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Investigation of the ^{236}U/^{238}U isotope abundance ratio in uranium ores and yellow cake samples

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Summary. Uranium ores and yellow cake samples of known geographic origin were investigated for their $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ isotope abundance ratio. Samples from four different uranium mines in Australia, Brazil and Canada were selected. Uranium was separated by UTEVA® Resin and was measured by Accelerator Mass Spectrometry (AMS) at the Vienna Environmental Research Accelerator (VERA). The measurement of the isotope abundance ratio $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ will be used to investigate possible correlations between the original mineral (uranium ore) and the intermediate product (yellow cake). Such correlations are useful indicators for nuclear forensic or for non-proliferation purposes.

1. Introduction

Illicit trafficking of nuclear and other radioactive material is a subject of serious concern due to the radiological hazard and the proliferation risks associated with such material. Nuclear forensics is a scientific discipline interfacing law enforcement, nuclear science and non-proliferation. Through nuclear forensic analysis, information on the history and on the potential origin of intercepted nuclear material can be obtained. The methods being used essentially focus on the measurements of the isotopic composition of the nuclear materials, the chemical composition, physical sample properties as well as their structure and microstructure. The nuclear forensic signatures established so far are the best suited for identifying final products (*e.g.* pellets) while attributing intermediate products (*e.g.* yellow cakes) poses more challenges due to the lack of reference data [1]. Additionally, studies were performed to evaluate the applicability of chemical impurities of nuclear material as a characteristic signature for providing hints on the origin of the material [2]. This work focused on the determination of the isotopic ratio $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ in uranium ores and uranium ore concentrates ("yellow cakes") from Australia, Brazil and Canada. Uranium ores differ significantly in their chemical composition. This allows the pattern of chemical impurities to be used as characteristic signature. During ore processing, however, the accompanying elements are removed to a large extent and thus the respective signature tends to be wiped out. Uranium mining involves the removal from the ground of large quantities of ore containing uranium and its decay products. Depending on the geological specifics and on local environment conditions, mining can be carried out through either open pit, underground (main techniques) or in situ leaching. Open pit mining is used on ore bodies lying near to the surface. Underground mining becomes necessary for deposit depths of 50–200 m and more. In situ leaching does not require the removal of solid ore from the ground (compared to open pit and underground). For removing the uranium from the ore different leaching solutions can be used depending on the matrix composition of the rock [3, 4]. Also for the subsequent chemical concentration and purification of the uranium different flow charts (involving different chemical processes) are in place. The final product is colloquially called "yellow cake". Most frequently this term describes ammonium diuranate $[(\mathrm{NH_4})_2\mathrm{U_2O_7}]$, it may also be ammonium uranyl carbonate, uranium peroxide or uranium oxide. It contains 65–70% uranium and the radioactive uranium daughters have been removed to a large extent. Their concentrations in yellow cakes, however, depend on the chemical and physical processes used for uranium extraction and drying. Concentrations ranging from 0.06 to 5.3% depending on the radioisotope and chemical procedure (Momeni *et al.* [5] and references therein) have been observed. This intermediate product is further purified in order to obtain a final pure product of $\mathrm{U_3O_8}$ (> 99.98% pure) [6].

^{236}U with a half-life of 2.3×10^7 years [7] is produced *via* thermal neutron capture on ^{235}U. In an ore, the neutrons are produced by spontaneous fission of ^{238}U and by (α, n) reactions on light elements (*e.g.* F, Na, Mg and Al) [8]. Due to its low isotope abundance in nature [9], ^{236}U can only be measured by sophisticated mass spectrometry techniques. Isotopic ratios of $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ have been measured previously in several uranium ores [8, 10–16]; the obtained ratios vary between 10^{-11} and 10^{-9} (the latter, relatively high, value was found in ore samples from one of the natural fission reactors in Gabon [10]).

The present investigation focused on the uranium fraction in order to evaluate the possibility of using the minor

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abundant uranium isotopes as a nuclear forensic indicator. Reportedly, the $n(^{235}\mathrm{U})/n(^{238}\mathrm{U})$ ratio shows only extremely small variations in nature and is thus not well suited as indicator. The $n(^{234}\mathrm{U})/n(^{238}\mathrm{U})$ shows a larger variability, yet a number of coinciding values between different mines were observed [10]. The $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ ratio may be a similar marker of an uranium ore as the $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ depends very much on the actual characteristics of the ore body (U concentration, light element concentration, water content, *etc.*) an even larger variability between mines is to be expected. But on the other hand, it cannot be excluded that the same or similar ratios are achieved from different ore deposits. Up to now uranium ores were measured by several research groups [8, 10–16]. Coinciding values of $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ ratios were measured for ores originating from Cigar Lake and from Joachimsthal, while pitchblende from Czech Republic were similar but pitchblende from Italy showed different results (up to one order of magnitude). Furthermore it is known from the literature that the production and thus the occurrence of *e.g.* ^{36}Cl and ^{239}Pu (both produced like ^{236}U *via* neutron capture) is locally different and depends on the composition of the ore. It has been demonstrated in [17, 18] that due to the ore characteristics the production parameters of the rare isotopes can change within a single borehole.

Therefore yellow cakes appear much more representative of a total ore body than a single rock sample because typically, several tons of samples are used to produce a batch of yellow cake. Consequently, differences between these uranium ore concentrates are more important as they are less prone to inhomogeneity.

As mentioned above, the $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ ratio is fairly small and so a sophisticated mass spectrometric measurement technique like accelerator mass spectrometry (AMS) is necessary. The detection limit for $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ with conventional mass spectrometry is $\sim 10^{-10}$ (*e.g.* TIMS) [11]. AMS is the method of choice because of its very high abundance sensitivity and its ability to suppress background signals arising from molecular interferences. Only few specialized AMS facilities [8, 13] are set up for measuring natural uranium. This study was carried out at the VERA facility and addresses the main question whether the ore and the intermediate product, the yellow cake, show the same isotopic ratio. To this end, four pairs of samples (uranium ore and yellow cake, *i.e.* feed and product of the same milling facility) were studied by AMS. The uranium concentration of the yellow cake samples is $\sim 76\%$ and also the grade of the ores does not show large variations (*i.e.* 0.1–0.5% in general). According to Richter *et al.* [10] uranium ore samples comparable to ours from Rabbit Lake, Ranger and Olympic Dam were measured by thermal ionisation mass spectrometry (TIMS) and the obtained $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ isotopic ratios were in the range of 10^{-10}–10^{-8}.

2. Material and methods

For the chemical sample preparation all chemical reagents were prepared using deionized Milli-Q (18 MΩ cm) water (Millipore, USA). 65% Suprapure $\mathrm{HNO_3}$ was further purified by sub-boiling. To prevent cross-contamination (with natural uranium) during the chemical treatment, all laboratory equipment was leached with 10% sub-boiled $\mathrm{HNO_3}$ at least for one day and washed three times with Milli-Q water, subsequently dried in a laminar flow bench and stored in clean zip bags. 30% HCl, 30% $\mathrm{H_2O_2}$ and 25% $\mathrm{NH_3}$ were of Suprapure grade. The procedure for the separation of uranium in ores and yellow cakes using UTEVA® Resin is available from Eichrom Technologies, Inc. [19] and was modified to our needs. After leaching the uranium ore for 3 h in 8 M $\mathrm{HNO_3}$, the solution was filtered through a blue ribbon filter paper. The sample was evaporated to dryness, fumed three times with $\mathrm{HNO_3/H_2O_2}$ and the residue was taken up in 3 M $\mathrm{HNO_3}$. The yellow cake samples could be dissolved in 8 M $\mathrm{HNO_3}$ with gentle heating. Then the solution was taken to dryness and dissolved in 3 M $\mathrm{HNO_3}$. The chromatographic column was loaded with 1.6 ml UTEVA® Resin (Eichrom Technologies, Inc.) and pre-conditioned with 3 M $\mathrm{HNO_3}$. The sample solution was transferred to the column, washed with 3 M $\mathrm{HNO_3}$ and 5 M HCl to remove Th and finally U was eluted with 1 M HCl in a 50 ml vial. This solution was evaporated gently to dryness, taken up in 2 ml conc. HCl and the wall was washed with 7 ml deionised water. Finally, 1 ml $\mathrm{Fe^{3+}}$ solution containing 1 mg Fe/ml was added. By slowly adding 25% $\mathrm{NH_3}$ uranium was co-precipitated with $\mathrm{Fe(OH)_3}$. The solution was heated gently on a hot plate for about 15 min; the pH was reduced to 9. After centrifuging, the precipitate was dried at 100 °C in an electric furnace for about 2 h. The dried precipitate was calcined at 800 °C in small Quartz tubes. The resulting oxides samples were pressed into aluminium sample holders suitable for the ion source of VERA. Blank samples were prepared in parallel to the actual ore and yellow cake samples in order to verify the absence of tracers of uranium in the reagents or in the laboratory ware.

The VERA setup as used for actinide measurements is described in Vockenhuber *et al.* [20] and Steier *et al.* [21]. The uranium is sputtered with a $\mathrm{Cs^+}$ beam, and in a first analysis $^{236}\mathrm{UO^-}$ (mass ~ 252 amu) is selected; this ion is accompanied by molecular isobars, most abundant $^{235}\mathrm{U}^{16}\mathrm{OH}^-$. While conventional mass spectrometry (ICP-MS) cannot suppress this background sufficiently, AMS destroys the molecules by acceleration to high energies (3 MeV at VERA) and stripping to a high positive charge state in a dilute gas cell. After another stage of acceleration, $^{236}\mathrm{U}^{5+}$ at 18 MeV is selected in a second mass analysis. The ions are detected in a time-of-flight spectrometer and a final ionization chamber. Other ions can reach the detectors only on irregular trajectories, *e.g.* due to charge exchange on residual gas or scattering on surface in the spectrometer. VERA is optimised to suppress the most common types of such background [20]. However some complex processes – despite having very low possibilities – can be significant at the very low abundance of natural ^{236}U (10^{-10} and below). One such background, $^{235}\mathrm{U}^{5+}$ from $^{235}\mathrm{U}^{16}\mathrm{OH}^-$, is discussed in [21]. The occurrence of such background depends on the chemical composition of the sputter material. While generally AMS is much less sensitive than other methods to the composition of the sample matrix, preparation of pure material prevents unexpected background. The time-of-flight and the particle energy measured by the detectors allowed in the cases observed so far to distinguish the background ions from $^{236}U^{5+}$; however, the occurrence of an unexpected chemical element at high concentration in the matrix might raise new background processes, requiring dedicated systematic investigations.

To allow the determination of the ratio $n(^{236}U)/n(^{238}U)$, the first mass spectrometer is switched to measurements of $^{238}U^{16}O^{-}$ once per second; this ion is measured as a beam current in a Faraday cup (typically 200 nA). Every five minutes $^{238}U^{16}O^{-}$ is injected into the accelerator, and the $^{238}U^{5+}$ current is measured in a Faraday cup in the second, high-energy mass spectrometer. By these means, the stripping yield of U^{5+} can be determined (typically 5%). The efficiency of the time-of-flight/ionisation detector (typically 25%) is determined using our in-house standard Vienna KkU ($^{236}U/^{238}U = (6.98 \pm 0.32) \times 10^{-11}$) [16].

3. Results and discussion

Ore samples from four different uranium mines (Olympic Dam, Ranger, Lagoa Real and Rabbit Lake) were analysed for their $n(^{236}U)/n(^{238}U)$ isotopic ratio (Table 1). Additionally, two further samples from the Ranger mine were investigated. The grade of the latter was approximately 0.35% and the three samples from Ranger mine were sampled independently over a period of several weeks. The ore from Olympic Dam is geologically described as polymetallic hematite breccia complex deposit. The ore is exploited by open pit mining. Ranger is an unconformity-Proterozoic fracture-bound deposit. The deposit consists of several ore bodies which were exploited subsequently. Lagoa Real is a metasomite; the ore is monometallic and the principal uranium mineral is uraninite. The deposit is exploited by open pit and underground mining. Rabbit Lake is also an unconformity-Proterozoic fracture-bound deposit. Several deposits in the area are exploited, mostly by open pit mining (with Eagle Point operating in underground mining). The mill (where the yellow cake is produced) accepts also ore from Cigar Lake.

The obtained values were all in the range of 10^{-11}–10^{-9} which can be still attributed to natural uranium. According to Richter *et al.* [10] an uranium ore sample from Rabbit Lake was measured by Thermal Ionisation Mass Spectrometry (TIMS) with a detection limit of 1.2×10^{-10}. The obtained $n(^{236}U)/n(^{238}U)$ isotopic ratio for Rabbit Lake was $(2.8 \pm 1.7) \times 10^{-10}$. Uranium ore samples from Ranger and Olympic Dam were investigated by Ovaskainen *et al.* [22] also *via* TIMS with an abundance sensitivity of few parts in 10^{-8} and the obtained values of the ratio were less than 1.45×10^{-8}. At the VERA facility, systematic investigations suggest a detection limit for $n(^{236}U)/n(^{238}U)$ below 5×10^{-12} for samples of 0.5 mg U [16]. Therefore more accurate results are to be expected. The samples from Australia have shown the lowest isotopic ratio while in Brazil (Lagoa Real) the ratio was two orders of magnitude higher. The obtained value with $(4.12 \pm 0.04) \times 10^{-9}$ is surprisingly high. Up to now only from the natural reactor near Oklo (Gabon) a ratio in this range (9.32×10^{-9}) is known [10]. But we observe for this uranium ore from Lagoa Real a very high ^{236}U count rate (compared to all other samples) which would suggest an anthropogenic contamination during the sample collection, sample storage, *etc.* However, an anthropogenic contamination during the chemical separation and sample preparation in our laboratories can be excluded because no significant blank contribution or cross-contamination could be noticed in the blank samples. The results from Ranger_2 to Ranger_4 show no significant differences (consistent within $2s$). The $n(^{236}U)/n(^{238}U)$ ratio measured in the Rabbit Lake ore sample is significantly lower than the values measured from the two Australian ores (Olympic Dam and Ranger mine). Yet, the value is in the same order of magnitude as previously measured by Zhao *et al.* [15] and Berkovits *et al.* [11] and their values were $(5.6 \pm 1.5) \times 10^{-10}$ and $(3.3 \pm 0.5) \times 10^{-10}$, respectively.

In Table 2 the $n(^{236}U)/n(^{238}U)$ ratios of the yellow cake samples are shown. All of them are at the level of few parts in 10^{-11}. The results of the chemically treated yellow cake sample fit with the order of magnitude of the isotopic ratio of the uranium ore (Table 1) with the exception of Lagoa Real. Here the yellow cake seems to be more reliable and confirm our suggestion of a contamination of the ore sample investigated above. The isotopic ratio of the uranium ore from Rabbit Lake is a factor of 2 higher than the ratios obtained from the yellow cakes (Tables 2 and 3). This can be explained by the fact that the mine exploits several deposits and the mill accepts also ore from other mines. This leads to an isotopic inhomogeneity of the material being processed. In consequence, the single sample of uranium ore which was investigated does not necessarily correspond to the yellow cake sample provided by the same facility. The same might be true for the uranium ore from Olympic Dam which shows a slightly smaller value than the yellow cake samples. For the samples from Ranger mine an excellent agreement of the $n(^{236}U)/n(^{238}U)$ ratios between the ore and yellow cake material can be noticed.

Complementary to the measurements described above (with chemical separation of the uranium prior to AMS measurement), the four yellow cakes were subjected to two

Table 1. Results of $n(^{236}U)/n(^{238}U)$ isotope ratio measurements in uranium ores. The measurement uncertainties are given in $\pm 2s$.

Sample No.	Mine	Country	$n(^{236}U)/n(^{238}U)$
73971	Olympic Dam_4	Australia	$(1.51 \pm 0.44) \times 10^{-11}$
73951	Ranger_4	Australia	$(1.92 \pm 0.28) \times 10^{-11}$
73950	Ranger_3	Australia	$(1.73 \pm 0.30) \times 10^{-11}$
73947	Ranger_2	Australia	$(1.76 \pm 0.30) \times 10^{-11}$
BR11	Lagoa Real	Brazil	$(4.12 \pm 0.04) \times 10^{-9}$
1425	Rabbit Lake	Canada	$(1.12 \pm 0.06) \times 10^{-10}$

Table 2. Results of $n(^{236}U)/n(^{238}U)$ isotope abundance ratio measurement in yellow cake samples (with chemical sample preparation prior to AMS measurement). The measurement uncertainties are given in $\pm 2s$.

Sample No.	Mine	Country	$n(^{236}U)/n(^{238}U)$
28	Olympic Dam	Australia	$(4.46 \pm 0.48) \times 10^{-11}$
24	Ranger	Australia	$(1.48 \pm 0.26) \times 10^{-11}$
15	Lagoa Real	Brazil	$(3.09 \pm 0.38) \times 10^{-11}$
51	Rabbit Lake	Canada	$(5.17 \pm 0.48) \times 10^{-11}$

Table 3. Results of $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ isotope abundance ratio measurement in yellow cake samples (without chemical sample preparation prior to AMS measurement). The measurement uncertainties are given in $\pm 2s$. March 2010 and May 2010 denote two independent measurements at different times.

Sample No.	Mine	Country	$n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ March 2010	$n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ May 2010
28	Olympic Dam	Australia	$(3.76 \pm 0.52) \times 10^{-11}$	$(3.90 \pm 0.70) \times 10^{-11}$
24	Ranger	Australia	$(1.14 \pm 0.40) \times 10^{-10}$	$(1.30 \pm 0.78) \times 10^{-10}$
15	Lagoa Real	Brazil	$(2.66 \pm 1.30) \times 10^{-11}$	$(1.54 \pm 0.46) \times 10^{-11}$
51	Rabbit Lake	Canada	$(5.40 \pm 0.56) \times 10^{-11}$	$(4.70 \pm 0.48) \times 10^{-11}$

additional measurements (independent sample preparation, two separated beam times, Table 3) directly by AMS without any chemical separation of the uranium; the samples were only calcined at 800 °C and pressed into aluminium sample holders.

The $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ ratios as measured in the (unseparated) replicate samples from Olympic Dam and Ranger agree very well and also the isotopic ratios of the two independent measurements on the Rabbit Lake sample agree within the stated uncertainty. The results obtained for Lagoa Real suggest small differences though statistically not significant (Table 3). By comparing the separated uranium fractions with the untreated samples one can notice that all samples, except Ranger, are in good agreement. The isotopic ratios of the separated uranium fraction of Olympic Dam and Rabbit Lake agree very well with the directly measured samples. The value for $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ from the chemically treated Ranger samples is an order of magnitude lower than the directly measured samples. This could be explained by an isobaric interference arising from $^{232}\mathrm{Th}$; a significant current of $^{232}\mathrm{Th}^{16}\mathrm{O}^-$ (about one percent of the $^{238}\mathrm{U}^{16}\mathrm{O}^-$ current) was indeed observed in the first mass spectrometer of VERA for the Ranger samples without chemical treatment. Further investigations would be required to rule out that $^{232}\mathrm{Th}$ can interfere with the $^{236}\mathrm{U}$ measurement.

4. Conclusion

The $^{236}\mathrm{U}$ production in uranium ores depends on the neutron flux as well as on the probability of neutron capture by uranium isotopes. The main sources of neutrons are the spontaneous fission of $^{238}\mathrm{U}$ and (α, n)-reactions on light elements (*e.g.* Li, Be, Na, etc). The presence of water and neutron absorbers like Gd and Sm influence the build up of $^{236}\mathrm{U}$ [14]. In consequence, a mine-to-mine variability of the $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ ratio should be expected and it might serve as candidate parameter for tracing the origin of natural uranium. The set of uranium ore samples we studied by means of AMS measurements indeed showed a mine-to-mine variability of the $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ ratio. Uranium ores are, however, not homogeneous and analysing one ore sample per mine results only in a local isotopic composition. For one mine (Ranger), three different samples were investigated and they agree within $2s$. Four pairs of samples (uranium ore and yellow cake, *i.e.* feed and product of the same milling facility) were studied by AMS for their $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ isotope abundance ratio and the possible usage of $^{236}\mathrm{U}$ as an indicator for establishing a relation between the ore and the yellow cake. In this study, yellow cake samples with and without chemical treatment prior to AMS measurements were investigated. The isotopic ratio $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ of the yellow cake samples after the chemical separation reflects a typical profile (average value) of the yellow cake sample and isobaric interferences are not to be expected. The $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ ratio obtained on the samples without chemical treatment (prior to measurement) confirm those observations, with the exception of the Ranger material. The latter is apparently suffering from an isobaric interference, which increases the $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ ratio by one order of magnitude. Comparing the $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ ratios measured in the ore samples to those obtained for the respective yellow cake samples, we observe a good correlation only for the materials from Ranger mine. The three other sample pairs do not correlate. This observation, though obtained for a small sample population only, suggest that for tracing the origin of natural uranium, yellow cake samples should be taken as starting point rather than uranium ores. This eliminates to some extent the variability arising from inhomogeneity in the ore body, which reflects in the $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$. Moreover, the fact the uranium mills may process material from various deposits and often accept ore from different mines leads to blending of material and in consequence to an average value of the isotope ratio.

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