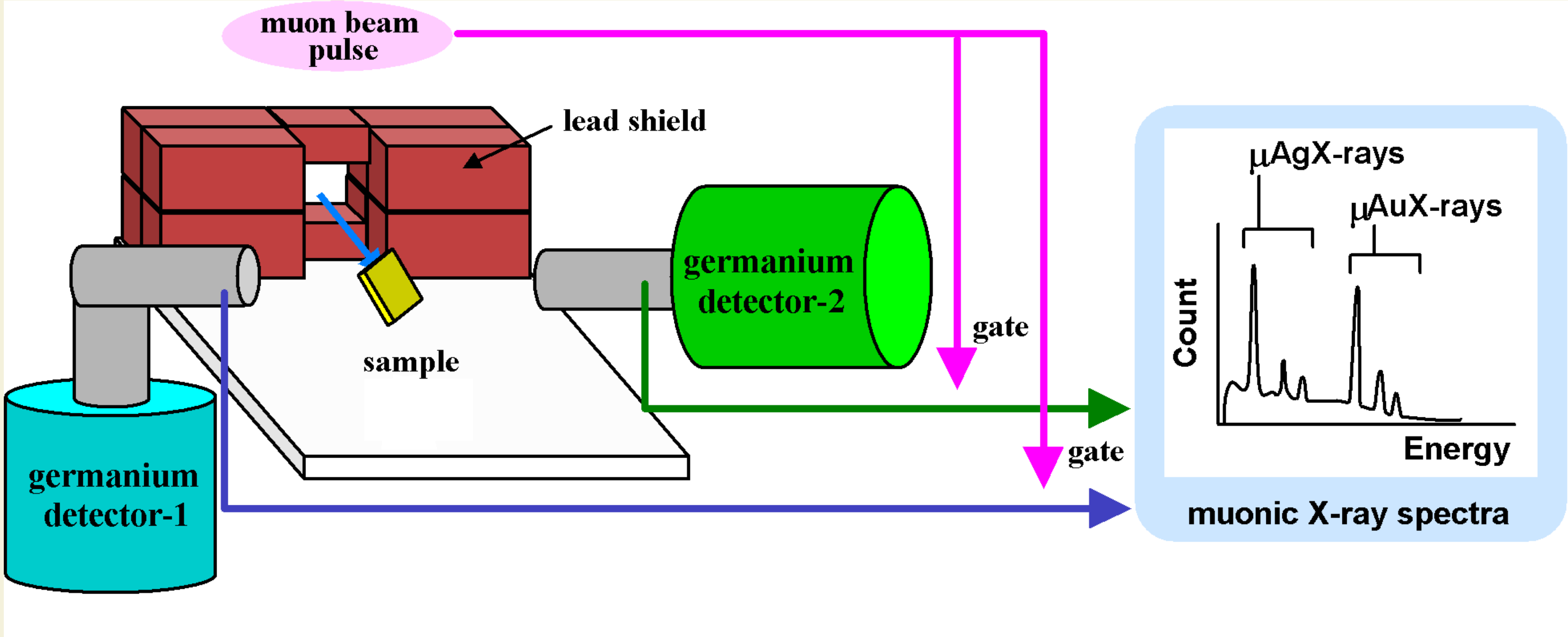
J-PARC muon facility

Experiments were performed at D2-area in J-PARC/MUSE, Tokai, Ibaraki, Japan



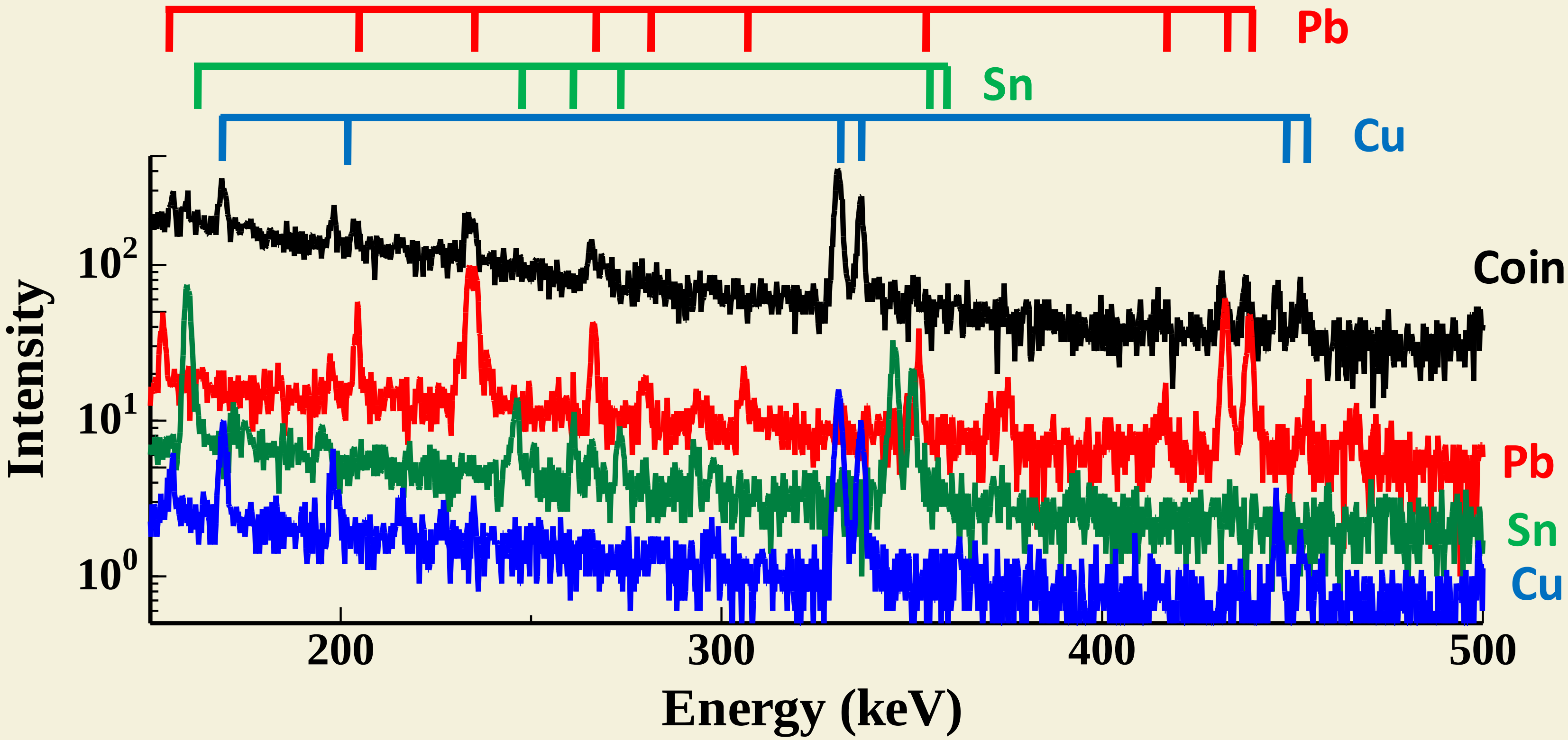
MLF, Muon Science Establishment (MUSE), D2-area

Experimental setup



Experimental setup at J-PARC/MUSE D2-ares in 2009

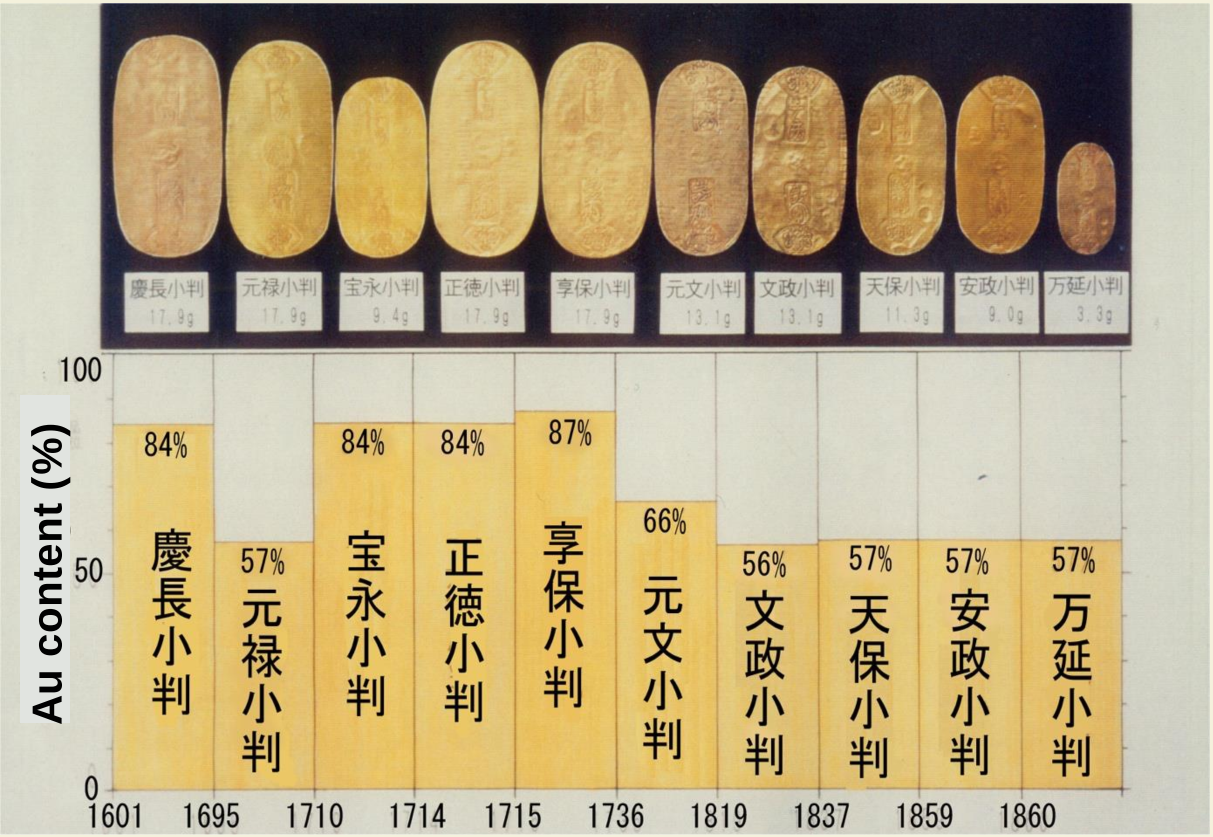
Chinese bronze coin



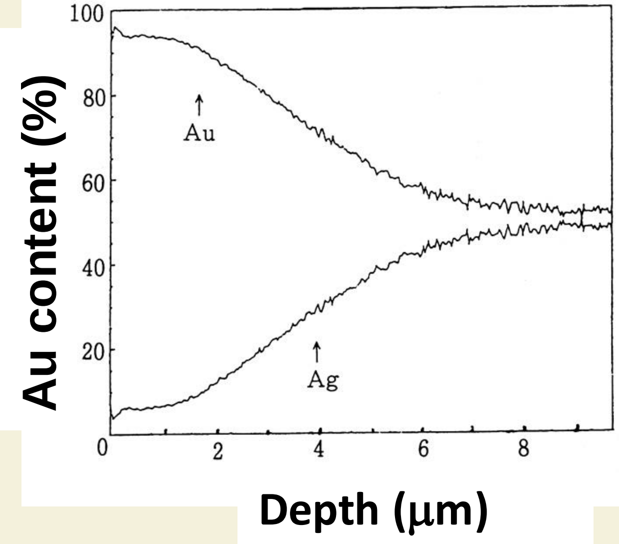
Muonic X-ray spectra of a Chinese Bronze Coin (55000s) and Pb, Sn and Cu metal.

Sensitivity (in weight) is almost same for all elements.

Japanese Gold Coin (Edo Period, 1601 - 1867)



Tempo Koban



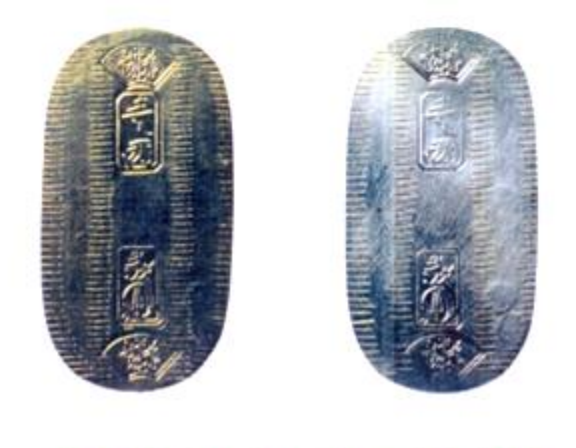
Tempo Koban

Auger Electron Spectroscopy

Chemcal surface treatment of gold coin

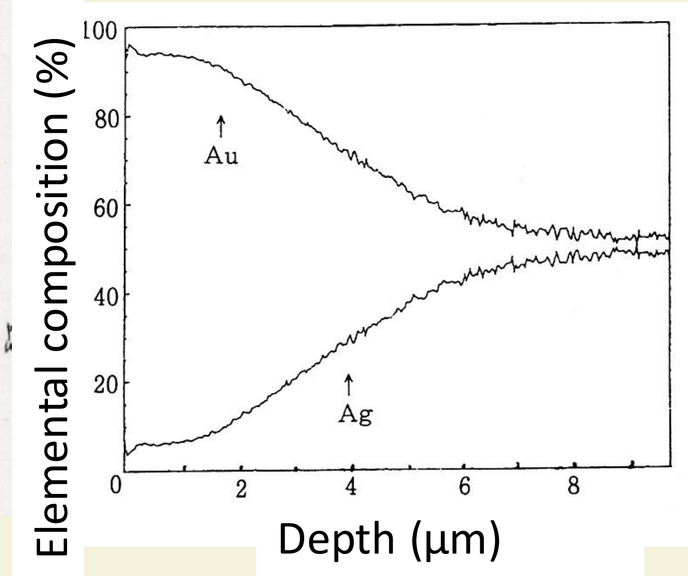
Iroage (色揚げ, color dressing)

Elemental composition differs depending on depth



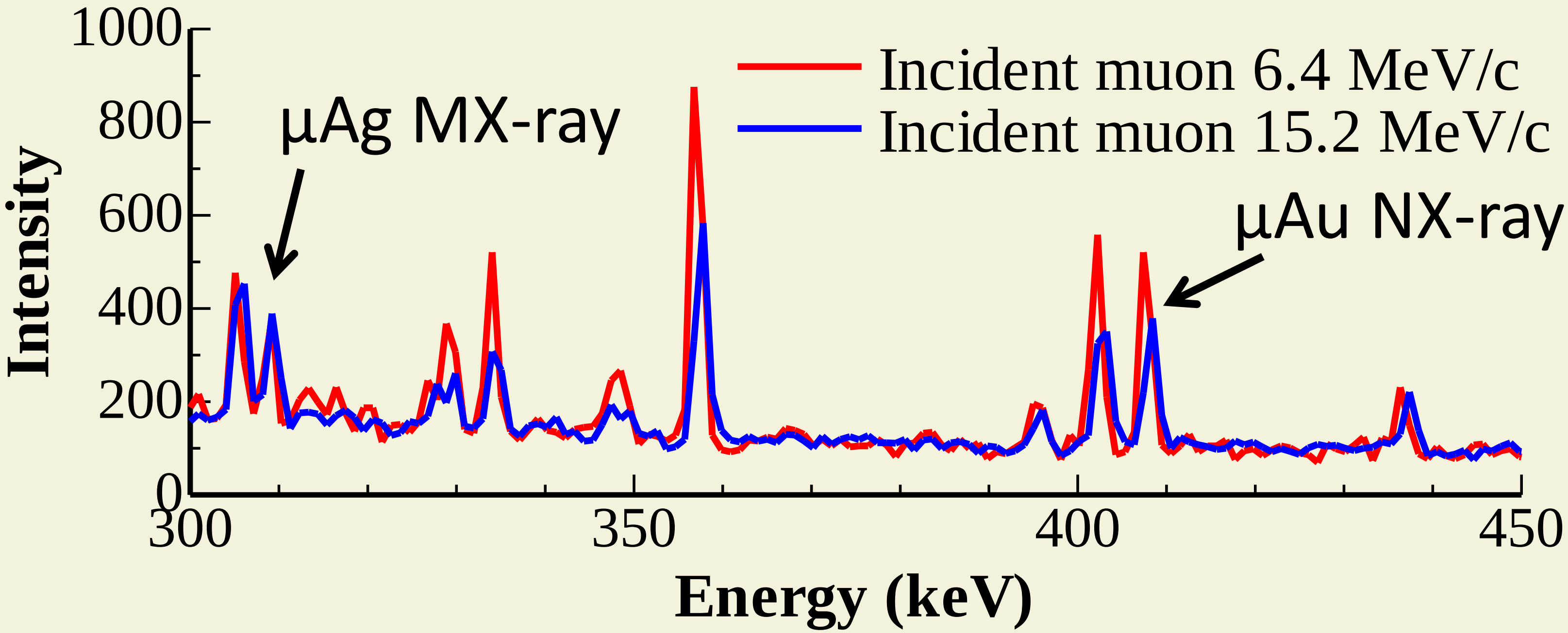
色揚げ前（右）と後（左）の小判（複製）

小判の製造工程一色揚げ（小判所絵図）



Gold coin analysis by muonic X-ray

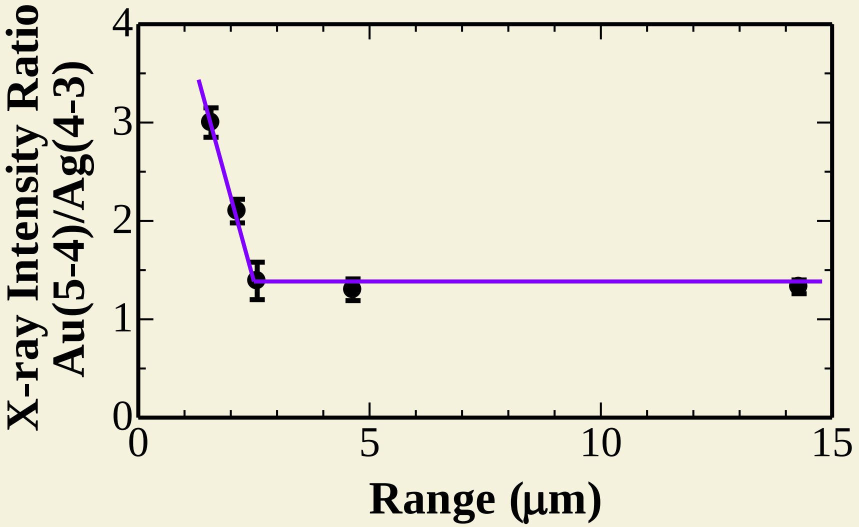
Muonic X-ray spectra for gold coin with different incident muon momenta

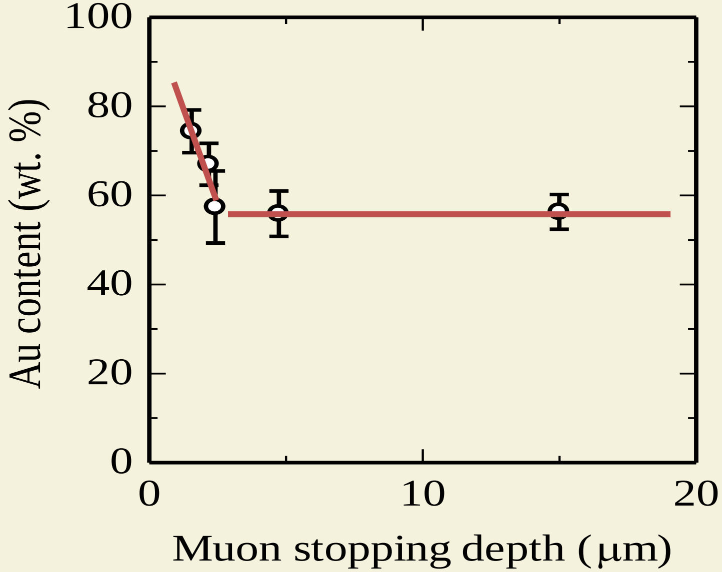


Different μAu/μAg X-ray intensity ratio was obtained

⇨ **Elemental composition varies with depth**

Gold coin analysis by muonic X-ray





Muon momentum (MeV/c)	Range in gold (μm)	Mass % of gold
6.4	2.2	74.4 ± 4.8
7.5	3.0	67.0 ± 4.7
8.0	3.6	57.4 ± 8.1
10.2	6.5	55.9 ± 5.1
15.2	20	56.3 ± 3.9

Bull. Chem. Soc. Jpn., 85. 228 (2012)
Anal. Chem., 87, 4597 (2015)

Depth selective analysis of bronze artefact

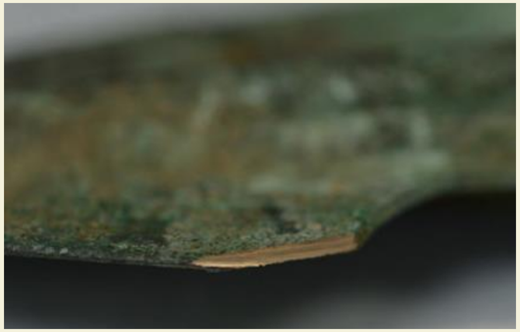
Originally brilliant metal of Cu, Sn, Pb

Covered with rust when excavated

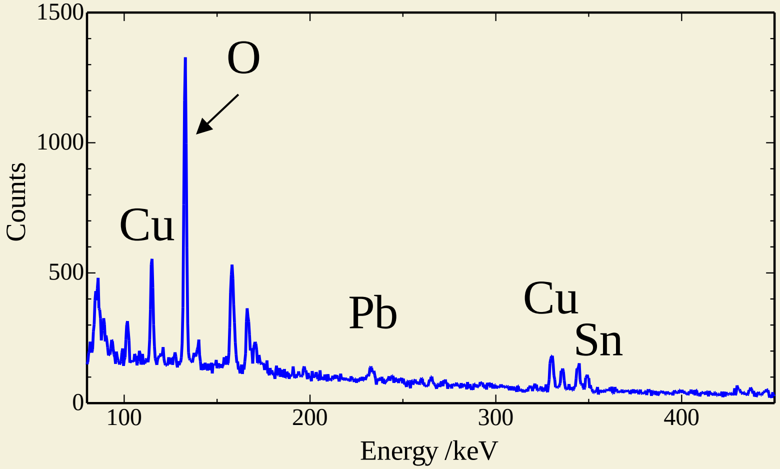
original
(reconstruction)

from webpage of Rekihaku

rusted



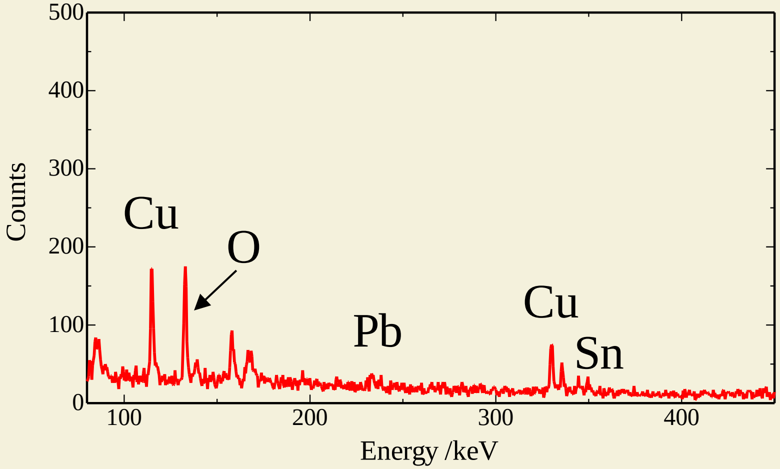
rust covers metallic layer



Abundant O
Low Cu

→ Rust

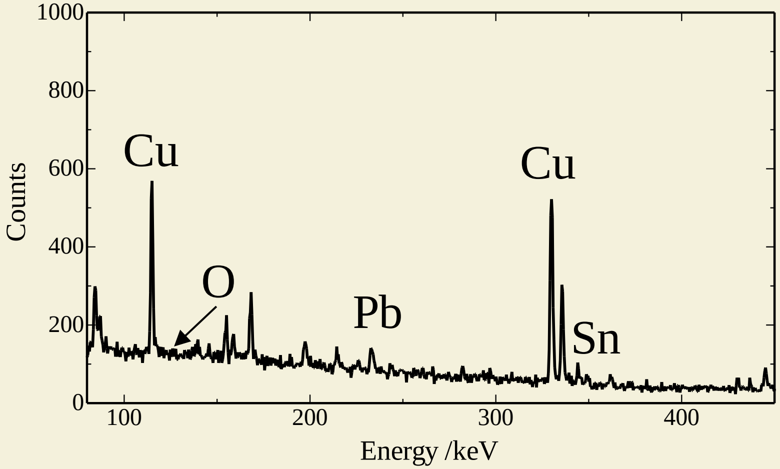
depth 0.04 mm
(**17** MeV/c)



Abundant O
Low Cu

→ Rust

depth 0.04 mm
(**22** MeV/c)



No Oxygen
High Cu

→ metal layer

depth 0.04 mm
(**40** MeV/c)

Depth selective analysis of bronze artefact

Elemental composition（%）

Depth（mm）	Cu	Sn	Pb	O
0.04（rust）	24.4	10.4	4.9	60.3
0.10（rust）	49.6	8.1	6.6	35.7
0.69（metal）	**83.1**	**9.1**	**7.9**	**ND**

Original composition

齋藤努　文化財科学 87（2024）17
T. Saito et al., *Bunkazai Kagaku*

Asteroid sample

Multi-elemental ･ Non-destructive ･Depth-selective

Negative muon is the answer

- Wide variety of samples:
  historical, archaeological,,,etc.
  e. g. asteroid sample by MUSES-C probe

(Our research plan around 2000)

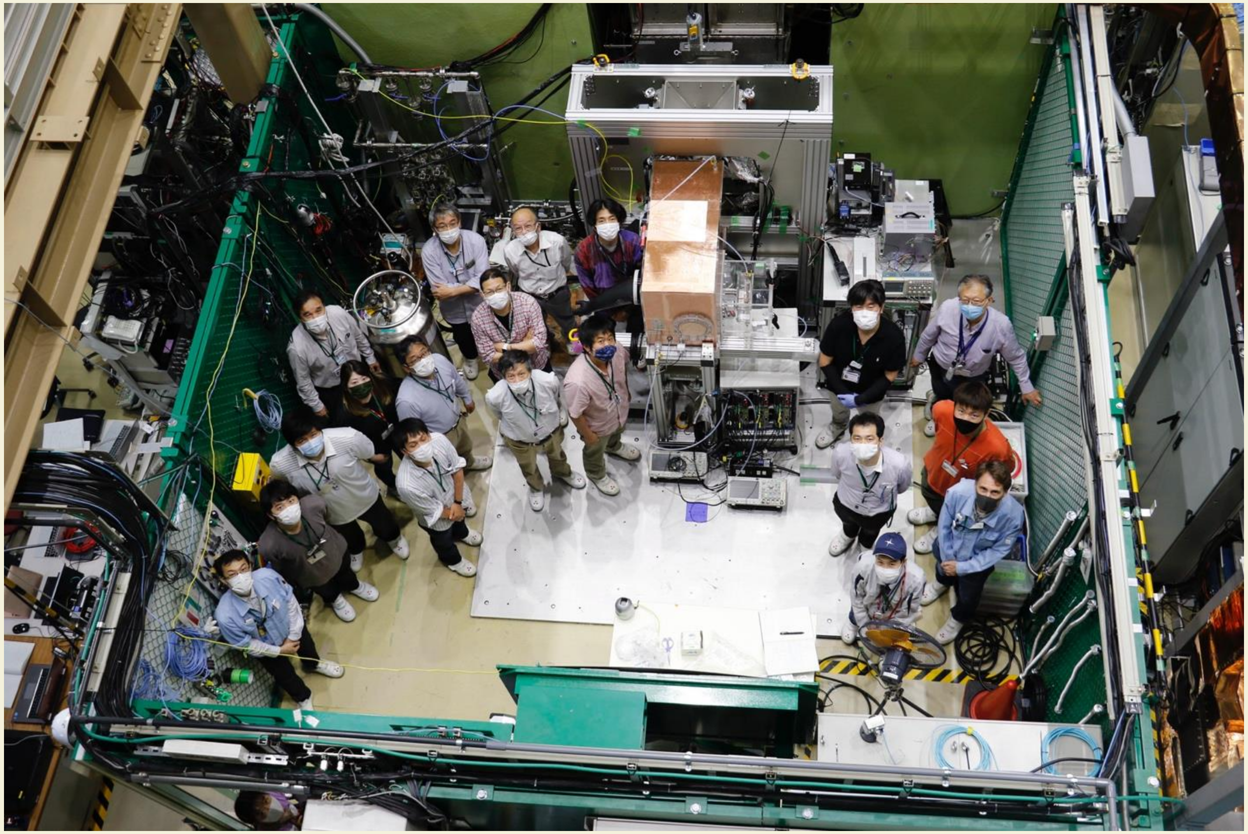
MUSES-C (renamed as Hayabusa) returned in 2010 with a few hundreds of micrograms of dust samples of the asteroid

Planetary scientists joined.
Hayabusa-2 space craft was traveling to asteroid 1999JC3(Ryugu) supposed to have abundant of B, C, N, O, and to return with samples in 2020.

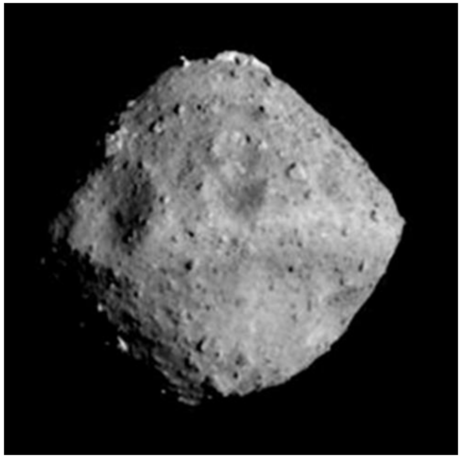
Asteroid Ryugu Sample Returned by Hayabusa-2

Non-destructive Carbon, Oxygen, Nitrogen analysis

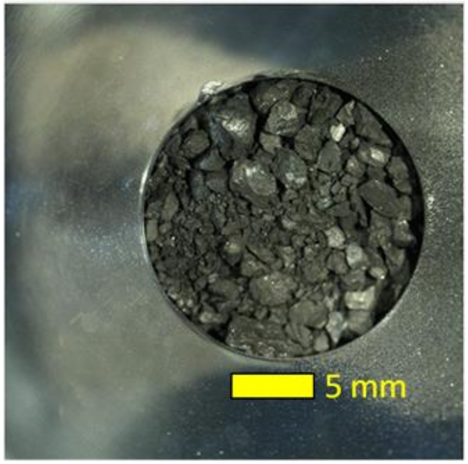
June - July, 2021



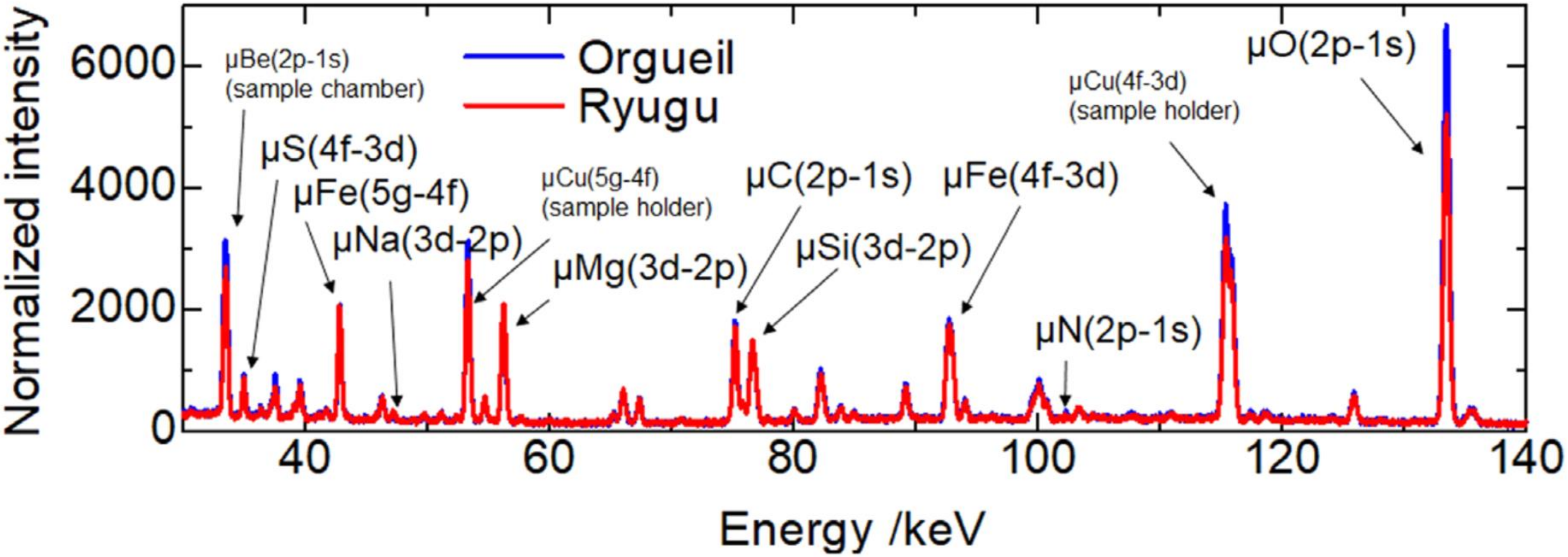
Asteroid sample



JAXA

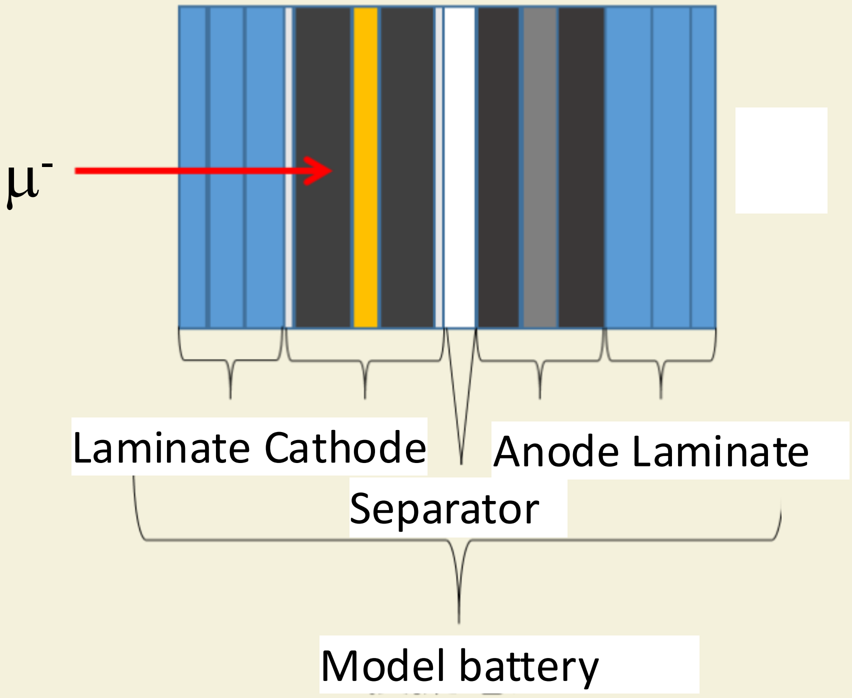


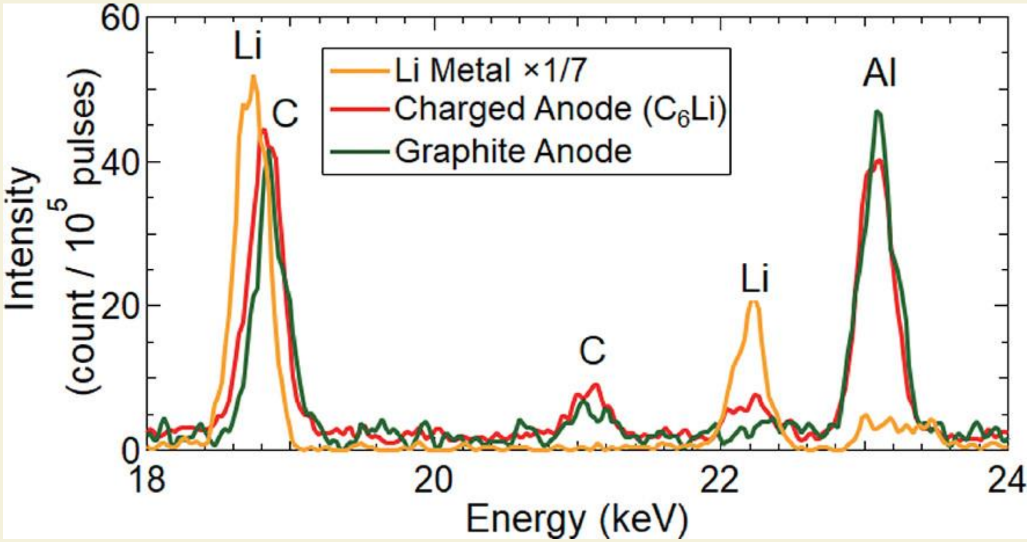
JAXA



https://www.science.org/doi/10.1126/science.abn8671

Lithium ion battery

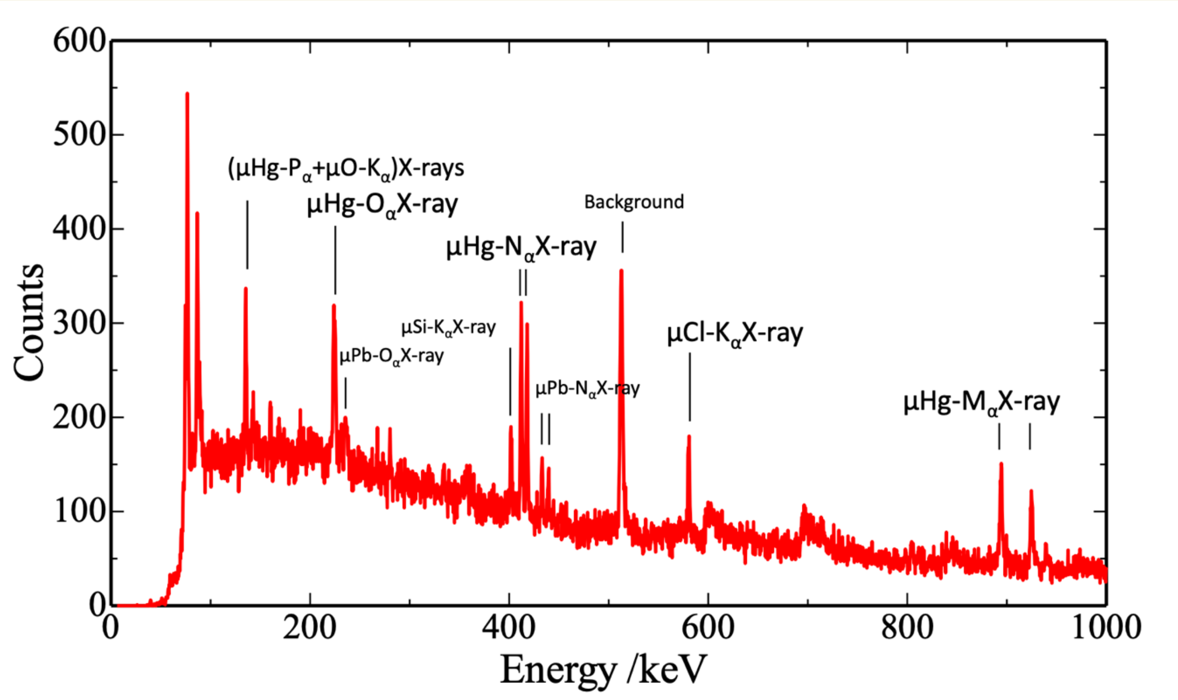




Anal. Chem. 92 (2020) 194-8200

cathode･･･graphite（C）、coll. electrode（Cu）
alumina coating（Al_2O_3）
anode･･･active material $Li(Ni_{0.85}Co_{0.15})O_2$
coll. electrode（Al）
separator･･･organic（C, H）
laminate･･･Al,nylon（C, H）

Unopenable Glass Bottle of Dr. OGATA Koan (1810 – 1863)

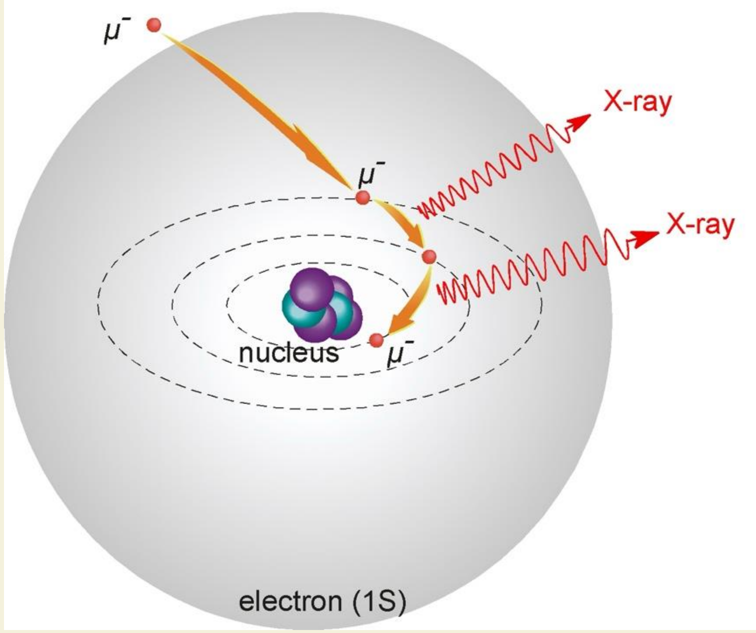


https://j-parc.jp/c/press-release/2021/03/17000664.html

Isotope analysis

m:a reduced mass containing the contribution from the atomic nucleus

Muonic X-ray shows isotope shifts



$$E_n = -\frac{Z^2 m e^4}{8 n^2 \varepsilon_o^2 h^2}$$

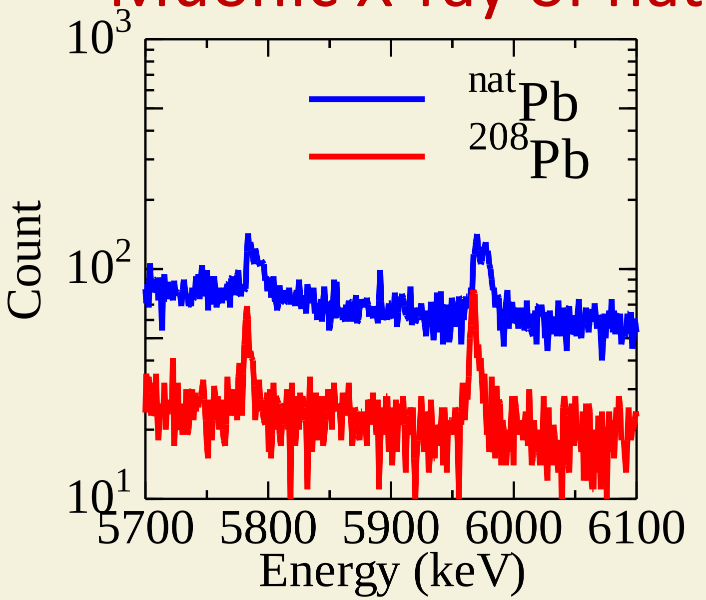
$$r_n = -\frac{4\pi\varepsilon_o n^2 \hbar^2}{Z m e^2}$$

$$\frac{m_\mu}{m_e} \approx 207 \approx \frac{E_\mu}{E_e} \approx \frac{r_e}{r_\mu}$$

Bohr model

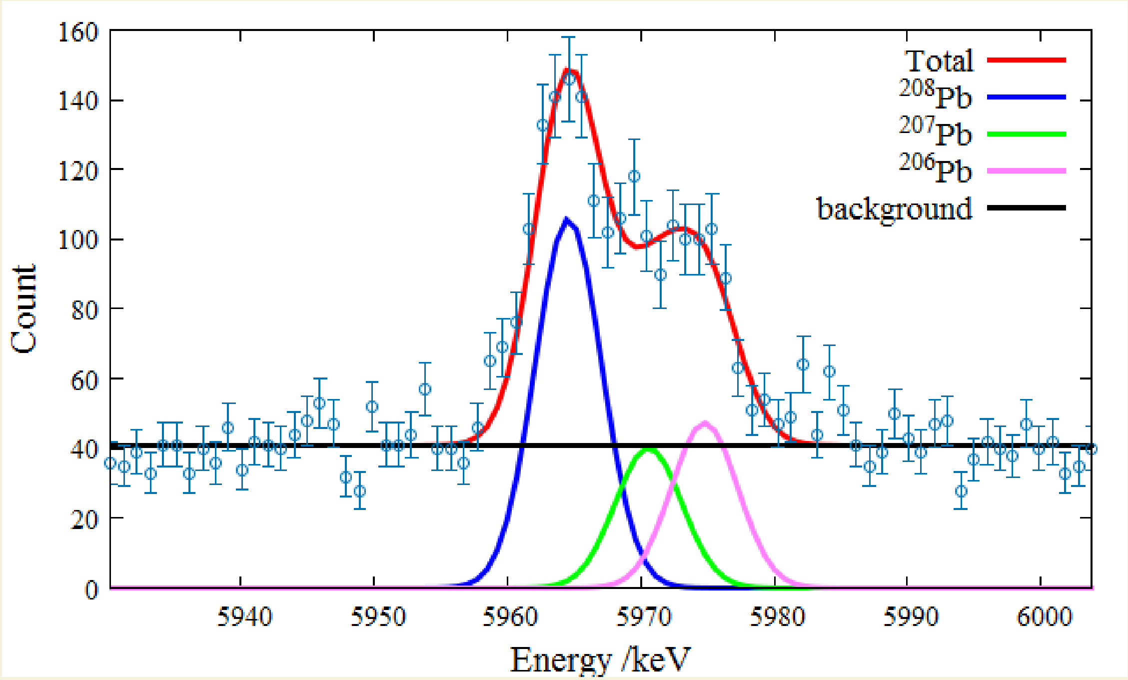
Isotope analysis

Muonic X-ray of natural Pb and ^{208}Pb



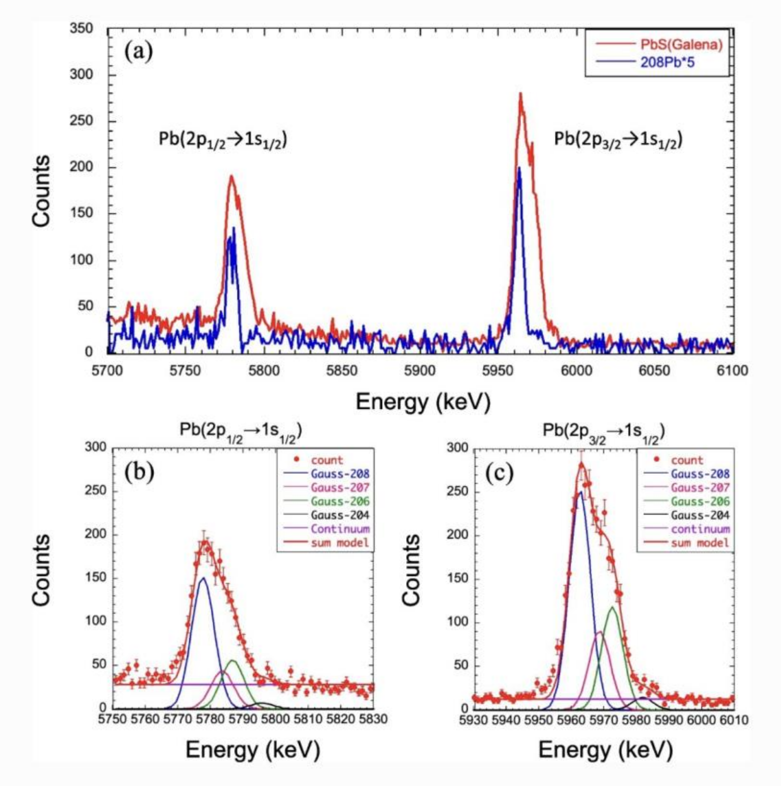
Isotope	abundance(%)
^{204}Pb	1.4(6)
^{206}Pb	24.1(30)
^{207}Pb	22.1(50)
^{208}Pb	52.4(70)

Decomposition of Muonic ^{nat}Pb X-ray



Isotope analysis

Galena: PbS

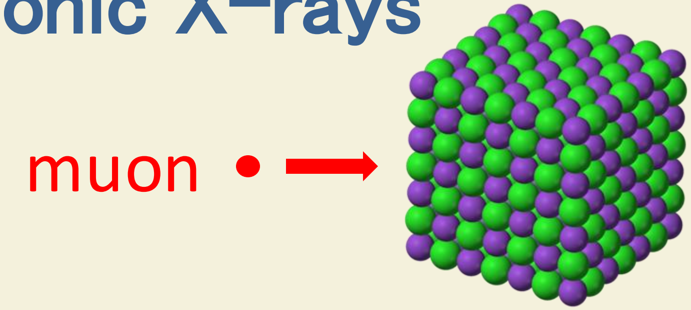


	Average of Muon analysis	LA-ICP mass
^{207}Pb/^{208}Pb	0.343 ± 0.040	0.396 ± 0.005
^{206}Pb/^{208}Pb	0.487 ± 0.041	0.472 ± 0.007
^{204}Pb/^{208}Pb	0.076 ± 0.018	0.026 ± 0.004

Measured with a DC muon beam at Research Center for Nuclear Physics, Osaka University

Muon Lifetime Method

Elemental Analysis with Muonic X-rays



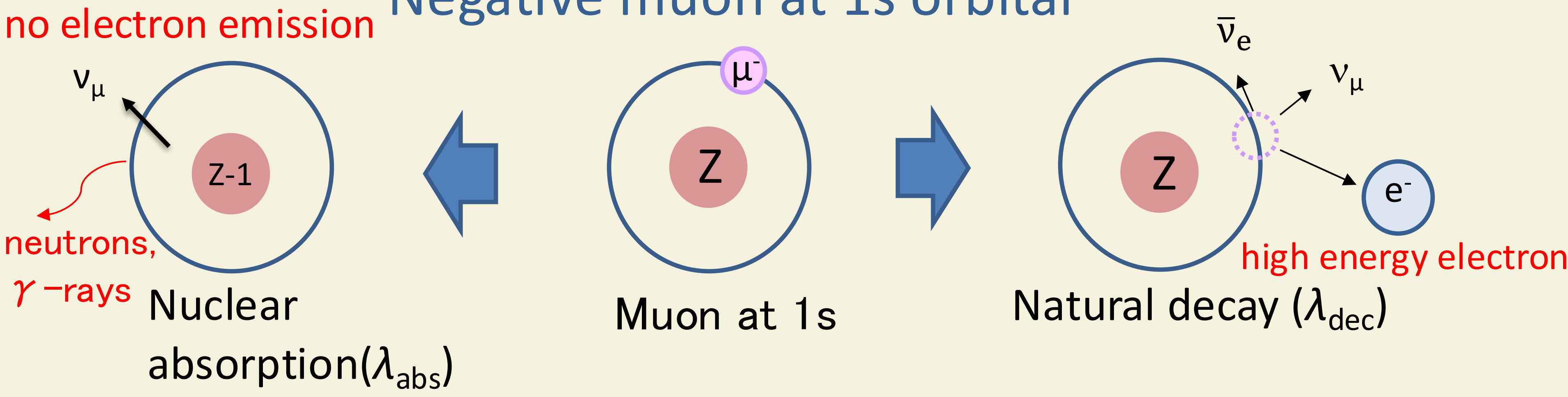
Atomic capture rate is roughly proportional to the atomic number (*Z*) of the muonium capturing atom
→ muonic X-ray yield is proportional to the weight content of the element

Components with more than 1% are easily quantified.

For analysis of minor components･･･
longer beam time for good statistics
accurate evaluation of small peaks

Muon Lifetime Method

Negative muon at 1s orbital



no electron emission

neutrons, γ −rays

Nuclear absorption(λ_{abs})

Muon at 1s

high energy electron

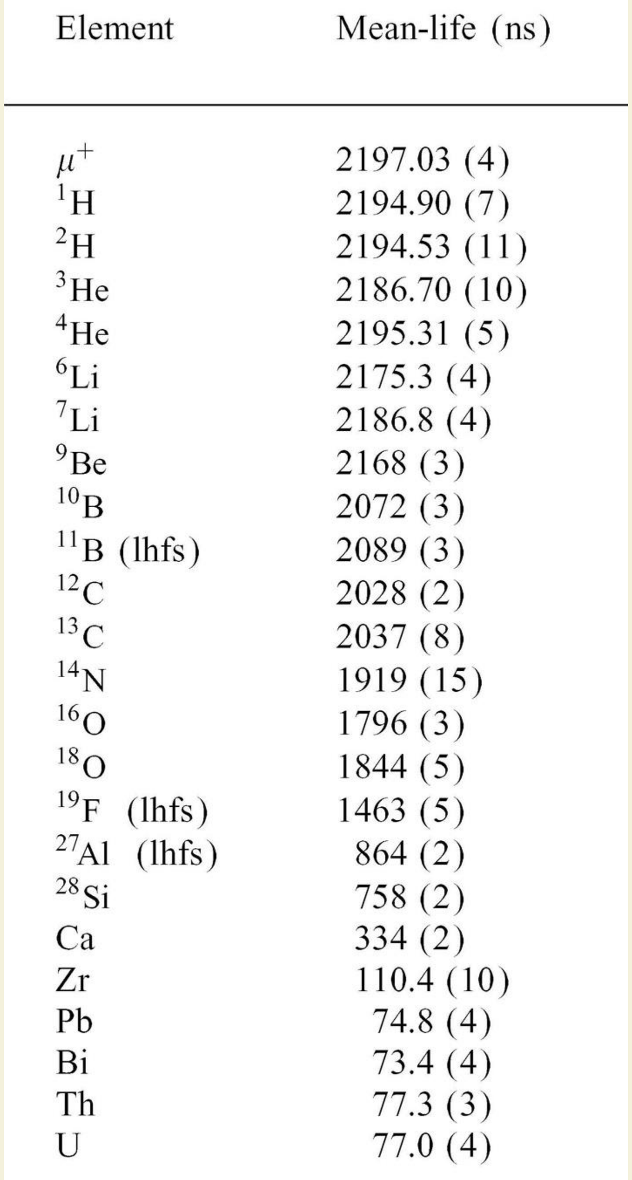
Natural decay (λ_{dec})

Addition of the nuclear absorption process shortens apparent muon lifetime

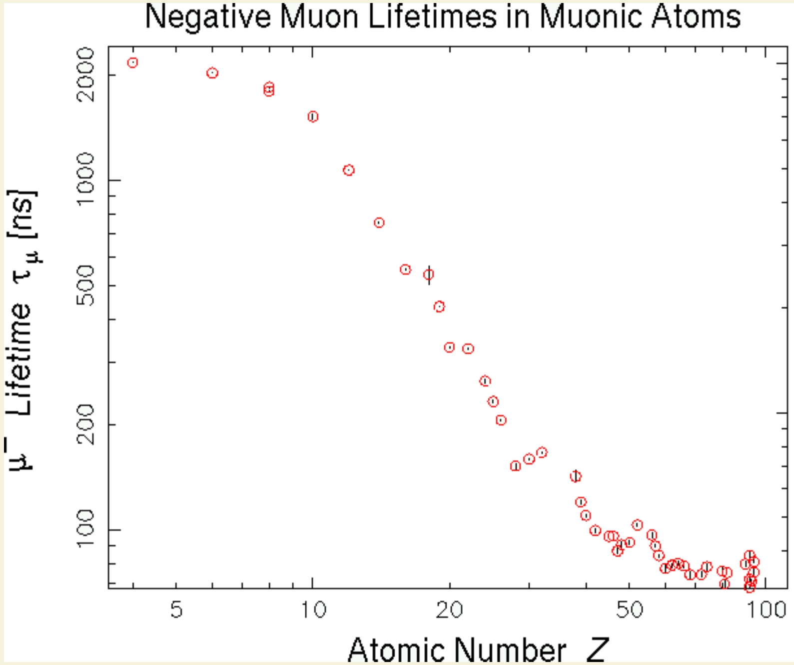
$$\tau = \frac{1}{\lambda_{\mathrm{dec}} + \lambda_{\mathrm{abs}}} < \frac{1}{\lambda_{\mathrm{dec}}} (= 2.2\ \mu s)$$

Muons in high Z atoms exhibits shorter apparent lifetime
→ distinguishable from a long-lived component

lifetime of negative muon in matter



Element	Mean-life (ns)
μ^+	2197.03 (4)
^{1}H	2194.90 (7)
^{2}H	2194.53 (11)
^{3}He	2186.70 (10)
^{4}He	2195.31 (5)
^{6}Li	2175.3 (4)
^{7}Li	2186.8 (4)
^{9}Be	2168 (3)
^{10}B	2072 (3)
^{11}B (lhfs)	2089 (3)
^{12}C	2028 (2)
^{13}C	2037 (8)
^{14}N	1919 (15)
^{16}O	1796 (3)
^{18}O	1844 (5)
^{19}F (lhfs)	1463 (5)
^{27}Al (lhfs)	864 (2)
^{28}Si	758 (2)
Ca	334 (2)
Zr	110.4 (10)
Pb	74.8 (4)
Bi	73.4 (4)
Th	77.3 (3)
U	77.0 (4)

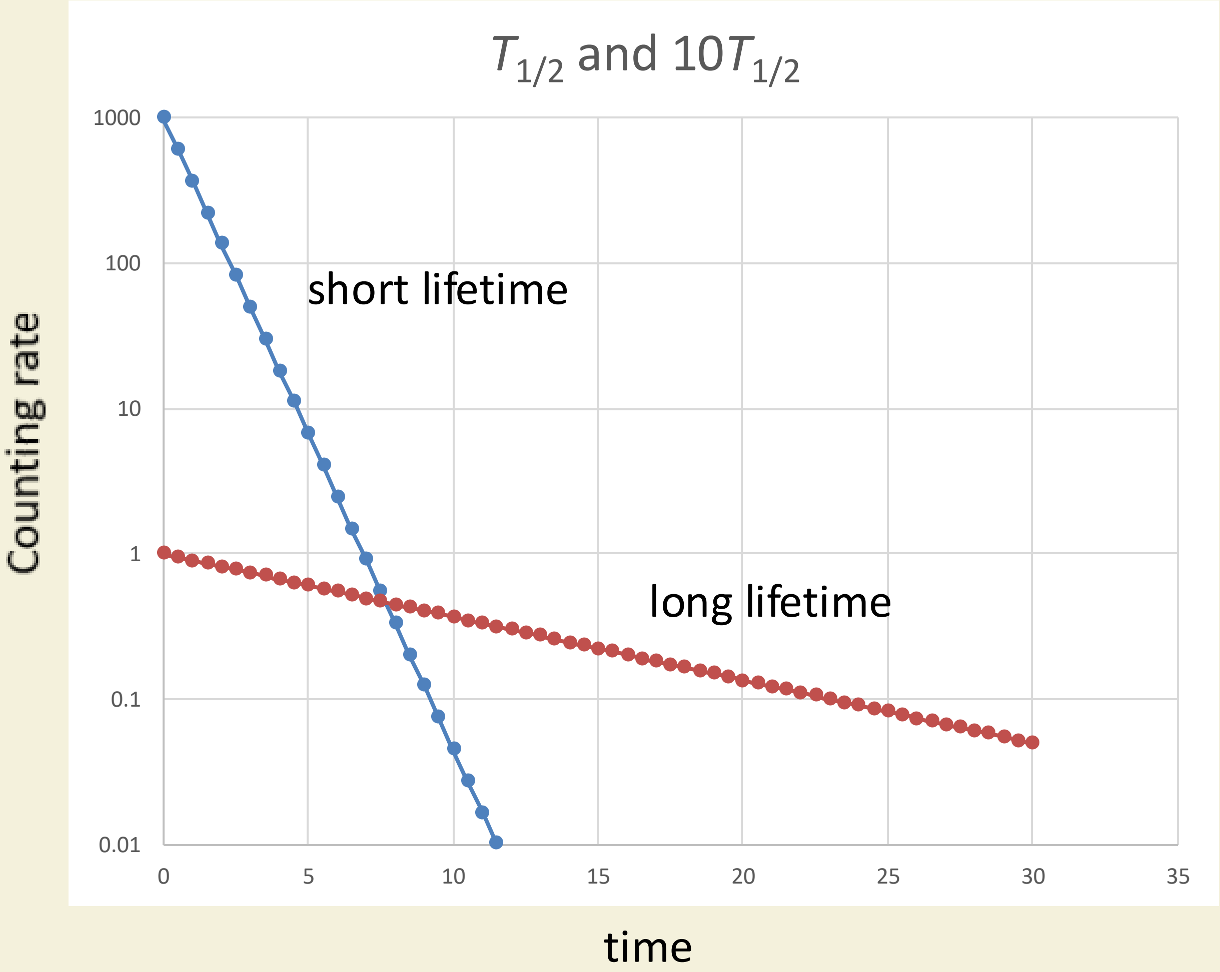


Muon lifetime / μs	
in vacuo	2.197
Carbon	2.03
Iron	0.206

D. F. Measday, Phys. Rep., 354 (2002) 243

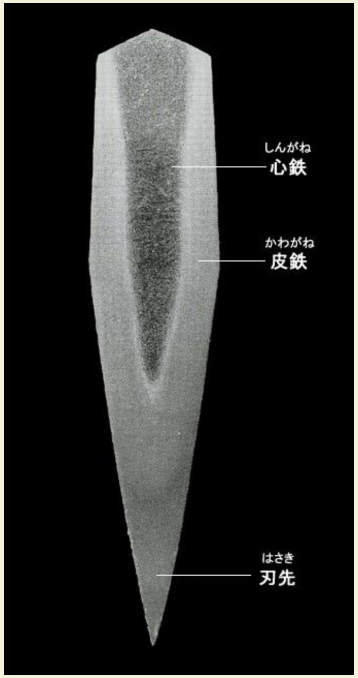
Lifetime method

Major component with a short half-life and long-lived minor component



Muon lifetime / μs	
in vacuo	2.197
Carbon	2.03
Iron	0.206

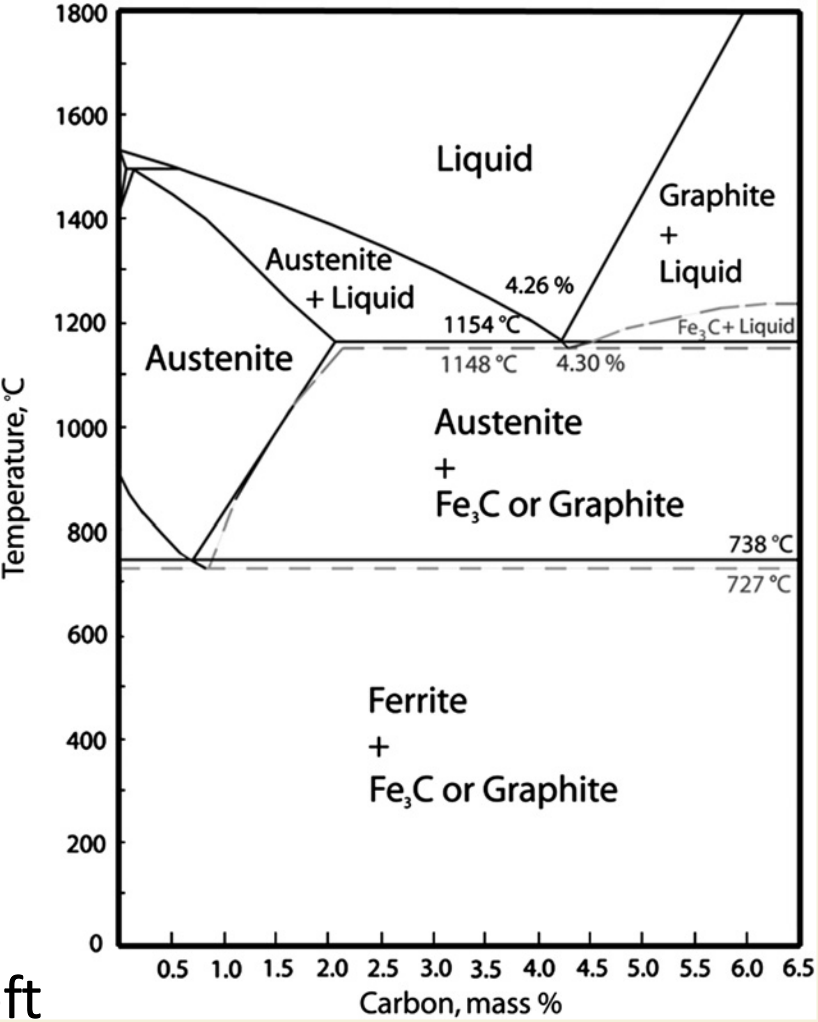
Japanese sword



Cross section of a Japanese sword

from Kyoto National Museum web page

Carbon in Iron



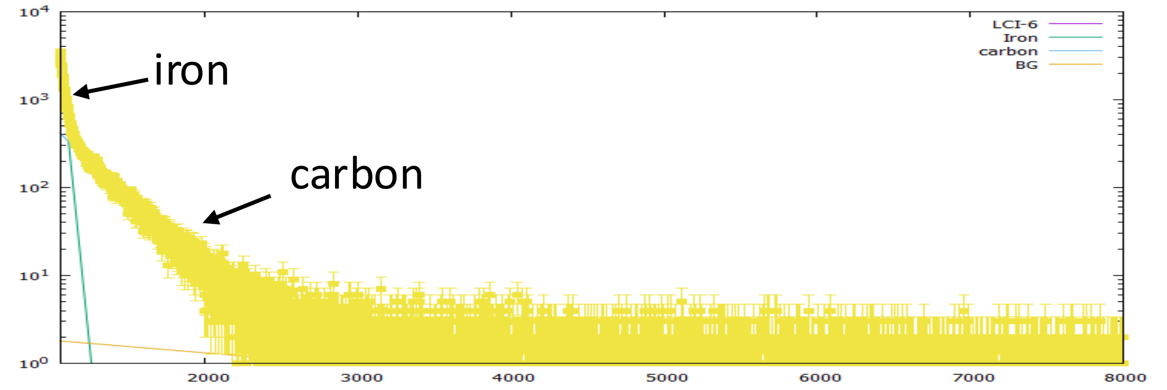
Soft

hard

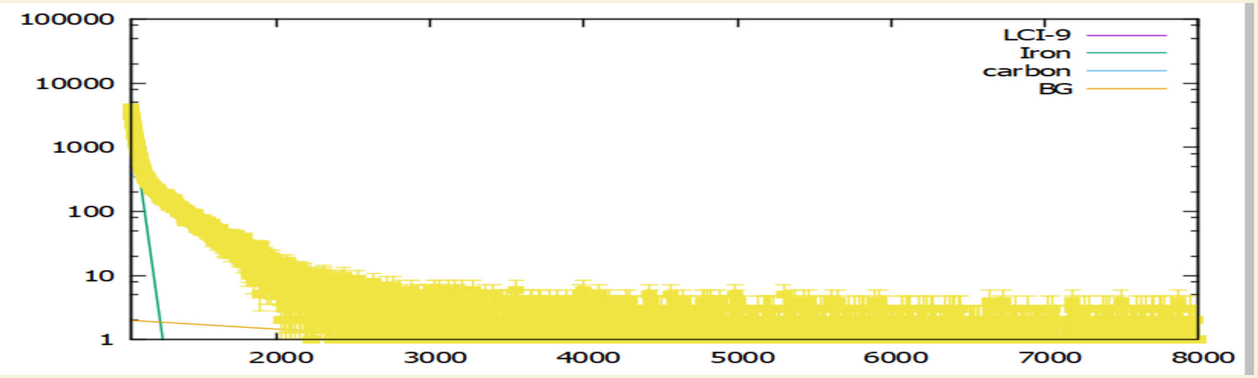
C. N. McCowan et al., Mat. Character., 62 (2011) 807

Standard iron samples

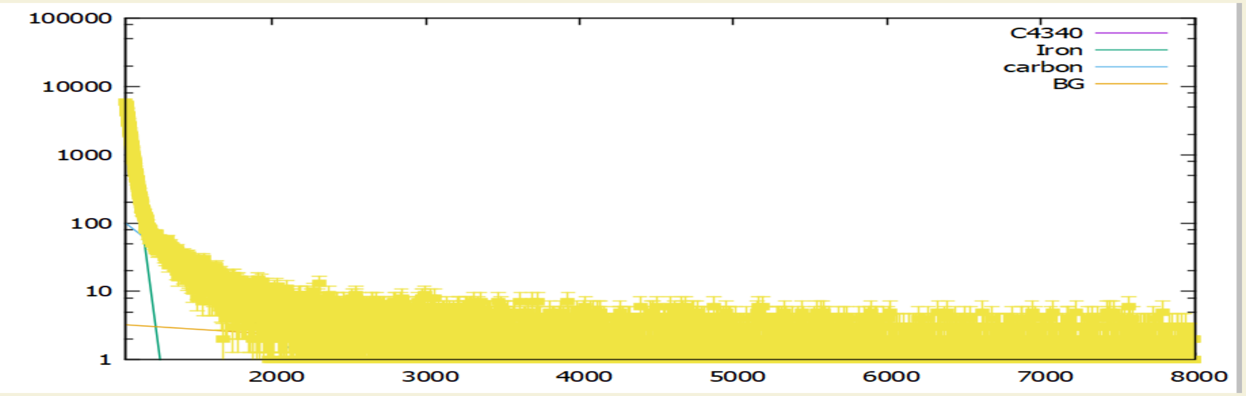
carbon
4.46%



carbon
3.22%



carbon
0.42%



Depth selective analysis

Stacked sample of iron plates (0.5 mm) with different carbon content

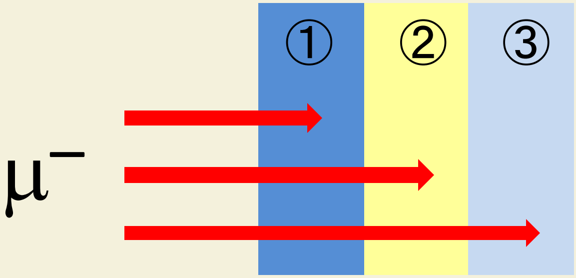


plate	momentum	chemical analysis	muon
①	30 MeV/c	1.03%	1.09(2)%
②	37 MeV/c	0.20%	0.19(1)%
③	43 MeV/c	0.51%	0.50(1)%

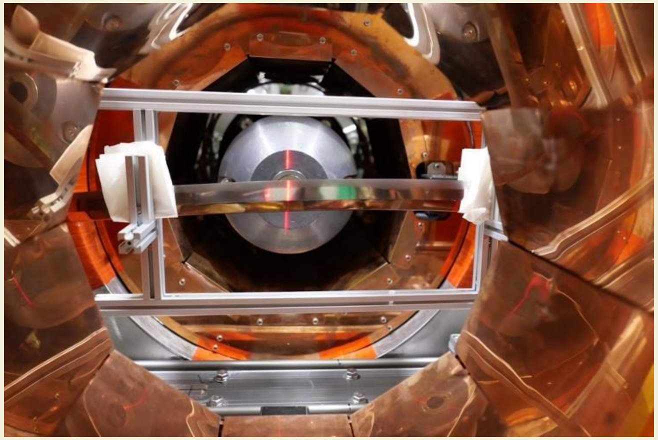
Sci. Rep. 17, 1797 (2024)

Japanese Sword

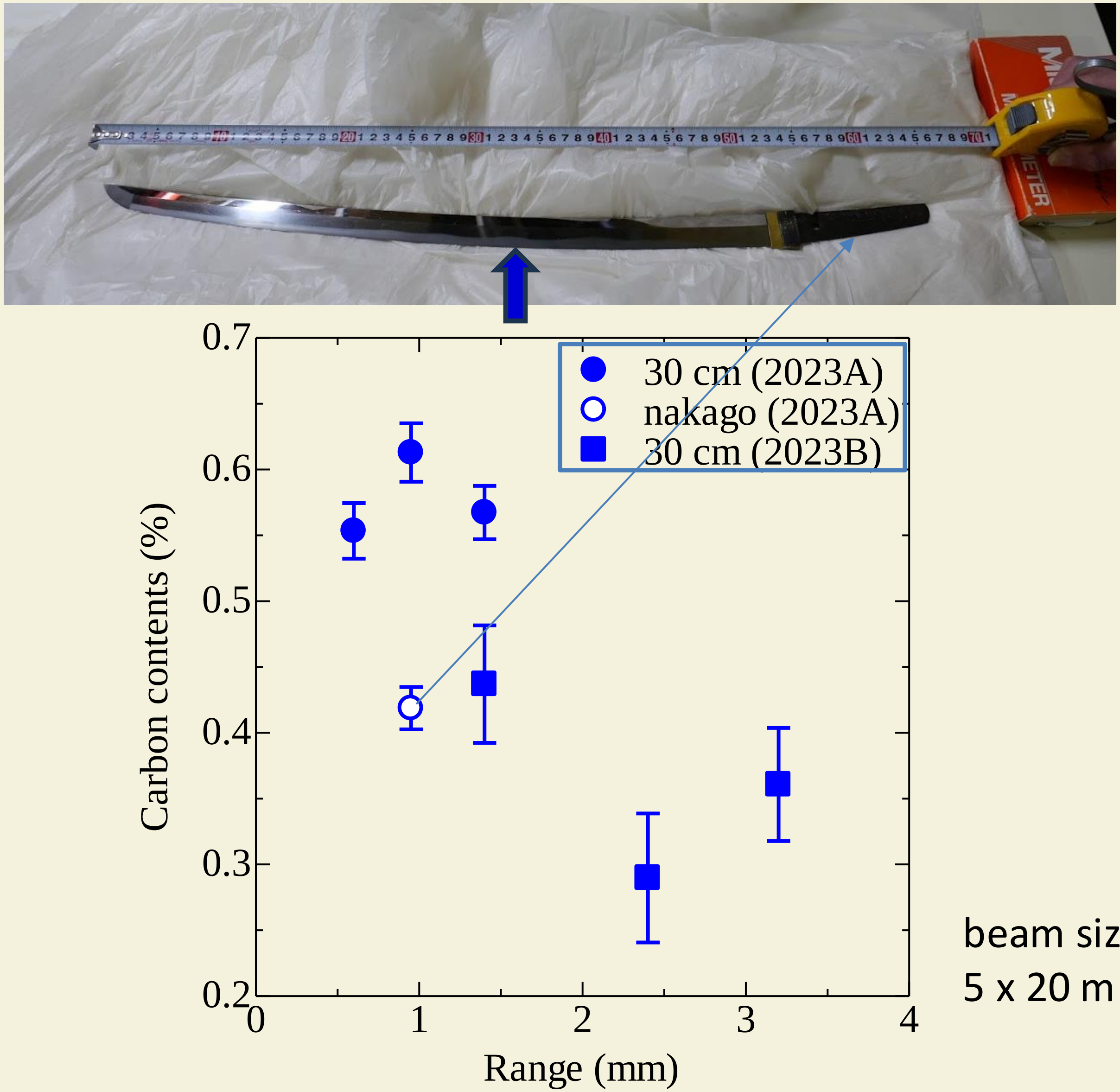
銃砲刀剣類登録証

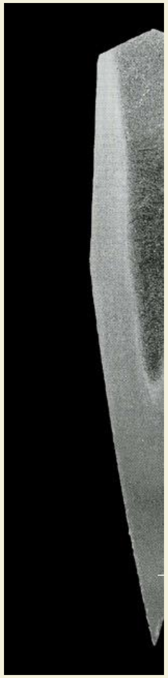
登録記号番号 東京都 第 282466 号

length 51.8 cm
made in around 16c



Japanese Sword





beam size
5 x 20 mm^2

Summary

・Muonic X-ray is a powerful and useful tool for non-destructive, multi-elemental and depth selective elemental analysis.

・Light elements and isotope analysis are possible.

・Uncertainty in depth is about 10% (+- 5%).

・Lifetime method is an alternative for a minor component of a binary element system.

J-PARC MUSE is now accepting proposals from non-science researchers.

ISIS(UK) and PSI(Swiss) group are enjoying Muonic X-ray.

FUME is planned as one of the satellite meetings.