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ARTICLE

Spectroscopic measurements of L X rays using a transition-edge sensor microcalorimeter for nondestructive analysis of transuranium elements

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The TES microcalorimeter was employed for spectroscopic measurements of L X rays emitted from ^{237}Np, ^{239}Pu, ^{241}Am and ^{244}Cm sources of typical transuranium (TRU) elements. Peaks of major L X rays of TRU elements were clearly identified in experimental energy spectra. The feasibility was considered for a nondestructive analysis of TRU elements based on experimental L X ray spectra measured by the TES microcalorimter.

Keywords: TES microcalorimeter; L X ray spectroscopy; Transuranium elements; nondestructive analysis

1. Introduction

Management of transuranium (TRU) elements is one of important tasks in nuclear cycle facilities. Currently, complicated chemical process is required to quantify TRU elements with the α ray spectroscopy because the emission probability of γ rays is lower than 0.03 % in α decay of Pu isotopes. On the other hand, most of TRU elements emit L X rays following internal conversion after their α-decay. The energy of L X rays emitted by TRU elements ranges from 10 to 25 keV. Accurate L X ray spectroscopy is expected to be useful for non-destructive assay of TRU elements. High purity germanium semiconductor (HPGe) detectors have been used in spectroscopic measurement of L X rays emitted from TRU elements so far. The accurate identification of L X ray peaks is difficult due to the insufficient energy resolution of the HPGe detector because small energy difference of L X ray lines. For identification of L X ray peaks of TRU elements, the energy resolution of the detector is required to be better than 100 eV of the full width at half maximum (FWHM). Recently several types of transition edge sensor (TES) microcalorimeters have been developed for X-ray and γ ray spectroscopy and achieved the energy resolution an order of magnitude better than that of typical semiconductor detectors [1,2]. Therefore, we had developed TES microcalorimeters for spectroscopic measurements of L X rays emitted by TRU elements [3,4].

In this work, the TES microcalorimeter was employed for spectroscopic measurements of L X rays emitted from ^{237}Np, ^{239}Pu, ^{241}Am and ^{244}Cm sources of typical TRU elements. After identification of major L X ray peaks in experimental spectra, the feasibility was considered for a nondestructive analysis of TRU elements based on experimental L X ray spectra obtained by the TES micro-calorimeter.

2. TES microcalorimeter and signal readout

A microcalorimeter consists of an energy absorber and a thermometer and measures the energy of an incident photon as a temperature rise [5]. The thermometer is an important component of the microcalorimeter for a precise measurement of the temperature rise. A TES microcalorimeter utilizes superconducting thin film thermometer which has the strong temperature dependence of the electric resistance in the phase transition region between superconducting and normal conducting states. **Figure 1** shows the concept of the operation of a TES microcalorimeter. The TES microcalorimeter chip is thermally connected to a cold heat sink of temperature T_b through a heat link of conductance G. A coil L is connected in series with the TES, while a shunt resistor R_S is connected in parallel with the TES–L line. A constant dc bias current I_0 supplied to the TES electric circuit is divided into the TES current I_{TES} and the shunt current I_S as shown in Figure 1. The energy of incident X ray photons is

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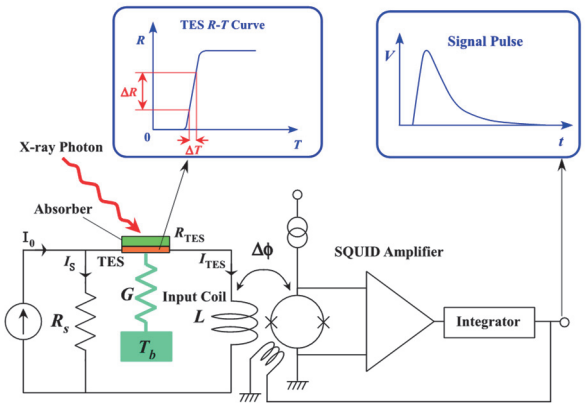

Figure 1. Concept of TES microcalorimeter operation.

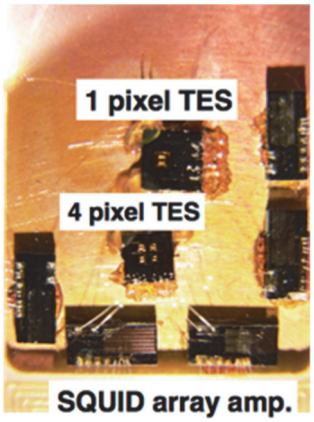

Figure 2. Photograph of single-pixel and 4-pixel-array TES calorimeters and the SQUID array amplifier chips were glued on the holder.

converted into a temperature rise ΔT in the absorber. As shown in the inset of Figure 1, the temperature rise ΔT induces a rapid increase of ΔR in the electric resistance of the TES. The increase in resistance ΔR causes a reduction in the TES current I_{TES}. The coil L induces a change in the magnetic flux $\Delta\varphi$ across a superconducting quantum interference device (SQUID). The magnetic flux change $\Delta\varphi$ is converted into a voltage signal pulse by the SQUID array amplifier with the flux-locking loop which maintains a constant magnetic flux across the SQUID [6] as shown in the inset of Figure 1.

In this work energy spectra of L X rays emitted from sources of typical TRU elements were measured with single-pixel and 4-pixel-array TES microcalorimeters which had been previously developed [3,4]. Transition temperature of the TES of Ti/Au bilayer structure ranged from 100 to 200 mK. A Au layer of 5.0 μm thickness was deposited on the TES to achieve an absorption efficiency of 50% in the detection of L X ray photons with energy from 10 to 20 keV. The single-pixel and 4-pixel-array TES calorimeters and the SQUID array amplifier chips were glued on the holder as shown in **Figure 2**.

3. Spectroscopic measurements

3.1. Setup

Because it is difficult to use liquid helium in a laboratory for handling TRU elements, a Gifford-McMahon cryocooler split-type dry ^{3}He-^{4}He dilution refrigerator (DR) using a remote helium cooling loop system [7] was employed to maintain the operating temperature of the TES microcalorimeters below 200 mK. The TES calorimeter holder of Figure 2 was attached to the cold stage of the DR.

Since the handling of plutonium isotopes is strictly controlled under various safety regulations, it is difficult to use plutonium isotopes with strong radioactivity for this experiment. In this work, checking sources of Pu isotope for calibration of a lung counter were used for spectroscopic measurement of L X ray. For the L X ray source of ^{239}Pu isotope, we used three lymph node blocks containing ^{239}Pu with total activity of 184.63 kBq. A sealed ^{241}Am source with activity of 370 kBq was used for spectroscopic measurements of L X rays emitted from ^{241}Am isotope. The L X ray source of ^{239}Pu or ^{241}Am isotope was placed in front of X ray window of the DR for preventing damage in cryogenic temperatures. The X ray window was made of a disk-shaped beryllium plate of 25 mm in diameter and 1 mm thick.

For spectroscopic measurements of L X rays emitted from ^{237}Np and ^{244}Cm isotopes, electroplated radioactive sources were used for placing inside the vacuum chamber of the DR. A ^{237}Np source with activity of 200 Bq or ^{244}Cm with activity of 4 kBq was wrapped with a polyimide tape of 150 μm thick for stopping α-rays and placed in front of the TES microcalorimeter chips.

3.2. Detection signal pulse analysis

Output signal pulses of the SQUID amplifier were digitized and recorded in a PC. Different values of the rise and decay time constant were observed in recorded signal pulses. By analyzing relationship between pulse shape of recorded signals and absorbed position of incident L X ray photons, events of L X ray photon detection in the absorber were found to correspond to signal pulses with the rise and the decay time constant ranging from 15 to 20 μs and 150 to 200 μs, respectively. Pulse height distribution was obtained by analyzing selected signal pulses of L X ray detection in the absorber with the optimal filtering method [8]. In the optimal filtering method, a shape of detection signal pulses is assumed to be same. However, a pulse shape of detection signals was found to slightly change with the energy of L X rays. Therefore, signal pulses of L X ray photon detection in the absorber were divided into three groups with respect to their height corresponding to L_α-, L_β- and L_γ-X rays emitted from the TRU source for obtaining the pulse height distribution by analyzing with the optimal filtering method.

A fluctuation in the holder temperature was observed

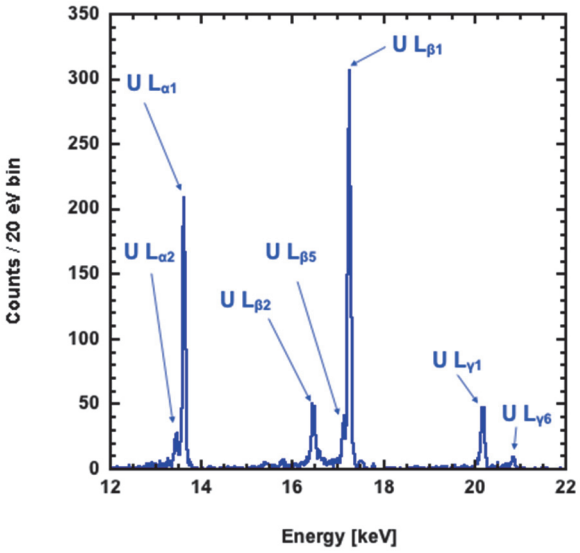

Figure 3. Energy spectrum of U L X-rays emitted from the ^{239}Pu source.

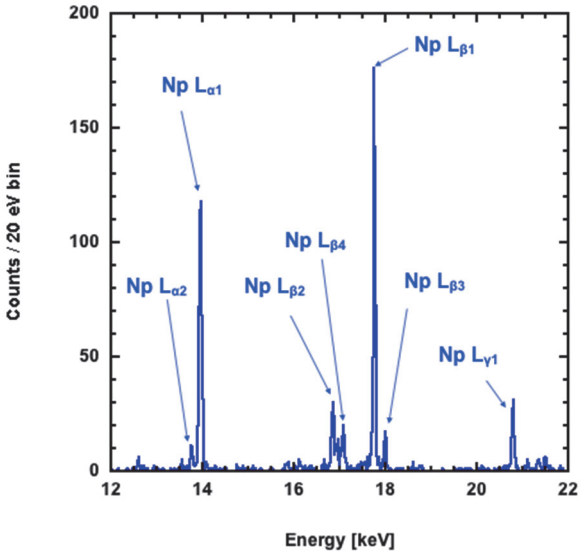

Figure 5. Energy spectrum of Np L X-rays emitted from the ^{241}Am source.

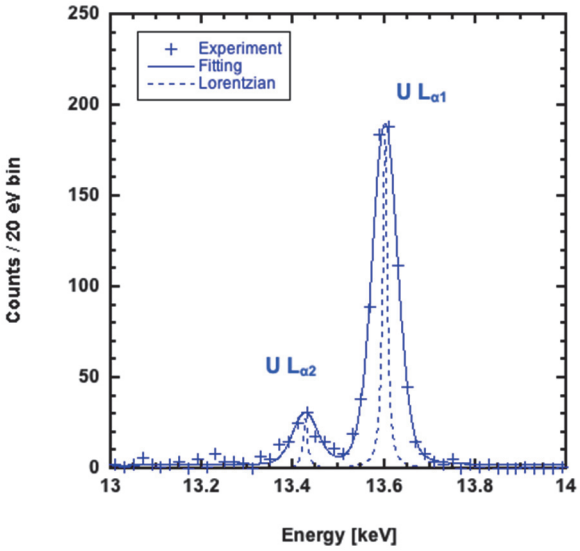

Figure 4. Fitting result of experimental U $L_{\alpha 1}$ and $L_{\alpha 2}$ peaks with Voigt function.

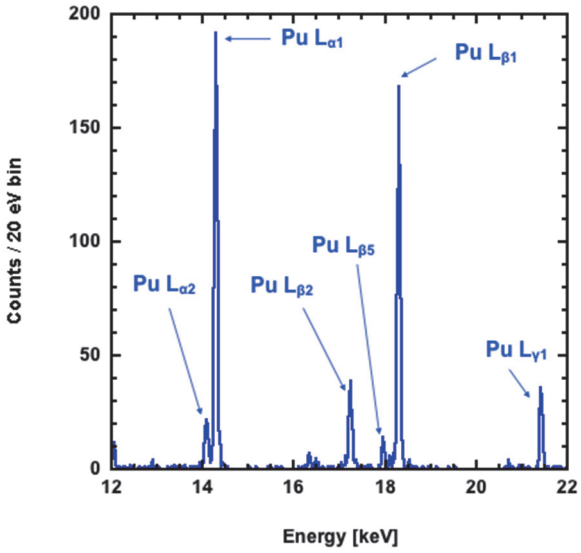

Figure 6. Energy spectrum of Pu L X-rays emitted from the ^{244}Cm source.

in long term operation of the DR. Because the TES operation temperature strongly depends on the bath temperature, the bath temperature fluctuation affects the value of pulse height of detection signals. The values of pulse height were corrected with using the base line voltage of the signal pulse which indicates the operating temperature of TES.

Width of obtained L X ray peaks are composed of the energy resolution of the TES microcalorimeter and the natural width of each L X ray line. The energy resolution of the TES microcalorimeters is the same order of magnitude of the natural width of major L X ray lines of TRU elements. In this work, Lorentzian function and Gaussian function was assumed to represent a profile of the L X ray line natural width and the instrumental energy resolution of the detector, respectively. To evaluate the energy resolution of the TES microcalorimeter, L X ray peaks in the obtained spectrum was fitted with Voigt function, which is convolution of Lorentzian function and Gaussian function.

3.3. Energy spectrum of L X rays

The single-pixel TES microcalorimeter was operated for spectroscopic measurement of L X rays emitted from the ^{239}Pu source which emit U L X rays following α decay of ^{239}Pu isotope. **Figure 3** shows the obtained energy spectrum of U L X rays emitted from the ^{239}Pu source. Major peaks of U L X ray are labeled in Figure 3.

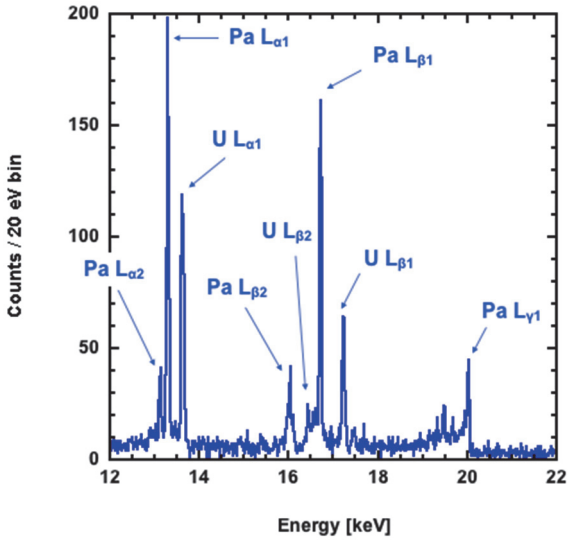

Figure 7. Energy spectrum of Pa L X rays and U L X rays are emitted from 237Np source.

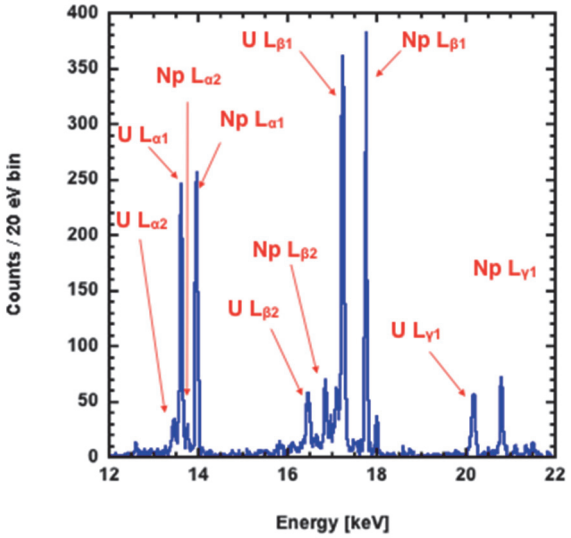

Figure 8. Predicted energy spectrum L X rays emitted from the Pu solution sample measured by the TES microcalorimeter.

Figure 4 shows a fitting result of experimental U $L_{\alpha 1}$ and $L_{\alpha 2}$ peaks with Voigt function. In Figure 4 the solid- and the dashed- line indicates fitting of experimental detector response and the Lorentzian function corresponding to the natural line width, respectively. The FWHM value of the energy resolution of the TES microcalorimeter was obtained to be 59 eV at U $L_{\alpha 1}$ peak of 13.618 keV with the natural width of 11.7 eV.

Selected one-pixel of 4-pixel array TES microcalorimeter was operated for spectroscopic measurement of L X rays emitted from the ^{241}Am source which emit Np L X rays are emitted following α decay of ^{241}Am isotope. **Figure 5** shows the obtained energy spectrum of Np L X rays emitted from the ^{241}Am source. Major peaks of U L X ray are labeled in Figure 5. The FWHM value of the energy resolution of the TES microcalorimeter was obtained to be 34 eV at Np $L_{\alpha 1}$ peak of 13.946 keV with the natural width of 11.8 eV.

Table 1. Values of radioactivity- and L X ray yield-ratio for ^{237}Np, Pu isotopes, ^{241}Am and ^{244}Cm estimated in the case of empirically assumed waste-solution.

	Radioactivity ratio	L X ray yield ratio
^{237}Np	0.01	0.17
Pu isotopes	0.25	0.71
^{241}Am	1	1
^{244}Cm	0.25	0.058

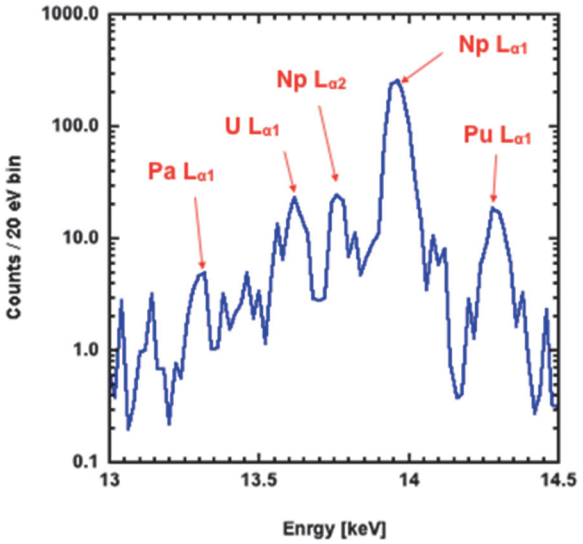

Figure 9. Predicted energy spectrum of L X rays emitted from the waste-solution measured by the TES microcalorimeter.

Selected one-pixel of 4-pixel array TES microcalorimeter was operated for spectroscopic measurement of L X rays emitted from the ^{244}Cm source which emit Pu L X rays are emitted following α decay of ^{244}Cm isotope. **Figure 6** shows the obtained energy spectrum of Pu L X rays emitted from the ^{244}Cm source. Major peaks of Pu L X ray are labeled in Figure 6. The FWHM value of the energy resolution of the TES microcalorimeter was obtained to be 43 eV at Pu $L_{\alpha 1}$ peak of 14.279 keV with the natural width of 12.2 eV.

Selected one-pixel of 4-pixel array TES microcalorimeter was operated for spectroscopic measurement of L X rays emitted from the ^{237}Np source. Following α decay of ^{237}Np isotope, ^{233}Pa isotope is generated and then ^{233}U isotope is generated following β^- decay of ^{233}Pa isotope. Because a decay of ^{237}Np and β^- decay of ^{233}Pa exist as radiative equilibrium, Pa L X rays and U L X rays are emitted from ^{237}Np source. **Figure 7** shows the obtained energy spectrum of Pa L X rays and U L X rays are emitted from ^{237}Np source. Major peaks of Pa L X rays and U L X rays are labeled in Figure 7. Background counts were caused by bremsstrahlung generated by β^- rays. The FWHM value of the energy resolution of the TES microcalorimeter

was obtained to be 65 eV at Pa $L_{\alpha 1}$ peak of 13.291 keV with the natural width of 11.1 eV. As shown in Figures 3–7, peaks of major L X rays of TRU elements were clearly identified in experimental energy spectra.

4. Feasibility of nondestructive analysis

The feasibility was considered for a nondestructive analysis of TRU elements based on experimental L X ray spectra in Figures 3-7.

For the case of a Pu solution in a nuclear fuel reprocessing facility for 10 years, ^{238}Pu, ^{239}Pu and ^{240}Pu and ^{241}Am were assumed to be major TRU elements contained in the Pu solution. The yield of L X rays emitted by the Pu isotopes was approximated to be equal to that by ^{241}Am isotope. **Figure 8** shows a predicted energy spectrum L X rays emitted from the Pu solution sample measured by the TES microcalorimeter. Pu isotopes and ^{241}Am are expected to be identified by clear peaks of L_α, L_β and L_γ series of U and Np elements in Figure 8.

For the case of a high radioactivity waste-solution in a nuclear fuel reprocessing facility, the waste-solution is assumed to contain ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Am and ^{244}Cm. Values of radioactivity- and L X ray yield-ratio for each TRU element were estimated in the case of empirically assumed waste-solution. Estimated results are listed in **Table 1**. **Figure 9** shows a predicted energy spectrum of L X rays emitted from the waste-solution measured by the TES microcalorimeter. Major TRU elements are expected to be identified by analyzing L X ray peaks in Figure 9.

5. Conclusion

The TES microcalorimeter was employed for spectroscopic measurements of L X rays emitted from ^{237}Np, ^{239}Pu, ^{241}Am and ^{244}Cm sources of typical transuranium (TRU) elements. Peaks of major L X rays of TRU elements were clearly identified in experimental energy spectra with the energy resolution better than 100 eV FWHM. The feasibility was considered for a nondestructive analysis of TRU elements based on experimental L X ray spectra measured by the TES microcalorimter.

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