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EMPLOYEE RADIATION EXPOSURE LEVELS IN ZIRCON MILLING PLANTS

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Abstract

Zircon has many properties which make it an industrially useful material. It does however contain small quantities of naturally occurring uranium and thorium which may result in it being designated a radioactive substance under recent standards promulgated by the International Atomic Energy Agency. This raises the matter of whether processing of zircon could deliver radiation doses in excess of levels suggested by the IAEA which require workers to be designated radiation workers.

The processing of zircon often involves grinding it to a fine powder known as zircon flour. As the particles are small they may be inhaled if they become airborne and deliver radiation doses to workers. Whilst estimates have been made of potential radiation doses, no reports have been made of measured doses from dust inhalation in zircon flour mills. Similarly no gamma dose measurements have been reported for workers in zircon milling plants

Theoretical and measured radiation exposure from zircon are reported in this paper. Theoretical exposures are based on worst case scenarios. Measurements of dust and gamma radiation have been made at a number of zircon milling plants in Australia.

Theoretical doses indicate a maximum exposure of 5.5 mSv per year. Measured doses vary in different plants from 0.66 mSv to 1.03 mSv per year and indicate that in the dustiest Australian zircon mill the maximum dose would be of the order of 1 mSv per year if workers spent their entire working year in the dustiest occupation. Based on the theoretical and measured doses workers would not be designated as Category A, exceeding 6 mSv per year, and probably would not be designated category B workers, exceeding 1 mSv per year, under the guidelines of the IAEA Basic Safety Standards adopted by the Euratom Directive 96/29.

Introduction

The heavy mineral sands industry is an important one for Western Australia, where significant deposits exist. Zircon is one of the principal components of the mineral sands and is separated from other mineral sands usually by physical separation techniques. All mineral sands contain the radioactive elements uranium and thorium mostly in low concentrations, except for the mineral monazite which typically contains six percent thorium.

Recent revision of the International Atomic Energy Agency's Basic Safely Standards [6] have drawn attention to a range of materials which contain low levels of radioactivity. Zircon has been identified as a material which would be designated radioactive as the activity concentrations can exceed the lower limit of 1 $Bq.g^{-1}$ for thorium and uranium. It can be expected that regulatory bodies will look more closely at materials such as zircon which could be designated radioactive according to the latest standards which propose that;

- where the possibility of exposure is below a dose of 1 mSv per year only minimal surveillance is required,
- between 1 mSv and 6 mSv workers would be classified Category B workers and surveillance needs be sufficient to demonstrate workers are correctly classified, and
- above 6 mSv per year classification as Category A workers and more extensive monitoring is necessary.

The level of radioactivity in Australian commercial zircon is between 150 and 300 ppm uranium and 150 and 250 ppm of thorium [9]. Table 1 gives a summary of activity levels reported for zircon for different places of origin.

Table 1: **Typical uranium and thorium content of zircon ores from various sources [3], [8], [9].**

	Typical U and Th concentrations in commercial zircon ores	
Country of Origin	**Uranium ppm (Bq.g^{-1})**	**Thorium ppm (Bq.g^{-1})**
Australia[9]	150-300 (1.8-3.6)	150-250 (0.62-1.03)
Eastern Australia[8]	170-190 (2.0-2.3)	160-180 (0.66-0.74)
Western Australia[8]	200-250 (2.4-3.0)	180-220 (0.74-0.90)
South Africa[8]	230-260 (2.8-3.1)	200-220 (0.82-0.90)
USA[8]	200-250 (2.4-3.0)	100-150 (0.41-0.62)
China [3]	1190 (14.3)	2000 (8.2)

The further processing of zircon may require that it be ground to a fine powder known as zircon flour where physical particle sizes are in the range from one to several microns diameter. The milling of zircon has been suggested as posing a potential risk of radiation exposure from inhalation of airborne dust, particularly during bagging of product or in maintenance operations on the grinding mills. Despite the identification of this as a possible radiation exposure pathway, few data have been published which verify or otherwise the reality of the risk.

Review of Previous Measured Doses

A small number of publications have dealt directly with exposure of workers in zircon separation and milling plants. None deal directly with the potential hazard of zircon dust inhalation and most report low exposures to gamma radiation from the zircon. Reported dose estimations are for the handling and processing of zircon sand rather than of the milling of the material for further industrial processing. No reports of measured exposure to dust in milling plants have been made.

Boothe et al [1] discuss potential radiation exposure from the use of zircon in the United States. They identify a number of exposure pathways and examples of the use zircon but do not identify inhalation of dust as a significant exposure pathway. They analyse the potential environmental contamination from the use of zircon sand, principally from its use as a foundry sand. Gamma measurement adjacent to large stockpiles of sand were 1.7 μSv.hr^{-1}. No annual dose estimates were made.

The Italian National Group for Studying Radiological Implications in the use of Zircon [7] report gamma radiation levels and dust levels in a zircon refractory plant. The gamma levels were a maximum of 0.4 μSv.hr^{-1} but generally much lower in other working areas. High volume dust samples were also taken and measured for alpha activity. They conclude that the radon levels in the plant were comparable to those found in dwellings and that doses due to inhaled dust were of the order of 2.0×10^{-5} Sv for an eight hour shift. This may be translated to a full years working dose of about 5 mSv. This dose estimation is based on ICRP 30 inhalation model parameters and from the highest activity reported in dust in a zircon smelting plant. The activity inhaled was from smelting fume and not from zircon dust.

Deng et al [3] report significant potential exposure could arise from the use of zircon in ceramic glazes on tiles and point out a potential of a beta radiation hazard in close proximity to the glazes. They identify gamma radiation as a potential hazard where stockpiles of glazed tiles were stored. The reported radioactivity in the zircon sand of Chinese origin reported by Deng is much greater than that reported from other commercial sources. The activity concentrations of uranium, thorium and potassium were up to three times those reported for Australian zircon. The high activity in their zircon does not seem to be typical of other commercial zircon which, whilst having variable activity, does not commonly reach the level reported for the Chinese material.

Summary of Predicted Doses

A broad sheet published by the National Radiological Protection Board [12] indicates potential exposure of 6 mSv per year from exposure to zircon dust and gamma radiation based principally on desk top studies and use ICRP 26 and ICRP 30 models.

Hartley [4] has also estimated maximum potential doses to zircon based on conservative assumption as discussed below. These indicate a maximum potential total dose of 5.5 mSv per year based on the processing of Australian zircon.

The potential exposure to gamma radiation was estimated by Hartley from the concentration of uranium, thorium and potassium in the mineral. Conservative assumptions of 2000 hours working on a semi infinite plane of zircon material with both uranium and thorium concentrations of 200 ppm were used. This level of natural activity is within the range found in Australian zircon product. The maximum radiation dose using this scenario was calculated to be 2.2 mSv per year.

The potential radiation doses from inhaled zircon dust were also based on a worst case scenario. In an industrial situation zircon would be considered a nuisance dust and subject to controls for that type of contaminant. In Australia the limit for airborne nuisance dust is 10 $mg.m^{-3}$. Again it was assumed that the zircon contains 200 ppm of both uranium and thorium. Using these assumptions and a dose conversion factor of 4.5 $\mu Sv.Bq^{-1}$ for uranium and 9.7 $\mu Sv.Bq^{-1}$ for thorium, measured as alpha activities, the worst case dose was calculated as 3.3 mSv per year.

Papers by Mobbs et al [10], by Degrange et al [2], and by Hipkin and Shaw [5], presented at the NORM I conference, also discuss exposure to zircon. Degrange et al estimate exposure to zircon to be as high as 270 mSv per year using their normal assumptions and if the levels of uranium and thorium are high ie 40 $Bq.g^{-1}$ for thorium and 74 $Bq.g^{-1}$ for uranium. These levels seem improbably high for commercial zircon when compared with the data presented in Table 1. Hipkin and Shaw, however estimate typical doses of zero for both gamma radiation and inhaled dust assuming much lower concentration of uranium and thorium in zircon.

On the basis of the uncertainties in the reported data relating to radiation exposure, this work was initiated to clarify the exposures to radiation from zircon by conducting measurements in zircon milling plants.

Doses Measured in Australian Zircon Mills

In order to verify or otherwise the above estimates, measurements of gamma radiation and dust levels in a number of zircon milling plants in Australia have been made. This work is continuing but results are given here of comprehensive measurements taken at four sites.

The mills which have been monitored included modern plant with autogenous grinding facilities to plants at least twenty years old with ball mills and roller mills used for grinding the zircon. The specifications for the zircon flour particle sizes differed from plant to plant but the typical size was less than 10 µm physical diameter. It would be expected that the airborne dust would be smaller than the average physical particle size as the finer dust is more likely to become airborne. The aerodynamic diameter will however be greater than the physical diameter by the square root of the density of zircon; ie by about a factor of about two. No measurements of mass median aerodynamic diameter have been made and it is assumed that the dust particle size is 5 µm.

Gamma radiation levels were measured using Xetex portable radiation monitors calibrated for radiation from monazite by the Radiation Health Section of the Health Department of Western Australia. These monitors were worn by workers in the zircon processing section of the zircon milling plants. Gamma radiation levels were corrected for background radiation, determined from broad scale environmental radiation measurements.

Dust levels were measured with Dupont model P2500A dust pumps which are frequently used for personal dust monitoring in Australian mine sites. Samples were taken on PVC filters with a pore size of 5.0 µm. Standard seven hole samplers were used and the flow rate was nominally two litres per minute. The flow rate was measured at the start and end of the sampling period and an average flow rate used to determine the dust concentrations in the air. For any individual worker dust sampling was usually undertaken for a full eight hour shift. The dustiest operations were targeted for monitoring and this was usually the bagging of the zircon flour product.

Radioactivity in the dust was determined by alpha particle counting by different counting systems at two sites. The uranium and thorium concentrations in the product being milled was used to apportion the alpha activity between the two principal decay chains. Standard breathing rate of 1.2 $m^3.hr^{-1}$ and annual 2000 working hours were used to estimate the annual radiation dose.

The working patterns in the plants were not considered in determining the annual radiation doses. In all cases it was assumed that a worker would be occupied for the full year doing the work done during the time the sample was taken. This would give an overestimate of the radiation dose as the dustiest operations were targeted. In some plants the grinding of zircon occupied less than ten percent of the use of the grinding mill. These factors would produce an overestimate of the doses.

On the other hand average values of the dust and gamma radiation levels were used to determine exposure levels. This could lead to an underestimate of dose for an individual workers as work patterns will differ. This averaging is considered to be acceptable in a survey such as this as doses are low.

***Table 2:* Annual radiation doses calculated from measurements in zircon milling plants**

	Gamma dose mSv	Activity in dust $Bq.m^{-3}$	Inhaled Dose mSv	Total Dose mSv
Plant A	0.4	0.0187	0.27	0.67
Plant B	0.3	0.0525	0.73	1.03
Plant C	0.1	0.0407	0.56	0.66
Plant D	0.4	0.0400	0.56	0.96

Plant A is a relatively modern plant which uses autogenous grinding to produce zircon flour. Dust extraction and enclosed bagging facilities are installed. The plant is visually clean with evidently good industrial hygiene practice.

Plant B is an old plant more than twenty years of age. Bagging is done without special dust extraction. The grinding plant is a roller mill and the working schedule for the plant means that the zircon processing occupies less than ten percent of the total operating time.

Plant C is an old plant more than 25 years old and uses a ball mill for grinding. There are no special dust suppression measures taken. Grinding zircon occupies less than 25 percent of the plant time.

Plant D is also a 25 year old plant which uses a ball mill for grinding. It uses semi automatic bag filling system and uses 50 Kg paper bags as well as one tonne bulk bags. There is no special dust collection system.

Discussion

Although further monitoring is being conducted at zircon milling plants in Australia, doses to workers as a result of their working in the zircon milling plants are probably less than 1 mSv per year. Gamma radiation levels are reported as measured but at these low levels of exposure the precision cannot be high and could include a component of self radiation from the person wearing the monitoring device as well as a contribution from the building material of the milling plant.

Measured dust levels are converted to exposure without any corrections for respiratory protection. Should such protection be used a typical protection factors of five may be considered appropriate.

The dust levels and gamma radiation levels measured are typical of well controlled work places. The mines and factories inspection system in Australia is fairly strong with a system of checking for plant condition and industrial hazards. Based on the measurements and estimates of worst case conditions, it is not considered possible that doses from zircon, typical of Australian product, could reach 6 mSv per year and probably do not exceed 1 mSv per year, these being the doses delineating the requirements for progressively greater worker monitoring under the Euratom 96/29 directive.

The radiation levels stated represent average levels for a number of workers employed in similar tasks within a plant. Individual levels will vary about that average and individual work practices could be responsible for those variations as well as the different work schedules. In most plants surveyed the work schedules are such that the exposures to zircon dust will be much lower than is stated here as grinding of zircon is a small part of the overall plant production.

The gamma radiation doses as well as those for inhaled dust will depend on the concentration of uranium and thorium in the feedstock and product. The estimates of dose are based on typical levels in Australian zircon. Doses would be approximately proportional to the levels of activity in the product.

Conclusion
Although zircon may be designated a radioactive material under the IAEA Basic Safety Standards it does not seem possible for workers to exceed 6 mSv per year as a result of exposure in zircon milling plants. This conclusion is based on theoretical estimates of potential exposure given worst case scenario and uranium and thorium concentrations typical of Australian zircon. Measured doses indicate that the maximum dose would be of the order of 1 mSv if workers spend their entire working year in the dustiest occupation in a zircon mill. Based on these estimates we can be confident that workers would not be designated as Category A workers, with the potential to exceed 6 mSv per year. Depending of the working conditions and the uranium and thorium content of the zircon, worker exposure should not require that workers be designated at category B workers exceeding 1 mSv per year.

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