Measurement of 238U and 232Th radionuclides in ilmenite and synthetic rutile

M I Idris1a), K K Siong1 and S M Fadzil1

1School of Applied Physics, Faculty of Science and Technology, Universiti Kebangsaan Malaysia, 43600 UKM, Bangi, Selangor Darul Ehsan

a)aidzat@ukm.edu.my

Abstract. The only factory that currently processes ilmenite to produce synthetic rutile is Tor Minerals in Ipoh, Perak, Malaysia. These two minerals contain radioactive elements such as uranium and thorium. Furthermore, this factory was built close to the residential areas. Thus, the primary issues are radiation exposure attributed to the decay of the radionuclides. Hence, the objectives of this study are to measure the dose and to evaluate activity levels of uranium and thorium. Dose rates from surrounding area of factory indicate the normal range for both on the surface and 1 meter above the ground (0.3-0.7 $\mu$Sv/hr) lower than the global range of 0.5-1.3 $\mu$Sv/hr set by UNSCEAR. The mean activity levels of uranium and thorium for ilmenite are 235 Bq/kg and 503 Bq/kg while for synthetic rutile are 980 Bq/kg and 401 Bq/kg, respectively. The result shows that uranium activity levels of synthetic rutile is 4 times higher than ilmenite but it is still lower than the regulatory exemption limit of 1000 Bq/kg set by IAEA Basic Safety Standards. Even though the dose rates at the factory and the activity levels are within safe limits, safety precautions must be followed by the factory management to prevent any unwanted accident to occur.

1. Introduction

Tor Minerals International, was founded by Benelite Corporation of America in 1973. “Benelite Process” was developed by Benelite and patented for producing synthetic rutile. Synthetic rutile is the raw material used in producing HITOX TiO$_2$ pigment. Benelite licensed and helped to build several plants around the world, which have utilized this process, including Tor Minerals in Ipoh, Malaysia [1].

Synthetic rutile can be produced by reducing the iron oxide in ilmenite to metallic iron using carbon monoxide, followed by reoxidation and separation from the TiO$_2$ rich fraction (Becher process) or leaching with hydrochloric acid (Benelite process) [2]. Both synthetic rutile and ilmenite contain radioactive elements such as Uranium-238 and Thorium-232. The probability of exposure to radiation due to decay of radioactive products is high depending on the dose rate and the activity levels during processing of these materials. Hence, a safety measure should be taken to ensure that radiation exposure to the public and workers does not exceed the limit. Therefore, the objectives of this study are to measure the dose rates surrounding the factory area which is located near the residential area and to evaluate the activity levels of uranium and thorium in the ilmenite and synthetic rutile samples.
2. Methodology

2.1. Study area
The study areas were located around Tor Minerals factory (Figure 1 and 2). The Tor Minerals factory is in the Kinta district in the state of Perak (Jalan Bukit Merah). However, it was built near residential area.

![Location of Tor Minerals, Ipoh.](Source: Google Map, 2017)

**Figure 1.** Location of Tor Minerals, Ipoh.

![Sampling Points at Tor Minerals factory.](Source: Google Map, 2017)

**Figure 2.** Sampling Points at Tor Minerals factory

2.2. Dose rates measurement
“LUDLIM Rate Meter” (Geiger-Mueller Counter) was used to measure the radiation dose around the study area. At each sampling point, two radiation dosage readings were recorded, one at the soil surface and the other at one meter above the soil surface [3]. Coordinates of each location were recorded using Silva Multi-Navigator equipment.

2.3. Radioactivity measurement
Samples of ilmenite and synthetic rutile were taken directly from the factory’s stockpile. Each sample was weighed, sealed and left for a month to achieve a secular equilibrium [4]. The samples were measured using a gamma spectrometry system with a High-Purity Germanium (HPGe) detector and analyzed with Genie 2000 software. The HPGe detector is enclosed inside CANBERRA 747 shielding
with 10 cm thickness lead coated with 1 mm and 1.6 mm of tin and copper respectively, in order to reduce the background radiation from building and cosmic rays. Energy resolution for the 1332.5 keV energy peak was 1.80 keV and the relative efficiency of the detector was 30%. The energy calibration of gamma spectrometry system was done using a mixture of $^{22}\text{Na}$, $^{51}\text{Cr}$, $^{57}\text{Co}$, $^{60}\text{Co}$, $^{85}\text{Sr}$, $^{88}\text{Y}$, $^{109}\text{Cd}$, $^{113}\text{Sn}$, $^{137}\text{Cs}$, $^{123}\text{mTe}$, and $^{241}\text{Am}$ radionuclides. The efficiency calibration was done using an in-house preparation silica standard spiked with mixture of $^{51}\text{Cr}$, $^{57}\text{Co}$, $^{60}\text{Co}$, $^{85}\text{Sr}$, $^{88}\text{Y}$, $^{109}\text{Cd}$, $^{113}\text{Sn}$, $^{137}\text{Cs}$, $^{123}\text{mTe}$, $^{210}\text{Pb}$ and $^{241}\text{Am}$ radionuclides. Activity levels of Thorium ($^{232}\text{Th}$) were determined through the $^{228}\text{Ac}$ (338.32, 911.20, and 968.97 keV) and $^{208}\text{Tl}$ (583.19 and 2614.51 keV) [5,6].

2.4. Determination of specific activities of radionuclides

The specific activity of radionuclide was determined using comparative method with the following equation [7]:

$$W_s = \left( \frac{M_p \times A_s}{M_s \times A_p} \right) \times W_p$$  \hspace{1cm} (1)

where,

- $W_s$: Specific activities in samples (Bq/kg).
- $W_p$: Specific activities in certified reference material (Bq/kg).
- $M_s$: Mass sample (g).
- $M_p$: Mass certified reference material (g).
- $A_s$: Activities of samples (cps)
- $A_p$: Activities of certified reference material (cps)

3. Results and discussion

The surface dose rates from surrounding areas of Tor Minerals factory show normal range reading for both, on the surface (0.3-0.7 μSv/hr) and 1 meter above the ground (0.3-0.7 μSv/hr).

The results indicate that sampling points number 3, 4, 9 and 10 show higher dose rates than the other locations because they are located near ilmenite storage and Beh Minerals factory which has monozite storage. However, the dose rates from surrounding areas are below the global range of 0.5-1.3 μSv/hr [4]. In general, dose rate on soil surface depends on the weather. Based on Assessment of radiation doses to the population in the environment of a nuclear power plant, calculations may statistically consider the variability of weather-dependent dispersion conditions as well as any seasonal differences in radiation doses. For example in dry weather, radionuclides are deposited on the surface of the soil. However, the radionuclides quickly penetrate into the first centimeter of soil after rainfall. Therefore, if there is no further rainfall, the radiation will return to normal within a few hours or days as the soil saturation disappears. This will influence the radiation measurement at the surface of the soil. One meter from the soil surface is generally used as reference for surface radiation dose measurement [4].
Table 1. Coordinates of sampling points and dose rates measurement.

| Sampling point | Location | Surface Dose Rate (μSv/hr) |
|----------------|----------|-----------------------------|
|                | North    | South | On the surface | 1 m above |
| 1              | 4°32.800 | 101°2.441 | 0.3 | 0.3 |
| 2              | 4°32.805 | 101°2.561 | 0.4 | 0.3 |
| 3              | 4°32.794 | 101°2.532 | 0.6 | 0.5 |
| 4              | 4°32.801 | 101°2.549 | 0.7 | 0.6 |
| 5              | 4°32.772 | 101°2.555 | 0.3 | 0.3 |
| 6              | 4°32.801 | 101°2.548 | 0.4 | 0.3 |
| 7              | 4°32.751 | 101°2.537 | 0.4 | 0.3 |
| 8              | 4°32.701 | 101°2.505 | 0.5 | 0.4 |
| 9              | 4°32.662 | 101°2.506 | 0.7 | 0.6 |
| 10             | 4°32.681 | 101°2.514 | 0.7 | 0.7 |
|                | Range    |       | 0.3-0.7 | 0.3-0.7 |
|                | Mean     |       | 0.5     | 0.43    |

The results indicate that sampling points number 3, 4, 9 and 10 show higher dose rates than the other locations because they are located near ilmenite storage and Beh Minerals factory which has monozite storage. However, the dose rates from surrounding areas are below the global range of 0.5-1.3 μSv/hr [4]. In general, dose rate on soil surface depends on the weather. Based on Assessment of radiation doses to the population in the environment of a nuclear power plant, calculations may statistically consider the variability of weather-dependent dispersion conditions as well as any seasonal differences in radiation doses. For example, in dry weather, radionuclides are deposited on the surface of the soil. However, the radionuclides quickly penetrate into the first centimeter of soil after rainfall. Therefore, if there is no further rainfall, the radiation will return to normal within a few hours or days as the soil saturation disappears. This will influence the radiation measurement at the surface of the soil. One meter from the soil surface is generally used as reference for surface radiation dose measurement [4].

The mean activity levels of uranium and thorium for ilmenite are 235 Bq/kg and 503 Bq/kg while for synthetic rutile are 980 Bq/kg and 401 Bq/kg, respectively as shown on Figure 3.
Figure 3: Activity levels for $^{238}$U and $^{232}$Th in ilmenite and synthetic rutile samples.

From figure 3, the result shows that uranium activity level of synthetic rutile is 4 times higher than activity level of ilmenite but it is still below than the limit of 1000 Bq/kg set by IAEA Basic Safety Standard [8]. Comparing with Chinnaekksaki et al. [9], the mean activity levels of Uranium in synthetic rutile taken from production plant in Tamil Nadu, India is very low (38.23 Bq/kg). Additionally, Cooper [10] showed that synthetic rutile processed in Australia also has low activity level of Uranium at 210 Bq/kg and it is lower than uranium activity in its corresponding ilmenite (300 Bq/kg). In this study, the activity level of Uranium in synthetic rutile is shown higher than ilmenite may be caused by the incomplete Uranium extraction from ilmenite. Based on Iluka’s Synthetic Rutile Production Reports [11], all ilmenites consist low levels of Uranium and Thorium. However, in some mineral sands deposits these levels can be elevated near 500ppm U plus Th. The elements tend to remain with the TiO$_2$ during the synthetic rutile process. Therefore, they are found in even higher levels in the synthetic rutile product.

In the ilmenite chemical processing, it has been reported that 77% of the initial Thorium activity end up in the iron oxide waste and 18% will go into synthetic rutile [12]. Apparently from figure 3, the activity level of thorium in synthetic rutile is not 18% of the initial but it is still lower than ilmenite sample.

4. Conclusion
It is observed that the dose rates surrounding Tor Minerals factory are higher than the background dose due to the presence of thorium and uranium in ilmenite and synthetic rutile. Moreover, it is observed that
activity level of uranium in synthetic rutile is higher than activity level of thorium. It is concluded that the residential area surrounding Tor Minerals factory are safe in the context of public health with respect to radioactivity. Future studies shall also include the interior of the factory in order to assess the radiation risks to the plant workers. Radon level reading shall also be included.

5. References
[1] Tor Minerals International 2007 Tor Minerals History (http://www.torminerals.com/history.html. 10 May 2012).
[2] McNulty G S 2007 Production of titanium dioxide (IAEA TECDOC-NORM-V Vienna IAEA)
[3] UNSCEAR 2000 Sources and effects of ionizing radiation (New York United Nation).
[4] Ismail B, Nasirian M and Pauzi A 2007 Radioactivity and radiological risk associated with effluent sediment containing technologically enhanced naturally occurring radioactive materials in amang (tin tailings) processing industry J. of Environmental Radioactivity 95 161-170.
[5] Hassan A M, Abdel W M, Nada A, Walley E N and Khazbak A 1997 Determination of Uranium and Thorium in Egyptian monazite by gamma-ray spectrometry Appl. Radiation Isotope 48 149-152.
[6] Aznan F I, Yasir M S, Amran, Ab M, Ismail B, Redzuwan Y and Irman, Abd R 2009 Radiological studies of naturally occurring radioactive materials in some Malaysia’s sand used in building construction The Malaysian J. of Analytical Sciences 13 29-35.
[7] Yasir M S, Majid A Ab, Ibrahim F, Tap M S Q and Abidin M R Z 2006 Analisis $^{238}$U, $^{232}$Th, $^{226}$Ra dan $^{40}$K dalam sampel amang, tanah dan air di Dengkil, Selangor menggunakan spektrometri sinar gam Resist. Protection Dosimetry 140(4) 378-382.
[8] Cooper M B 2005 Naturally Occurring Radioactive Materials (NORM) in Australian Industries-Review of Current Inventories and Future Generation (Radiation Health and Safety Advisory Council).
[9] Iluka’s Synthetic Rutile Production Reports 2012 (Product and Technical Development in Australia).
[10] Haridasan P P, Pillai P M B, Tripathi R M and Puranik V D 2007 Thorium in ilmenite and its radiological implications in the production of Titanium Dioxide Rad. Protection Dosimetry 49 220-227.

Acknowledgement
The authors wish to thank to Ministry of Higher Education for the research grant given to this project (reference number UKM-ST-07-FRGS0235-2010) also to the technical support from Nuclear Science Programme, UKM.