Radiological Impacts Associated with Zircon Sand Processing Plant in Brazil

M.A. Pires do Rio, P.R. Ferreira, D.C. Lauria, V. Paula Melo

Instituto de Radioproteção e Dosimetria/IRD/CNEN
Rio de Janeiro
Brazil

Abstract. Aimed at assessing radiological impacts of zircon sands processing in Brazil, a survey of zircon sand and flour production and of the ceramic and porcelain industries has been performed in order to identify the most important industries that should be investigated. As a starting point, an important processing plant located in a coastal area of Rio de Janeiro State has been investigated. Mining and milling of heavy mineral sands are performed in the plant to produce zirconite, ilmenite and rutile. The zirconite fraction is then sent to milling companies involved in zircon silicate flour production, which will be used in the ceramics and porcelain industries. The contents of natural radionuclides in raw materials, residues and the end products of the plant milling were measured by gamma spectrometry. The radionuclide flux in the process showed the highest concentrations of ^{238}U and ^{232}Th in the monazite fraction, respectively, 7300 and 44 500 Bq/kg, followed by zirconite (2300 and 500 Bq/kg), rutile (1240 and 1600 Bq/kg) and ilmenite (130 and 620 Bq/kg). The only discharge to environment is a liquid effluent of the plant, which presented concentrations of U and Th in the order of 10^{-2} and no environmental impact was observed. Previous studies pointed out that individuals are occupationally exposed to uranium via inhalation and estimated a value of effective dose due to uranium of 0.4 mSv/a. Regarding inhalation exposure, the results indicated the existence of operational areas with dose values above the maximum recommended limits. The bioassay monitoring data indicated that the implemented actions have decreased the worker's dose in a significant way. This paper presents an overview of previous studies that were carried out in order to evaluate radiological impacts associated with zircon sand processing plant in Brazil.

1. Introduction

Brazilian monazite sand is composed of four minerals: ilmenite ($FeTiO_3$); zirconite ($ZrSiO_4$); rutile (TiO_2) and monazite, an orthophosphate of rare earths containing up to 6% ThO_2 and 0.3% U_3O_8. The most important deposits of monazite sand in Brazil are located on the Atlantic coast of the country, in the northern part of the State of Rio de Janeiro.

Buena, a small village of this region, is the place where an important processing plant has been in operation for more than 25 years. The only industry activity of the region is the mining and milling of the heavy mineral sand and many Buena inhabitants work at the mill (Buena Unit). An important characteristic of the people living in this area is their low mobility. Most of the people have been living in Buena and its surroundings for more than 40 years [1]. The food consumed by the population is basically composed of local products [2].

Because the levels of thorium, monazite was considered as a strategic mineral and the Buena Unit was classified as a nuclear facility according to Brazilian standards [3]. Thus for years it has been occupationally and environmentally monitored. As the mill´s process area was classified as a controlled area, the access to the area is controlled, the workers are continuously monitored by EPIs and they use for protection, overalls, boots, gloves, hardhats and masks. Over time, and according to the surveys and studies conducted in the area, new control methods have been introduced.

The process begins with the concentration of the minerals by a hydrogravimetric process. After the separation, the ore concentrate is sent to the mill and the sterile material (sand) is then returned to the pit. The heavy minerals content in the sand and concentrate are shown in Table 1. It can be observed that the minerals in the ore are around ten times more concentrated than in the original sand. In the mill, the heavy minerals are separated by dry separation (physical separation through electrostatic, magnetic and gravimetric processes). The industrial process is described in the simplified flow chart in Fig. 1.

The final phase of the dry separation process produces zirconite (zirconium silicate), ilmenite (iron titanate) and rutile (titanium dioxide) that are sold directly from the mill,. Nowadays, monazite is stored for a new process in the future, which is now being developed.

TABLE 1. CHARACTERISTICS OF THE HEAVY MINERAL SAND AND CONCENTRATE [4]

	Content (%)	
	Heavy mineral sand	Concentrate
Ilmenite	3.9–4.3	39.1–43.0
Zirconite	2.3	23.3–23.6
Monazite	0.28–0.65	2.8–6.5
Rutile	0.18–0.30	1.8–3.0
Silica	60–85	6.0–8.5
Other	2.1–2.5	20.5–25.0

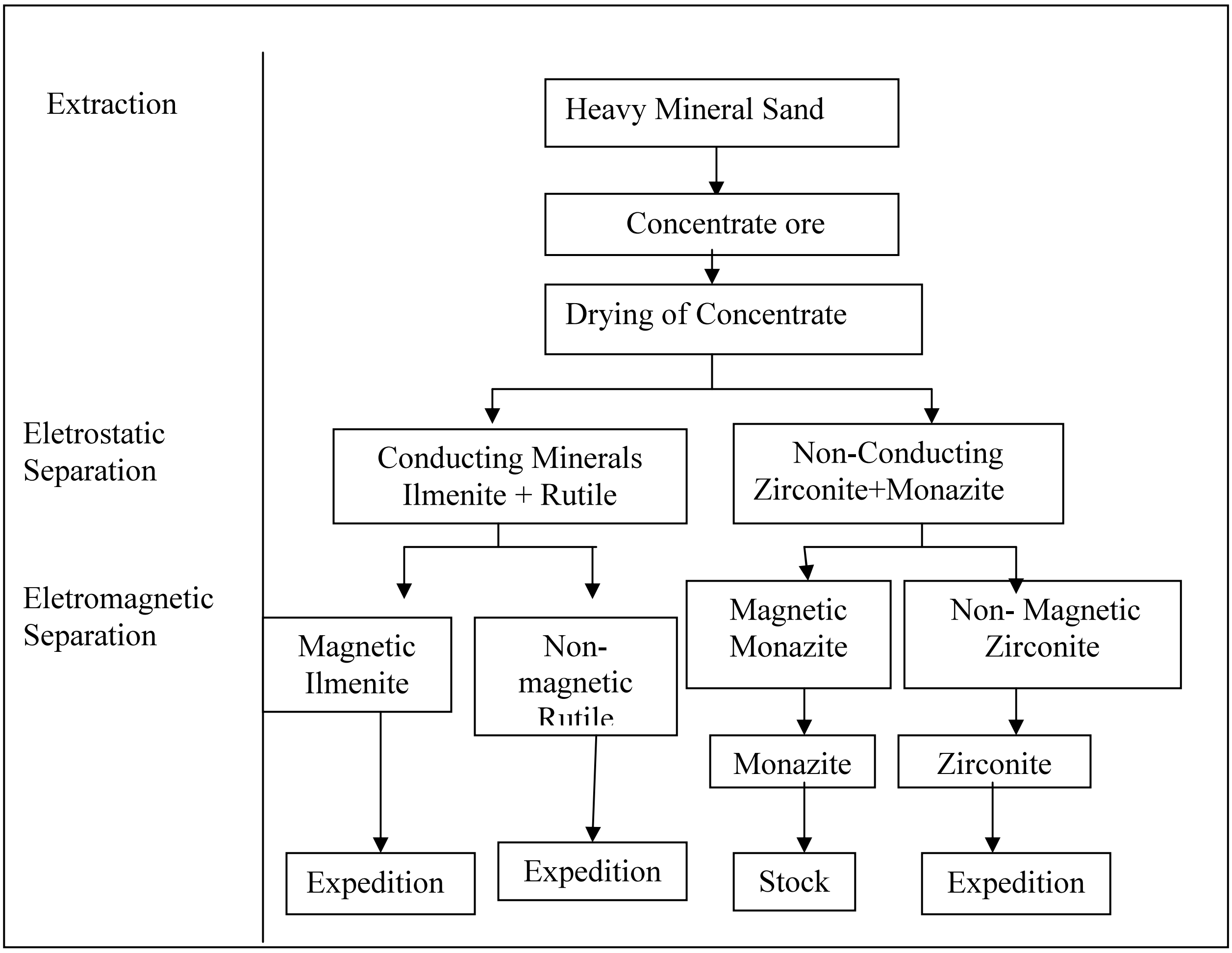

FIG. 1. Simplified flow chart of the mineral separation process

The zirconite fraction is then sent to milling companies for the production of zircon flour, which will be used in the ceramics and porcelain industries. The assessment of the radiological impacts associated with these industries will be carried out in the near future.

The former chemical processing of monazite concentrate was performed in other plants and produces rare earth chlorides. The process generated two byproducts: the mesothorium cake and the thorium concentrate cake, both of which contain most of the initial thorium and uranium of the monazite concentrate. The radioecological questions concerning the monazite sand cycle waste in Brazil has already been published elsewhere [5] [6].

2. Methodology

A summary of the main results of previous studies and surveys carried out in Buena region aimed at assessing the radiological impacts associated with zircon sand processing plant in Brazil are presented below. These studies have included public and occupational exposures to uranium and thorium due to operation of the processing plant.

3. Results

3.1 Public exposure

The contents of natural radionuclides in raw materials, residues and the end products of the plant milling were measured by gamma spectrometry and are presented in Table 2. These results are part of the environmental radiological programme that was carried out by the Institute of Radiation Protection and Dosimetry/IRD/CNEN from 1996 to 2002.

TABLE 2. RADIONUCLIDE CONCENTRATIONS IN RAW MATERIALS, RESIDUES AND END PRODUCTS OF THE MILLING PLANT

	Radionuclide activity concentration (Bq/kg)[a]	
	^{238}U	^{232}Th
Heavy mineral sand	99	520
Heavy mineral concentrate	2100	7580
Monazite fraction	7300	44500
Zirconite	2300	500
Rutile	1240	1600
Ilmenite	130	620
Silica (sand)	10	44
Overflow (waste)	1350	2090

[a] Secular equilibrium assumed.

The radionuclide flux in the process showed the highest concentrations of ^{238}U and ^{232}Th in the monazite fraction, followed by zirconite, rutile and ilmenite. The only discharge to the environment is a liquid effluent from the hydrogravimetric process. Water used in the process is pumped from a local lagoon and after its utilization in the hydrogravimetric process it is returned to the lagoon. The data from the liquid effluent monitoring programme showed concentrations of U, Th, ^{226}Ra and ^{228}Ra of the order of 10^{-3}, 10^{-2}, 10^{-2} and 10^{-1} Bq/L, respectively. It is interesting to observe that higher radionuclide concentrations in the water of the lagoon were found upstream instead of downstream from the effluent discharge point [7]. This unexpected result led to research that concluded that the radionuclide concentrations originated naturally from springs at the lagoon head area. Curiously, the levels of radionuclides in the mill effluent were lower than the levels of this natural environment and no environmental impact was observed due to the effluent discharge of the mill.

Significant concentrations of thorium, uranium and other nuclides from the respective radioactive series have been reported in the standard diet consumed by the inhabitants of Buena village [2]. The results indicated that the major route of intake of ^{226}Ra and ^{210}Pb by workers and inhabitants is ingestion (alimentary diet and water).

Characterization of the airborne particle aerosol and the origin of the aerosol source were performed in samples collected in the vicinity of the mill [8, 9]. The airborne particle characterization has suggested that the inhabitants of Buena were exposed to mineral sands particulate (mainly from the mineral sands process plant and a natural deposit of mineral sands located in the village), marine aerosols and particles from anthropogenic sources in the respirable fraction of the aerosols [8]. Thus, as the inhabitants of Buena are also exposed to aerosols containing natural sources, no direct impact of the aerosols from the mill could be quantified.

3.2 Occupational exposure

The main radiological problem in this mill concerns the dust generation during the gravitational separation steps of the process (vibrating tables are used to sieve the minerals and the products are handled and bagged in the area). The main studies and surveys performed in the mill concern a survey of the potential exposure of the workers and studies of radionuclides incorporated by them.

Occupational Measurements

In a study of the potential occupational exposure, a survey was carried out in five different locations within the mill, relating to different steps of the process, by aerosol sampling with AGF followed by gross alpha counting [10]. Evaluating the ^{232}Th activity by the long-lived alpha emitters, the ^{232}Th concentration in the fine fraction of the aerosols (2.5–10 µm) was 0.026–0.26 Bq/m^3, while the finest fraction of the aerosols (below 2 µm) had a ^{232} Th concentration of 0.0016–0.00076 Bq/m^3. Thus, using the data of the respirable fraction of aerosol, the assessed value of the inhalation dose ranged from 8 to 125 mSv/a.

Regarding external occupational exposure, which was estimated by thermoluminescent dosimeters, (TLDs) during one year using 17 workers, an average of 2.7 ± 0.3 mSv/a was observed [10]. Additionally, gamma radiation dose rates were measured at the five different points of the mill. The values of the dose rate was 0.07–1.90 µSv/h (0.1–3.4 mSv/a). Considering the external and inhalation dose, the total dose was estimated to be 8–128 mSv/a. The highest value was found in the drying area, while the smallest value was found in the magnetic separation of non-magnetic minerals area.

Bioassay Monitoring

Because of Buena´s location in a high background radiation area (HBRA) the workers of the mill are exposed to radiation as inhabitants of a HBRA and as workers in a NORM facility. Then, the evaluation of occupational radiological exposures are not straightforward, esspecially considering all uncertainties involved in measurements, models, in vivo samples and so on. Aiming to discriminate the background internal contamination (Buena) from the mill contribution from the mill, research has been carried out taking in account data of excreta samples from the mill workers and from the Buena inhabitants [11, 12].

Regarding ^{232}Th internal contamination, excreta samples of 29 workers in the mill and 21 inhabitants of Buena, where most of workers live, were collected and analyzed [6]. The average ^{232}Th concentration in feaces of the Buena non-exposured population (6.7 ± 5.9 mBq/g_{ash}) was about one third of the average ^{232}Th concentration in feaces samples from workers (25.9± 18.4 mBq/g_{ash}) and twice the average concentration in feaces from inhabitants of the city of Rio de Janeiro (2.3± 1.5 mBq/g_{ash}), the control group. The results have shown the higher intake of ^{232}Th by the workers than their wives, which can be caused by dust ingestion. It is suggested that measures to diminish dust ingestion in the mill should be reinforced. Besides, the results pointed out the importance in characterizing the region where workers live when occupational control is carried out [1].

Concerning occupational exposure to uranium, the intake of radionuclides by the workers and the inhabitants of the mill vicinity was studied by excreta sample analysis. The results indicated that workers might be exposed by inhalation to compounds of uranium type S with an AMAD of 1 or 5 µm and exposed by ingestion to compounds of uranium type M. A value of effective dose due to uranium inhalation of 0.4 mSv/a was estimated [12].

Only one of the sixty mill workers monitored in the whole-body counter for measurements related to intakes of long-lived nuclides showed activity in the lung slightly above the detection limit [1].

4. Conclusions

- The highest concentrations of ^{238}U and ^{232}Th were observed in the monazite fraction, followed by zirconite, rutile and ilmenite.
- The liquid effluent discharge of the plant to the environment presented a concentration of radionuclides of the order of 10^{-2}, which is below the radionuclide levels found in the water from the region, and no environmental impact was observed due to liquid discharge.
- The airborne particle characterization has suggested that the inhabitants of Buena village can be slightly exposed to mineral sands particulate from the mineral sands process. However, other natural sources can have a greater influence than the mill for this exposure route. In conclusion, no environmental impact from the mill process could be observed.
- Significant concentrations of thorium, uranium and other nuclides from the respective radioactive series in the standard diet consumed by the inhabitants of village of Buena were reported. Thus, to evaluate the internal contamination of the workers, the intake of radionuclides by food consumption has to be take into account.
- Concerning bioassay monitoring, an effective dose of 0.4 mSv/a due to inhalation of uranium was estimated and the concentrations of ^{232}Th in workers' feaces were three times higher than those for their wives.
- Regarding occupational exposure measurements, the results indicated the existence of operational areas where effective doses were above the maximum recommended limits. The results showed that these areas have to be continuously monitored in order not to allow unnecessary exposure of the workers.
- The comparison of the potential exposure data by occupational measurements and the bioassay monitoring data indicated that the implemented actions to decrease the dose (such as the use of EPIs and masks) had significantly contributed to the reduction of the dose to the workers.

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