Classification of ores used for the radiation source in NORM as consumer products by PGAA

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Summary. NORM means Naturally Occurring Radioactive Material. Our object in this study is only consumer products in several kinds of NORM. The existence of NORM in our living-environment raises the radiation exposure dose of the users. This has become a world-wide issue. The sources of NORMs are mainly radionuclides in the thorium (Th) and uranium (U) series. However, we have little published information about the kinds of ores added to NORMs and there are little indications on the NORMs' labels. It is well-known that Th and U usually exist with rare earth elements. So we sought a classification method for identifying the kinds of ores added to the consumer products from the correlation of the concentration ratios of the Th/U and the Gd/Sm in NORM and ores. The conventional γ -ray spectrometry by a Ge detector for analysis of Th and U and the prompt gamma-ray activation analysis for the determination of Sm and Gd were employed. It was found that the radiation sources of NORM in the consumer products in recent markets were mainly monazite. This classification method is found to be useful to identify the ores used for the radiation sources in consumer products of NORMs.

1. Introduction

Radium-226 was added to consumer products in Europe in the early 20th century [1]. These consumer products are called Radioactive Consumer Products (RCP). Fluorescent paint of radium added for writing figures on clock-faces, gas lantern mantles [2] of thorium added and some uranium added cosmetics of cream and face-powder are famous as RCP. However, production of almost all these RCP in Europe was stopped in the late 1970s. These RCP are not sold in the European markets today. However, there might be still small numbers of RCP sold on the antique markets [1].

In Japanese markets today, many RCP are sold and many manufactures are claiming some beneficial effects. For example, there are cosmetics [3] claiming hormesis effects (hormesis cosmetics) through Japanese home pages (HP) of the Internet. Also some wall papers [4] are sold with claim

that the emission of “negative ion” from the products is healthy for users. The radionuclides in these RCP have been clearly identified by measuring their gamma-rays. However, the kinds of ores added in these RCP have not been identified yet.

These RCP are under legal regulation. For example, no radioactive substances should be added in foodstuffs (food, drinking water, feed), toys, cosmetics and personal ornaments in the EURATOM Council Directive [5]. Germany and 5 other countries prohibit the addition of radioactive substances to these consumer products by their laws.

In Japan, radioactive materials may be added to some consumer products, but they are exempted by the criteria for quantity and concentration by the Law Concerning Prevention of Radiation Hazards based on the International Basic Safety Standards. Moreover, when the radionuclides are uranium-238 or thorium-232, the criteria for their quantity and concentration in the regulation are stipulated in the law of the Nuclear Reactor and Fuel Regulation Law. Furthermore, “The Guideline for Ensuring Safety of Raw Materials and Products Containing Uranium or Thorium” took effect in June, 2009 [6]. However, the addition of radioactive materials to products is not prohibited in Japan.

In this way, the regulation on RCP and naturally occurring radioactive materials are different in each country. In any case, because of the fact that the radiation exposure doses to the public from the naturally occurring radioactive materials have been raised in each country, the existence of the naturally occurring radioactive materials in consumer products is an international issue. Various research and published information about naturally occurring radioactive materials have been reported by domestic and international organizations including IAEA. However, there is little comprehensive information. It is reported in the Japanese “guideline” that monazite, bastnesite, zircon, uranium ores, thorium ores and titanium ores are given as the raw materials of naturally occurring radioactive materials of consumer products. However, the actual materials used by industry are not publicized. Also it is not clear how much radioactivity is added in each naturally occurring radioactive materials of consumer product. The radioactivity concentration and kinds of ores as a radiation source in many naturally occurring radioactive materials of consumer products should

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be investigated in order to make the industrial route open. For this purpose, the National Institute of Radiological Sciences (NIRS) has been recently investigating the current extent of the industrial use of naturally occurring radioactive materials by acquiring data from references in literature and some experimental results. With the provided information, they created a “NORM database” [7]. However, as the database is not completed, it can not be applied to all kinds of naturally occurring radioactive materials of general consumer products.

The object in this study is only consumer products in naturally occurring radioactive materials (NORM). Furuta *et al.* have analyzed NORMs (RCP) qualitatively and quantitatively for many years, and estimated the radiation exposure doses from many kinds of NORMs [2–4, 8–13]. We used the prompt gamma-ray activation analysis (PGAA) method to analyze hormesis cosmetics [3]. In all hormesis cosmetics, high concentration of samarium (Sm) and gadolinium (Gd) were detected in a short measurement time.

It is well known that Th and U coexist with rare earth elements in ores. The ratios of Gd/Sm in ores might be different depending on the kind of ore and geographic area. Furthermore, the concentration of radioactivity in general consumer products is totally different because of what kinds of ores used and what kinds of consumer products are produced. However, as the chemical characteristics of Sm and Gd are similar, a little difference of the ratio of Sm and Gd can be considered to be a new classification method of NORMs. The informations of Th/U ratios plus Sm/Gd ratios in NORMs can become a new identification method of the radiation sources of NORMs.

The aim of this study is to propose a new identification method of the radiation sources of naturally occurring radioactive materials in consumer products (NORMs) by paying attention to 4 kinds of elements; Sm, Gd, Th and U.

2. Experimental

2.1 Samples

Twenty-six kinds of NORMs were chosen as investigation samples with a random-selection in markets, which are commonly sold in Japan and whose activities can be measured by gamma-ray spectrometry by using a Ge detector. NORMs of very low activity were not investigated as samples.

Nine kinds of titanium (Ti) ores and 2 kinds of zirconium (Zr) ores, which were offered from NIRS (explained above), were used as samples of raw materials for industrial use (ind-NORMs). The total number of these ind-NORMs was 11 (samples of Code Nos. of 23–33 shown in Table 2).

For comparison, the producers of 4 ores of monazite were unknown. Others were a monazite ore (xenotime mixture) of the Iranian product which was sold under the name of “radium ore” (Code No. 50), an allanite ore (zircon mixture) of the Korean product which was sold under the name of “germanium ore” (Code No. 62) and a tourmaline powder which was a mixture of tourmaline produced in China, South Africa, and Brazil (Code No. 59). Four kinds of “monazite” were “Sand” samples gathered at the Indian shores (Code No. 51–54). Monazite, bastnasite, xenotime, uranium ores, thorium ores, titanium ores and other

9 kinds of minerals (Code Nos. of 60, 61, 63–69) were investigated as mineral raw materials for comparison with NORMs. The total number of these mineral samples was 28. The total number of these ores and mineral samples together was 50.

The NORM samples were crushed to small pieces with a crusher. Then they were sealed in plastic containers of the same size as the ones used for calibration of Ge detector and put directly on an end-cap of the Ge detector. The ore samples were crushed into pieces with a hammer to become as uniform as possible. The samples which looked non-uniform were made homogeneously smaller with a mortar. Samples were enclosed in FEP (fluoroethylene-propylene) film by measuring their weights. Standard samples for Sm and Gd were those commercially available for atomic absorption spectrometry (Wako Pure Chemical Industries) and two kinds of reference rock samples of JR-2 and JA-2(GSJ; Geological Survey of Japan) [14]. Triplicate samples and standards were prepared for each irradiation.

2.2 Measurement

The prompt gamma-ray activation analysis was carried out at JRR-3 of JAEA. The samples of 5×5 mm in dimension were continuously irradiated with a beam of thermal neutron flux of $1.6 \times 10^8 \text{ n cm}^{-2} \text{ s}^{-1}$ with the cross section of 20×20 mm. The neutron flux was found to be constant during the irradiation time. Prompt gamma-rays were observed for 300–50 000 s depending on the intensity of the gamma-rays with a Ge detector and the observed spectra were analyzed by using the spectra analysis software of Spectrum Navigator (SEIKO EG&G). The quantitative analysis of elements were carried out by the so called “comparison method”, *i.e.* comparison of photo peak intensities observed for the sample and those for a known weight of the Sm and Gd standard solution. The triplicate samples were measured and the counting statistic was used for evaluation of the error associated with each determination.

Radioactivities of Th and U series contained in the samples were determined by gamma-ray spectrometry using a high purity Ge detector (HPGe; CANBERRA, Genie2000) with the detection efficiency of 35% relative to NaI and the photo peak energy resolution of a FWHM = 1.85 keV at $E_\gamma = 1330$ keV. The gamma-ray spectra in the energy range 0–4 MeV were displayed in the 8192 ch MCA (CANBERRA). Each sample was measured for 1–96 h depending on their activities. The spectra from the Th and U series gamma-rays were corrected for those coming from the surroundings by the software of LabSOCS (CANBERRA) [15].

3. Results

The total radioactivities of the Th and U series in NORMs and ores are shown in Tables 1 and 2, respectively. The total radioactivities were estimated by assuming radio equilibrium among the nuclides in the decay series except for the Code No.16 of NORM. As ^{228}Ac activity was not detected in the Code No. 16 of NORM of the insole (foot deodorant) for shoes, its Th series activity shown in Table 1 is the total activities of a descendant nuclides below ^{224}Ra

Table 1. Concentrations of Th and U and their ratios in 22 kinds of radioactive consumer products (NORM).

Code	NORM Kinds: Name	Effect claimed by maker	Concentration(Bq/g)		Ratio of Th/U	Additives specified in the label (Specification)
1	Jell-1	Hormesis	4.1×10 ¹	1.5×10 ¹	2.7×10 ⁰	Zirconium Oxides
2	Liquid (red)		2.1×10 ¹	3.9×10 ⁰	5.4×10 ⁰	Tourmaline
3	Cream		2.1×10 ¹	7.1×10 ⁰	2.9×10 ⁰	Xenotime
4	Face powder		6.8×10 ¹	1.3×10 ¹	5.2×10 ⁰	Dolomite, mica, Si and so on
5	Jell-2		4.1×10 ¹	1.5×10 ¹	2.7×10 ⁰	(unknown)
6	Liquid	Minus-ion	3.0×10 ⁻²	1.7×10 ⁻¹	1.7×10 ⁻¹	Tourmaline
7	Toothpaste	Healthy	3.4×10 ⁻³	1.5×10 ⁻²	2.2×10 ⁻¹	Tourmaline
8	Bedrock-bath (red)	Relax and healthy	1.3×10 ³	1.9×10 ²	6.8×10 ⁰	(Hearsay information: Monazite from China)
9	Spring-bath		1.3×10 ³	2.1×10 ²	5.6×10 ⁰	
10	Bedrock-bath (white)		5.4×10 ³	8.6×10 ¹	6.3×10 ¹	(Rn spring)
11	Bakelite	Lightning	2.6×10 ⁻¹	2.0×10 ⁻¹	1.3×10 ⁰	(Ra powder)
12	Ra spring		1.7×10 ⁻²	4.2×10 ⁰	4.1×10 ¹	Th (Canada patent)
13	Gas lantern mantle		2.9×10 ⁻²	5.8×10 ⁰	5.0×10 ¹	Th (China patent)
14	Car goods	Mileage improvement	2.0×10 ⁰	4.6×10 ⁰	4.3×10 ⁻¹	Mineral (unknown) (China patent ; brown)
15	Deodorant	Deodorant	1.7×10 ⁻²	2.1×10 ⁻¹	8.0×10 ⁰	Monazite
16	Shoes-box		1.6×10 ⁻²	1.6×10 ⁰	9.9×10 ¹	Mineral (unknown)
17	Foot deodorant	Relax and healthy	1.2×10 ²	2.0×10 ¹	6.0×10 ⁰	Mineral (unknown)
18	Pillow		4.1×10 ⁰	9.9×10 ⁻¹	4.2×10 ⁰	Mineral (unknown)
19	Wall-paper	Minus-ion	8.1×10 ²	1.4×10 ²	5.8×10 ⁰	(The name is "monazu-bracelet".)
20	Ornament	Bracelet	1.1×10 ⁰	1.1×10 ⁻¹	1.0×10 ¹	Ge
21	Cloth	Under wear	1.0×10 ⁻²	1.5×10 ⁻²	7.2×10 ⁻¹	Mineral (unknown)
22	T-shirt	Minus-ion	1.5×10 ⁻²	9.5×10 ⁻³	1.5×10 ⁰	Tourmaline
	Wide stomach band					

in the decay series. In addition, three kinds of mileage improvement materials for the car and the cloisonné glaze are not shown in Table 1, as either Th or U series nuclides was detected although they were also investigated. Moreover, the concentration of mascara was not shown in Table 1 because the contents could not be transferred quantitatively to another container for the correct evaluation of the gamma-ray measurement efficiency. Furthermore, although in some NORMs, inclusion of ⁴⁰K was indicated in the gamma-ray spectra; however as their concentrations were of the order of natural range, they were not given in Table 1.

The radioactive concentrations of NORMs were from 0.01 to 5400 Bq Th g⁻¹ and from 0.01 to 210 Bq U g⁻¹ (see Table 1), and they differed by a factor of 20 000 to 500 000. The amounts of Th and U in the unit of weight; mg kg⁻¹ were from 0.1 to 320 000 and from 0.8 to 17 000, respec-

tively. It is surprising that, for example, in one bottle of the jell-cosmetics of Code No. 1, as large as 0.2 g of Th has been added.

All the ore and mineral samples, except Ti, Zr, thoriumite and uranite (samples of Code Nos. 28–30, 11 shown in Table 2), included rare earth ores. The radioactivity concentrations of Th and U were from 0.3 to 23 000 Bq g⁻¹ and from 0.4 to 86 000 Bq g⁻¹, respectively. The concentrations in ore and mineral samples were 10 times higher than those in NORMs.

The relationships among the concentrations of Th and U in NORMs and ore and mineral samples are shown in Fig. 1 in units of mg kg⁻¹. The figure shows that these investigated NORMs can be grouped into 3; 1. only U added, 2. only Th added, 3. the Th concentrations were higher than those of U, and the number of samples in the third group were larger than in the other groups. The Code Nos.

Fig. 1. Relationship between Th and U in NORM and ores. The error was based on counting statistics of radioactivity for each determination. The overall error of triplicate samples was calculated by the general method of error propagation. However, they were not shown by an error bar attached to each data points, as they were small enough to fall within the size of the symbol of the data point shown in the figure.

Table 2. Concentrations of Th and U and their ratios in 50 kinds of ores, sand and minerals.

Ore			Concentration (Bq.g ⁻¹)		Ratio	Ore			Concentration (Bq.g ⁻¹)		Ratio
Code	Kinds	Name	Th	U	Th/U	Code	Kinds	Name	Th	U	Th/U
23	Ore	Ti-ore* ¹	1.6×10 ²	5.4×10 ²	3.0×10 ⁻¹	48	Ore	Monazite	3.6×10 ⁻¹	4.4×10 ⁻¹	8.3×10 ⁻¹
24			2.8×10 ²	2.2×10 ²	1.3×10 ⁰	49		Monazite	2.8×10 ⁻¹	4.7×10 ⁻¹	5.9×10 ⁻¹
25			1.7×10 ²	7.4×10 ¹	2.2×10 ⁰	50* ²		Monazite(+Xenotime)	6.5×10 ⁻¹	5.0×10 ⁻¹	1.3×10 ⁰
26			2.3×10 ²	1.6×10 ²	1.5×10 ⁰	51	Sand (Beach)	Monazite	5.6×10 ¹	1.3×10 ¹	4.2×10 ⁰
27			2.3×10 ²	5.3×10 ¹	4.4×10 ⁰	52			2.9×10 ²	5.8×10 ¹	5.0×10 ⁰
28			9.1×10 ³	1.7×10 ³	5.5×10 ⁰	53			2.7×10 ²	5.6×10 ¹	4.9×10 ⁰
29	1.3×10 ³	5.4×10 ²	2.3×10 ⁰	54	3.6×10 ²	7.5×10 ¹			4.9×10 ⁰		
30	6.2×10 ⁴	7.5×10 ³	8.2×10 ⁰	55	Mineral	Xenotime	6.8×10 ¹	1.8×10 ¹	3.9×10 ⁰		
31	7.1×10 ⁴	6.4×10 ³	1.2×10 ¹	56			5.0×10 ¹	1.1×10 ¹	4.5×10 ⁰		
32	Sand	Zr-sand* ¹	5.9×10 ⁻¹	2.7×10 ⁰			2.2×10 ⁻¹	57	1.9×10 ¹	8.0×10 ¹	2.4×10 ⁻¹
33			5.5×10 ⁻¹	2.5×10 ⁰	2.2×10 ⁻¹	58	2.9×10 ⁴	3.2×10 ³	8.9×10 ⁰		
34	Mineral	Zircon	2.0×10 ⁰	7.2×10 ⁰	2.8×10 ⁻¹	59	Ore	Tourmaline (powder)	1.3×10 ⁻¹	2.0×10 ⁰	6.5×10 ⁻²
35			1.4×10 ¹	5.7×10 ¹	2.4×10 ⁻¹	60	Mineral	Aeschynite	1.6×10 ²	1.5×10 ⁴	1.1×10 ⁻²
36			1.7×10 ¹	7.1×10 ¹	2.4×10 ⁻¹	61		Allanite	7.4×10 ³	9.3×10 ¹	8.0×10 ⁰
37			3.3×10 ⁻¹	3.5×10 ⁰	9.4×10 ⁻²	62* ²	Ore	Allanite(+Zircon)	1.1×10 ⁻¹	8.6×10 ⁻²	1.3×10 ⁰
38			9.0×10 ⁰	5.6×10 ¹	1.6×10 ⁻¹	63		Betafite	1.1×10 ⁴	5.8×10 ⁻²	1.5×10 ⁵
39			8.0×10 ⁻¹	5.8×10 ⁰	1.4×10 ⁻¹	64		Davidite	2.2×10 ²	5.5×10 ²	4.0×10 ⁻¹
40	9.5×10 ⁰	-	-	65	Euxenite	8.0×10 ²		1.9×10 ⁴	4.3×10 ⁻²		
41	Mineral	Bastnesite	4.8×10 ¹	6.7×10 ⁻¹	7.2×10 ¹	66	Mineral	Euxenite	1.0×10 ³	2.2×10 ⁴	4.7×10 ⁻²
42			5.0×10 ¹	4.2×10 ⁻¹	1.2×10 ¹	67		Samarskite	1.4×10 ³	9.6×10 ³	1.5×10 ⁻¹
43	Mineral	Monazite	2.7×10 ³	2.9×10 ²	9.4×10 ⁰	68		Samarskite	1.2×10 ²	1.5×10 ³	8.3×10 ⁻²
44			2.5×10 ³	3.5×10 ²	7.2×10 ⁰	69		Stillwellite	1.8×10 ⁰	5.1×10 ¹	3.5×10 ⁻²
45			4.0×10 ²	4.2×10 ¹	9.5×10 ⁰	70	Thorianite	2.3×10 ⁴	8.6×10 ³	2.6×10 ⁰	
46	Ore	Monazite	6.1×10 ⁰	9.7×10 ⁻¹	6.3×10 ⁰	71	Uraninite	1.1×10 ²	8.6×10 ⁴	1.2×10 ⁻³	
47			2.9×10 ⁻¹	4.5×10 ⁻¹	6.5×10 ⁻¹	72	Uraninite	6.9×10 ²	6.7×10 ⁴	1.0×10 ⁻²	

*1; These ores and sand are used as industrial materials for paint, ink, plastic and so on. They are imported by some big companies and their radioactivities are controlled by the companies. They are called “industrial NORM”.

*2; The two ores of Code Nos. 50 and 62 were sold at a normal handcraft shop and the name of each ore shown in the Name column are analyzed by Dr. Miyawaki.

2, 6, 7, 22 of NORM that claimed “tourmaline” as raw materials showed relatively low concentration of both Th and U.

There is the tendency that the ore and mineral samples having the same names exist close to each other as indicated by marks in Fig. 1 under the name of Ti, Zr, xenotime and monazite ores (rocks, sands, minerals) and show a roughly the same ratio of Th vs. U. However, it is found not possible to identify the radiation sources of NORMs only from the relations of Th and U in ore and mineral samples as shown in Fig. 1.

The relationships among the concentrations of Gd and Sm in all samples analyzed by PGAA are shown in Fig. 2. The concentrations of Sm and Gd in many NORMs are found to follow approximately the relation shown by power function such as given below:

$$y = 0.86x^{0.959}$$

where x is the concentration of Sm (mg kg⁻¹) and y is the concentration of Gd (mg kg⁻¹). The Code Nos. 6, 7, 22 of NORMs which were claimed tourmaline as raw materials showed very low Gd and Sm concentrations, and their data

points are not shown in Fig. 2 as no meaningful counts were accumulated even with the measuring time of 50 000 s. On the other hand, although the Code No. 2 of NORM which was claimed to contain tourmaline as raw materials was found to include Gd and Sm, their concentrations were 1–3 times lower than those of other NORMs. Furthermore, relatively very low concentrations of Gd and Sm were found in the “mascara” (NORM).

Among the 11 kinds of industrial raw materials, Gd and/or Sm were detected in 7 kinds of samples of Ti ores (Code Nos. 23–27, 31) and Zr sand (Code No. 32). There is a tendency for the ore and mineral under the same name to have much similar concentrations of Sm and Gd as shown in Fig. 2 when compared to the case of the Th/U ratios in Fig. 1. However, from Fig. 2 it was also not possible to find easily a correlation between ore and mineral samples and NORMs.

Fig. 3 shows the correlation between the Th/U ratio (vertical axis) and the Gd/Sm ratio (horizontal axis). The ratio of some of zirconium mineral and bastnesite mineral are very large and they scaled out the limit of horizontal and vertical axis of the figure by 2 orders of magnitude, respectively. In most of NORMs, the Th/U ratios were around 10 and

Fig. 2. Relationship between Sm and Gd in NORM and ores. For errors associated each data points, please refer to the caption of Fig. 1. The solid line in the figure represents the correlation between Sm and Gd contents; power function $y = ax^b$ to log-log for the data points of open circle of NORMs.

Fig. 3. Correlation between Gd/Sm and Th/U. The data of monazite, 2 allanite minerals, titanium ores and all the NORMs except Code No. 16 (Deodorant) fall within an ellipse shown by solid curve. A; Gd/Sm ratio for zirconium mineral exceed the horizontal scale. B; Th/U ratio for bastnesite mineral exceed the vertical scale. They are indicated by arrows.

the Gd/Sm ratios were around 1. From Fig. 3, the raw materials for the radiation sources of NORMs studied in this work were deduced to be as monazite group, Allanite mineral and/or Ti ores. However, Ti ores could be excluded as discussed below.

As examples of prompt gamma-ray spectra in between 65–1565 ch (46.3–1078 keV) observed by PGAA are shown in Fig. 4. The gamma-ray spectrum of the Ti ore of Code No.26 showed the main large 2 titanium peaks (Ti-48; 341.7, 1381.7 keV). However, the gamma-ray spectrum by the NORM of Code No.15, a deodorant for shoes-box, did not show the titanium peaks. As the gamma-ray spectra of other NORMs and Ti ores were similar to the respective

Fig. 4. PGAA gamma-ray spectra of the NORM of shoes-box deodorant (A) and a Ti ore (B). The spectrum of the NORM was shifted to the upper side by 200 counts to help the readers to see the gamma ray peaks clearly. A; the upper side spectrum is the NORM; Code No. 15. B; the lower side spectrum is the Ti ore; Code No.26. There are large gamma-ray peaks of Gd (Gd-157; 181.93 keV) and Sm (Sm-149; 333.97 keV) and no gamma-ray peaks of Ti (Ti-48; 341.7 keV) in the NORM.

spectra of Fig. 4, it is clear that the raw materials which became the radiation source of NORMs were not Ti ores.

4. Discussion

For the aim of identifying the raw radioactive substances added to consumer products (NORMs), the classification of the NORMs and ore was attempted by seeking correlation among 4 elements of Th, U, Sm and Gd. The absolute radioactive concentrations of Th and U cannot be used for this purpose because the concentrations of Th and U included in each NORM would be different depending on the material of NORM. On the other hand, by paying attention to the ratio of Th/U and Gd/Sm, which are not generally included in general consumer products, it was considered that these ratios would show values peculiar to the ores added. The relationships between the radiation sources of NORMs and ores could not be revealed clearly only by Th/U ratio or the Gd/Sm ratio. On the other hand, it was possible to iden-

tify that the radiation sources of NORMs should be monazite by analyzing both ratios of Th/U and Gd/Sm together. We conclude that:

- Among the monazite ores including minerals and sands, the correlation between the ratios of Th/U and Gd/Sm was somewhat different from those of other ores.
- Because minerals including rare earth elements are rather expensive and not easy to get in a large quantity, for instance allanite, these minerals may not be used as the radiation sources of a large amounts of NORMs.
- The NORMs that tourmaline was mentioned as raw materials had low concentration of these four elements in comparison with other NORMs. Though the concentration was low, the detected Th/U ratio and Gd/Sm ratio were approximately the same both in the ore and in NORMs, and it may be easy to understand that the radiation sources for these NORMs (Code Nos. 2, 6, 7, 22) were tourmaline ores.
- In the Ti ores, which are used for many industrial products and called *ind-NORMs* (Code Nos. 23–31), the correlation among the concentration ratios of Th/U and Gd/Sm were similar to those of NORMs other than Code Nos. 2, 6, 7, 22. However, the PGAA gamma-ray spectra of those NORMs did not necessarily show the Ti peaks. Furthermore, in the PGAA gamma-ray spectra, the peak heights of ^{149}Sm (333.97 keV) and ^{157}Gd (181.93 keV) in Ti ores were always lower by a factor of 10 or more in comparison with that of ^{48}Ti (341.7 keV). However, the sensitivities of Gd and Sm in comparison with those of Ti in PGAA are about 1,000 and 450 times higher, respectively [16]. Thus, Ti ores of *ind-NORMs* were found not to be the radiation sources of NORMs.
- There are 4 kinds of “monazite” in the geology. In addition, there might be various kinds of minerals coexisting in an ore. Actually it was confirmed by electron probe micro analysis (EPMA) that the coexistence of minerals in the ore samples of Code Nos. 50 and 62 in Table 2 was xenotime in the monazite ore and zircon in the allanite ore, respectively. The ores are named from the major mineral ingredients. However, the Gd/Sm ratio did not change greatly if it was “monazite”. Although a mineral of zircon, xenotime, and allanite may be included in a monazite ore, the ore of radiation source for NORMs is regarded as monazite. In addition, it was considered that the spread of the Gd/Sm ratios *vs.* Th/U ratios of the monazite ores in Fig. 3 was caused by the inclusion of other minerals.

In 26 samples of NORM, 13 samples strongly reacted to a radiation survey meter compared with other samples. The existence of NORM is becoming the global issue. For *ind-NORMs*, only a major company can control the handling of the radiation source for naturally occurring radioactive materials in most cases. On the other hand, a small company which makes NORMs (consumer products) might not have a control over the radiation sources as well. So, workers of a factory and its neighboring inhabitants and users of NORMs are affected by the gamma-ray radiations of NORMs, the fact of which can be a big health issue. This study clarified that the radiation sources of NORMs were “monazite”. In 13 NORM

samples which strongly reacted to a radiation survey meter, there were cosmetics and personal ornaments to which addition of radioactive materials is prohibited in EU. The investigation of the handling of the imported monazite is very important for protection of the workers, its neighboring inhabitants and users of NORMs from the radiation exposure.

As a conclusion, the classification method proposed in this study is useful for an investigation of the source of NORMs which is general consumer products spreading in many countries all over the world.

5. Conclusions

Gadolinium (Gd) and samarium (Sm) were analyzed by PGAA for the studying the radiation sources of NORMs (consumer products) including thorium and uranium. The existence of NORMs has become a world-wide issue. It was not possible to find the relationship between NORMs and the ores from only the concentration of Gd and Sm because Sm *vs.* Gd showed constant relations in many things include NORMs. By combining the PGAA with the gamma-ray spectra measurement by HPGe, it was found that a group of “monazite” was almost all the radiation sources added in NORMs (consumer products). Also the radiation sources of some NORMs of low radioactivity were considered “tourmaline”. By investigating and clarifying into the route of the import of monazite, we believe that health damage for workers, its neighboring inhabitants and users of NORMs can be decreased. The proposed classification method of analyzing the correlation between the Th/U and Gd/Sm ratio was found to be a useful method for identifying the radiation source of NORMs.

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