High-Energy Resolution Microcalorimeter EDS System for Electron Beam Excitation

*K. Tanaka, A. Nagata, N. Sasayama, M. Ikeda, A. Odawara,
S. Nakayama and K. Chinone.

SII NanoTechnology Inc.,563 Takatsuka-shinden, Matsudo-city, Chiba 270-2222, Japan

*keiichi.tanaka@sii.co.jp

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We have developed a microcalorimeter energy dispersive spectroscopy (EDS) system with high-energy resolution to analyze the composition in the materials with low voltage excitation. This system is based on the dilution refrigerator and has a snout like conventional EDS system. The energy resolution was 14.2 eV for Al-Kα line (1.5 keV) and the estimated count rate was 333 cps. We demonstrated the measurement of the energy spectra for three samples. A conventional EDS cannot separate between the Si-Kα (1470 eV) and W-Mα (1775 eV) lines. This separation is important in the semiconductor application and is one of the most difficult separation for the composition analysis. Our microcalorimeter EDS system could separate these peaks clearly for the WSi_2 film (300 nm thickness) deposited on the silicon wafer when the excitation voltage was 5 kV.

INTRODUCTION

A Scanning Electron Microscope (SEM) using low acceleration voltage is a powerful tool to image the surface details of delicate and/or uncoated specimens, and users of low voltage SEM usually need to analyze the elemental composition under low voltage (5 kV) without having to increase the excitation voltage as typically required in conventional x-ray analysis (20 kV). The EDS system with high energy resolution is desired to analyze the materials with low voltage excitation.

A Transition Edge Sensor (TES) is an Energy Dispersive X-ray Spectrometer with high-energy resolution (4.5 eV at 6 keV) and can analyze a wide energy range (0-10 keV) by using only one detector [2]. A TES consists of an absorber to detect x-rays, a thermometer to measure the change in temperature, and a thermal link to regulate the heat flow from the thermometer to the heat sink. A superconductor is used as a thermometer utilizing the steepness in the transition curve from normal to the superconducting state. We previously developed a TES and a fabrication method suitable for TES format arrays [3]; this TES uses a dilution refrigerator and has an energy resolution of 6.6 eV at 6 keV [4]. This TES is usually cooled below 100 mK to decrease the phonon noise and to improve the energy resolution. A commercial dilution refrigerator cannot introduce x-rays from outside the refrigerator. A sample is

Fig. 1 SEM- microcalorimeter EDS system. This EDS system is based on the dilution refrigerator that can cool the x-ray detector below 100 mK, and has a “snout” to minimize the distance between the TES on the cold stage and sample, similar to that in a conventional semiconductor EDS system.

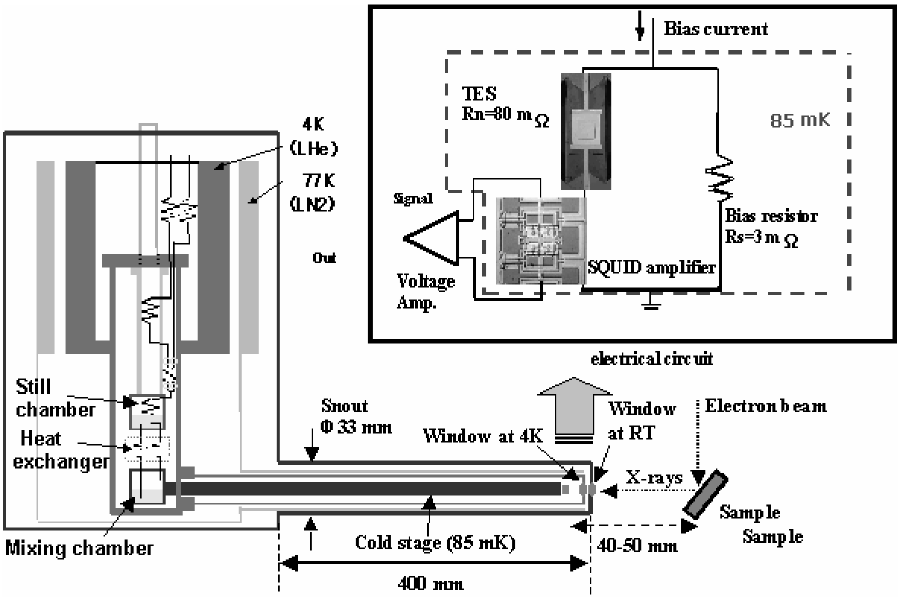

Fig. 2 Schematic cross-section of the microcalorimeter EDS system shown in Fig. 1. The microcalorimeter EDS system consists of LN2 tank, LHe tank and a dilution refrigerator. The dilution refrigerator consists of a mixing chamber, still chamber and heat exchanger. Inserted figure is electrical circuit to detect x-rays. The electrical circuit is integrated on top of the cold stage attached to the mixing chamber.

located outside the refrigerator like a conventional EDS system. We must develop the EDS system that introduces x-rays into the refrigerator to analyze the materials. In this study, we developed a microcalorimeter EDS system that can introduce x-rays from outside the refrigerator. Here, first we describe the system and its basic characteristics (e.g., minimum temperature, temperature stability). Then, we report the performance evaluation of the system (i.e., energy resolution, estimated count rate), and energy spectra for different samples measured using the system.

MICROCALORIMETER EDS SYSTEM

The microcalorimeter EDS system developed here is based on a dilution refrigerator that can be cooled below 100 mK and has a “snout” to minimize the distance between the TES and sample, such as the minimized distance in a conventional Si(Li) EDS system. Figure 1 is a photo of this system attached to a SEM, and Fig. 2 is a schematic cross-section. This system consists of 77 K cold stage, 4K cold stage, and a dilution refrigerator. A dilution refrigerator consists of a mixing chamber, still chamber and a heat exchanger. The minimum temperature is achieved at the mixing chamber of the dilution refrigerator, and a copper rod (cold stage) is attached to the mixing chamber. The liquid N_2 and He tanks act as thermal shields to insulate the cold stage. The snout is 400 mm long and 33 mm in diameter. The minimum temperature (50 mK) of the EDS system is determined by the balance between the cooling ability and the heat penetration from the electrical wires connected between the electrical terminals on the cold stage and that on the He tank. The TES and a SQUID amplifier are mounted on the top of the cold stage. The SQUID amplifier is a superconducting device with low noise ($<10\,\mathrm{pA}/\sqrt{\mathrm{Hz}}$) for a wide frequency band (DC-1 MHz) and converts the small current signal (1 μA) from the TES to high voltage (10 mV) [5]. X-ray windows are located in the cold stage (not shown here), 4K shield, and room-temperature shield of the snout to detect x-rays radiated from the sample located outside this system.

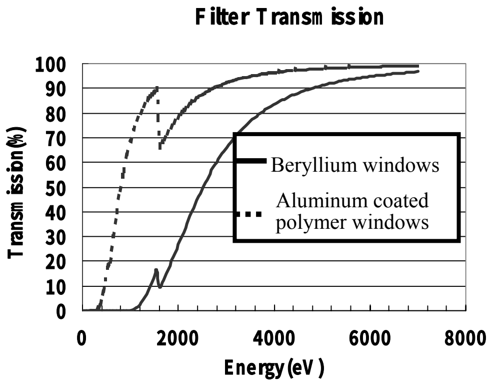

Fig. 3 Transmittance curves for beryllium windows (solid line) used in the current version of the microcalorimeter EDS system and for aluminum-coated polymer thin film windows (dashed line).

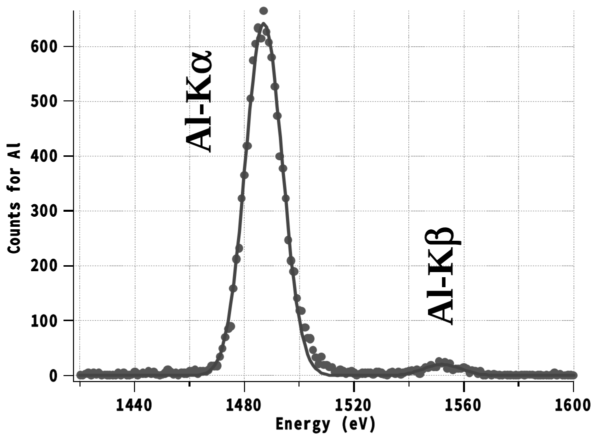

Fig. 4 Energy spectra for pure aluminum when the excitation voltage is 10 kV.

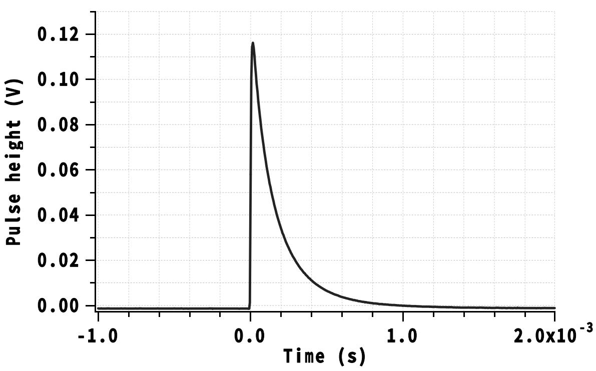

Fig. 5 Average pulse for Al-Kα for both samples measured using the microcalorimeter EDS system.

Figure 3 shows the calculated transmittance of these x-ray windows, revealing that the transmittance in this EDS system is inadequate to detect x-rays less than 2 keV because this system uses a beryllium window at room temperature. The transmittance can be improved by replacing beryllium by an aluminum-coated polymer thin film (Fig. 3). The dilution refrigerator of our EDS system achieves a minimum temperature of about 50 mK. The cold stage is stabilized at $85\,\mathrm{mK} \pm 10\,\mu\mathrm{K}$ by a PID control system, and the distance between the TES and the sample is 40-50 mm.

SYSTEM EVALUATION

Energy resolution and count rate

First, the energy resolution of the microcalorimeter EDS system was evaluated. Figure 4 shows the energy spectra for pure aluminum and measured using this system. The Al-Kα line and the Al-Kα lines are clearly observed. The average current pulse height for the both Al-Kα lines was 1.2 μA and the decay time (τ_{eff}) was 150 μs (Fig. 5). Based on Figs. 4, the energy resolution for the both Al-Kα lines was 14.2 eV. The measurement time was 500 s (45 cps) with the SEM beam current 10 nA. This low count rate was not due to inability of the EDS system but to the low solid angle. The estimated maximum count rate of this EDS system was about $1/20\tau_{eff}$ =333 cps without degrading the energy resolution [1]. In future versions of this EDS system, the measurement count rate can be improved from 45 cps to about 300 cps by increasing the solid angle by use of a poly capillary x-ray lens. For example, we can estimate the count rate about 300 cps when the optical gain is about 700 times and the distance between the TES and the sample is 85 mm.

Measured spectra for different samples

Three different samples were measured to evaluate the performance of the microcalorimeter EDS system: a tungsten silicide film (WSi_2), a mixture of five rare earth metals and three transition metals, and stainless steel (SUS 304). Here, we define the upper limit of the low voltage material analysis under 5 kV because the all elements can be analyze by using K lines for light elements (atomic

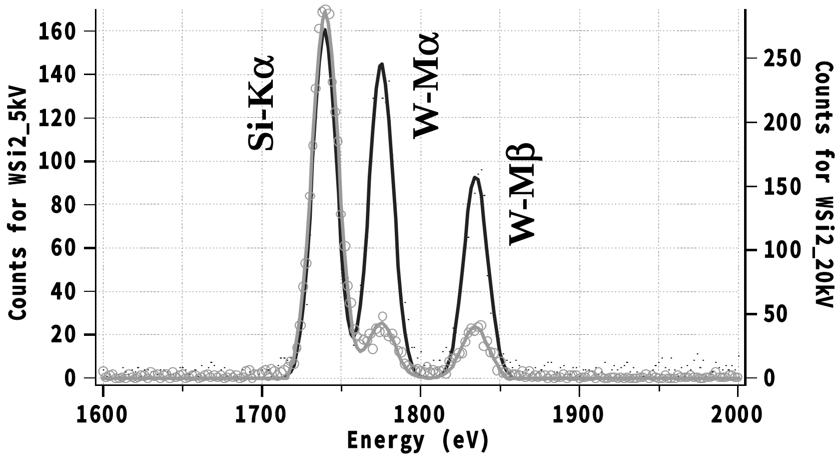

Fig. 6 Comparison between energy spectrum for 5 kV excitation and that for 20 kV (open circles) measured using the microcalorimeter EDS system.

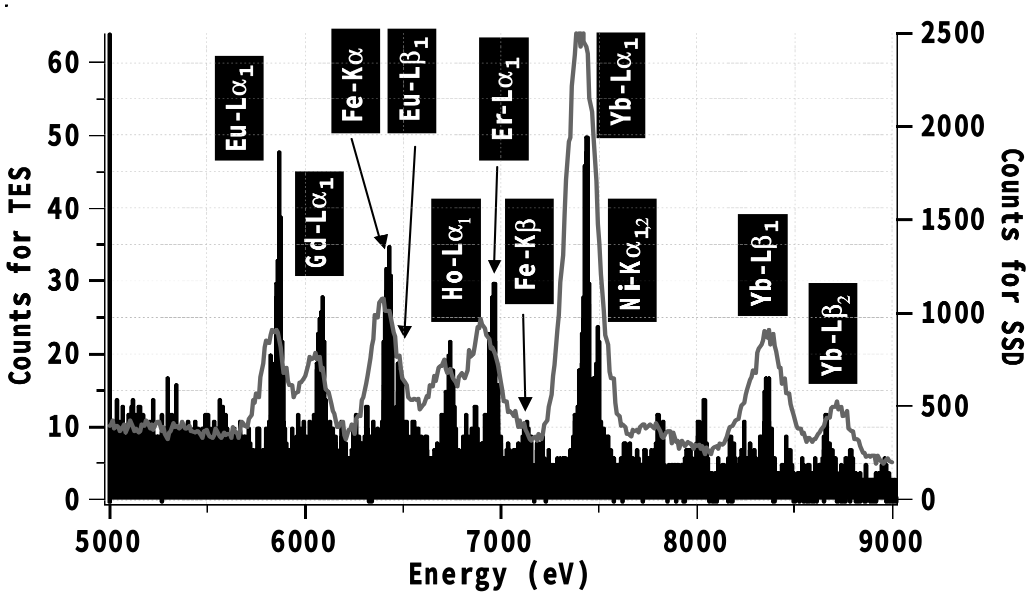

Fig. 7 Comparison between energy spectrum measured using the microcalorimeter EDS system and the SSD (solid line) for a mixture of five rare earth and three transition metals.

number 23) and L, M lines for other elements.

A WSi_2 film is commonly used as a LSI wiring material. A Si-Kα line (1740 eV) and W-Mα, βlines (1775 eV, 1835 eV) are emitted at the same time. A conventional EDS system cannot separate these three lines. Figure 6 shows the energy spectra for a WSi_2 (300 nm) film for excitation voltage 5 kV and 20 kV measured using this microcalorimeter EDS system. We chose the 5 kV and 20 kV as a typical excitation voltage in analyzing the material with a TES and a SSD. The intensity of the Si-Kα line was stronger than that of the W-Mα, Mβ lines for 20 kV because the electron beam penetrates deeply into the bulk and x-rays are generated in the bulk rather than around the surface. The peak intensity ratio of the Si-Kα line, W-Mα, and β lines for 5 kV were nearly equal to those of bulk WSi_2 (data not shown here), indicating that the x-rays are emitted mainly from around the surface. Such low voltage excitation is useful for analyzing the composition around the surface of a sample.

Figure 7 shows a comparison of the peak

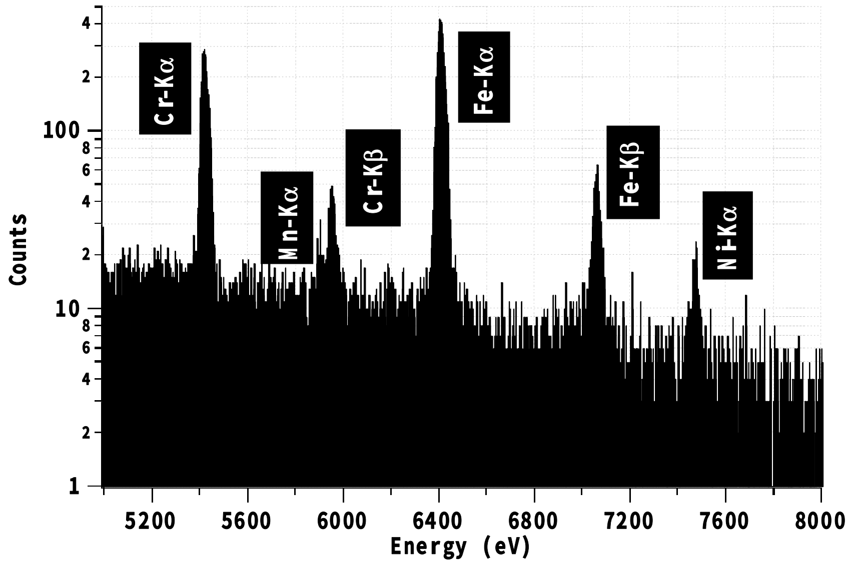

Fig. 8 Energy spectrum for stainless steel SUS 304 (containing 1.2% manganese) measured using the microcalorimeter EDS system.

separation for a mixture of 5 rare earth elements and 3 transition metals for an excitation voltage of 15 kV determined using our EDS system and using an solid silicon detector (SSD). The SSD showed three unresolved peaks, Fe-Kα (6403 eV) and Eu-Lβ_1 (6456 eV), Er-Lα_1 (6948 eV) and Fe-kβ (7057 eV), and Yb-Lα (7414 eV) and Ni-Kα (7477 eV). In contrast, the TES could easily separate these spectra.

Figure 8 shows the energy spectrum for an SUS-304 sample with our EDS system. We had measured the manganese content about 1.2 % using EPMA. In the measured spectra, Mn-Kα (5898 eV) was clearly observed and distinguished from the Cr-Kβ (5946 eV).

CONCLUSION

A microcalorimeter EDS system was developed based on a dilution refrigerator that can cool the system from room temperature to 4 K by using liquid helium. This system achieved an energy resolution of 14.2 eV for the Al-Kα line and an estimated count rate of 333 cps. This energy resolution is adequate to separate the K lines for light elements and the L, M lines for heavy elements below 5 kV. In the future, we will develop the EDS system without using a liquid helium free system.

REFERENCES

[1] D. A. Wollman, K. D. Irwin, G. C. Hilton, L. L. Dulcie, D. E. Newbury and J. M. Martinis, *Journal of Microscopy* **188** 196-223, December 3, 1997.

[2] K. D. Irwin, G. C. Hilton, J. M. Martinis, S. Deiker, Bergren, S. W. Nam, D. A. Rudman, and D. A. Wollman, Nucl. Instr. Meth. A **444**, 184 (2000).

[3] K. Tanaka, T. Morooka, K. Chinone, M. Ukibe, F. Hirayama, M. Ohkubo, and M. Koyanagi, Appl. Phys. Lett. **77**, 4196 (2000).

[4] Y. Takei, K. Tanaka, R. Fujimoto, Y. Ishisaki, U. Morita, T. Morooka, T. Oshima, K Futamoto, T. Hiroike, T. Koga, N. Iyomoto, T. Ichitsubo, K. Sato, T. Fujimori, K. Shinozaki, S. Nakayama, K. Chinone, N. Y. Yamasaki, T. Ohashi, and K. Mitsuda, Nucl. Instr. and Meth. A **523**, 134 (2004).

[5] T. Morooka, H. Myoren, S. Takada, and K. Chinone, Jpn. J. Appl. Phys. **42**, L1375 (2000).