Development of Correlation for the Determination of $^{234}\text{U}/^{238}\text{U}$ atom ratio in Nuclear Fuel Samples

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Abstract

A correlation has been developed for the determination of $^{234}\text{U}/^{238}\text{U}$ atom ratio in Nuclear Fuel Samples using the atom ratios $^{234}\text{U}/^{238}\text{U}$ and $^{235}\text{U}/^{238}\text{U}$ determined by Thermal Ionization Mass Spectrometry. The relation is linear over the range of atom ratios of $^{234}\text{U}/^{238}\text{U}$ that would be encountered in the nuclear fuel samples. This correlation is extremely useful for the determination of $^{235}\text{U}/^{238}\text{U}$ in fuel samples using the cheaper alternative method such as alpha spectrometry for $^{234}\text{U}/^{238}\text{U}$ without resorting to the capital intensive Thermal Ionization Mass Spectrometric measurements.

Introduction

Determination of isotopic composition and concentration of Uranium (U) is very important in nuclear industry due to its strategic importance. Moreover, Low enriched uranium is used as a nuclear fuel in light water based reactors. The isotopic composition and the trace elemental composition in the nuclear fuel is an important step in the chemical quality control of the reactor fuel. The data on isotopic composition are necessary for obtaining the fissile content of the fuel sample.

Thermal Ionization mass spectrometry (TIMS) is a well recognized technique for the determination of isotopic composition and concentration of Uranium in a wide range of matrices. Other mass spectrometric techniques such as Inductively Coupled Plasma Mass Spectrometry (ICP-MS) can also be employed for the isotopic analysis of uranium. In the event of non-availability of Mass Spectrometer, other radiometric techniques such as Alpha Spectrometry and Gamma spectrometry can also be employed. These techniques have a limited applicability since the data on all the U isotopes cannot be obtained.

Development of isotope correlations is promising since it is possible to obtain the data on the different isotopes with minimal experimental effort and can also be used by employing simple experimental techniques such as alpha spectrometry. The correlations are also useful for obtaining the isotopic composition data when the amount of sample available is small, which is sufficient to obtain the data on major isotopes namely, $^{235}\text{U}$ and $^{238}\text{U}$ using mass spectrometry. Moreover, since the sample preparation steps are minimized in alpha spectrometry, these correlations can be employed for obtaining the data in a short time.

Experimental

The uranium samples were treated with concentrated nitric acid twice. The U sample was then loaded onto the sample filament, which is made of high purity Rhenium, of the double filament assembly of TIMS. The $^{234}\text{U}/^{238}\text{U}$, $^{235}\text{U}/^{238}\text{U}$ atom ratios were then obtained using a TIMS equipped with multi Faraday cup detection system, by employing static mode of multicollection.
Results and Discussion

Isotopic composition data of 20 samples were used for developing a correlation between $^{235}\text{U}/^{238}\text{U}$ Vs $^{234}\text{U}/^{238}\text{U}$ atom ratios. Fig-1 shows the correlation obtained between these $^{234}\text{U}/^{238}\text{U}$ atom ratio Vs Atom Percent Abundance of $^{235}\text{U}$. As can be seen from this figure, the correlation is linear over the range under study. The isotope ratio involving the minor isotope namely $^{234}\text{U}$, can be obtained with a reasonable accuracy of about 5%, which is promising especially when the sample available is small. These correlations are also quite useful, in instances, when the sensitivity of the mass spectrometer is not upto the mark, and hence the data on $^{235}\text{U}/^{238}\text{U}$ can only be obtained from the instrument, with a reasonably good precision and accuracy.

Since the alpha specific activity of $^{234}\text{U}$ is much higher in comparison with that of $^{235}\text{U}$ (~ $10^3$), it can be noted that this correlation can be used for obtaining the $^{235}\text{U}/^{238}\text{U}$ atom ratio using the $^{234}\text{U}/^{238}\text{U}$ alpha activity ratio which can be determined by alpha spectrometry. It is however to be noted that this method may have some limitations when the enrichment of $^{236}\text{U}$ is higher, which will result in the contribution from the alpha particles from $^{235}\text{U}$ isotope at the alpha energy peak of $^{238}\text{U}$ isotope.

The correlation once developed can then be employed for the determination of isotopic composition of U samples on a regular basis. Similar correlations can be developed using the data from dissolver samples and involving the other U isotope namely $^{236}\text{U}$. It may be noted that the data on minor isotopes $^{234}\text{U}$ and $^{236}\text{U}$ can be obtained with a reasonable accuracy using the secondary electron multipliers as detectors in TIMS. The correlations can then be used for checking the internal consistency of the data on U samples from reactors of similar type. The correlations are also useful in case of old TIMS instruments, in which, the adjustment of the Faraday cups is usually manual and can be affected due to the wear and tear of the instruments.

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Fig. 1 Correlation between $^{235}$U Atom Percent Abundance and $^{234}$U/$^{238}$U Atom Ratio in U Fuel Samples

\[ Y = B \times X \]

\[ B = 9741.3 \pm 256.5 \]

\[ R = 0.98 \]

\[ SD = 0.09 \]