

Development of non-destructive and depth-selective quantification method of sub-percent carbon contents in steel by negative muon lifetime measurement

I-Huan Chiu¹, M. Kenya Kubo², Kazuhiko Ninomiya¹, Shunsuke Asari¹, Makoto Inagaki³, Go Yoshida⁴, Soshi Takeshita⁴, Koichiro Shimomura⁴, Naritoshi Kawamura⁴, Patrick Strasser⁴, Yasuhiro Miyake⁴, Takashi U. Ito⁵, Wataru Higemoto⁵, Tsutomu Saito⁶

¹Osaka University, ²International Christian University, ³Kyoto University, ⁴KEK, ⁵Japan Atomic Energy Agency, ⁶National Museum of Japanese History

Abstract

A negative muon captured in an atom decays into electron or is absorbed into a nucleus with a mean life depending on the atomic number of the muon capturing nucleus. It is possible to identify the element that captured a muon by measuring the lifetime. We have conducted a feasibility study on the non-destructive identification of carbon contents in a stacked steel sample (iron-containing carbon) consisting of three steel plates with different carbon contents of 0.2-1.0 %. We successfully quantified the elemental composition of each layer of the sample by controlling incident muon momentum, that is, we successfully demonstrated the non-destructive and depth-selective determination of sub-percent carbon contents in steel by muon lifetime measurement.

Introduction

Motivation

Steel is one of the most important materials for humankind. Steel is mainly composed of iron and contains smaller amounts of other elements. Among these minor elements, carbon is one of the most important that controls properties of steel such as hardness. However, it is not easy to quantify carbon in steel; destructive analysis has been generally applied in present. This study focused on a negative muon as a probe for quantification of sub-percent carbon in steel non-destructively and depth-selectively.

Muonic atom

1. "Negative" muon stop in a material
2. Muon capture in muon atomic orbit, muonic atom formation
3. Muon de-excitation to muonic 1s state with emission of muonic X-rays
Elemental analysis by muonic X-ray measurement (for example [1,2])
4. Muon absorption into the nucleus or muon decay into an electron

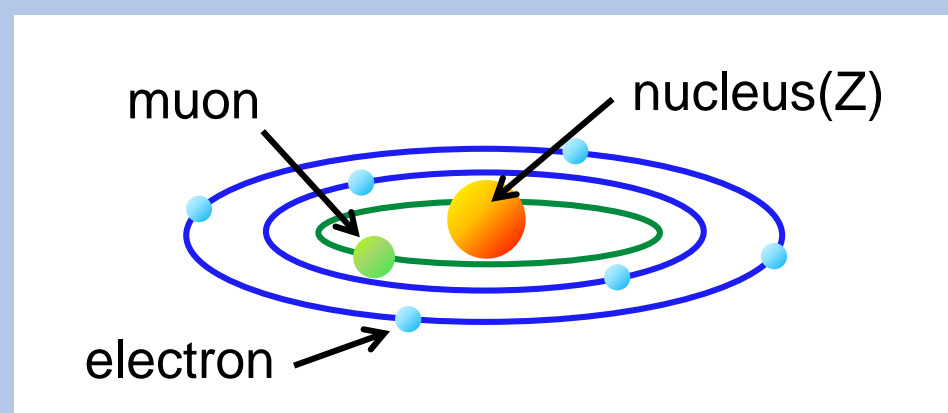


Figure 1: Muonic atom

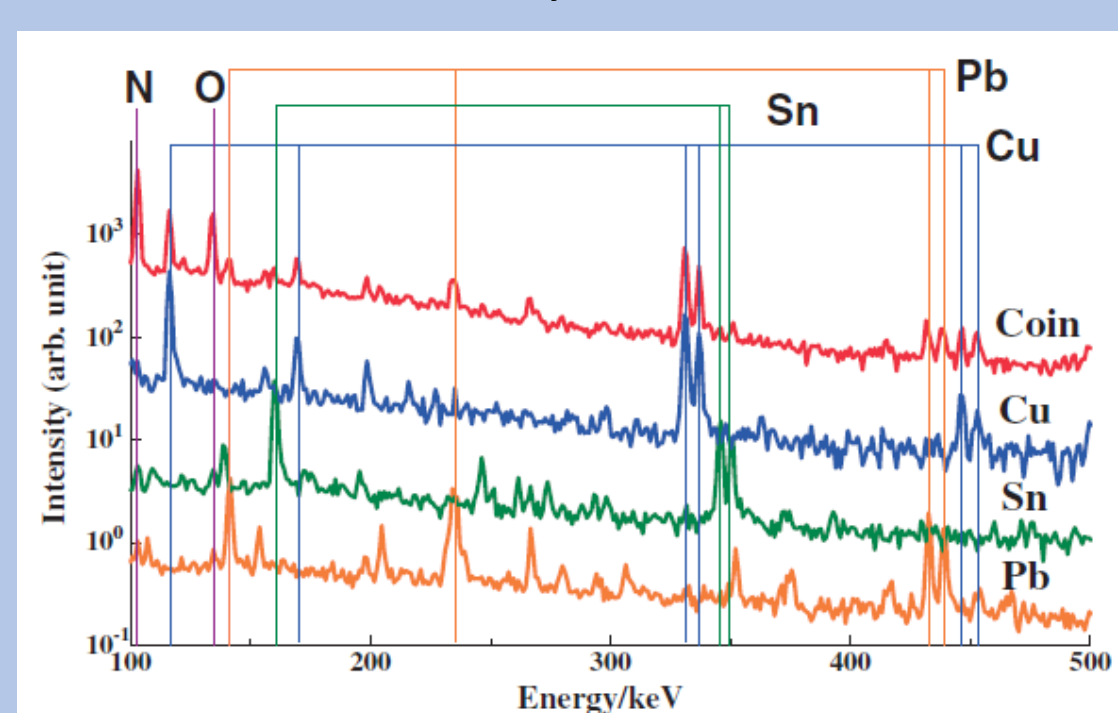


Figure 2: elemental analysis for bronze coin [1]

Elemental analysis method by lifetime measurement

- Lifetime of the muon in the muonic atom is different by element, that means **identification of element by lifetime measurement of the decay electron is possible**
The rate of nuclear absorption becomes large with increasing atomic number; 2.0 μ s in carbon and 0.20 μ s iron [3].
- **Quantitative analysis is possible** because there is a linear relationship between the probability of muon capture and the elemental composition over a wide concentration range [1, 4].
- The muon decay electron from light elements are emitted even after all the muons captured by the heavy element have decayed out. **High sensitivity analysis for light element is possible** by paying attention to long-lived components even if the material coexists with large amounts of heavy elements.
- Muon stopping position (depth) can be controlled by adjusting incident muon momentum. **Non-destructive and depth-profile analysis is possible.**
- Large-solid angle μ SR apparatus are available for decay electron measurement. It is **no need to develop additional counting system.**

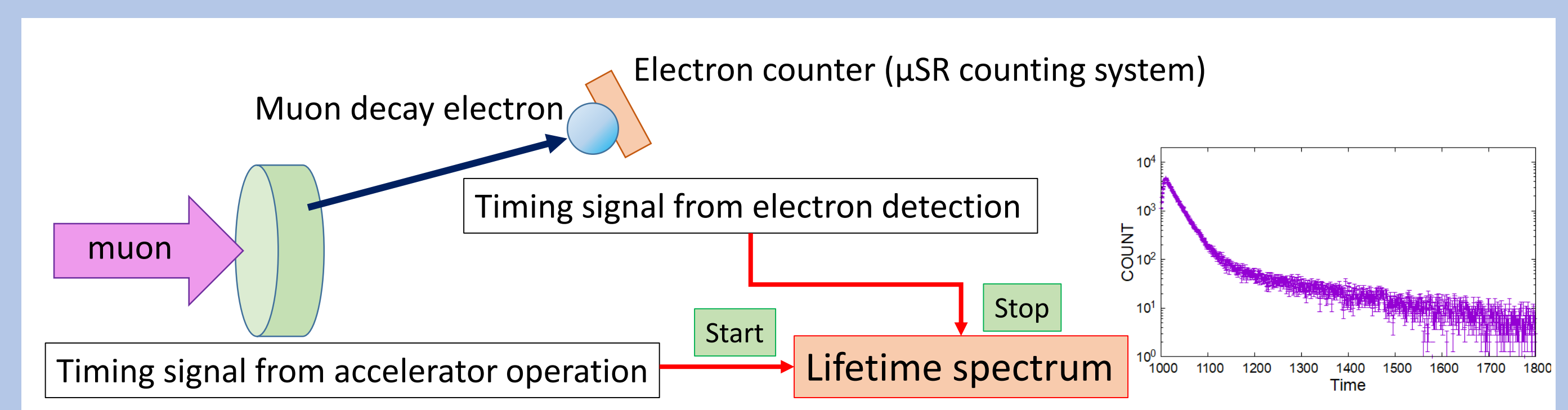


Figure 3: Conceptual diagram of muon lifetime analysis method

Experimental

- The muon experiment was conducted at the D1 area of the Muon Science Establishment in the Japan Proton Accelerator Research Complex (J-PARC MUSE).
- The decay electrons were measured by the large-solid angle plastic scintillation counter system, Kalliope, developed for μ SR [5].
- Standard steel samples with carbon contents of 20ppm, 0.42% and 4.46% were used for obtaining a calibration curve.
- A stacked steel sample consisting of three steel plates with carbon contents of 0.51%, 0.20% and 1.03 % of 0.5 mm in thickness was irradiate with three momentum conditions for depth-profile analysis.

Table1: Summary for muon irradiation conditions

Sample	Momentum (MeV/c)	Pulse	Irradiation time (h)
Fe STD (C: 20 ppm)	40	190621	2.1
Fe STD (C: 0.42%)	40	187829	2.1
Fe STD (C: 4.46%)	40	81264	0.9
Fe STD (C: 4.46%)	47	46353	0.5
Stacked Sample (C: 1.03%, 0.20% and 0.51%)	43	161764	1.8
Stacked Sample (C: 1.03%, 0.20% and 0.51%)	37	256441	2.8
Stacked Sample (C: 1.03%, 0.20% and 0.51%)	30	563337	6.3

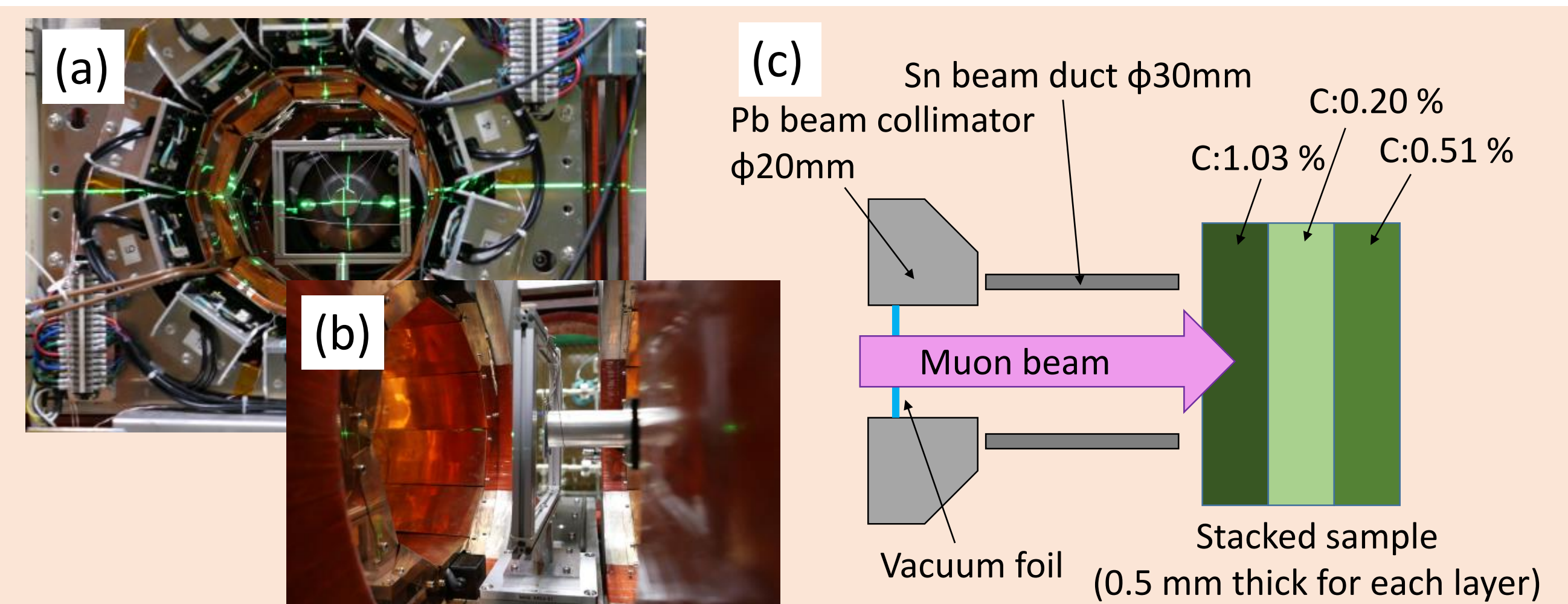


Figure 4: (a) Photo of counting system Kalliope (b) Photo of muon irradiation sample (c) Schematic view of muon irradiation for a stacked sample

Results and Discussion

Spectrum analysis

- Four components, iron, carbon, air, and long-lived background, were considered in the analysis of muon lifetime spectrum (see Figure 5).
- All fittings were done after 1050 ch region because we found decay electron contribution from Sn as the beam duct just after muon beam arrival timing.
- The component of air was considered as a mixture of three molecules, that is, 78% nitrogen, 21% oxygen, and 1% argon.
- The lifetime of each element was fixed in the analysis; 206 ns for Fe, 2026 ns for C, 1907 ns for N, 1795 ns for O and 537 ns for Ar [3].
- The origin of long-lived background component was unknown, but the component is not originated muon because the lifetime is longer than the muon lifetime (2.2 μ s).
- The intensity of air was fixed by the analysis result of pure iron (steel containing 20 ppm carbon) sample.

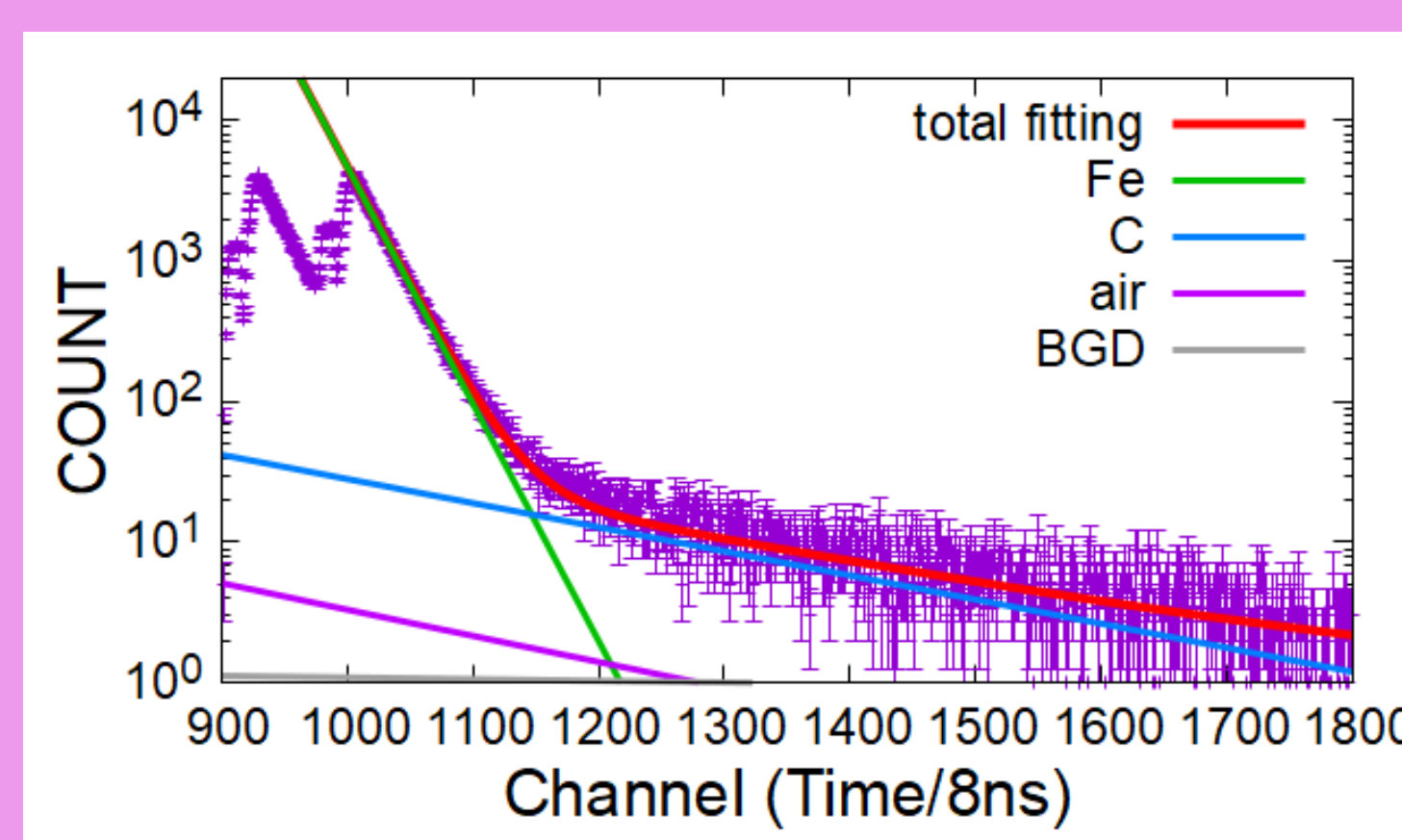


Figure 5: Lifetime spectrum obtained of standard steel sample containing 0.42% carbon together with the fitting line.

Calibration curve and detection limit

- Figure 6 shows the calibration curve obtained from muon experiments for standard steel samples.
- Linear relationship was obtained between carbon contents in steel samples and signal ratios of C to Fe. From the relation from Figure 6, we can quantify carbon contents in steel sample.
- The detection limit was estimated as 180 ppm for 2 hour measurement with 40 MeV/c incident muon momentum from the 3 sigma deviation of air intensity of pure iron sample.

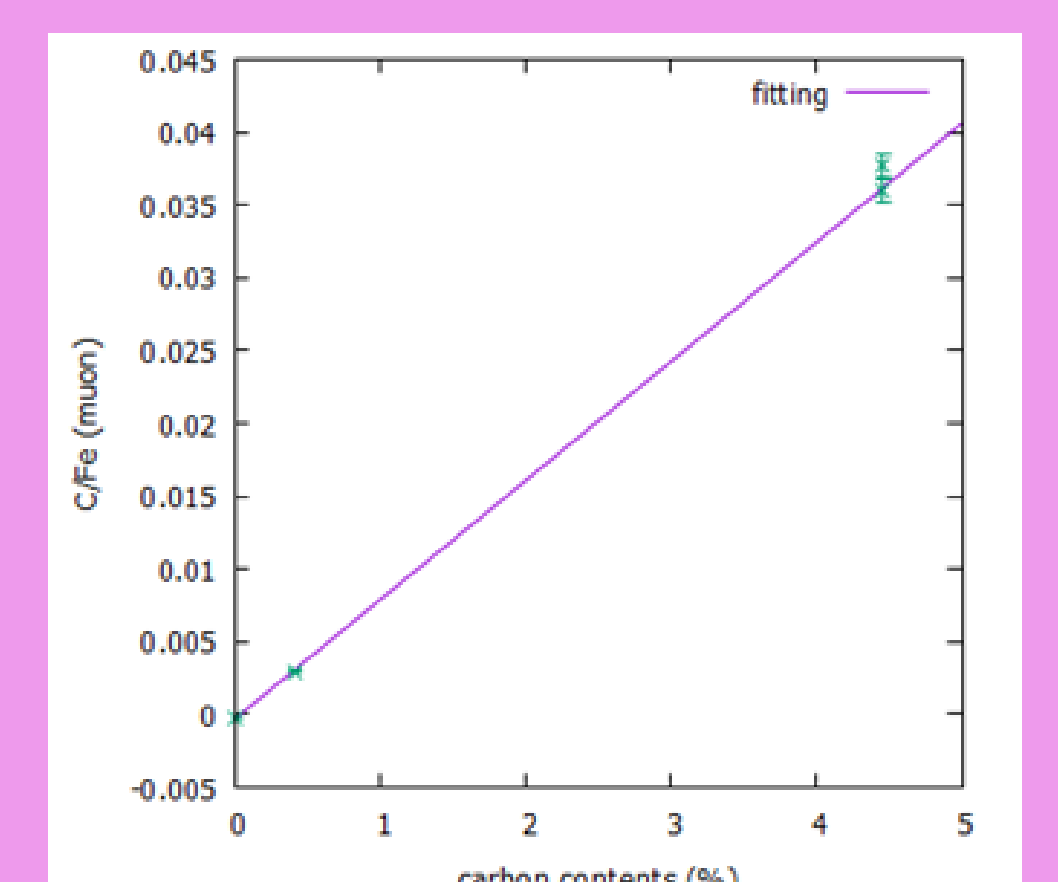


Figure 6: Relationship between carbon contents in steel and signal ratio of C to Fe

Depth profile analysis

- The muon stopping depths with incident momenta of 30, 37 and 43 MeV/c were estimated as 0.35, 0.75 and 1.2 mm, respectively. These stopping depths correspond to muon stop in the first, second and third layers in the stacked sample.
- As shown in Table 2, different C/Fe values were obtained by muon incident momenta. Using the calibration curve shown in Figure 6, elemental composition of each muon irradiation condition was determined.
- The obtained carbon contents in the stacked sample with different muon momenta were well agreed with the analysis results using chemical method. **We successfully quantified sub-percent carbon contents in steel non-destructively and position selectively by the muon lifetime measurement.**

Table 2: Analysis results of standard steel and stacked steel samples

Sample	Carbon intensity (/10000 pulse)	Iron intensity (/10000 pulse)	C/Fe (x 1000)	Carbon contents from muon analysis	Carbon contents from chemical analysis
Fe STD1	-	222.4 (19)	-	-	20 ppm
Fe STD2	0.76 (2)	244.6 (23)	3.12 (9)	-	0.42%
Fe STD3 40MeV/c	7.87 (7)	216.8 (41)	36.3 (8)	-	0.51%
Fe STD3 47MeV/c	13.14 (12)	347.3 (70)	37.8 (8)	-	0.51%
Stacked Sample 43 MeV/c	1.15 (2)	294.9 (26)	3.89 (9)	0.49 (2) %	0.51%
Stacked Sample 37 MeV/c	0.27 (1)	185.2 (15)	1.45 (7)	0.19 (1) %	0.20%
Stacked Sample 30 MeV/c	0.75 (1)	85.5 (7)	8.64 (13)	1.08 (4) %	1.03%

Summary

We clarified that the sub-percent carbon contents in steel can be quantified non-destructively by the muon lifetime measurement. Due to high-efficient measuring system and high-intensity beam, the detection limit of carbon is 180 ppm by 2 hour measurement. A calibration curve prepared from standard samples showed good linearity, making it clear that quantitative analysis is possible. Depth profiling of carbon contents was also achieved by controlling incident muon momenta for a stacked sample of three steel with different carbon contents.

References

[1] K. Ninomiya et al., Bull. Chem. Soc. Jpn., 85 (2012) 228
[4] H. Daniel et al., Fresenius' Z. Anal. Chem., 321 (1985) 65

[2] I-Huan Chiu et al., Scientific Reports 12 (2022) 5261
[5] K. M. Kojima et al., J. Phys. Conf. Ser., 551 (2014) 012063

[3] T. Suzuki et al., Phys. Rev. C, 35 (1987) 2212