Exposure and radiation protection for working areas with enhanced natural radioactivity

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Abstract. Work activities with materials containing enhanced content of natural radioactivity may lead to increased exposures of workers or members of the public. Thus they are subject to radiation protection regulations if reference levels are exceeded as described in the German Radiation Protection Ordinance which is in force since July 2001. To optimize monitoring and/or possible radiation protection measures, reliable methods for measurements and exposure assessments have to be developed taking into account material matrices, radionuclides involved, workplace conditions and exposure pathways. In this context, case studies have been carried out for both radiation exposure of workers at several work places and exposure of members of the public due to the disposal of industrial residues. In the paper, practical results and conclusions are summarized for two cases: (i) exposure of workers during handling and/or milling of zircon sand, and (ii) exposure of members of the public who live close to a red mud disposal site of an aluminium oxide production plant.

1. Introduction

In the German Radiation Protection Ordinance (RPO) [1], Part 3, work activities are described as operations involving the (unintended) presence or increase of natural radiation sources leading to a significant radiation exposure of workers or members of the public and/or to radioactive contamination. Work activities which may lead to elevated exposures of workers are given in Appendix XI of the RPO. Elevated exposures of members of the public may result from the use or disposal of residues from industrial processes with enhanced natural radioactivity (ENORM) as listed in Appendix XII. Both work activities and the disposal of the listed residues are subject to radiation protection regulations if reference levels are exceeded. These levels are dose limits that apply to workers involved in work activities. Control of residues is required if the processing or disposal of these residues could result in the reference effective dose (Richtwert) of 1 mSv in a calendar year being exceeded. Based on this reference effective dose a list of residues to be considered is given in Appendix XII, and appropriate action levels in terms of specific activities of natural radionuclides in the residues have been deduced. Furthermore, the authorities can prescribe protective measures for unlisted materials or residues in cases of elevated exposures to workers or members of the public that cannot be disregarded from the viewpoint of radiation protection.

Practically, protective measures should be implemented only after a careful assessment of the radiological situation. Specific measurements have to be carried out to assess whether the reference levels of the RPO are exceeded. Contrary to the monitoring of workers in the field of practices, generally applicable methods for measurements and assessments are not yet available for work activities and industrial residues containing ENORM. Therefore, a Coordinating Office has been established at the Federal Office for Radiation Protection to investigate typical work activities and industrial residues with the objective of developing optimized methods for measurements and exposure assessments, and to provide guidance for practical solutions [2]. In this context, a study of the exposure paths of workers and members of the public during the disposal of residues (sludges) from pig iron production was carried out previously [3]. In the present paper, the possible exposure of members of the public from red mud of an aluminium oxide production plant which is explicitly listed in Appendix XII of the RPO is discussed. Furthermore, results of our zircon sand study are presented. This material is known to have enhanced natural radioactivity [4–6], but is not listed in the RPO since a previous estimate using a generic model [7] did not indicate a significant exposure to workers. To verify this exclusion, practical measurements have been carried in cooperation with a zircon milling company in Germany.

2. Exposure of workers during handling and/or milling of zircon sand

For different kinds of industrial applications, for example for the production of zircon-based opacifiers used in the ceramic industry (glazes of tiles and sanitary ware), fine and dry flour of iron-free zircon sand is needed as an ingredient. Zircon sand is used also with increasing extent for the production of iron-free grinding balls, cylinders, bricks for linings of mills, funnels and special parts exposed to abrasion. It is well known that zircon sand has an approximate chemical composition of ZrO_2 (66%), SiO_2 (33%), Al_2O_3 (≤0.2%) and TiO_2 (≤0.2%), and contains small amounts of enhanced natural radioactivity, i.e. the radionuclides of the ^{238}U and ^{232}Th decay chains [4–6]. Work activities related to zircon sand handling or milling, as well as the raw zircon sand, the products or components containing such sand and the production residues, are not included in the German RPO, Appendices XI and XII. The omission of zircon sand residues is due to an earlier estimate based on a generic model [7] that the airborne zircon dust in a foundry may lead to an internal exposure of workers of up to 0.5 mSv/a only. To verify this estimate, a case study has been initiated together with a zircon sand milling company in Germany. Thus an assessment of the possible exposure paths (external exposure, finger exposure and internal exposure due to inhalation of dust) of workers handling zircon sand was carried out to evaluate the radiological situation. The measurements are specific to the working areas considered.

Different kinds of sample materials were analysed in the laboratory using gamma-ray spectrometry. The samples originated from different places around the world and reflect the most commonly used raw materials in Germany. About 500 g of sample material was filled in gas-tight containers to keep the radon and its decay products inside the sample container. After waiting for almost three weeks to reach the equilibrium between them, the emitted gamma radiation was measured with an n-type high-purity Ge detector (GMX-Detector, 40% relative efficiency) placed inside lead shielding. The GMX-detector provides high detection efficiency also at low gamma-ray energies such as the 46.54 keV gamma ray originating from ^{210}Pb decay and the 63.28 keV gamma ray from ^{234}Th decay which have, in addition, low emission probabilities of 4.25% and 4.1%, respectively. A systematic study was performed to deduce correction factors for the self-absorption of these gamma-rays in the zircon sand samples. Selected results are listed in Table 1. The highest ^{238}U activity concentration of about 3700 Bq/kg was found in a sample from South Africa.

TABLE 1. ACTIVITY AND MASS CONCENTRATIONS IN ZIRCON SAND
(2-sigma uncertainties are displayed in parentheses)

Origin	Density (g/cm^3)	Activity concentration (Bq/kg)			Mass concentration (μg/g)	
		^{238}U	^{226}Ra	^{232}Th	U_{nat}	Th_{nat}
South Africa	2.7	3700 (700)	3500 (700)	645 (130)	300 (6)	160 (30)
USA	2.7	1320 (260)	1500 (300)	190 (40)	107 (21)	47 (9)
Australia	2.85	2300 (450)	2300 (450)	530 (110)	185 (40)	130 (25)

Ambient dose rate measurements were performed at several working places in the milling plant, where the mobile dose rate meters were placed on a tripod about 1 m above ground. Enhanced values were measured, for example, in the storage and milling rooms. A pile of several thousand tons of zircon sand generated an ambient dose rate of about $\dot{H}*(10) = 0.9$ μSv/h, measured in front of the pile. The highest values measured were about 1.5 μSv/h, in fair agreement with published values of 1.7 μSv/h [4]. Assuming 2000 hours of working time per year in the storage or milling rooms, which is a conservative assumption, an effective dose of about 3 mSv would be delivered to the workers. This estimate is based on a conversion factor of 1 between the measured ambient dose and the effective dose according to the recommendations given in Appendix D of the Guidelines [2]. It should be noted that for workers (>17 years of age) in opencast mining areas, a conversion factor of 0.6 is recommended [8] for the estimate of the effective dose which would result in an effective dose of 1.8 mSv/a.

Usually, the zircon sand is milled according to the specifications of the user, and a grain size down to 1 μm is achievable. In the milling room, airborne dust is unavoidable which may be inhaled by the workers. Thus, the size distribution of the dust grains in the breathing air was measured using a Berner

Impactor placed on a tripod. The maximum of the distribution was found for a grain size between 3 and 6 μm, and for the range from 0.3 to 3 μm the dust contained the highest alpha activity. In particular the small grain size fraction of the dust may be deposited in the lung causing an internal exposure. Therefore, the exposure path due to the inhalation of dust was investigated in more detail.

The dust measurements were carried out during the milling of a large amount of zircon sand using two standard air-sampler systems (GSA-GSP). The two dust samplers were placed in the milling room with the air sucking heads positioned about 1 m above ground. The amount of dust deposited on a filter placed into the air stream was measured over a period of about 8 h. An average dust concentration of about 3.2 mg/m^3 air was estimated. The alpha activity of the dust collected by the filter was measured using a commercially available and calibrated gross alpha counter. As a result, an alpha activity of 29 mBq/m^3 air was determined. Using the conservative assumption of 2000 working hours per year at this place, a standard breathing rate of 1.2 m^3/h and an effective dose coefficient of 8.0 μSv/Bq, a lung dose of about 0.65 mSv per year can be calculated due to this alpha activity. The effective dose coefficient has been deduced from a weighted average (according to the amounts of U_{nat} and Th_{nat}, see Table 1) of the dose coefficients given for inhalation of uranium and thorium and their decay products in Appendix C of Ref. [2].

The milling process and the partitioning of the product are only partly remotely controlled. Thus, there are situations where the workers have to perform some tasks by hand and have contact with zircon sand, zircon flour, or zircon dust layers, i.e. the hands and fingers may get contaminated. Therefore, measurements of the exposure of the fingers of workers handling the zircon sand and/or operating the milling machines were carried out using authorized fingering dose meters containing thin thermo-luminescent detectors on the basis of ^{7}Li:Mg,Cu,P (TLD-700). These detectors are sensitive to beta and gamma radiation. Six detectors were used for a relatively short period, about ten working days. On these days, standard working tasks were performed by the workers. Using the assumption of 2000 h of working time (which is a quite conservative assumption), $H_P(0.07)$ dose values between 0.3 and 9 mSv have been estimated for the fingers. These values need to be verified via additional measurements, i.e. over a longer period of time to improve the statistical significance of the results; however, they are far below the organ dose limit (500 mSv for fingers) specified in the RPO.

In conclusion, possible exposure paths were identified and studied during the handling and/or milling of zircon sand. The estimated effective doses do not exceed the reference level of the RPO regulation (6 mSv/a for workers). Thus, the omission of these work activities from Appendix XI of the RPO is justified and authorities do not need to intervene. Dedicated measures to improve the radiation protection of workers are not necessary. The implementation of working conditions and general safety standards according to the guidelines of the professional organizations and associations of the mineral industry contributes in this particular case also to the compliance with the radiation protection regulations.

3. Exposure of workers and/or members of the public from deposited red mud

Bauxite is the raw material for the production of aluminium oxide. Nowadays, the raw material is imported in large amounts into Germany. The material contains, besides aluminium, many compounds of medium and heavy elements as well as natural radionuclides in low concentrations. In the primary production process, the aluminium is extracted as aluminium hydroxide via a chemical method. The bauxite is washed, ground and dissolved in sodium hydroxide at high pressure and temperature (Bayer Process, 1887). The resulting liquor contains, besides aluminium compounds, undissolved bauxite residues containing Fe_2O_3 (30–60%), SiO_2 (3–50%), Na_2O (2–10%) and TiO_2 (Trace–10%) [9, 10] — the so-called red mud. About 900 000 t of red mud are produced every year in Germany [11]. The colour reflects the high amount of iron hydroxide in the red mud. It has been subjected to sodium hydroxide treatment, i.e. red mud is highly caustic with pH values in excess of 13, but chemically stable and non-toxic. Despite many efforts, red mud is not used for any important industrial processes, and thus is disposed of in special areas according to the German disposal regulations. The areas are usually fenced and closed to the public. Red mud is explicitly listed in the RPO, Appendix XII, due to the occurrence of enhanced natural radioactivity. Thus, a possible exposure of members of the public has to be assessed.

At the beginning of the study of a large disposal site in Germany, the possible exposure of workers through external gamma-radiation was estimated. The dose rate ($\dot{H}_x$) was measured at several places on the fenced disposal area with a mobile dose rate meter positioned about 1 m above the ground. Values between 180 and 275 nSv/h were obtained. The contribution from the background, measured outside the area, was about 50 nSv/h. Since the annual amount of working hours directly on the deposit is low, of the order of 100 h, the exposure of workers is not significant and can be neglected.

The activity concentrations of natural radionuclides in bauxite and red mud samples were determined in the laboratory using gamma-ray spectrometry. The bauxite samples were milled to a grain size of <2mm using a laboratory mill to obtain homogeneous samples. Subsequently, the sample material was filled into gas-tight containers. After a waiting period of about three weeks, as was done for the zircon sand samples, the gamma rays were measured with the GMX-detector setup. Typical results are given in Table 2.

The activity concentrations in the bauxite samples are relatively low, and the radionuclides ^{238}U and ^{226}Ra are found to be in radioactive equilibrium (within uncertainties). For the red mud, the measured activity concentrations are generally higher compared with the values for the bauxite samples. Furthermore, the activity concentrations of ^{228}Ra and ^{228}Th (radionuclides in the ^{232}Th decay chain) are about twice as high as the values for ^{238}U and ^{226}Ra of the ^{238}U decay chain.

TABLE 2. ACTIVITY CONCENTRATIONS IN BAUXITE AND RED MUD
(2-sigma uncertainties are given in parentheses)

	Activity concentration in dry mass (Bq/kg)					
	^{40}K	^{228}Ra	^{228}Th	$^{22}6Ra$	^{238}U	^{210}Pb
Gove-bauxite	<20	140 (20)	140 (40)	95 (15)	93 (18)	80 (15)
Boke-bauxite	<20	135 (20)	135 (40)	65 (10)	65 (13)	62 (12)
Red mud	<30	370 (50)	380 (50)	190 (30)	190 (40)	150 (30)

The enhanced activity concentrations in bauxite and red mud samples found in the investigation are in agreement with previous work [9]. Since red mud is listed in Appendix XII of the RPO, it has to be assessed and handled according to the regulations. The total activity concentration in the red mud sample was deduced to be 0.19 Bq/g (^{238}U decay chain) + 0.38 Bq/g (^{232}Th decay chain) = 0.57 Bq/g according to the sum rule given in Appendix XII where the representatively determined highest activity concentrations of any member of the decay chains of ^{238}U and ^{232}Th are added. Subsequently, this value is compared with a control limit which depends on the groundwater conditions and on the amount of residues to be deposited. In this particular case, the deduced value of 0.57 Bq/g is lower than the control limit of 1.0 Bq/g as given in the regulations for a disposal site outside the catchment area of an exploitable ground water reservoir. Thus, the disposal of red mud on this site is not subject to regulations. However, a reduced control limit of 0.5 Bq/g has to be applied if more than 5000 t of residues are deposited in an area of exploitable groundwater. In any case, a detailed analysis of the groundwater conditions is required, which is, however, outside the scope of the present study.

The red mud disposal site investigated in this case had been established many years ago, at an area with a thick clay (waterlogged soils) basement and sidewalls which provide a natural shield against water flow due to their low permeability. Surface water and groundwater samples were taken at several control points and analysed. For this purpose, the water samples were treated in the radiochemical laboratory according to specific procedures [12], and subsequently, the alpha and beta activities were measured. Some typical results are given in Table 3. The measured activity concentrations are very low and close to the upper end of the natural background concentrations measured for undisturbed groundwater in Germany.

TABLE 3. ACTIVITY CONCENTRATIONS IN GROUNDWATER AND SURFACE WATER (2-sigma uncertainties are given in parentheses)

	Activity concentration (mBq/L)				
	^{210}Pb	^{210}Po	^{226}Ra	^{228}Ra	^{238}U
Groundwater 4	2.5 (1.1)	2.8 (0.6)	59 (7)	14.0 (3.8)	<0.7
Groundwater 6	3.4 (1.2)	2.9 (0.6)	11.0 (1.9)	12.0 (3.5)	<0.7
Well A (surface)	4.5 (1.2)	4.6 (0.8)	5.3 (1.8)	15 (4)	2.8 (0.8)

Another possible scenario would be an internal exposure of members of the public due to inhalation of airborne dust generated from the surface of the red mud deposit. During the production of aluminium hydroxide, the continuously generated red mud is suspended in process water and pumped through long pipes to the disposal site. Subsequently, it precipitates and part of the water is recycled. On sunny days, parts of the surface are not covered by water so that airborne dust could be produced and transported to the population living in the neighbourhood. To estimate a possible internal exposure of members of the public due to inhalation, the model developed for uranium mining sites has been applied [8]. Based on the specific activities given for the red mud in Table 2, radioactive equilibrium in each decay chain, an assumed dust concentration of 50 $\mu g/m^3$ and time of residence outdoors 2000 h and indoors 7000 h, an effective dose of about 10 µSv/a has been estimated for a person of age >17 years. Similar values have been obtained for children. Hence, this exposure path is not significant and can be neglected.

In summary, based on the measurements and exposure assessments, the red mud disposal site is in compliance with the German regulations as outlined in the RPO. No additional radiation protection measures have to be implemented.

Since July 2001, Germany has had in place, for the first time, dedicated regulations to enforce radiation protection measures for workers and members of the public to limit exposure to natural radiation sources. The regulations are based on generic model considerations and partly on measurements and encompass specified work activities as well as a detailed list of industrial residues. The results of the present study confirm the validity and omission of these regulations for the bauxite residue and zircon sand milling, respectively.

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