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Applying Nuclear Forensics in Nuclear Material Accounting and Control of Radioactive Material out of Regulatory Control from a Facility

Nuclear forensic has found a niche in Nuclear Security and a tool for attribution evidence. This is normally applied when nuclear material gets out of regulatory control and then interdicted. Nuclear Forensics provide a Laboratory technique for material accounting and control. The signatures can be in the form of Nuclear material age dating, Lead isotopic ratios, Nuclear material processing mechanism (impurity data), sample morphology or a combination of at least any two of the above. At the Center for Applied Radiation Science and Technology, we used both Gamma spectrometry (High purity Germanium Detectors) and ICP-MS (NexION 2000), to resolve the nuclear forensics signatures of a Uranium Ore Concentrate (Yellow cake). The aim of the research was to determine the nuclear forensics signatures of a yellow cake powder for attribution. The objective is to provide a systematic validated Nuclear Material accounting and control that answers to the question of interdicted materials out of Regulatory control.

First recovery processing water yellow cake production stage of the Processing Plant was analyzed by ICP-MS Uranium Provenance and age of the ore sample before crashing. Results show that the Recovery Process water from has a Uranium ranging from (145.68 \pm 33.43) mgL-1 (1.81 \pm 0.41) kBqL-1 to 239.54 \pm 22.78 mgL-1 (2.97 \pm 0.28) kBqL-1), at the Processing plant. Thorium levels were at mean concentration of (16.88 \pm 2.48) µgL-1 (0.21 \pm 0.03) BqL-1.

The absence of Ca, Mg, Al, Mn, Fe, Na, Zn and Cd shows that the Mine does not utilize acid leaching as its uranium processing mechanism (leaching). In acid leaching, the oxidation of the uranium compounds is typically achieved using manganese dioxide (MnO2), sodium chlorate (NaClO3), and Fe(II) salts. The major impurities are iron, thorium, phosphate and rare earths, molybdenum as well as zirconium. Other major impurities include thorium, vanadium, phosphate and sulphate.

The high levels of Vanadium with a mean value of (125.88 ± 0.68) mgL-1; Cr, As (0.384 ± 0.038) mgL-1, Se (0.030 - 0.096) mgL-1, and Mo $(0.471 \pm 0.030$ mgL-1) indicates that the Mine operators used alkaline leaching followed by hydrogen peroxide to remove the high concentrations of Vanadium and Molybdenum. Selenium and Uranium increased linearly (strongly correlated) with the Sample ID

Then we determined the lead isotopic ratios of the yellow cake from the same batch of samples, which was then used to calculate the age of the original sample and found to be around 1.3 Ga yrs. This is expected as the yellow cake investigated was from an Open pit uranium mine, which are characterized by younger ore deposits.

Finally we evaluated the REE Signature patterns from the two process stages (Ore and yellow cake). Results show that in the ore the REE patterns are all the LRRE are depleted by the Acid processing mechanism of the mine, except Ce which is much high. The HREE are however slightly enriched towards Lu, showing very minimal fraction of the HREE due to processing. The observed deviations may be due to heterogeneous distribution of REE in different mineralogical phases (such as phosphates) in some CI-Chondrites. We concluded that

i. analyzing the recovery process water gives distinct indicators of impurities that are in the ore. Hence the solvent extraction process can be decided upon (eg. alkaline leaching followed by hydrogen peroxide),

ii. Calcination of the Uranium precipitate needs to be done to remove Vanadium and other similar impurities.iii. at least three different nuclear forensics signatures could be clearly resolved by ICP-MS, viz: Processing mechanism, REE patterns and the age of the samples.

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