Practical advice on the evaluation and control of radiation exposure of workers in NORM industries – leaflets

Preface

Radiation exposures of workers or members of the public can arise when materials containing naturally occurring radioactive materials are processed without using their radioactive, fissile or fertile properties. For such materials, the term NORM (Naturally Occurring Radioactive Materials) is commonly accepted\(^1\). In some cases, radiation exposures can be sufficient to require radiation protection controls. For such processes involving NORM or other natural radiation sources in the European Basic Safety Standards (Directive 96/29 of the Council of the European Union) [1] the term ‘work activities’ is applied.

The platform for Europe-wide discussions of radiation protection issues in NORM industries, and for promoting good radiation protection practice, is the European ALARA Network for Naturally Occurring Radioactive Material (EAN\textsubscript{NORM}). In the Network, the exchange of experience derived from radiation protection practice in workplaces is of particular importance. More information is available at http://www.ean-norm.net or http://www.ean-norm.eu. Outputs from the Network are summarized in leaflets for individual NORM industries by the organizers of EAN\textsubscript{NORM}. These include information on legal aspects, measurement methods, estimation and evaluation of radiation exposures, and the integration of radiation protection measures into the workplace health and safety system. The leaflets are designed to support the harmonization of operational radiation protection in NORM industries in the Member States of the European Community. They are aimed primarily at persons who are responsible for health, safety and radiation protection in the relevant industries.

A) Practical information about the procedure for the evaluation and control of radiation exposure of workers processing zirconium minerals - Leaflet for the zircon industry

1 Zirconium minerals - naturally occurring radioactive materials

Economically important zirconium minerals are zircon (zirconium silicate), and zirconia (zirconium oxide), which occurs in nature as the mineral baddeleyite. Zirconium silicate can be found as an accompanying mineral in igneous rocks, or in secondary deposits of zircon sand in rivers and beaches, which have been formed by natural processes of separation due to the high density of zirconium silicate. Baddeleyite is found only in a few deposits of carbonate minerals.

The grain sizes of zircon are in many cases too large and therefore, the raw material is milled (micronized). The resulting intermediates (micronized zirconium silicate - micronized zircon, zircon - zircon flour) will be mainly used in the ceramic industry, in foundry casting (mould construction) and the refractory industry. In recent years, the production of cubic zirconia has gained importance.

\(^1\) IAEA Safety Glossary Terminology Used in Nuclear Safety and Radiation Protection, 2007 Edition
Zircon and other naturally occurring zirconium minerals contain naturally occurring radionuclides, particularly the radionuclides of the uranium (U-238) and the thorium (Th-232) decay series. U-238 and U-235 are present in zirconium minerals in natural isotope ratio (99.3% to 0.7%). From a radiation protection perspective, the radionuclides of the U-235 decay series are of minor importance.

The radionuclides are strongly bound within crystal structure of zirconium minerals. This structure is extremely stable and, if the minerals are milled the radionuclides retain in the crystal lattice. Even if the minerals are heated (e.g. in the production of glazed ceramic tiles and sanitary ware) the crystal lattice remains stable and the volatilisation of radionuclides is unlikely to occur. Only in a few cases (e.g. manufacturing zirconia by fusion of zirconium minerals, chemical dissolution of zirconium minerals) is the crystal lattice destroyed and the radionuclides released into the process media and end up in the products (e.g. U-238 and Th-232 in the zirconia product) or in the residues (e.g. Ra-226 in silica or in the scales of pipes and fluid handling systems, Pb-210 and Po-210 in dust in off-gas collection systems).

The activity concentrations of U-238 in commercially available zirconium minerals are typically from 1 Bq/g to 4 Bq/g. In some mineral deposits significantly higher values have been found (e.g. in China 15 Bq/g, in Malaysia up to 60 Bq/g). The activity concentrations of Th-232 are lower and range from 0.5 Bq/g to 1 Bq/g. There are however, considerably higher values (e.g. in China 8 Bq/g, in Malaysia 40 Bq/g).

In baddeleyite, the activity concentrations of U-238 and Th-232 are slightly higher and typically range from 3 Bq/g to 10 Bq/g for U-238. Extreme values (e.g. as in some zircon) have not been found in baddeleyite. For Th-232, activity concentrations are lower and typical values range from 0.1 Bq/g to 2 Bq/g.

In zirconium minerals, the radionuclides of the decay chains are approximately in radioactive equilibrium (secular equilibrium). For experimental studies and for estimates of radiation exposures this is of importance.

Because of the elevated content of natural radionuclides in zirconium minerals, the radiation exposure of workers during production and processing should be considered. Radiation protection problems can occur in industries in which zirconium minerals, zirconium compounds or intermediates are processed. There may be a need for special radiological attention especially in situations where large amounts of material are used (e.g. milling of zirconium minerals).

2 European Basis Safety Standards for the radiation protection of workers – application to the processing of zirconium minerals

2.1 Council Directive 96/29 EURATOM

Council Directive 96/29 EURATOM [1] specifies the basic principles of radiation protection for workers and the public. These principles should also be applied to work activities which involve the presence of natural radiation sources and lead to significant increase in the exposure of workers or members of the public which

2 The state of a radioactive decay chain where the activity of each radionuclide is the same. The state is achieved when the parent radionuclide (here U-238 or Th-232) has a much longer half-life than any of the progeny
cannot be disregarded from the radiological point of view (Article 2, paragraph 2 of [1]). Member states are obliged to identify such work activities (Article 40, paragraph 2 of [1]) and if necessary, to initiate measures in order to reduce radiation exposures and to control unavoidable exposures (Article 41 of [1]). In [2] the production and processing of zirconium minerals are explicitly mentioned as an industry that can give elevated radiation exposures to workers (i.e. annual effective doses greater than 1 mSv).

Exposures above 1 mSv per year are potentially possible from processing zirconium minerals with activity concentrations greater than 1 Bq/g U-238 or Th-232 (each in secular equilibrium) – as indicated above, this includes zirconium minerals. The annual radiation exposure of employees from work activities involving such minerals should be estimated and, if above 1 mSv regulatory controls should be applied in a graded manner. There is, however, much discretion on the part of national authorities, as to how the standards are applied (Article 41 of [1]). In practice, this has resulted in different decisions in Member States and some (still) do not regulate the processing of zirconium minerals to control radiation exposures.

2.2 Draft of the revised European Basic Safety Standards
The European Basic Safety Standards [1] are being revised and the draft BSS [3] include more specific requirements for NORM industries. In Annex V of the draft the relevant industries are specifically listed, and these include the processing of zirconium minerals (zircon and zirconium industry). The revised standards also include activity concentration exemption levels (e.g. 1 Bq/g U-238 or 1 Bq/g Th-232, each in secular equilibrium), and also require that graded regulatory controls (for radiation protection purposes) are applied where doses to workers exceed 1 mSv per year.

3 Radiation protection during transport of zirconium minerals
The regulations for transport of radioactive materials [4] apply only if the sum of the U-238 and Th-232 activity concentrations exceed 10 Bq/g, in which case the requirements for transporting Low Specific Activity (LSA) material have to be applied. Typical commercially available zirconium minerals with activity concentrations of 1 Bq/g to 4 Bq/g for U-238 and 0.5 Bq/g to 1 Bq/g for Th-232 are not, therefore, within the scope of these regulations.

Systematic studies have been conducted to determine the radiation exposure of workers in the transport of NORM [5, 6, and 7]. From these studies the annual effective dose of a truck driver is estimated to be below 1 mSv and therefore radiation protection controls, as required by the European BSS are not necessary.

4 Evaluation of radiation exposure in the processing of zirconium minerals
4.1 Exposure scenarios and exposure pathways
Radiation exposure can occur
- by external radiation (gamma radiation from the material),
- due to inhalation of dust or
- by inhalation of short-lived radon decay products.
Workers in the zircon and zirconium industry can be exposed to gamma radiation from a range of work activities. Scenarios that give the highest radiation exposures...
are the extended occupancy by employees in areas where large amounts of material (e.g. feedstock, intermediates, or products) are stored. In some cases, significantly enhanced radionuclide concentrations can occur in residues such as scales, and similarly enhanced localised gamma radiation levels can exist in the workplace.

Several work activities can result in radiation exposure due to the inhalation of dust and have to be considered. Relevant scenarios are the transfer of feedstock, feeding mills and furnaces, bagging products and dusty operations such as plant maintenance. In processes that take place in closed systems with good workplace ventilation (e.g. firing ceramic tiles, fusion of zirconium silicates) the radiation exposure due to inhalation is often also well controlled, and of secondary importance in terms of radiation protection.

Overall, external exposure tends to be the most significant pathway in the zircon industry, especially where the standard of dust control is good. The inhalation of short-lived radon decay products is generally of minor importance, and exposures from ingestion of dust can be ignored if good occupational hygiene standards are met.

4.2 Calculation and measurements to evaluate the radiation exposure

4.2.1 Calculation of radiation exposure from the specific activity

It is possible to broadly calculate the radiation exposure of workers from the specific activity of the material. The activity concentrations are often known and if this information is not available, the activity concentrations of the U-238 and Th-232 decay series should be determined. The "Method for rapid determination of the specific activity of NORM materials" [8] can be used.

In [9] a simple calculation rule for determining the annual effective dose from the activity concentrations of the material is given, as follows:

\[
E = \frac{t_A}{2000} (c_{ MU} \cdot K_U + c_{ MTh} \cdot K_{Th})
\]

where

- \( E \) annual effective dose (mSv),
- \( c_{ MU} \) activity concentration of U-238 and Th-232 in the material (Bq/g),
- \( K_U \) and \( K_{Th} \) nominal annual effective dose for U-238 (0.78 mSv/(Bq/g)) and Th-232 (0.98 mSv/(Bq/g)) and
- \( t_A \) exposure time in hours per year.

The nominal dose values above assume occupancy in the vicinity (approximately 1 metre) of a large amount of material. Therefore, estimates using these values often lead to an overestimation of actual exposures. If the annual effective dose determined in this way exceeds 1 mSv, workplace monitoring should take place in accordance with chapter 4.2.2 and 4.2.3 of this leaflet.

In [10] dose estimates for employees during milling (or other work activities) of zirconium minerals calculated with the model Microshield\textsuperscript{TM} are discussed. This model allows the calculation of radiation exposure for different work areas and may be useful in the identification of the work areas which require further investigation, e.g. through workplace measurements.
4.2.2 Measurements of external radiation exposure

The external gamma radiation exposure can be determined by measuring the ambient dose rate \( \dot{H}^*(10) \), and combining the results with estimated residence times, as follows:

\[
E = f \cdot \dot{H}^*(10) \cdot t_A \cdot 10^{-3} \tag{2}
\]

where

- \( E \) annual effective dose (mSv),
- \( f \) conversion factor of ambient equivalent dose to effective dose for workers (in this case the coefficient is approximately 1.0),
- \( \dot{H}^*(10) \) measurement of ambient equivalent dose rate in \( \mu \text{Sv/h} \) and
- \( t_A \) residence time in the workplace (working time) in hours per year.

To achieve a realistic estimate of annual dose it is important to use dose rate measurements from locations that workers actually occupy, and also representative (not pessimistic) assumptions about occupancy time. If several workplaces for a person are relevant, the sum of the individual dose at each workplace should be calculated.

The radiation exposure from external gamma radiation can be determined directly by issuing individual dosimeters to workers. National dosimetry services can provide this service. Although requiring more effort to arrange and operate, individual dosimetry provides the most reliable estimate of external effective dose, and avoids the over-estimation that is inherent in estimates based on modelling and assumptions about occupancy.

4.2.3 Measurements of radiation exposure from the inhalation of dust

Handling of zirconium minerals can generate airborne dust and lead to the inhalation of zircon particles by workers. The annual radiation exposure due to the inhalation of dust can be determined from the air concentration of radionuclides as follows:

\[
E = V \cdot t_A \cdot \left( g_{\text{inh,U-238}} \cdot c_{\text{U-238}} + g_{\text{inh,Th-232}} \cdot c_{\text{Th-232}} \right) \cdot 10^{3} \tag{3}
\]

where

- \( E \) annual committed effective dose (mSv),
- \( V \) the breathing rate (for light activity = 1.2 m\(^3\)/h),
- \( g_{\text{inh,U-238}}, g_{\text{inh,Th-232}} \) inhalation dose coefficients for U-238, Th-232 (Sv/Bq),
- \( c_{\text{U-238}}, c_{\text{Th-232}} \) activity concentrations of U-238, Th-232 in the air (Bq/m\(^3\)),
- \( t_A \) residence time in the workplace in hours per year.

Inhalation dose coefficients for the U-238 and Th-232 decay series in secular equilibrium, taken from Table 23 of [11], are:

- \( 3 \cdot 10^{-5} \text{ Sv/Bq for U-238(sec)}^3 \)
- \( 4.8 \cdot 10^{-5} \text{ Sv/Bq for Th-232(sec)} \)

\( ^3 \) The dose coefficient for U-238 also takes into account the natural isotopic ratio of U-238 and U-235.
A simple way to calculate the air concentrations $c_{U-238}$ and $c_{Th-232}$ is to determine the dust loading (in g/m$^3$) using methods commonly employed in industrial dust monitoring, and combine this with the activity concentrations $c_{MT}$ and $c_{MU}$ of the material (in Bq/g), to give the air concentrations $c_{U-238}$ and $c_{Th-232}$. This method does pessimistically assume that airborne dust consists entirely of the material being processed - however, this is a reasonable assumption in workplaces such as zircon mills.

A simple and reliable way of estimating inhalation doses is to determine the activity deposited on respiratory protective equipment (RPE) filters worn by workers. The efficiency of the filter materials usually used is usually above 90% and the activity collected is approximately the same as that which would have been inhaled by the worker. The annual effective dose is then calculated by

$$ E = \frac{1}{T} \left( a_{U-238} \cdot g_{U-238} + a_{Th-232} \cdot g_{Th-232} \right) \cdot 10^3 $$

(4)

where

- $E$ annual committed effective dose (mSv),
- $T$ length of time the RPE is worn (hours),
- $t_A$ annual working hours (hours per year),
- $a_{U-238}, a_{Th-232}$ activity on the respirator filter of U-238 and Th-232 (Bq),
- $g_{U-238}, g_{Th-232}$ inhalation dose coefficients for U-238 or Th-232 in Sv/Bq (see above).

Additional guidance on the evaluation and measurements of the exposure from dust inhalation may be found in [12].

### 4.2.4 Measurements of radiation exposure by the inhalation of radon / radon decay products

When evaluating the radiation exposure of workers, the assessment of exposures from the inhalation of radon/radon decay products is often neglected. In [13] a simple mathematical model is described for estimating such exposures. The Rn-222 release rate (measured in Bq/m$^2$sec) necessary for the calculation of the Rn-222 concentration at the workplace can be determined from the emanation factor.

For most applications the release of radon is very low and an emanation factor of 0.002 can be used. In practice, high radon concentrations normally only occur in the presence of large amounts of material (or high activity concentration radium scales) in poorly ventilated rooms.

### 5 Radiation protection measures in the processing of zirconium minerals

The basic requirements for the radiation protection of workers are defined in Title VI, Article 18 of the European Basic Safety Standards [1]. Radiation protection measures are required if the exposure of a worker (the annual effective dose) can exceed a value of 1 mSv per year. The measures must be appropriate to the situation, taking into account the process and plant, the magnitude of the exposures received and the potential for reducing them (the “graded approach”). In practice, the following should normally be considered:

1. It is preferable to use materials with a lower activity concentration, since the potential doses often depend on the activity of the process materials.
2. It is important to identify the main sources of dust. Routine inspection and maintenance of plant and dust control equipment should be undertaken to keep airborne dust levels under control.

3. As required, containment and ventilation to control workplace dust levels should be used. This should be backed up with working procedures (e.g. cleaning programmes) and (where still necessary to control radiation exposures from dust inhalation), the use of individual respiratory protective equipment (RPE). Use should be made of existing RPE programmes (e.g. to protect against nuisance dust) – in most cases this is also sufficient for radiation protection purposes. In some cases, RPE may be the only practical protection option, for example during maintenance work inside the plant.

4. The location of bulk materials, and the working time in such areas should be reviewed and optimised. External doses can often be reduced significantly through this process.

The above list is not exhaustive. Specific measures will depend on the situation in each particular workplace. For reasons of optimisation, the measures should be integrated into the general health and safety procedures.

Independently of radiation protection aspects, inhalation of crystalline silica may constitute a health hazard and the good practices developed with respect to this hazard and detailed described in [14 ] should also be taken into account.

6 Residues
Residues may be generated during the processing of zirconium minerals and derived intermediates, although these are typically only in small quantities, i.e. because the operational processes are designed to avoid loss of materials for economical reasons. In some cases, it is possible to separate zirconium minerals from residues and return them into the process – however the potential for inadvertently producing residual high activity concentrations should be considered.

In many cases, residues (normally in small quantities) are disposed of in landfills. The release of radionuclides from the deposited materials in groundwater is normally extremely low and any exposures that might occur from this pathway are generally negligible. The most significant exposure pathway is typically the external exposure of the workers at the landfill site. However, since the number of disposals and the amounts of waste are low, the radiation exposure is much lower than those that occur during processing. The annual doses to landfill workers are not expected to exceed the exemption criteria of 1 mSv, and regulatory radiation protection measures are not considered necessary.

Special attention has to be paid to processes (e.g. fusion of zirconium minerals, manufacture of zirconium compounds by chemical dissolution on zirconium minerals) that can produce residues with much higher activity concentrations of radionuclides. Although the quantities of these residues are often small, specific radiation protection controls (on handling and disposal) may be required.
7  References


[2] European Commission: Reference levels for workplaces processing materials with enhanced levels of naturally occurring radionuclides. EUR-STR-095


Further information on radiation protection management in the NORM industry is given in papers presented at the conferences NORM I, II, III, IV and V and in the report of the IAEA ‘Radiation Protection and NORM Residue Management in the Zircon and Zirconia Industries’ Safety Report Series No. 51.

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