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# DETECTION OF URANIUM MINING ACTIVITIES

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In undisturbed natural uranium ore the $^{238}U$ decay chain isotopes appear in secular decay equilibrium with activity ratios equal to *one*. In the course of ore processing the bulk of the uranium decay products is separated from the uranium product and concentrated in the tails. Therefore the disturbed activity ratios of short-lived daughters to long-lived parents can be indicators of ore processing.

Using $^{234}Th$ and $^{238}U$ activities (the short-lived daughter with $T_{1/2}$=24.1 days and the long-lived parent respectively) one can roughly estimate how much time has elapsed since ore processing occurred. Equilibrium is reached in about three months after processing and the $^{234}Th$ and $^{238}U$ activity levels are approximately equal (taking into account the error of measurements). Higher or lower $^{234}Th$ activity levels, relative to $^{238}U$, indicate the material has been recently processed. Assuming the product is depleted in Th and the tails are enriched, the activity of $^{234}Th$ in fresh product should be lower than $^{238}U$ and higher in fresh tails.

The $^{234}Th/^{230}Th$ activity ratio can also be used for age estimations ($^{230}Th$ is a long-lived nuclide). In non-processed ore and immediately after processing this ratio should be equal to *one*. After that the $^{234}Th/^{230}Th$ ratio for a product increases from *one* to a higher equilibrium value and for tails it decreases from *one* to a corresponding lower equilibrium value. As a result for undisturbed uranium ore the $^{234}Th/^{230}Th$ activity ratio will be equal to *one*, for uranium product - significantly higher than *one*, and for tails - significantly lower than *one*. The deviation of the ratio from *one* for product and tails depends on the U/Th separation coefficient. The relationship between the $^{234}Th/^{230}Th$ activity ratios at a given time after processing ($R_t$) and for equilibrium ($R_{eq}$) may be expressed as

$$R_t = R_{eq} + (1 - R_{eq})\exp(-\lambda_{234}t).$$

The relationship between the U/Th separation coefficient ($K_{U/Th}$) and $R_{eq}$ may be expressed as

$$K_{U/Th} = R_{eq}(\text{product})/R_{eq}(\text{tails}).$$

Five samples were taken from the Ranger Uranium Mine and Concentration Plant in Australia, and one sample was taken from the Jabiluka mine (10 km far from the Ranger Mine). The samples included non-processed ore, coarse ore from the stockpile, final crushed ore, fresh and old tails, and fresh product ($U_3O_8$). All the samples were analyzed by HRGS to measure the activities of gamma emitting nuclides. XRF and IDMS were used to measure uranium content and isotopic composition.

Subsamples of about 0.5 - 1.5 g were analyzed using XRF (Philips 1480 XRF analyzer) and IDMS ($^{233}U$ spike, VARIAN MAT-262 mass spectrometer) techniques. The $^{238}U$ activity was

calculated from these measurement results. The $^{234}$Th activity was measured by HRGS with a planar HPGe detector and a calibrated low activity $^{241}$Am solution as an internal standard.

The $^{234}$Th/$^{230}$Th activity ratio was measured using the 60 keV energy region where both isotopes have gamma lines. Use of gamma lines with close energies (63.29 keV for $^{234}$Th and 67.67 keV for $^{230}$Th) allows one to reduce the large contribution of geometry and absorption effects on the measured ratio. This is clear advantage of the method. Disadvantages of the method include the low intensity of analytical lines (long measurement time) and possible interference with the 63.9 keV line of $^{232}$Th, which can be significant for uranium ores with a high Th content. The intensity of the 63.9 keV line is about 6 times lower than the 63.29 keV line if concentrations of both thorium isotopes are equal. The Table below shows the measurement results.

| Sample ID | Material | $^{234}$Th±σ, Bq/g 21 July | $^{238}$U±σ, Bq/g 18 August | ($^{234}$Th/$^{230}$Th)±σ 10 July | ($^{234}$Th/$^{230}$Th)±σ 20 December |
|---|---|---|---|---|---|
| 97-01 | Fresh tails | 16 ± 3 | 3.0± 0.1 | 0.43 ± 0.06 | 0.08 ± 0.01 |
| 97-02 | Old tails | 4.3± 0.9 | 3.4± 0.9 | 0.10 ± 0.01 | 0.15 ± 0.02 |
| 97-03 | Coarse ore | 5.4± 1.1 | 6 ± 5 | 0.84 ± 0.17 | |
| 97-04 | Fine ore | 20 ± 4 | 19 ±10 | 0.64 ± 0.13 | |
| 97-05 | Product | 6500 ±1300 | 10200 | 30 ±15 | |
| 97-06 | Jabiluka ore | 1.0± 0.2 | 1.1± 0.1 | 0.70 ± 0.40 | |

For samples 97-02, -03, -04 and -06 (old tails and ore samples), the difference between $^{234}$Th and $^{238}$U activities is not significant, therefore one can conclude that these samples were not processed during the three months time period before the analysis date. Sample 97-01 (fresh tails) has a $^{234}$Th/$^{238}$U activity ratio of about 5 that certainly indicates that the tail material is fresh. Sample 97-05 (product) has a $^{234}$Th/$^{238}$U activity ratio of about 0.6 which indicates fresh product material.

One can see a significant difference in $^{234}$Th/$^{230}$Th activity ratios between non-processed ore, tails and product, and between the fresh and old tails. By using sample 97-01 $^{234}$Th/$^{230}$Th ratios measured in July (non-equilibrium ratio of 0.43±0.06) and in December (equilibrium ratio of 0.08±0.01), one can estimate the age of the fresh tails. The estimated age is equal to 34±6 days at the time the non-equilibrium sample was analyzed (10 July ). Therefore, the estimated date the fresh tails were produced is about 6 June.

The $^{234}$Th/$^{230}$Th activity ratios, measured by HRGS in uranium mining samples provide the inspector with information on the type of material measured (non-processed ore, uranium product or tails) and can be used for the verification of the operator data on the date of ore processing. A portable gamma spectrometer with a planar HPGe detector can be used to measure the $^{234}$Th/$^{230}$Th activity ratio in the field.