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ENHANCED RADIOACTIVE CONTENT OF 'BALANCE' BRACELETS

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During a routine whole body counting measurement of a worker at the Nuclear Research Center Negev, abnormal activities of ^{232}Th and ^{238}U were measured. After a thorough investigation, it was found that the radioactivity was due to a rubber bracelet ('balance bracelet') worn by the worker during the measurement. The bracelet was counted directly by an high pure germanium gamma spectrometry system, and the specific activities determined were 10.80 $\pm$ 1.37 Bq g^{-1} for ^{232}Th and 5.68 $\pm$ 0.88 Bq g^{-1} for natural uranium. These values are obviously high compared with normally occurring radioactive material (NORM) average values. The dose rate to the wrist surface was estimated to be $\sim$3.9 μGy h^{-1} and $\sim$34 mGy for a whole year. The dose rate at the centre of the wrist was estimated to be $\sim$2.4 μGy h^{-1} and $\sim$21 mGy for a whole year. The present findings stresses a more general issue, as synthetic rubber and silicone products are common and widely used, but their radioactivity content is mostly uncontrolled, thus causing unjustified exposure due to enhanced NORM radioactivity levels.

INTRODUCTION

Individual monitoring of internal dose at the Nuclear Research Center Negev (NRCN) includes routine whole body counting (WBC) check. In one of these checks, an abnormal high activity of ^{232}Th and natural uranium was found. An investigation was set up, and it was found out that the worker did not take off his rubber bracelet before entering the WBC chamber (although it was required according to the WBC procedure), claiming that it was 'made of rubber and had no chance of containing any radioactivity'. According to the bracelet producers, it was used as a 'holographic and negative ion technologies' to 'improve body balances'.

The bracelet was counted directly by an high pure germanium (HPGe) gamma spectrometry system, supporting beyond any doubt the fact that it was the source of the detected enhanced ^{232}Th and uranium activities. This was supported by an additional WBC counting of the worker, without the bracelet, which gave results under the detection levels for thorium and natural uranium. The present work summarises the results of the bracelet radioactivity analysis.

RESULTS

^{232}Th is a natural radioisotope present in rocks, soils and water. It decays by alpha emission with a very long half-life (1.4×10^{10} y) and is the head of a chain on its name containing daughters, which emit also gamma radiation. In nature, ^{232}Th is in secular equilibrium with its daughters, and its activity can be determined by the progeny activity. Additionally, ^{238}U is the head of a chain on its name and is in secular equilibrium with its daughters. The prominent gamma energy lines and the corresponding yields of progeny nuclides of ^{232}Th and ^{238}U are given in Table 1.

The whole body counter of NRCN consists of four Ge detectors mounted in an old iron-shielded chamber[1]. The system is a lung detector and is calibrated using a realistic phantom. The calibration assumes that the detected contamination is located in the lungs, following inhalation.

In order to ensure that there is no external contamination, the worker was instructed to take off his watch, rings etc. to have a shower and to wear clean clothes supplied by the laboratory, before the WBC measurement. The worker followed the instructions but decided not to remove his bracelet. During the WBC measurement of the worker, abnormal activities of ^{232}Th and ^{238}U were measured. When the bracelet was suspected as the source of radiation, a direct gamma spectrometry was performed by placing it on the face of a high-resolution HPGe detector. The resulting pulse-height spectrum is presented in Figure 1, on which the characteristic gamma lines of thorium and uranium series can also be seen. Other bracelets from the same manufacturer were counted and indicated a significant content of thorium and natural uranium. Bracelets of other manufacturers, which were randomly checked, indicated no enhanced radioactive content.

The gamma spectrometry system is not calibrated for the bracelet geometry; therefore, one of the bracelets (Bracelet 1) was cut into 1 cm pieces weighting 0.8 g each, and the pieces were placed on the detector for spectrometry measurements. A point source geometry was assumed for these samples. The specific ^{232}Th and uranium activities of the various daughters, calculated according to their corresponding gamma emissions, are presented in Table 2. Within the given

uncertainties, a secular equilibrium can be observed. Weighted averages of the activities of the decay products were used to estimate the activity concentrations of ^{232}Th and natural uranium. The specific activities obtained were 10.80 $\pm$ 1.37 Bq g^{-1} for ^{232}Th and 5.68 $\pm$ 0.88 Bq g^{-1} for natural uranium. By using the correction factor obtained from the counting of the whole Bracelet 1 and the 1 cm pieces, a quantitative evaluation for the other bracelets could be done. The results are given in Table 3. The ^{232}Th-specific activity of the different bracelets varies by a factor of $\sim$3 between 4.71 and 13.5 Bq g^{-1}, while for natural uranium, the specific activities are in the range from 3.23 to 6.67 Bq g^{-1}. These values are unusually high compared with normally occurring radioactive material (NORM) average values.

Table 1. Main gamma emissions of the ^{232}Th and ^{238}U radioactive series.

Parent nuclide	Daughter nuclide	E_γ [keV]	P_γ [%]
^{232}Th	^{228}Ac	209.39	4.18
	^{212}Pb	238.63	43.61
	^{228}Ac	338.42	11.26
	^{228}Ac	463.10	4.63
	^{208}Tl	583.02	30.98
	^{212}Bi	727.33	6.64
	^{208}Tl	860.30	4.32
	^{228}Ac	911.16	29.30
	^{228}Ac	964.64	5.85
	^{228}Ac	968.97	17.41
	^{208}Tl	2614.35	35.82
^{238}U	^{234}Th	92.60	5.41
	^{234m}Pa	1001.0	0.65
	^{226}Ra	186.11	3.28
	^{214}Pb	241.92	7.44
	^{214}Pb	295.09	19.20
	^{214}Pb	351.87	37.09
	^{214}Bi	609.31	46.10
	^{214}Bi	768.35	4.88
	^{210}Pb	46.52	4.05

Table 2. Activity determination of Bracelet 1 by different gamma energies.

Series	Isotope	Energy [keV]	P_γ [%]	Activity [Bq g^{-1}]
^{232}Th	^{212}Pb	238.6	43.50	12.45 $\pm$ 1.69
	^{224}Ra	241.0	3.97	13.63 $\pm$ 3.03
	^{228}Ac	338.3	11.26	10.89 $\pm$ 1.63
	^{208}Tl	583.2	30.57	10.10 $\pm$ 1.55
	^{212}Bi	727.3	6.65	10.21 $\pm$ 2.69
	^{228}Ac	911.2	26.6	11.92 $\pm$ 2.02
	^{228}Ac	969.0	16.23	13.65 $\pm$ 3.77
^{238}U	^{234}Th	63.3	4.49	2.03 $\pm$ 0.37
	^{214}Pb	351.9	37.1	2.56 $\pm$ 0.50
	^{214}Bi	609.3	44.81	1.79 $\pm$ 0.45
	^{234m}Pa	1001.0	0.59	2.79 $\pm$ 0.56
	^{214}Bi	1764.6	15.90	2.30 $\pm$ 1.52
^{235}U	^{235}U	143.8	10.96	0.145 $\pm$ 0.032

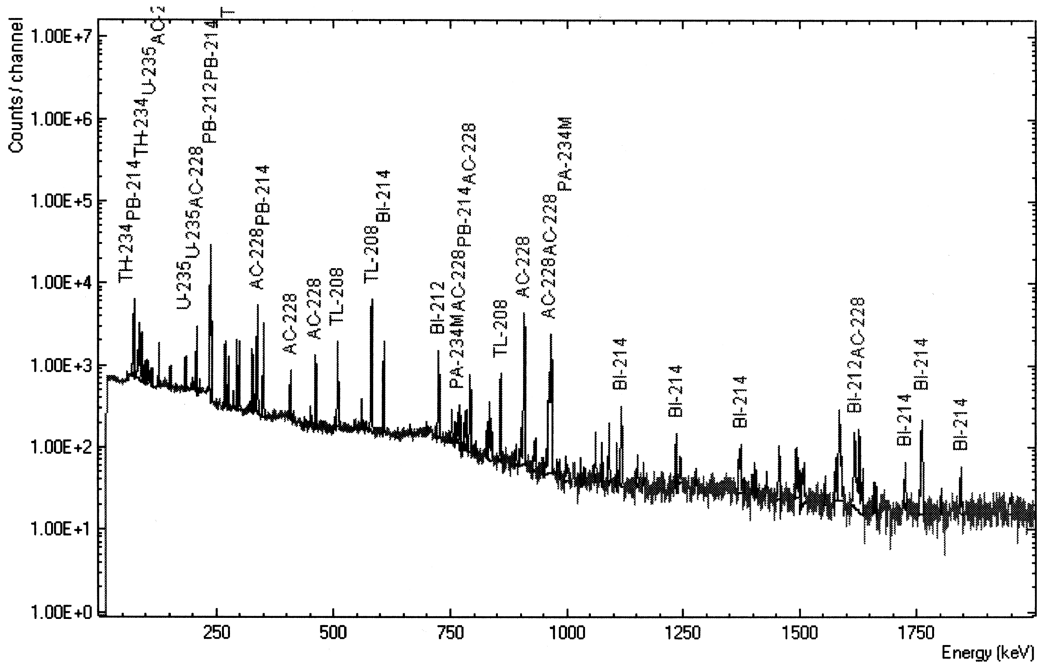

Figure 1. The pulse-height spectrum due to a bracelet placed on the HPGe detector.

In order to estimate the dose to the wrist when wearing the bracelet, a wrist phantom filled with water was set up and 3 × 3 × 0.38 mm TLD100 chips (LiF:Mg,Ti) were used, as seen in Figure 2. Six chips were placed on the inner surface of the wristband, and one chip was placed in the middle of the phantom, as seen in the figure.

A summary of the results is presented in Table 4. The dose rate to the wrist skin is estimated to be ~3.9 μGy h^{-1}, and the total local dose when wearing the bracelet for a whole year is estimated as ~34 mGy. The yearly dose at the centre of the wrist is estimated to be ~21 mGy.

DISCUSSION AND SUMMARY

The source of the abnormally high radioactive content in the bracelets is not clear. As the contamination consists of naturally occurring ^{232}Th and uranium, it seems to originate from a high-activity NORM source. Materials of natural origin, such as ores and minerals, often contain high levels of radioactivity caused by radionuclides of the uranium and thorium series.

The bracelets are made of rubber silicone, which may contain natural components. Natural materials as sands enriched in ^{232}Th and uranium were found in several locations[2]. This finding is mainly explained by variations in heavy minerals concentration (especially zircon) and the grain size effect, the finest fraction being enriched by about three orders of magnitude[3]. In another paper, activity concentrations of ^{232}Th ranged to ~10 kBq kg^{-1} were reported at Orissa beach sands in south-east India[4]. They were explained by the presence of significant amounts of monazite and zircon in the sand samples. The Monazite sands samples contained ~300 kBq kg^{-1} of ^{232}Th and ~22 kBq kg^{-1} of ^{238}U, while the zircon sands contained several kilobecquerels per kilogram of both radioisotopes.

Another source of enhanced natural radioactivity could be fly ash, used as filler in the production process of synthetic rubber[5, 6]. The average specific activity in coal is generally around 20 Bq kg^{-1} for both ^{238}U and ^{232}Th, but there are significant variations at different geographical locations. In general, the radionuclide enhancement factor for uranium and thorium in the ash is ~10, and the ^{238}U and ^{232}Th concentrations reported in the literature (<1 kBq kg^{-1}) are much lower than the concentrations found in the bracelet rubber.

As the production process of the bracelets and the origin of the raw materials are not known, no source can be identified for their enhanced radioactivity. Whatever the source is, according to the international standards[7], human radiation exposure must be justified. It is not the scope of the present work to discuss the potential effects or benefits of the aforementioned bracelets, but it seems hardly plausible that the radiation exposure involved can be justified in this case.

The present findings pose a more general issue. Synthetic rubber and silicone products are common and widely used, but their radioactivity content is mostly uncontrolled. Other products containing enhanced radioactivity may be in the market causing unjustified exposure due to enhanced radioactivity levels. Better control procedures are needed to avoid this source of radiation exposure of the public.

Table 3. Evaluation of ^{232}Th- and uranium-specific activities of several bracelets.

Bracelet serial number	Weight [g]	^{232}Th-specific activity [Bq g^{-1}]	Natural uranium-specific activity [Bq g^{-1}]
1	11.6	10.80 ± 1.37	5.68 ± 0.88
2	13.7	9.80 ± 1.22	4.77 ± 0.74
3	14.9	4.71 ± 0.59	3.23 ± 0.53
4	12.4	13.48 ± 1.39	6.67 ± 0.87
5	13.1	10.24 ± 1.27	4.34 ± 0.69

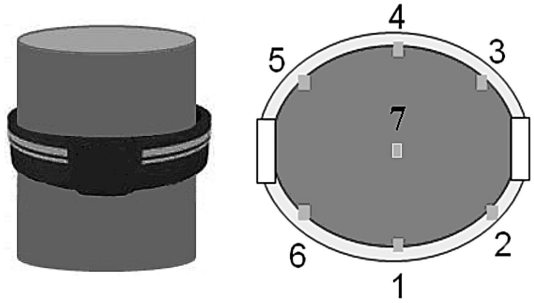

Figure 2. The setup for dose measurements from the bracelets.

Table 4. Radiation dose measured by TLD100 crystals during 91.6 h in contact with a bracelet and during 167.8 h in the centre of the phantom, when the bracelet was mounted on a cylindrical water phantom.

	Crystal position	Measured dose [mGy]	Dose rate [μGy h^{-1}]	Yearly dose [mGy y^{-1}]
On the bracelet	1	0.319	3.48	30.53
	2	0.344	3.76	32.94
	3	0.396	4.33	37.89
	4	0.376	4.11	36.00
	5	0.423	4.62	40.47
	6	0.287	3.14	27.50
	Average	0.36 ± 0.05	3.91 ± 0.55	34.22 ± 4.82
At phantom centre	7	0.402	2.40 ± 0.24	21.02

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