

Necessity of World Wide Regulation for Radioactive Consumer Products in Current Markets

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Abstract Many radioactive consumer products (RCP) are sold in the current market of Japan and some part of Asia; for example cosmetics and personal jewelry. The activities and concentrations of these RCP are under the regulation limit of each country, so purchase, use, transport and disposal of these RCP are perfectly freedom. However, some global regulations prohibit addition of radioactive materials to six items deliberately; food stuff, beverage, feed, toys, cosmetics and personal jewelry. Because there is little justification to add radioactive materials to consumer products, it is considered in the global regulations that these six items should be forbidden without taking a radiation exposure dose of radioactivity of RCP into consideration individually. The concentration of radioactivity of 28 kinds of RCP sold in Japan and Korea was measured and the highest concentration among samples was 1,300 Bq g⁻¹ of thorium and 200 Bq g⁻¹ of uranium for the powder of bedrock-bath. Also the radiation exposure doses from samples were estimated. The external exposures by using each RCP studied were less than 1 mSv per year with normal use. However, each user might use these RCP with too long time or more than one consumer product. Also, the internal exposures with radon or radioactive materials might be happen in some cases by using these RCP. The reality of the RCP in Japan and Korea which goes against such global regulations was reported in this study. It is considered that a regulation unified at world level is necessary for commercially-available RCP all over the world.

Key word; Radioactive consumer products, NORM, Radioactive concentration,
Unjustified exposure, Regulation

Introduction

Radioactive consumer products (RCP) are defined by OECD/NEA as “a manufactured item or product in which radionuclides are deliberately incorporated, generally for their radioactive properties and which can be supplied to members of the public without special surveillance or regulatory control after sale”.

The outline of the history of RCP and the regulation to them are as follows; the use of radioactive materials for consumer products had been tried right after the discovery of radiation, and many kinds of RCP were produced at that era. However, the health damage of the luminous

paint factory workers occurred and it was a very serious problem. Thereafter, the Radiation Protection 68 published the report entitled “Study on consumer products containing radioactive substances in the EU Member States”[1], the Documents of the NRPB published the report entitled “Board statement on approval of consumer goods containing radioactive substances” [2], the IAEA Safety Standards Series published the report entitled “Application of the concepts of exclusion, exemption and clearance” [3] and the Radiation Protection 146 published the report entitled “A review of consumer products containing radioactive substances in the EU”[4]. In Japan, the fact-finding circulation of consumer goods had been performed every ten years from 1978.

The International X-ray and Radium Protection Committee; IXRCP (Now; ICRP) were established in 1928. Until now, the regulation authorities of each country and/or international organizations gave some indicators for not to give health damage to public. For examples; The National Radiological Protection Board of UK (NRPB; now Health Protection Agency) reported that “To ensure that the annual doses to such individuals from all exempt practices are not more than a few tens of μSv , the annual effective dose to a member of a critical group should be less than 10 μSv for each product.”[5]. On the other hand, IAEA concluded that for the purpose of exemption, a level of an individual dose of some tens of μSv in a year could reasonably be regarded as trivial[6]. During these times, many studies had been done by many researchers and some international commissions.

Recently, the existence of naturally occurring radioactive materials (NORM) became a global social issue. It is considered that the issue of radiation exposure of the miner and public is mainly derived from industrial use of mineral. However, the existence of RCP in the Asian area [7] cannot be ignored in the case of intentionally addition of NORM for consumer products. Now, how to regulate NORM is examined in the international organizations such as ICRP, IAEA, WHO and the OECD/NEA. Especially, in the ICRP 2007 Recommendation (ICRP-103) [8], unjustified exposure is defined that “certain exposure which is increasing by deliberate addition of radioactive substances or by radiation, the activity of products such as food, beverage, cosmetics, toys, and personal jewelry or adornments, should be deemed to be unjustified without further analysis.” These 5 items are same as the addition prohibition items with IAEA [9] and EURATOM [10]. In the EURATOM case, there is one more item; feed. Though there is such a background, many kinds of RCP which include some of the six items mentioned above are sold in a current market of Asian area, particularly in Japan. Everyone can buy RCP from any place and area of every country through the Internet.

The purpose of this study is to analyze radioactive concentration in RCP and to estimate exposure doses from the normal use of them. Furthermore, a discussion on the justification of the existence of RCP was aimed for in this study.

Experimental

Samples of RCP

The kinds of RCP sold in current markets were sorted by two categories; 1. the RCP which mainly causes the external radiation exposure, 2. the RCP which mainly causes the internal radiation exposure by inhalation of Rn and/or the particles of radioactive materials. The number of samples was 28 in the category 1 and 10 in the category 2 with some overlaps. Table 1 shows a list of samples; the name for sale, the effects claimed by manufacturers, and their additives specified in the labels. The samples of Nos. 26 and 28 have been sold in Korea, and others have been sold in Japan.

Measurement

All the samples were put in plastic containers (U8; 65 mm in high \times 50 mm diameter) for measurement. The gamma-ray spectrometry of the samples was performed by using a HPGe coaxial detector with an efficiency of 30% relative to NaI, and with a FWHM=1.85 keV at gamma-line 1330 keV and a multi-channel analyzer (CANBERRA). The efficiency was calculated by using analysis software of LabSOCS [11]. Each sample was measured for 1-96 hours depending on their activities.

Also major elements in RCP were analyzed by instrumental neutron activation. Each sample of about 150-300 mg was enclosed in a high-purity polyethylene bag and irradiated in JRR-3 reactor (JAEA) for 10 min at a neutron flux of $5.2 \times 10^{13} \text{ n} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$. Two days after, they were analyzed with their gamma-ray spectrometry.

Radon (Rn) concentrations emitted from samples in category 2 were measured by using a scintillation Lucas cell (PMT-TEL, Pylon) with a flow meter, a radiation monitor (Model AB-5) and a printer (Model PPT-1), or a pico rad (PICO-RADTM; ACCUSTAR LABS Company). For a pico rad measurement, the sample was enclosed for 10 days in a sealed container of 50 L, and then a pico rad was put in it for 48 h. After 48 h, the pico rad was recovered, left it for 24h and then poured 10 mL of Insta-fluor (PerkinElmer). After 4 h, the Rn absorbed by the pico rad was measured for 60 min by liquid scintillation counter (Tri-Carb 3170TR/SL; PerkinElmer).

Estimation of radiation exposure by the RCP

External exposure: The samples in category 1 were estimated their external radiation exposures as follows, which was mentioned in “The Guideline for Ensuring Safety of Raw Materials and Products Containing Uranium and Thorium” (NORM Guideline) [12] by Japanese government;

$$Dose\ 1(mSv\ y^{-1}) = DEX [mSv/h/(Bq\ m^{-2})] \times C(Bq\ g^{-1}) \times M(g) \times T(h\ y^{-1}) \div [D(m)]^2,$$

$$Dose\ 2(mSv\ y^{-1}) = DSKIN [mSv/h/(Bq)] \times C(Bq\ g^{-1}) \times M(g) \times T(h\ y^{-1}).$$

where *Dose 1* means a radiation exposure dose of RCP which uses without adhering to skin (an

effective dose) and *Dose 2 means* a radiation exposure dose of RCP which uses within 1 meter from skin (an effective dose).

*DEX**: A conversion factor to effective dose per 1Bq at 1 m distance [mSv /h/(Bq/m²)]

For Th=1.8×10⁻¹⁰ and for U=2.7×10⁻¹⁰

*DSKIN**: A conversion factor to effective dose per 1Bq (mSv /h/Bq)

For Th=9.6×10⁻⁹ and for U=1.3×10⁻⁸

*; Conversion factors calculated from European Commission; *Radiation Protection 65* (1993)

C: Radioactivity concentration of RCP (Bq g⁻¹)

M: Mass of RCP (g)

T: Exposure time (h y⁻¹)

D: distance between a RCP and its user (m)

Each radiation exposure dose from the radioactivity of RCP was evaluated with the normal use scenario which estimated the use form of each RCP. (The details of scenarios are mentioned in each reference.)

Internal exposure: It was supposed that a woman used each cosmetic (Nos.1-5) for ten years continuously. Internal residual quantity (Bq) of a body according to the radioactivity in each sample was calculated using the residual coefficient and excretion rate function [13] for each nuclide. The conversion from the residual quantity by each radionuclide to radiation exposure doses (Sv) is used the Internal Dose Easy Calculation Code: IDEC ver.1.0 [14]. In the case of a trans-dermal intake, radioactivity infusion to blood was assumed 1% of consumption. In the case of an inhalational intake, an internal exposure dose of radioactivity was calculated by using 0.1% consumption of intake for the total lung. At the calculation, the particle size as the aerosol assumed it lognormality distribution of 2.5µm.

The internal exposures (*D_{in}*, µSv h⁻¹) by inhalation of Rn were calculated as follows;

$$D_{in}=k \times e \times MA$$

where *k* means an effective dose conversion factor for the radon equilibrium equivalent concentration, *e* means indoor equilibrium factor and *MA* (Bq m⁻³) means the Rn concentration measured. In this study, $k=9 \times 10^{-3} [\mu\text{Sv} (\text{Bq h m}^{-3})^{-1}]$ and $e=0.4$ were used [15].

Results

The total concentrations of ²³²Th and ²³⁸U in all RCP are shown in Table 2. Also main other elements included in each RCP are shown in Table 2, too. The concentrations of ²³²Th or ²³⁸U were estimated by assuming radioactive equilibrium among the nuclides in their decay series because the main progeny of decay series were detected in all samples of Table 1. The concentrations of ²³²Th and ²³⁸U added in RCP were extremely different among products; from

0.1 ppm to 32% of ^{232}Th and from 0.8 ppm to 1.7% of ^{238}U . Both of the concentration and the amount of radioactive materials were less than Japanese regulation limit values, and all of them were defined as “RCP”. Also main other elements showed that these RCP were added some kinds of minerals because some of elements do not exist in normal consumer products.

The external and internal radiation exposure evaluation values are shown in Table 3. The exposure doses of sample Nos.8-10 were not estimated because their concentrations were illegal and not sold in any current markets. Also the exposure doses of code Nos. 6-7, 11 and 22-24 were not estimated because it was considered they were too low to estimate them by their activity concentrations. All the external exposures were low enough to be not worried about health risks from the RCP because their exposure doses were under 1 mSv y^{-1} at their normal use. However, the concentrations of Rn emitted from the RCP (Nos.12, 19-20) were not low. When a user of No.19 used the pillow every day in a narrow Japanese room (25 m^3), the internal exposure was estimated almost 6 mSv per year. Furthermore, it is considered that there are risks of internal exposure from radioactive materials when use the toiletries and cosmetics of RCP (Nos.1-5), especially No.4.

Discussion

All external exposure doses by normal use of each RCP, which are sold in the current market of Japan and Korea, were estimated lower than the limits of law; 1 mSv per year. In Japan, there is NORM Guideline, which took effect in June, 2009 [12]. The samples were not defined as NORM consumer products by the NORM Guideline, and they did not need to take any regulations after sale, so all these samples matched a definition of RCP. However, the commission of ICRP, IAEA and EU consider that certain exposure by some RCP should be deemed to be unjustified without further analysis.

It is considered that there are mainly 4 issues in RCP sold in current markets, which were added mainly monazite [16].

1. Generally, all consumer products including RCP are required to prove the effects by manufactures themselves when they are sold claiming some effects. Almost all the RCP claim some effects which are extremely scientific catchphrases. However, it is not considered that the claimed effects by these RCP have been demonstrated by themselves, because their home pages (HP) of the Internet have connected with other HP of the claimed effects and there are no other evidence. Their effects claimed may have no evidence on their products. Perhaps the explanation of their effects may have borrowed different explanations for other cases.
2. In some cases, the RCP are used in abnormal ways; for examples, use too much quantity, put them too close by users' body, and use longer time than recommended usage time. The “NORM guideline” made with the premise that there is no misuse is imperfect.

3. When RCP was put within our living environment, the possibility of inhalation of gaseous Rn increases because Rn is the descendant nuclide of Th and U series which are added in the RCP. According to WHO, radon is one of the main causative agent of lung cancer. It is considered that there is no necessity to put these RCP within our living environment. It is doubtful that user of RCP can get some benefits which are higher than the risk of incidence of lung cancer.
4. Because some RCP as samples Nos. 6-8 were made in very high concentrations' sand, the RCP were stopped selling in a market by Japanese regulation authorities. However, there was a possibility they would be sold if no one noticed their high concentrations. This means that very high concentration's RCP might happen to be bought, used and put in our living environment.

Though there are the problems mentioned above, we can purchase many RCP through the Internet from all over the world. When the radioactivities of RCP are less than the regulation of law and the exposure from them is less than 1 mSv per year, the mainstream of tones of Japanese researchers is that the RCP has no problem and it is a narrow-minded thought to reject the RCP. Many Japanese researchers including Japanese government think that almost all the RCP are utilization of radioactive ores. However, it is considered that regulation of the addition of the radioactive materials in the consumer products is necessary, in particular for cosmetics, because they are used on the skin for a long time and there is the possibility of the surplus use and internal radiation exposure by them. We should notice that regulation unified at world level is necessary for RCP.

Conclusion

All the investigated objects in this study were radioactive consumer products (RCP) that the radioactive materials were added the quantities and amounts of less than the regulation limits. However, the EURATOM, IAEA and ICRP-103 adopt the way of thinking of the radiation exposure for RCP as the "Unjustified exposure" which have no justification. In other words, a way of thinking whether or not there is justification is important for a use of radioactive materials. It is considered that the regulation for radioactive materials having been in the raw materials of industrial products should differ from the regulation of RCP which intentionally added radioactive materials claiming some doubtful effects. It is considered that a regulation unified at world level is necessary for commercially-available RCP from all over the world.

References

- [1] Schnitt-Hannig A., Drenkard S. and Wheatly J., 1995, Radiation Protection 68, European Commission.
- [2] Paynter, R.A. , 1992, NRPB 3(2), UK.
- [3] IAEA, 2004, IAEA NoRS-G-1.7, Austria.

- [4] Shaw J., Dunderdale J. and Paynter R. A., 2007, Radiation Protection 146, European Commission, 2007.
- [5] Napier I.D. and Paynter R.A.,1997, NRPB-M836, p2, UK.
- [6] IAEA,1988, IAEA Safety Series No 89, Vienna.
- [7] Chang B-U, 2010, AOCR-3, proceeding, 25 May, Japan.
- [8] ICRP Publ.103, ICRP *Annals of the ICRP* 37, 2007.
- [9] IAEA Safety Guide, 2/10/2006, IAEA DS401.
- [10] EURATOM. Council Directive 96/29,1996, OJ L159, p7.
- [11] Bronson F.L.,2003, *J. Radioanal. Nucl. Chem.*, 255, p137-141.
- [12] MEXT of Japan, June,2009, The Ministry of Education, Culture, Sports, Science & Technology, Japan.
- [13] Akaishi J. et al., 1988, Atomic energy security technology center, Tokyo, Japan.
- [14] JAEA, 2000, IDEC Ver.1.0, JAEA, Japan.
- [15] UNSCEAR 2000 Report of the General Assembly, with scientific annexes Vol.1, 2000.
- [16] E.Furuta et.al., 2011, *Radiochimica Acta*, 1, 219-225.
- [17] E.Furuta et.al.,2007, *Japn. Health Phys.* 42, 341-348.
- [18] E.Furuta et.al.,2000, *J. Radioi.Prot.* 20, 423-431.
- [19] E.Furuta,2007, *Japn. Radiat. Saf. Manag.* 6, 31-36.
- [20] E.Furuta et.al., 2006, *Radioisotopes* 56, 443-453.
- [21] E.Furuta et.al., 1999, *Radioisotopes* 48, 107-112.
- [22] E.Furuta et.al.,2007, *Japn. Health Phys.* 42, 341-348.
- [23] E.Furuta et.al.,1998, *Radioisotopes* 47, 920-925.
- [24] E.Furuta et.al., 2010, *Jpn. Health Phys.* 45, 253-261.

Table 1 The list of Radioactive Consumer Products studied in this work.

No	Category*	RCP		Clamed effect by manufacture	Additives specified in the label (Specification)
		Kinds:	Name		
1	1,(2)		Jell-1		Zirconium oxides
2	1,(2)		Liquid (red)		Tourmaline
3	1,(2)	Toiletry	Cream	Hormesis	Xenotime
4	1, 2		Face powder		Dolomite, mica, Si and so on
5	1,(2)		Jell-2		(unknown)
6	1,(2)		Body lotion		Thorium oxides, Xenotime
7	1,(2)	Cosmetics	Mascara	Minus-ion	Mineral(unknown)
8	1, 2	Spring bath	Bedrock-bath; red	Relax and Healthy	Hearsay information: Monazite from China
9	1, 2		Bedrock-bath;white		(Rn spring)
10	1, 2		Bakelite		(Ra powder)
11	1, 2		Ra powder		Uranium residual substance
12	1, 2		Ra brick	Healthy	
13	1, 2	Gas	lantern mantle	Lightning	(Th ; Canada patent)
14	1, 2				(Th ; China patent)
15	1	Car goods	Engine sheet	Mileage improvement	Mineral (unknown) (China patent ; brown)
16	1		Ceramics		Mineral (unknown)
17	1	Deodorant	Shoes-box	Deodorant	Monazite
18	1		Foot (shoes)		Mineral (unknown)
19	1, 2	Pillow		Relax and healthy	Mineral (unknown)
20	1, 2	Wall-paper		Minus-ion	Mineral (unknown)
21	1	Glass	Optical microscope	High reflection	(unknown)
22	1	Cloth	Under wear	Detoxification	Germanium
23	1		T-shirt	Minus-ion	Mineral (unknown)
24	1		Stomach band	Minus-ion	Tourmaline
25	1		Accessory	Monaz-bracelet	Hormesis
26	1		Sports bracelet	Bio-radiation	(unknown;Korea)
27	1	Eye-mask	Ceramics	Minus-ion and	(unknown;Japan)
28	1		Rubber		Hormesis

*; The category 1 means it causes mainly external exposure.

The category 2 means it causes mainly internal exposure.

Table 2 Concentration of radioactivities and main other elements in the radioactive consumer products.

No	RCP Name	Concentration (Bq g ⁻¹)* ¹		Main other elements* ²
		²³² Th	²³⁸ U	
1	Jell-1	(4.1±0.3)×10 ¹	(1.5±0.01)×10 ¹	Ce, Zr, Yb, Eu, Hf
2	Liquid (red)	(2.1±0.01)×10 ¹	(3.9±0.3)×10 ⁰	-
3	Cream	(2.0±0.07)×10 ¹	(7.1±0.2)×10 ⁰	Zr, Ce, Hf, Yb
4	Face powder	(6.8±0.01)×10 ¹	(1.3±0.01)×10 ¹	Ce,
5	Jell-2	(4.1±0.3)×10 ¹	(1.5±0.01)×10 ¹	Ce, Zr, Yb, Eu, Hf
6	Body lotion	(1.8±0.2)×10 ⁻³	-	Ce, Hf
7	Mascara	(1.1±0.1)×10 ⁻¹	(5.6±1.2)×10 ⁻¹	Fe, Zn
8	Bedrock-bath; red	(1.3±0.07)×10 ³	(1.9±0.04)×10 ²	Fe, Ce, Mn, Zn
9	Bedrock-bath; white	(1.3±0.02)×10 ³	(2.1±0.1)×10 ²	Ce, Zn, Zr, Yb, Hf
10	Bakelite	(5.4±0.04)×10 ³	(8.6±0.3)×10 ¹	-
11	Ra powder	(2.6±0.1)×10 ⁻¹	(2.1±0.2)×10 ⁻¹	Ca, Mn, Ce, Hf
12	Ra brick	ND	(6.3±0.09)×10 ¹	-
13	Gas lantern mantle	(1.7±0.1)×10 ⁻²	(4.2±0.1)×10 ⁰	Mg, Pb, K, Al, Y
14		(2.9±0.1)×10 ⁻²	(5.8±0.1)×10 ⁰	Mg, Al, Na, Si, Ce
15	Mileage	(1.6±0.1)×10 ⁰	(3.7±0.5)×10 ⁰	-
16	improvement	(4.1±0.04)×10 ²	(7.3±0.2)×10 ¹	-
17	Deodorant	(1.6±0.1)×10 ³	(3.0±0.05)×10 ²	Ce, Zr, Zn, Cr, Tb
18		(1.6±0.07)×10 ²	(2.3±0.07)×10 ²	-
19	Pillow	(1.2±0.05)×10 ²	(1.8±0.4)×10 ¹	Ce, Zn, Zr, Cr
20	Wall paper	(4.4±0.01)×10 ⁰	(9.9±1.2)×10 ⁻¹	Fe, Zn, Zr, Hf
21	Optical microscope	(2.7±0.1)×10 ⁻²	ND	-
22	Under wear	(1.1±0.01)×10 ⁰	(1.3±0.01)×10 ⁰	Zr, Ce, Hf
23	T-shirt	(1.4±0.1)×10 ⁻²	(1.6±0.01)×10 ⁻²	-
24	Wide stomach band	(2.1±0.1)×10 ⁻²	(9.5±0.01)×10 ⁻³	-
25	Monaz-bracelet	(8.2±0.1)×10 ⁻²	(1.3±0.2)×10 ²	Ce, Zr, Zn, Tb
26	Sports bracelet	(5.0±0.6)×10 ⁻²	(1.0±0.02)×10 ²	-
27	Eye-mask (Japan)	(2.3±0.2)×10 ¹	(9.8±0.3)×10 ¹	-
28	Eye-mask (Korea)	(4.2±0.3)×10 ¹	(6.8±0.9)×10 ⁰	-

*1; The concentrations of ²³²Th or ²³⁸U were estimated by assuming radioactive equilibrium among the nuclides in their decay series because the main progeny of decay series were detected in all samples of Table 1.

*2; They were analyzed by Instrumental Neutron Activation Analysis or Inductively Coupled Plasma- Mass Spectrometry.

ND; Not detected

-; Not analyzed

Table 3 Effective dose from radioactive consumer product.

RCP No	External exposure ($\mu\text{Sv h}^{-1}$)	Annual dose (mSv per 2000h)	Internal exposure & Rn activity	Reference number
1	3×10^1	Skin ^{*1} ; 2	Born; $6 \times 10^1 \mu\text{Sv y}^{-1}$	[17]
2	3×10^{-1}	Skin ^{*1} ; 2×10^{-2}	Born; $2 \times 10^1 \mu\text{Sv y}^{-1}$	
3	9	Skin ^{*1} ; 3	Liver; $3 \times 10^1 \mu\text{Sv y}^{-1}$	
4	2×10^1	Skin ^{*1} ; 5×10^{-1}	Lung; 6 mSv y^{-1}	
5	3×10^1	Skin ^{*1} ; 2	Liver; $2 \mu\text{Sv y}^{-1}$	
12	2×10^{-1}	4×10^{-1}	$7 \times 10^2 \mu\text{Sv y}^{-1}$	* ³
13	8	2×10^1	* ²	[18]
14	1	2		
15	2×10^{-3}	5×10^{-3}	N.P.	[19]
16	3×10^{-3}	6×10^{-3}		
17	6×10^{-3}	1×10^{-2}	N.P.	[20]
18	8×10^{-6}	Skin ^{*1} ; 4		
19	3×10^{-1}	5×10^{-1}	6 mSv y^{-1} at 25 m^3	[21]
20	1×10^{-2}	2×10^{-2}	$2 \times 10^1 \mu\text{Sv y}^{-1}$	[22]
21	9	2×10^1	N.P.	[23]
25 ^{*4}	1×10^2	2×10^2	N.P.	[24]
26 ^{*4}	2×10^2	3×10^2	N.P.	-
27 ^{*4}	2×10^2	4×10^2	N.P.	-
28 ^{*4}	5×10^1	1×10^1	N.P.	-

N.P.; No Possibility at normal use.

Skin^{*1}: Annual skin equivalent dose of toiletries estimated for a face of 300 cm^2 at $70 \mu\text{m}$ depth.

*2; The progeny of Th series is released to the atmosphere by combustion. For example, ^{212}Pb is $8 \times 10^{-15} \text{ g}$ (410 Bq) per combustion.

*3; <http://www.dollstone.co.jp/> (Reference at 2012.02.18). The radon inhalation was estimated by the scenario that the user took a bath 1 h every day and the bath room was made from the 50 stones.

*4; The calculation doses were overestimated, because the D used in the Dose 1 was $7.0 \mu\text{m}$ and the radioactive materials added in the consumer products do not exist in one spot.