

DEPLETED, NATURAL AND LOW ENRICHED URANIUM VERIFICATION BY RECENT PORTABLE PASSIVE NON-DESTRUCTIVE ASSAY TOOLS

Sayed A. El-Mongy¹ and Humaid A.I²

*1- On-leave from Atomic Energy Authority, Cairo, Egypt
2- Main Chemical Lab., Armed Forces, United Arab Emirates*

Rec. 23/1/2007

Accept.22/7/2007

Employment and use of recent methods for verification of nuclear materials are milestone and key point for effective domestic and international safeguards.

Modern non-destructive assay (NDA) techniques based on CdZnTe, NaI and HpGe have been used in this work to verify depleted, natural and low enriched uranium. The ^{235}U activity of the samples was calculated based on the count rate of the 185.7 keV photopeak and compared with the certified values. The γ -lines 143.8 and 163.4 keV showed good correlation with the ^{235}U activity in the investigated samples. The ^{238}U and ^{234}U activity were also measured and calculated. The ^{235}U enrichment percentages (%E) of these samples were then calculated and found to be 0.58%, 0.72% and 1.57% respectively. The $^{235}\text{U}/^{238}\text{U}$ activity ratios of the depleted and low enriched samples were found to be sharply correlated with %E. Traces of the anthropogenic origin ^{236}U was identified by measurement in two of the verified samples. In spite of the $^{234}\text{U}/^{238}\text{U}$ ratio was almost one in the case of depleted and natural uranium, disequilibrium between ^{238}U and its daughter ^{234}Th was observed.

INTRODUCTION

The international atomic energy agency (IAEA) operates a large diversity of equipment to verify the nuclear materials in Member States within its mandate given by the non-proliferation treaty (NPT). Additional analytical capabilities for environmental samples and effective NDA methods are indispensable for the agency in its efforts to be able to detect undeclared nuclear materials and activities and to support Member States in the detection of and response to illicit trafficking [1,2].

The enrichment process creates small quantities of the man-made isotopes ^{236}U and ^{239}Pu . These isotopes are included in the depleted uranium mass as it too expensive to extract them [3]. Non-depleted uranium is uranium with $^{238}\text{U}/^{235}\text{U}$ isotopic ratio

comparable to natural uranium but having quantities of ^{236}U and presumably plutonium [3]. As a matter of fact, the presence of ^{236}U in a sample can also be used as a tracer for identifying uranium containing reprocessed fuel or uranium mixed with reactor by-products [3,4]. The lighter ^{234}U is proportionately enriched even more than ^{235}U by mass-based enrichment process. Highly enriched ^{235}U (HEU) typically contains 1.5 – 2.0% of ^{234}U [4].

This work deals with analysis of depleted (DU), natural (NU) and low enriched uranium (LEU) by recent portable non-destructive assay techniques based on the semiconductor CdZnTe and HpGe and scintillation NaI spectrometers. Origin and enrichment verification of these samples are also vital objectives to be achieved by this work.

EXPERIMENTAL WORK

Three samples (Analytics-USA) of 0.53 μCi , 0.57 μCi and 0.95 μCi were used in this work. They are U_3O_8 dissolved in nitric acid. The sample density is 1.15 g/cm^3 . The geometry of the assayed samples has the form of 500ml Marinilli beaker.

Portable Canberra HpGe with the Inspector 2000 Model 1N2K-USA and GenieTM2000 spectroscopy software was used for acquiring and analyzing the sample spectra through 16K Multichannel analyzer. The system has 52% relative efficiency and resolution of 0.91 FWHM at 122 keV of ^{57}Co . Due to its very thin Be window it has energy response down to $\sim 5\text{keV}$.

Portable semiconductor 10x10x2 mm CdZnTe model ICS-4000 -USA with ICS Explorer software was also used to identify both ^{238}U and ^{235}U in the samples. The system has 100% efficiency at low energies and 1.6% resolution at 661.6 keV of ^{137}Cs . This system is efficiently used for identifying radioactive isotopes in special nuclear materials (SNM) and naturally occurring radioactive materials (NORM).

Portable surveillance and measurement system SAM935 based on 3"x3" NaI with QuantumTM (quantitative analysis) software was used for analysis of the abovementioned samples. The system has 7.5% resolution at 661.6 keV of ^{137}Cs .

RESULTS AND DISCUSSION

The count rates of the gamma line 185.7 keV (57.5% γ -intensity) which is a direct function in ^{235}U enrichment were measured by using the three abovementioned systems. The results are presented in Figure 1.

As a matter of fact, the results obtained by the HpGe spectrometer are more reliable than those obtained by CdZnTe and NaI. This is mainly due to its distinguished resolving power.

Based on the count rate of the 185.7 keV, ^{235}U activity in the samples was calculated by using the following equation:

$$A_{sa} = A_{st} \cdot (C_{sa} / C_{st}) \tag{1}$$

where A_{sa} and A_{st} are ^{235}U activity of the sample and standard. The C_{sa} and C_{st} are the count rate (HpGe measurements) in the 185.7 keV γ - photopeak of ^{235}U of the sample and standard. The results are given in Table 1.

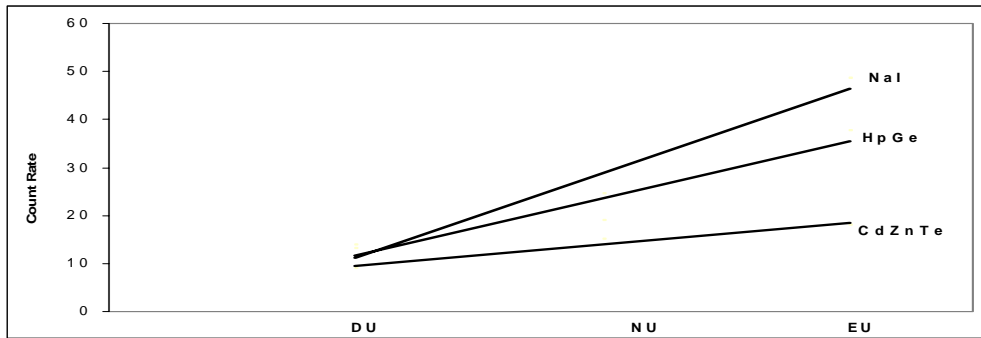


Figure 1. Analysis of the 185.7 keV of ^{235}U by CdZnTe, HpGe and NaI

Table 1. Results of ^{235}U activity calculations for DU, NU and LEU samples

Sample	Certified ^{235}U activity	Calculated ^{235}U activity	% diff.
DU	373.3 Bq	355.6 Bq	4.7%
NU	482.8 Bq	499.2 Bq	3.4%
LEU	994.0 Bq	961.3 Bq	3.3 %

As it is shown from the table, the bias between the measured and declared values does not exceed ~5%.

The enrichment percentage can also be identified and deduced by using the other ^{235}U γ -lines; 143.8 keV (10.9% γ -intensity) and 163.4 keV (5% γ -intensity). The measured count rates of these two γ -lines due to DU, NU and LEU samples are presented in Figure 2.

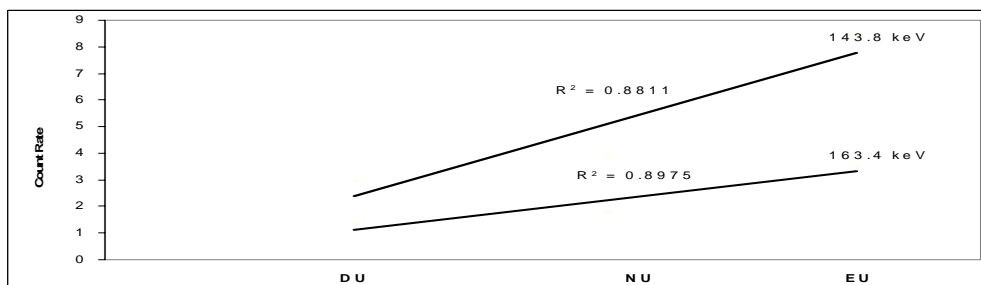


Figure 2. The count rates of ^{235}U based on its 143.8 keV and 163.4 keV γ -lines

The ^{235}U activity (A_{235}) calculations based on its 143.8 and 163.4 keV γ -transitions were carried out and given in Table 2.

Table 2. Results of ^{235}U activity (in Bq) calculations of the verified samples

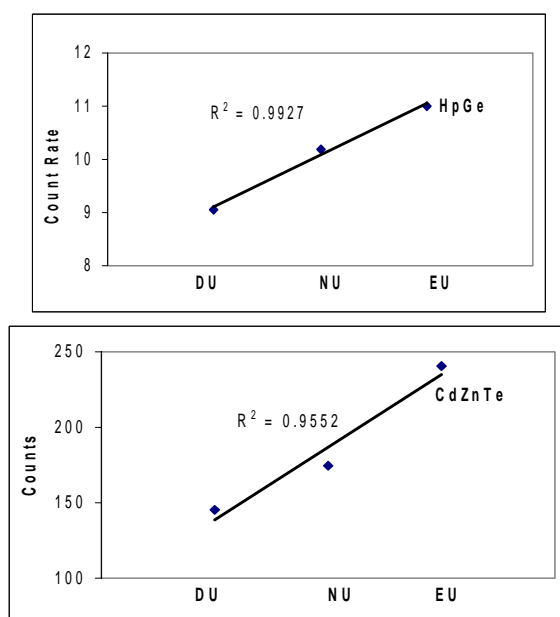
Sample	Certified A_{235} (Bq)	Calculated A_{235} (143.8 keV)	% diff.	Calculated A_{235} (163.4 keV)	% diff.
DU	373.3	362.4	2.9	362.1	3.0
NU	482.8	497.3	3.0	508.3	5.3
LEU	994.0	1024.6	3.1	944.1	5.0

The characteristic 16.2keV x-ray emission is used to identify uranium in unknown samples. The nuclear data of this x-ray line are given in Table 3.:

Table 3. The nuclear data of the 16.2 keV x-ray line [5]

Isotope	Energy (keV)	Intensity (%)	Assignment
^{234}U	16.2	4.1	ThL $_{\beta}$
^{235}U	16.2	14	ThL $_{\beta}$
^{236}U	16.2	3.7	ThL $_{\beta}$
^{238}U	16.2	5.2	ThL $_{\beta}$

The counts measured by HpGe and CdZnTe of the 16.2 keV x-ray of DU, NU and LEU samples are presented in Figure 3. It should be mentioned that the same x-ray line was not identified by NaI detector due to its relatively bad resolution and energy range response.

**Figure 3.** The analysis of the 16.2 keV x-ray of DU, NU and LEU samples

The ^{238}U activity (A_{238}) in the samples was calculated based on the count rate in the 16.2 keV peak and found to be as given in Table 4.

Table 4. Results of ^{238}U activity calculations by the 16.2 keV x-ray line

Sample	Certified A_{238} (Bq)	Calculated A_{238}	%diff.
DU	1.008×10^4	0.93×10^4	7.7
NU	1.045×10^4	1.10×10^4	5.3
LEU	9.801×10^3	11.3×10^3	15.3

The highest difference 15% is assigned to the enriched uranium sample. This may be due to the contribution of the elevated intensity of the 16.2 keV x-ray of ^{235}U .

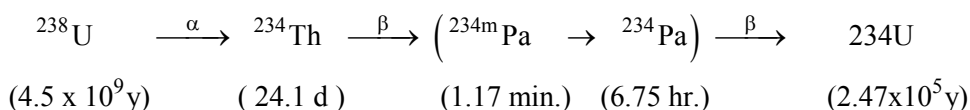
The $^{235}\text{U}/^{238}\text{U}$ activity ratio of the analyzed samples was then deduced from ^{235}U and ^{238}U activity and found to be 0.037, 0.046 and 0.1014. These values are typical for depleted, natural and slightly enriched uranium. In general, the $^{235}\text{U}/^{238}\text{U}$ ratio can be used as a signature indicator of anthropogenic uranium sources [6].

The isotope ^{234}U was identified in the samples through careful detection of its γ -lines 53.2 keV and 120.9 keV of 0.123% and 0.034% γ -intensity respectively. The ^{234}U activity (A_{234}) of the samples was calculated and compared with the certified values as given in Table 5.

Table 5. The calculated ^{234}U activity (in Bq) of the samples

Sample	Certified A_{234}	Calculated A_{234}	% diff.
DU	9.166×10^3	8.378×10^3	8.6 %
NU	1.024×10^4	1.0708×10^4	4.6 %
LEU	2.437×10^4	2.3347×10^4	4.2 %

It should be mentioned that ^{234}U is not an ancient survival from stellar nucleosynthesis and it is an indirect decay product of ^{238}U according to the following transformation chain [4];



The activity ratio percentages of ^{235}U to ^{234}U ($[A_{235}/A_{234}] \times 100$) were found to be 4.073%, 4.72% and 4.079% for DU, NU and LEU respectively. It is clear that both the DU and LEU samples have the same $^{235}\text{U}/^{234}\text{U}$ ratio.

The enrichment percentage (%E) was then calculated using the certified activity of uranium isotopes in the verified samples by the following equations:

$$A_{234} = N_{234} \lambda \tag{2}$$

$$A_{235} = N_{235} \lambda \tag{3}$$

$$A_{238} = N_{238} \lambda \tag{4}$$

where

$$N = (A \cdot T_{1/2}) / 0.693 \tag{5}$$

$$\%E = \{N_{235} / (N_{235} + N_{238} + N_{234})\} \cdot 100 \tag{6}$$

The A_{234} , A_{235} and A_{238} are the activity in Bq of ^{234}U , ^{235}U and ^{238}U in the samples. The N_{234} , N_{235} and N_{238} are the number of ^{234}U , ^{235}U and ^{238}U atoms and λ is the corresponding decay constants. The $T_{1/2}$ represents the half life time of ^{234}U , ^{235}U and ^{238}U in seconds. The results of enrichment calculations are given in Table 6.

Table 6. Results of the enrichment percentage calculations of the samples

Sample	Calculated N_{235}	Calculated (%E)
Depleted U	1.196×10^{19}	0.58
Natural U	1.547×10^{19}	0.72
Low enriched U	3.186×10^{19}	1.57

The calculations show that the analyzed U_3O_8 samples are depleted with 0.58 %, natural with 0.72 % and low enriched with 1.57 % of ^{235}U .

The calculated enrichment percentages (%E) were then depicted against the $^{235}\text{U}/^{238}\text{U}$ activity ratios as shown in Figure 4.

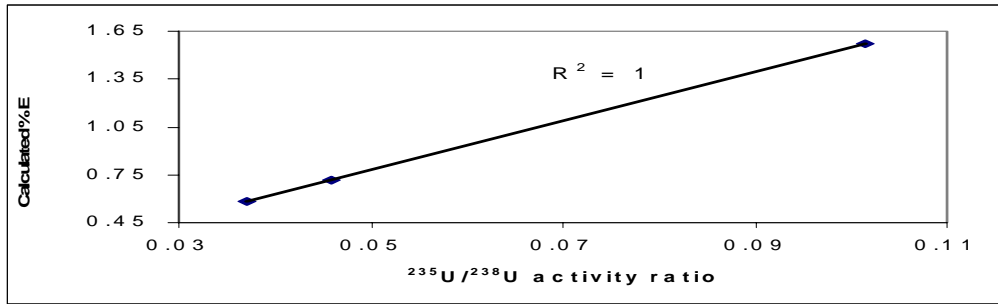
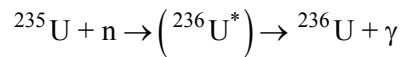


Figure 4. The correlation between the %E and $^{235}\text{U}/^{238}\text{U}$ activity ratios

This figure shows the strong correlation ($R^2 = 1$) between the calculated enrichment percentages and $^{235}\text{U}/^{238}\text{U}$ activity ratios of the samples.

Analysis of the depleted and low enriched uranium samples showed in trace count rate level the presence of the gamma line 112.8 keV (0.019% γ -intensity) of ^{236}U . The ^{236}U with half-life of 23.4 million years, is not found in nature in significant quantities. It produces as mentioned before in trace level from enrichment process. It also accumulates when uranium is exposed to neutrons in reactors according to the following radiative neutron capture; absorption without fission, by ^{235}U nucleus [3,4];



In this work, it was found that the ^{236}U activity in the DU and LEU samples is correlated to the ^{235}U activity concentration of the same samples. The ^{236}U activity of the samples was calculated based on the ^{235}U activity by the following equation;

$$A_{236}(\text{sa}) = A_{236}(\text{kn}) \cdot (A_{235}(\text{sa}) / A_{235}(\text{kn})) \quad (7)$$

where A_{236} (sa) and A_{236} (kn) are the ^{236}U activity of the sample and known. A_{235} (sa) and A_{235} (kn) are the ^{235}U activity of the sample and known. The result of calculations is given in Table 7.

Table 7. Results of ^{236}U activity calculations of the samples

Sample	Certified ^{236}U activity	Calculated ^{236}U activity	% diff.
DU	64.0 Bq	65.2 Bq	1.9%
LEU	173.5 Bq	170.4 Bq	1.8%

This table shows the low difference between the certified and calculated ^{236}U activity of the depleted and low enriched uranium samples. The presence of ^{236}U emphasizes that these samples are of anthropogenic origin.

The $^{235}\text{U}/^{236}\text{U}$ activity ratio of the DU and LEU was found to be almost the same; 5.83 and 5.73 respectively. This means that ^{236}U in the analyzed DU and LEU samples is directly proportional to the ^{235}U activity of the same sample. A sharp correlation ($R^2 = 1$) between the ^{235}U and ^{236}U activities was found and presented in Figure 6.

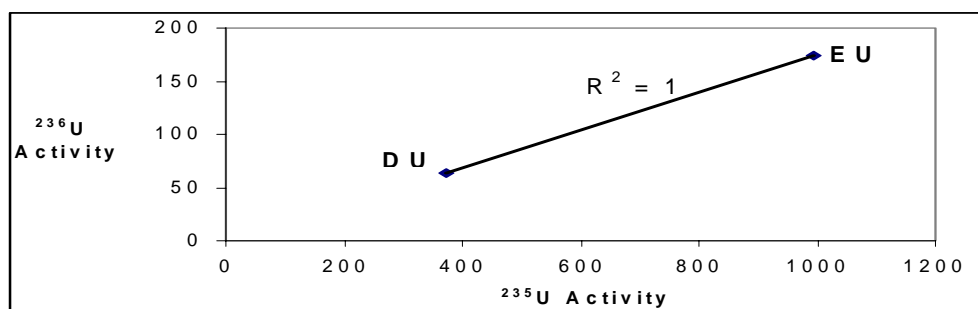


Figure 6. The correlation between ^{235}U and ^{236}U activities in the DU and LEU samples

The $^{236}\text{U}/^{238}\text{U}$ activity ratio was found to be 6.4×10^{-3} and 1.8×10^{-2} for depleted and low enriched uranium samples respectively. Recent work found this ratio greater than 2×10^{-5} [6]. They concluded that the $^{236}\text{U}/^{238}\text{U}$ ratio serves as a confirmation of contaminant even where the $^{235}\text{U}/^{238}\text{U}$ ratio appears normal [6].

The $^{234}\text{U}/^{238}\text{U}$ activity ratios were found to be 0.91 and 0.98 in the case of DU and NU, while it was 2.49 for LEU sample. In equilibrium, the $^{234}\text{U}/^{238}\text{U}$ activity ratio should be almost one. However physical differentiation process may result in disequilibrium in this ratio [6].

In spite of the observed equilibrium between ^{234}U and ^{238}U ($A_{238} \cong A_{234}$) in the case of DU and NU, the results of analysis based on the gamma lines 63.2 of ^{234}Th show disequilibrium between ^{238}U and its first daughter ^{234}Th . Almost similar count rates due to the 63.2 keV γ -transition of DU, NU and LEU samples were observed (Figure 7.).

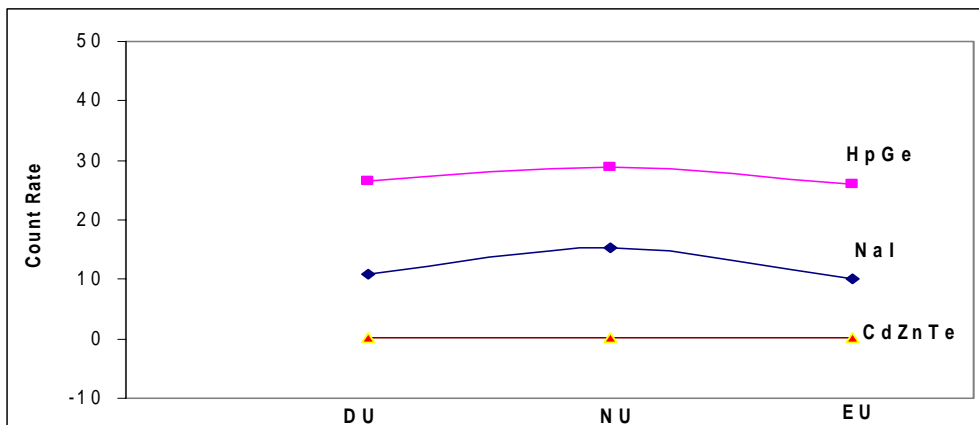


Figure 7. Results of analysis showing disequilibrium based on the 63.2 keV of ^{234}Th

This means that the investigated samples were freshly treated and prepared. The equilibrium between ^{238}U and ^{234}Th is usually achieved in 120 days (97%) to 168 days (99%) after chemical separation [7]. The difference in the chemistry of thorium and uranium leads them to be extracted from each other during separation, purification and preparation processes.

CONCLUSIONS

As a natural result of its distinguished resolving power, the HpGe analyses show reliable results more than those observed by CdZnTe and NaI. The declared and verified ^{235}U activities of the assayed samples are close to each other with 5% deviation. The enrichment percentage of the samples was accurately calculated based on the measured ^{234}U , ^{235}U and ^{238}U activities in the samples and assigned for depleted, natural and low (slightly) enriched uranium. Due to the presence of the ^{236}U in the investigated DU and EU samples, it was concluded that the samples are of anthropogenic nature. The $^{235}\text{U}/^{236}\text{U}$ activity ratio was found to be almost the same; 5.73 and 5.83 respectively. This ratio shows that ^{236}U is directly proportional to the ^{235}U in the same sample. It is also used as a tracer for identifying origin of enrichment process uranium and reprocessed uranium fuel. The 16.2 keV x-ray line was used for calculating of ^{238}U activity in the DU, NU and LEU samples. The results were found to be comparable with the declared values.

REFERENCES

- [1] Zendel, M., Khlebnikov, N., and Aparo, M., "Future safeguards Verification Tools", Symposium on international safeguards: Addressing Verification Challenges, Vienna, IAEA, 16-20 Oct. (2006).
- [2] "Safeguard Techniques and equipment", IAEA, *International Nuclear Verification Series No. 1* (2003).
- [3] Uranium medical research center, depleted and non-depleted Uranium, (2006) www.UMRC.net
- [4] MILNET Mirror: The nuclear weapons FAQ, Version 2.25: (Aug. (2001) www.MILNET Carey Sublette's nuclear weapons FAQ.htm – revised 2006
- [5] Chu S.Y.F., Ekstrom L.P., Richard B. Firestone, "Nuclear Data Dissemination Home page – Table of isotopes decay data", Lawrence Berkeley National Lab.(LBNL) isotopes project, March (2005).
- [6] Pottorff E.T., "Distinguishing Anthropogenic Uranium at the Rocky Flats Environmental Technology Site, Golden, Colorado", WM'04 Conference, WM-4035, Tucson, AZ, 29 Feb. – 4 March (2004).
- [7] Doug Reilly, Norbert Ensslin, Hastings Smith, Jr., and Sarah Kreiner, "Passive nondestructive assay of nuclear materials", NUREG/CR-5550, USA (1991).

التحقق من اليورانيوم المنضب والطبيعي ومنخفض الإثراء بطرق غير إتلافية وأجهزة محمولة حديثة

سيد علي المنجي , حميد اليماني

المختبر الكيميائي الرئيسي – القوات المسلحة – دولة الإمارات العربية المتحدة

يعد تطوير واستخدام تقنيات حديثة للتحقق من المواد النووية بمثابة حجر الزاوية ونقطة مفصلية في مجال الضمانات المحلية والدولية.
وفي هذا البحث تم استخدام أجهزة حديثة محمولة تعمل بكواشف الكاديوم زنك ويوديد الصوديوم والجرمانيوم بغرض التحقق من عينات يورانيوم منضب وطبيعي ومنخفض الإثراء.
هذا وقد حسب شدة النشاط الإشعاعي لليورانيوم 235 بالعينات اعتمادا على خطوطه الجامية المميزة له مثل 185,7 , 143,8 , 163,4 كيلو إلكترون فولت . وبالتحليل والحسابات تم تقدير قيمة تخصيب اليورانيوم 235 بالعينات ووجد أنها 0,58% , 0,72% و 1,58% . كذلك أظهرت النتائج عن وجود بقايا بتركيزات منخفضة من اليورانيوم 236 الانثروبولوجي الأصل في عينتي اليورانيوم المنضب ومنخفض الإثراء . كذلك أشارت النتائج من خلال رصد اللاتوازن بين اليورانيوم 238 وأول بناته الثوريوم 234 أن هذه العينات حديثة التحضير .