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	作成者: 阪上, 正信
	メールアドレス:
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Some Characteristics of Low Energy Photon Spectrometer (LEPS) and Its Applications to Radiochemical Studies of Zircon

Hiroshi MATSUDA and Masanobu SAKANOUE

Radiochemical Laboratory, Department of Chemistry,
Faculty of Science, Kanazawa University
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Abstract The characteristics of LEPS installed in Kanazawa University were examined in regard to the absolute counting efficiency in various low energy photon region and several spectra of standard sources.

The applications of this system to the radiochemical study of zircon crystal grains were made, that is, the measurements of its natural radioactivity, X-ray fluorescence analysis with external radioisotope sources and neutron activation analysis. The list of various elements identified by the methods applied was given.

1. Introduction

Spectrometric measurements of low energy photons are very valuable for the identification and quantitative determination of some radioactive nuclides which emit low energy γ-rays or X-rays. Recently, for this purpose, the low energy photon spectrometer, called as LEPS, has been made and several commercial instruments have become available. There are three types of semiconductor detector used as LEPS, that is, lithium drifted silicon 〔Si(Li)〕, lithium drifted germanium 〔Ge(Li)〕 and intrinsic germanium. In our laboratory, we have examined some charateristics of either a Ge(Li) detector or an intrinsic Ge detector, and these data are presented in this report with its applications to the radiochemical studies of zircon crystals which has been also used for fission track studies.

2. Characteristics of LEPS

2-1 Instrumentation

The detector has 16 mm active diameter and 4.95 mm active depth. This detector having 1000 $\mu g/cm^2$ gold contact on its surface is set in a vacuum container having 0.13

mm thick beryllium window. The cooling of this detector is made by liquid nitrogen contained in vertical type cryostat. The electronic pulse from the detector is led to the cooled preamplifier of ORTEC 117B. As shown in the block diagram (Fig.1) of this spectrometer system, the detector bias is supplied from ORTEC 459 bias supply and the pulse is amplified by linear amplifier of ORTEC 452, followed by a pulse height analysis with CANBERRA 8100/e multichannel analyser. The spectrum data can be displayed on a cathode ray tube and recorded by a teletype.

2–2 Energy resolution and counting efficiency

The energy resolution expressed by FWHM (full width at half maximum of peak height) of this Ge detector is 280 eV at 5.9 KeV of Cr K_βX–ray and 530 eV at 122 KeV of ^{57}Co γ-ray at 1000 volts of operation bias. Such a high resolution of this LEPS in low energy region is shown in some spectra for various standard sources (Fig.2a–2h). For quantitative analysis by photon spectrometry, it is essential to know the absolute efficiency of each full energy peak, namely the ratio of the peak counting rate to the emission rate of a definite energy photon from a nuclide. In order to know the peak efficiencies at various photon energies, the absolute calibration method was applied by using several standard sources which contain a known amount of radioactive nuclide, respectively. The γ reference sources having spectra shown in Fig. (2a–2f) were obtained as a set from the Radiochemical Centre Amersham and used for the measurement. Though the standard source of ^{203}Hg in this set was not used, all other standard sources were measured at the distance of 9 cm from a beryllium window. From disintegration rate of each radioactive nuclide, the emission rate of a definite energy γ-ray is calculated and compared with the counting rate of a peak in that energy region. The accuracy of the absolute efficiency depends on several factors, that is, the reproducibility of the geometrical situation of a source to the detector, the estimation of the exact peak area measured and the accuracy of the content of the standard radioactive nuclides. The results as for the absolute peak efficiency are summarized in

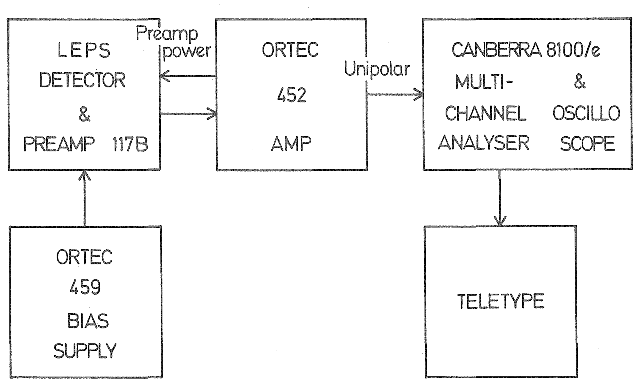


Fig. 1 Block diagram of the electronic components.

Fig. 2-a

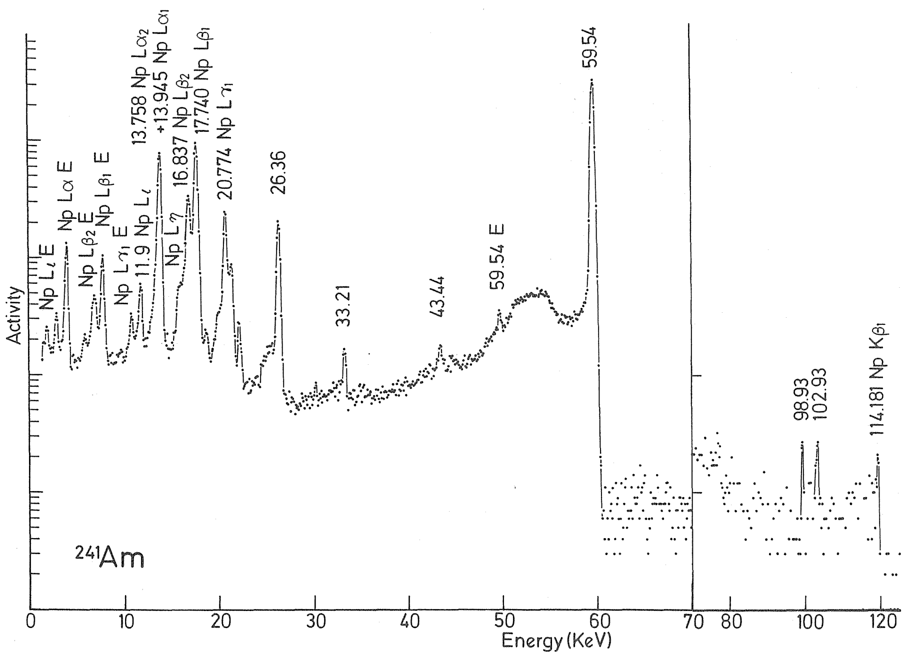


Fig. 2-b

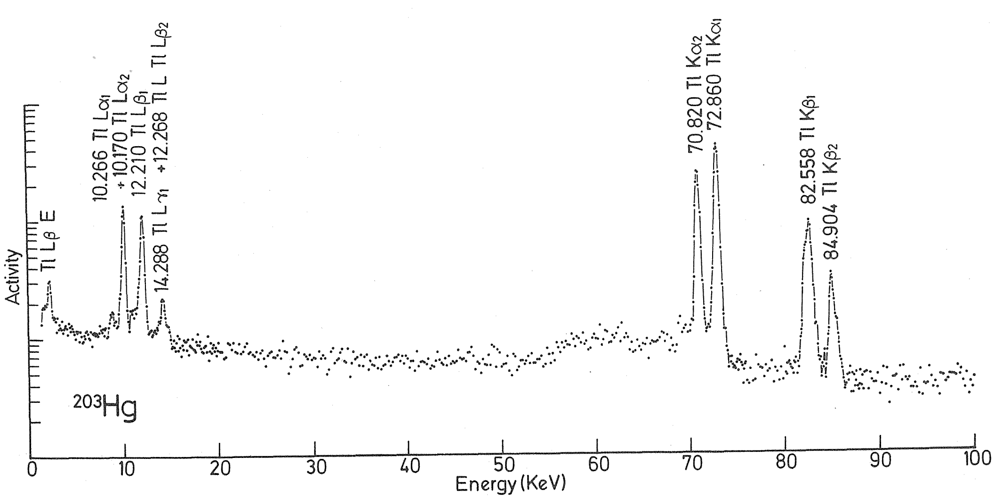


Fig. 2 Spectra of photon from standard sources with LEPS [Ge].
(a) ^{241}Am (b) ^{203}Hg (c), (c′) ^{133}Ba (d) ^{137}Cs (e) ^{88}Y
(f) ^{57}Co (g) "U"metal foil (h)"Th"metal foil

Fig. 2-c

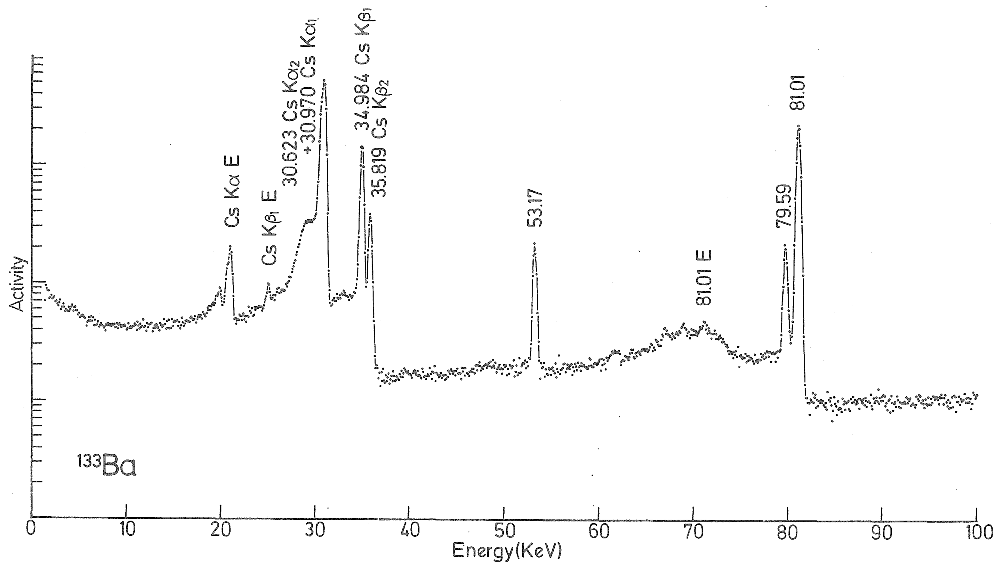

Cs Kα E
Cs Kβ1 E
30.623 Cs Kα2
+30.970 Cs Kα1
34.984 Cs Kβ1
35.819 Cs Kβ2
53.17
81.01 E
79.59
81.01
Activity
133Ba
0
10
20
30
40
50
60
70
80
90
100
Energy(KeV)

Fig. 2-d

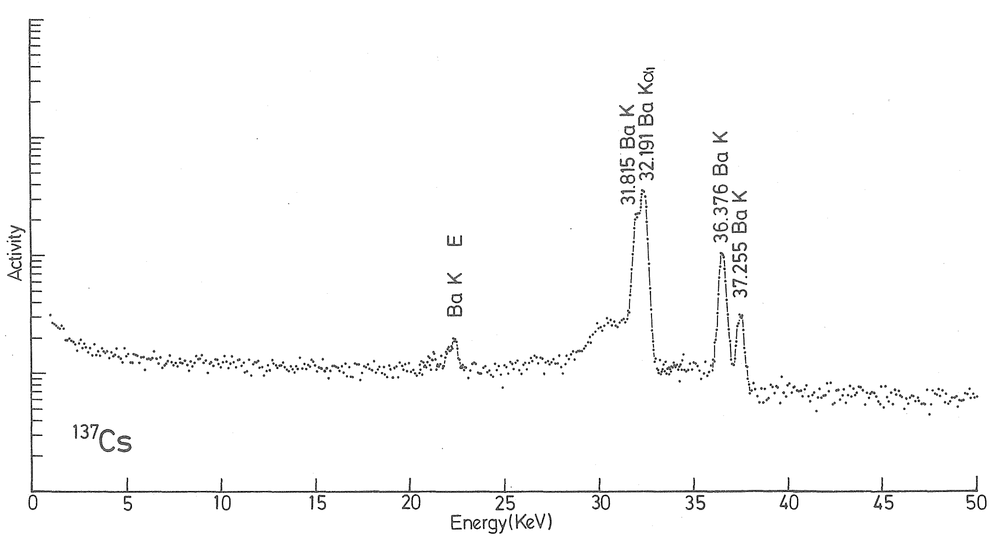

Ba K E
31.815 Ba K
32.191 Ba Kα1
36.376 Ba K
37.255 Ba K
Activity
137Cs
0
5
10
15
20
25
30
35
40
45
50
Energy(KeV)

Fig. 2-c′

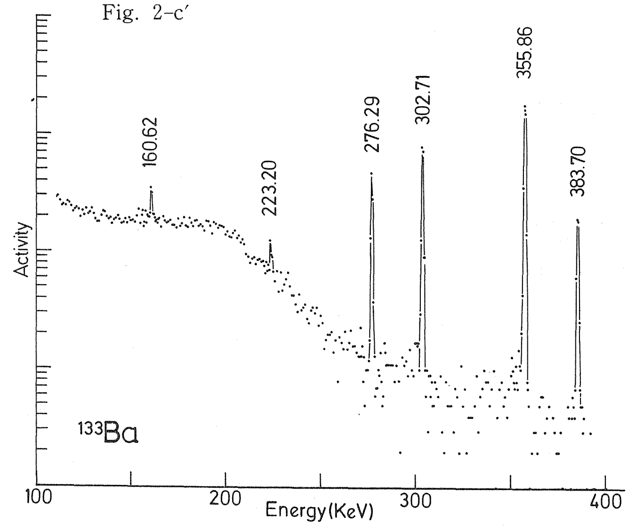


Fig. 2-e

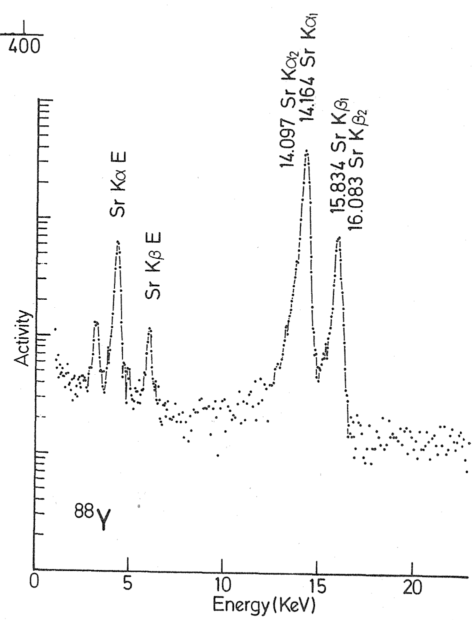


Fig. 2-f

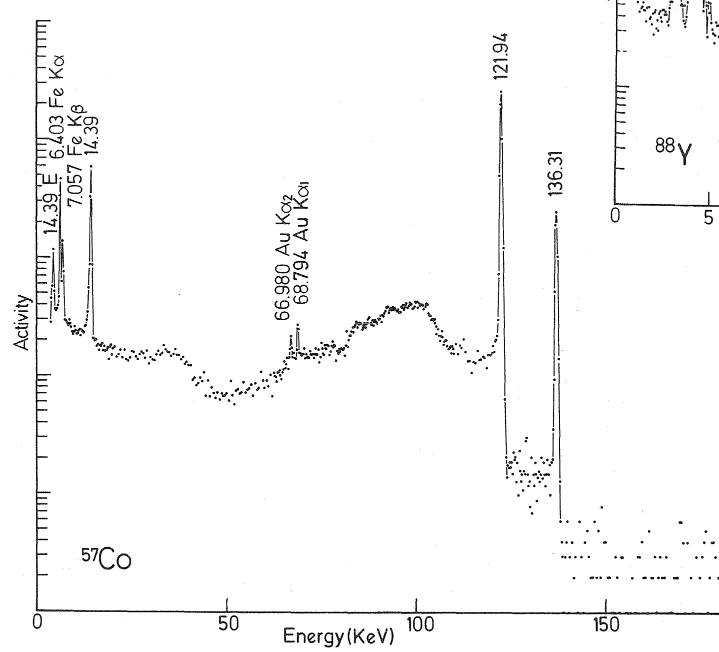


Fig. 2-g

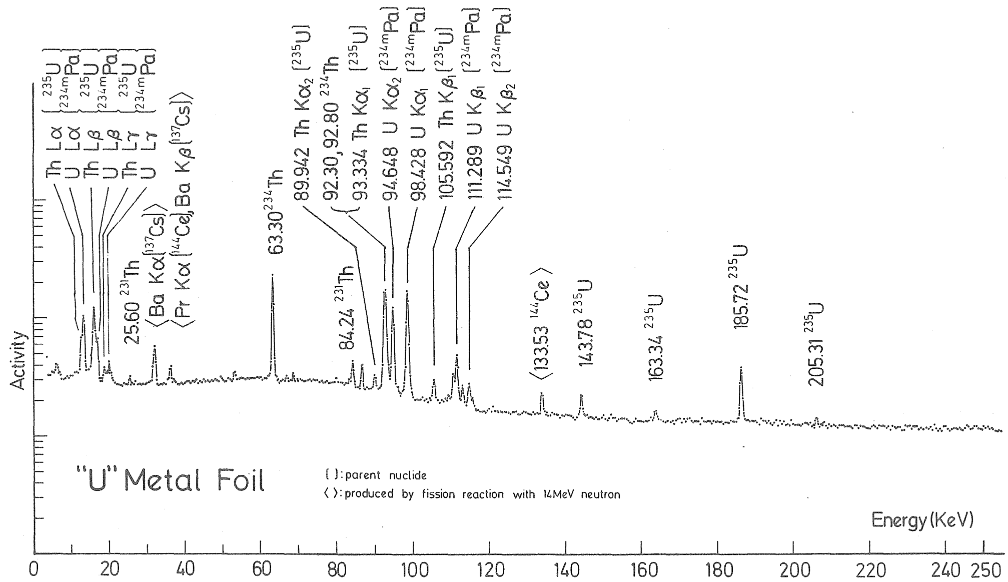

Activity
Th Lα [235U]
U Lα [234mPa]
Th Lβ [235U]
U Lβ [234mPa]
Th Lγ [235U]
U Lγ [234mPa]
25.60 231Th
⟨Ba Kα[137Cs]⟩
⟨Pr Kα [144Ce], Ba Kβ [137Cs]⟩
63.30 234Th
84.24 231Th
89.942 Th Kα2 [235U]
92.30, 92.80 234Th
93.334 Th Kα1 [235U]
94.648 U Kα2 [234mPa]
98.428 U Kα1 [234mPa]
105.592 Th Kβ1 [235U]
111.289 U Kβ1 [234mPa]
114.549 U Kβ2 [234mPa]
⟨133.53 144Ce⟩
143.78 235U
163.34 235U
185.72 235U
205.31 235U
"U" Metal Foil
[ ]: parent nuclide
⟨ ⟩: produced by fission reaction with 14MeV neutron
Energy (KeV)
0
20
40
60
80
100
120
140
160
180
200
220
240
250

Fig. 2-h

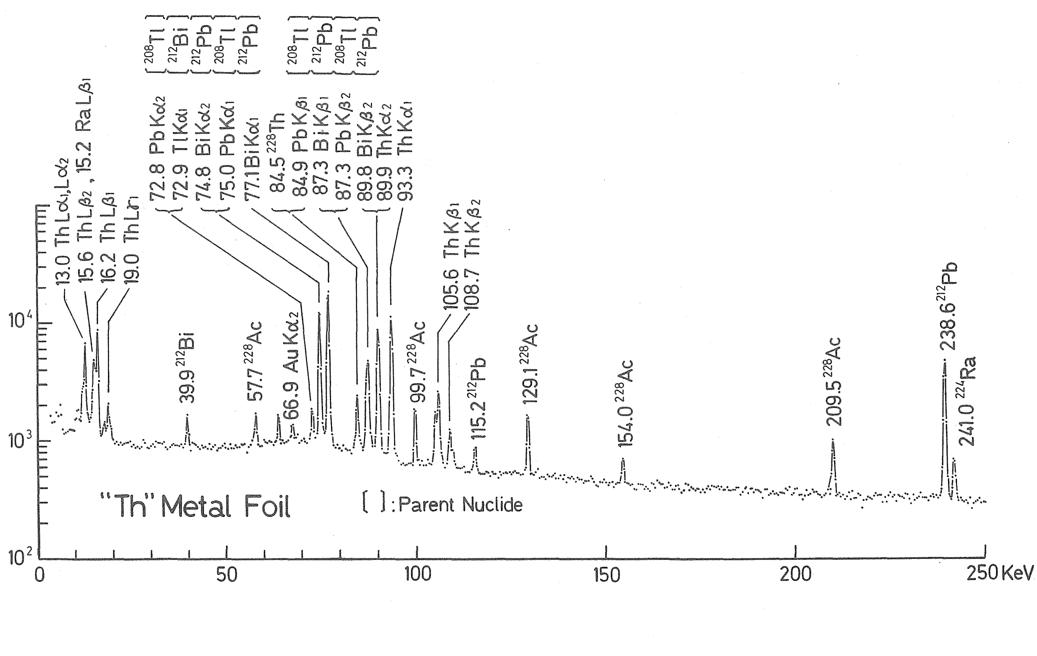

13.0 Th Lα1, Lα2
15.6 Th Lβ2, 15.2 Ra Lβ1
16.2 Th Lβ1
19.0 Th Lγ1
72.8 Pb Kα2 [208Tl]
72.9 Tl Kα1 [212Bi]
74.8 Bi Kα2 [212Pb]
75.0 Pb Kα1 [208Tl]
77.1 Bi Kα1 [212Pb]
84.5 228Th
84.9 Pb Kβ1 [208Tl]
87.3 Bi Kβ1 [212Pb]
87.3 Pb Kβ2 [208Tl]
89.8 Bi Kβ2 [212Pb]
89.9 Th Kα2
93.3 Th Kα1
39.9 212Bi
57.7 228Ac
66.9 Au Kα2
99.7 228Ac
105.6 Th Kβ1
108.7 Th Kβ2
115.2 212Pb
129.1 228Ac
154.0 228Ac
209.5 228Ac
238.6 212Pb
241.0 224Ra
"Th" Metal Foil
[ ]: Parent Nuclide
10^4
10^3
10^2
0
50
100
150
200
250 KeV

Fig.3. It is confirmed that the detector has the high efficiency between 10 KeV and 100 KeV.

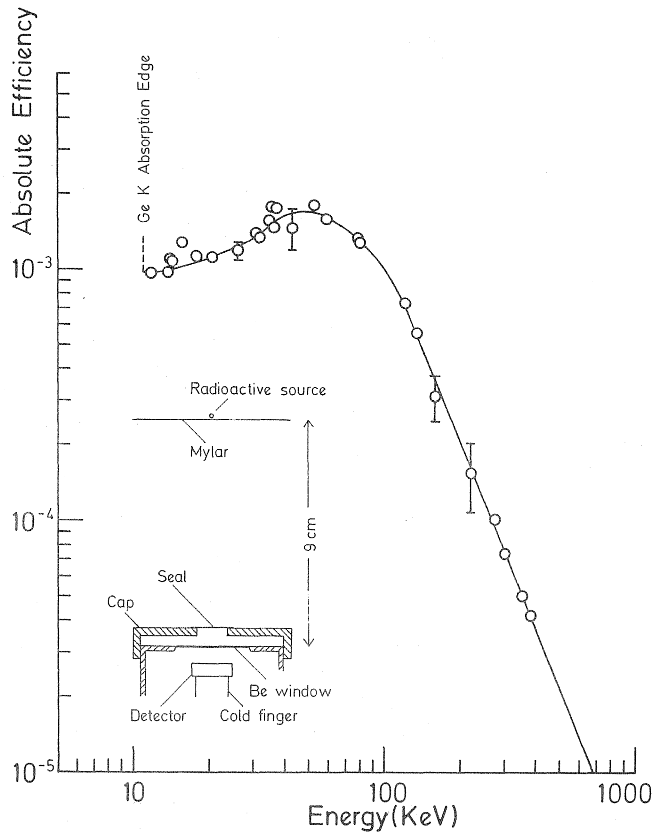


Fig. 3 Absolute efficiency curve of LEPS [Ge].

3. Application to zircon grains

Zircon crystals contain generally a trace ammounts of radioactive materials together with fairly high concentration of rare earth elements, though it has very low level radioactivity. Therefore, it is interesting to measure the radioactivity of zircon and identify the elements contained in this mineral by using various radiochemical techniques with LEPS. The sample used is the zircon crystal grains used for ceramics, which could be obtained from Tokyo Yogyo company.

3-1 Natural radioactivity

At first, the grain samples were packed in polyethylene tube and its natural radioactivity was measured with the normal Ge(Li) detector[1] having the effective volume of 20 cc. The γ-ray spectrum shown in Fig.4 was obtained. The sharp peaks obtained are those of ^{214}Bi and ^{214}Pb which belong to the uranium series. Beside these peaks, other peaks due to ^{228}Ac and ^{208}Tl were also observed, showing the existence of thorium series elements. The low energy spectum of the sample was also measured by spreading the grains on a mylar sheet and setting the mylar on the head of LEPS. The spectrum shown in Fig.5 was obtained. Each peaks observed in this spectrum is identified as follows; the peaks due to the nuclides which belong to uranium, thorium and actinium series, the characteristic X-ray peak accompanied with the decay of those radioactive nuclides and the fluorescence X-ray peak from zirconium and hafnium which is excited by these radioactivity. In this way, the existence of hafnium in zircon grains could be

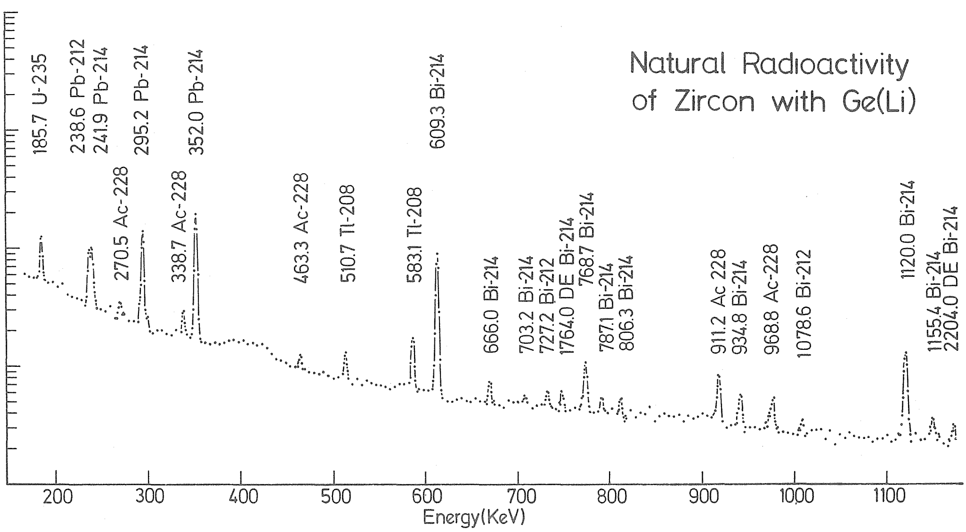


Fig. 4 Photon spectrum of zircon measured with normal Ge(Li) detector.

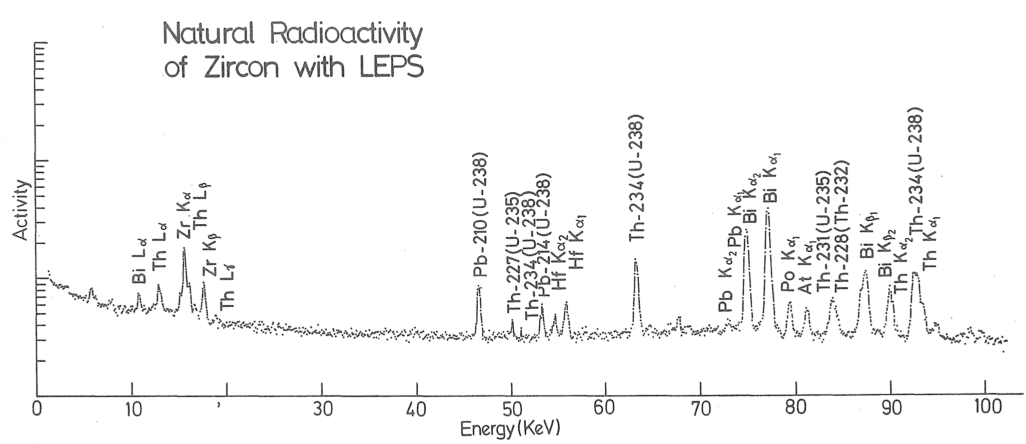


Fig. 5 Photon spectrum of zircon measured with LEPS [Ge(Li)].

easily confirmed. Besides the peaks mentioned above, the peaks of ^{234}Th (63KeV) and ^{235}U (185KeV) are also detected by the measurement with LEPS and can be used to determine the content of ^{238}U and ^{235}U in those zircon grains.

3-2 X-ray fluorescence analysis

The excitation of fluorescence X-rays can be made more intensely by the external sources having more radioactivity than natural one. The efficiency of such excitation may depend on various exciting radiations having different energies. To study this circumstance, the standard sources mentioned before were used also for the exciting sources. The results are shown in Fig.6a-6d. It was found that the most effective source among these nuclides was ^{241}Am for zirconium and ^{57}Co for hafnium. Otherwise, if the radioactive sources are more intense or the contents of the rare earth element is much higher, the identification of some rare earth elements may be also made by such fluorescence X-ray analysis. The variation of maximum peak height of zirconium K_{α} line according to the number of crystal grains was examined by using ^{241}Am γ-ray excitation as shown in Fig.7. In this case, samples were spread uniformly on the mylar sheet. On the other hand, a semi-quantitative determination of zirconium with ^{133}Ba source was tried by confining the diameter of the zircon grain sample as shown in Fig.8 which shows also the results of this experiment. The decrease of the integral counts of peak area in the case of large amount of samples is due to the absorption of zirconium KX-ray by the thick layer of the sample.

Fig. 6-a

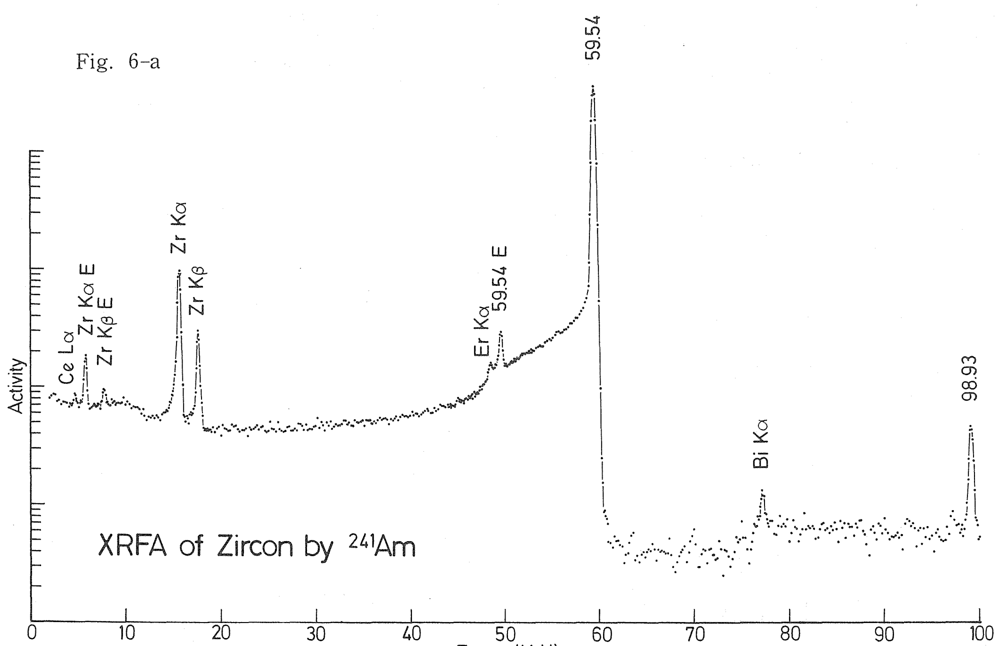


Fig. 6 Spectra of X-ray fluorescence analysis of zircon grains;
External excitation source; (a) ^{241}Am (b) ^{203}Hg (c) ^{133}Ba (d) ^{57}Co

Fig. 6-b

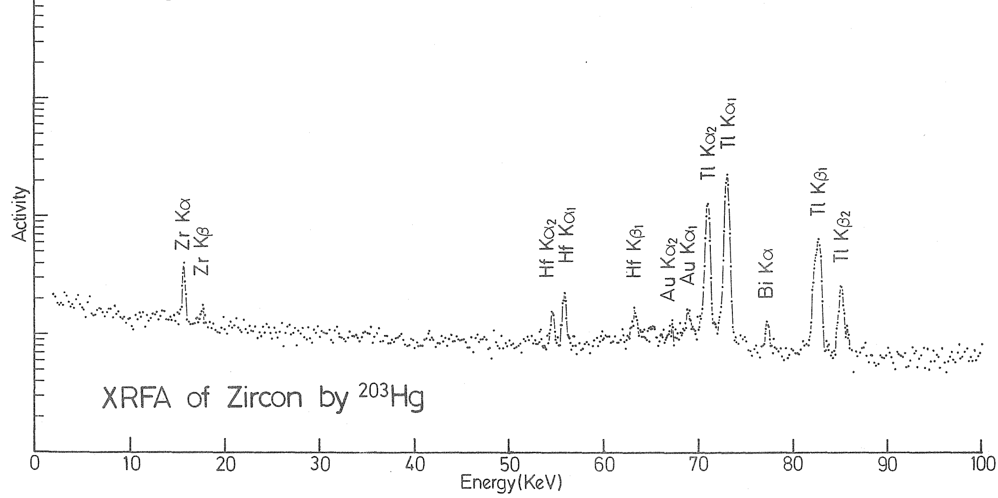


Fig. 6-c

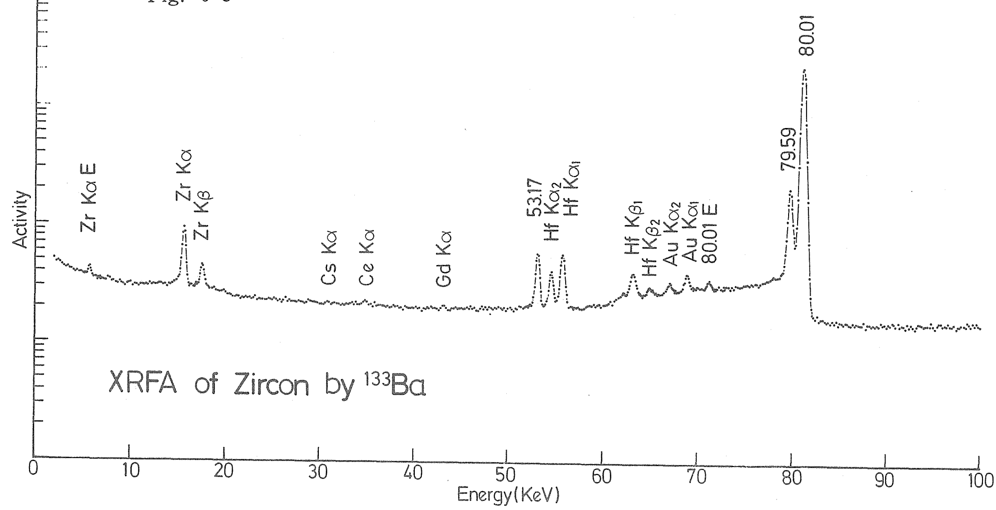
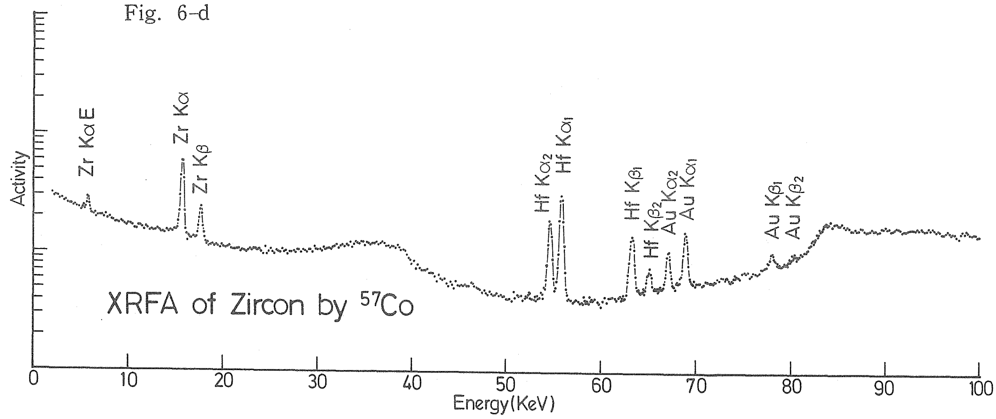


Fig. 6-d



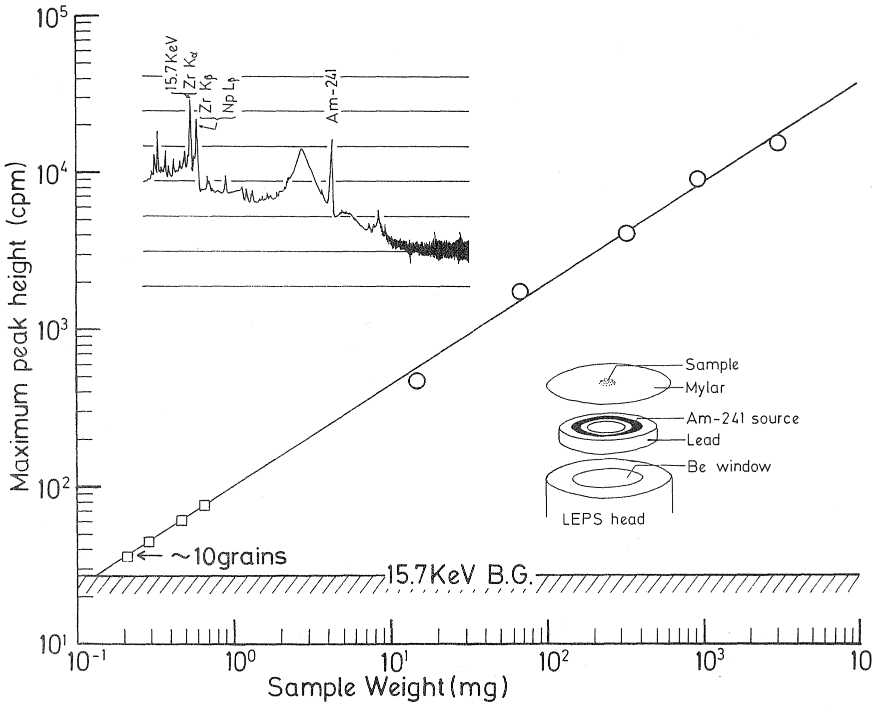


Fig. 7 The variation of maximum peak height of zirconium Kα line according to the amounts of zircon grains spread on a mylar sheet by using ^{241}Am as external excitation source.

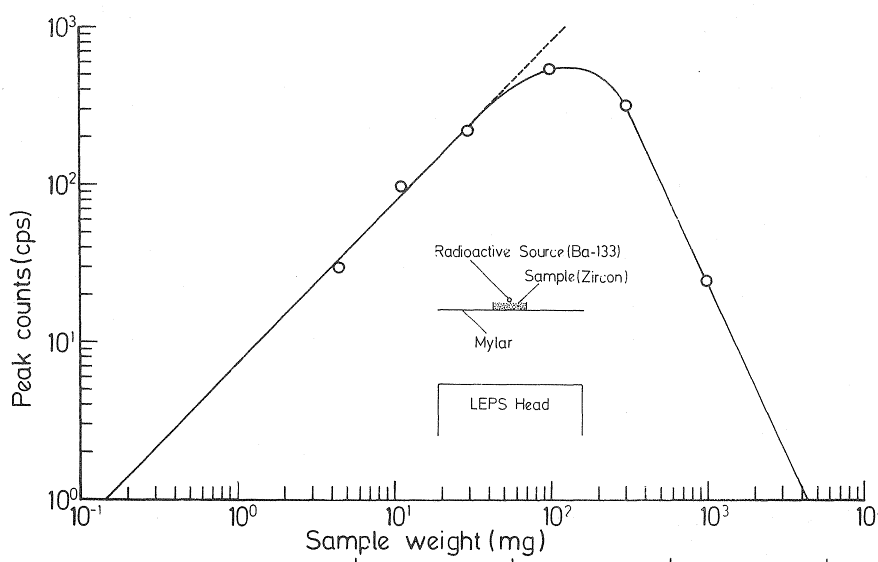


Fig. 8 The variation of peak area of zirconium Kα line according to the amounts of zircon grains in a definite geometrical configuration by using ^{133}Ba as external excitation source.

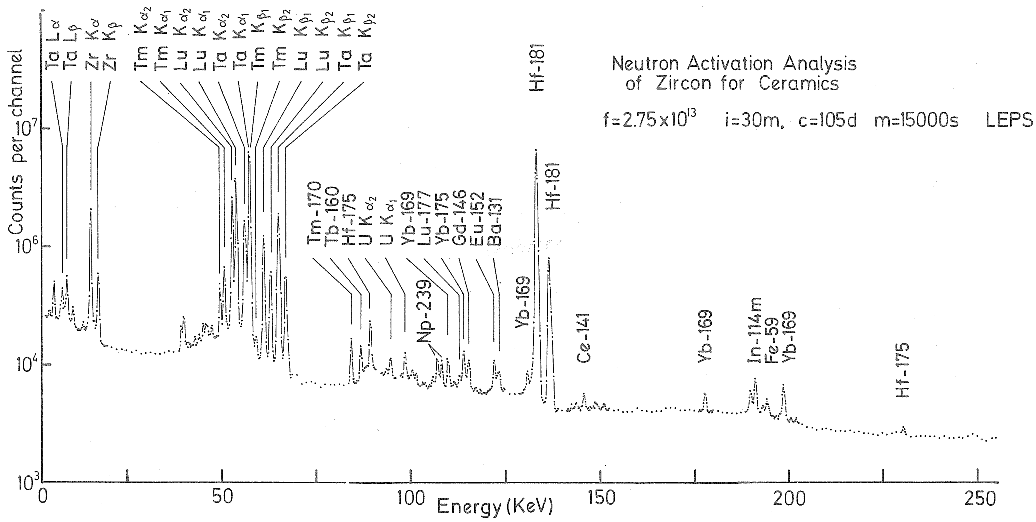


Fig. 9 Photon spectrum of neutron-activated zircon measured with LEPS.

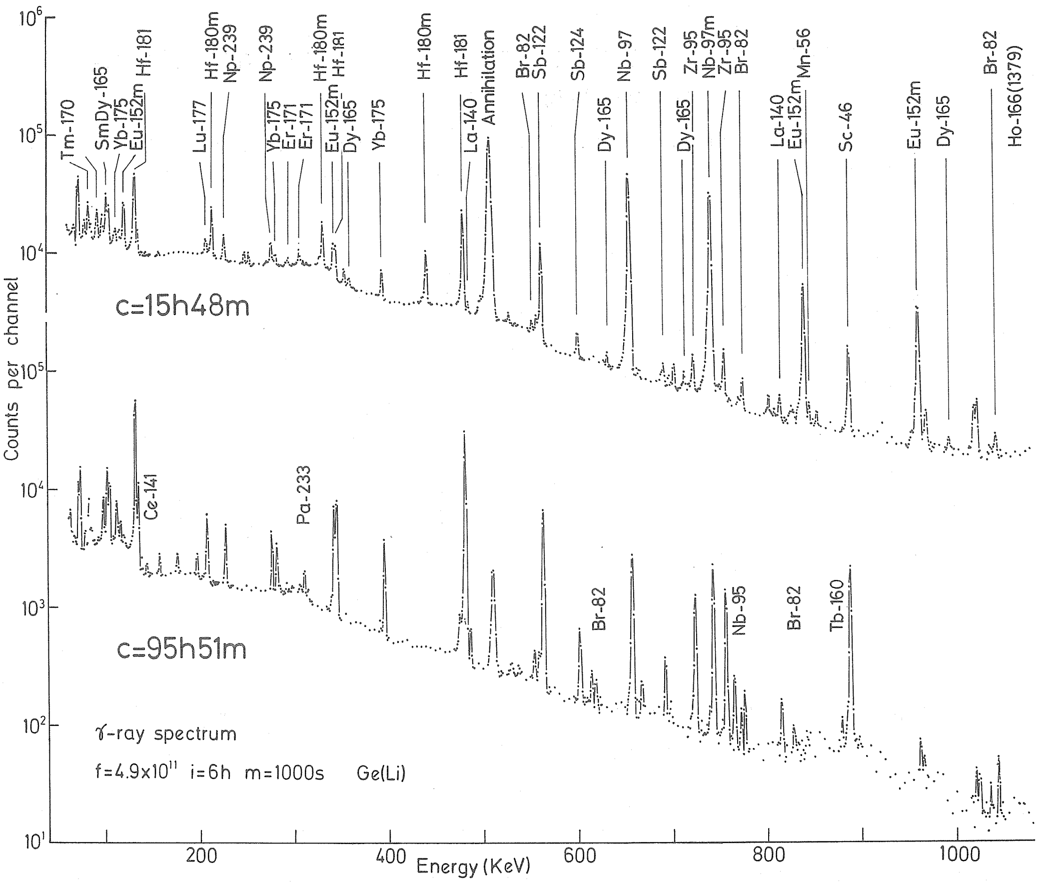


Fig. 10 γ-ray spectrum of neutron-activated zircon measured with normal Ge(Li) detector.
c : cooling time

3-3 Neuron activation analysis

Neutron activation analysis is more sensitive method than X-ray fluorescence analysis. The low energy photon spectrum shown in Fig.9 was obtained by irradiating the grain sample in the nuclear reactor of Kyoto University, the neutron flux density (f), irradiation time (i), cooling time (c) and measurement time (m) being shown also in this figure. The γ-ray spectra shown in Fig.10 was obtained by irradiating the zircon grain sample in the nuclear reactor of Rikkyo University. The conditions of the irradiation and measurement are also shown in that figure. Much more elements were identified in these spectra by using the difference of both energy and the half-life of each nuclide produced by neutron irradiation. Several peaks observed in the spectrum taken by the low energy photon spectrometer are due to the fluorescence X-ray which were excited by the radiation enitted from neutron activated nuclides. Such inner excitation should be remarked in the analysis using low energy photon spectrometer and it is interesting that these peaks show the existence of zirconium, ytterbium, hafnium and uranium in the sample.

The list of various elements identified by each method applied in this study is summarized in Fig.11 which shows the appropriate range of cooling time for activation analysis to identify each element. The fission track study will be reported in other report.

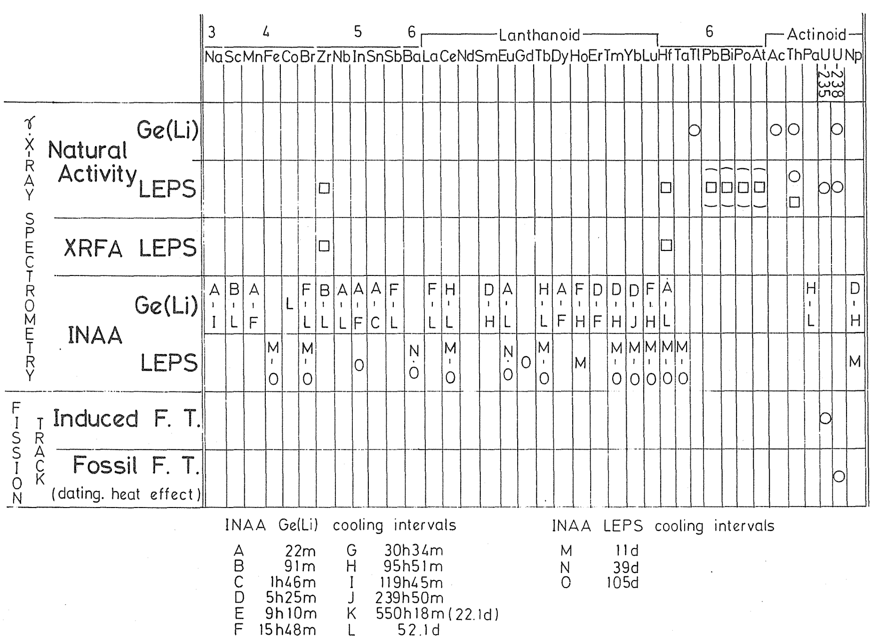


Fig. 11 The comparison of various methods applied to identify various elements in zircon.

Reference

1) Hideo KAWAZU and Masanobu SAKANOUE, Sci. Rep. Kanazawa Univ., **16**, 1. (1971).