A COMPARATIVE STUDY OF SOME NUCLEAR METHODS FOR $^{235}$U/$^{238}$U ISOTOPIC RATIOS DETERMINATION

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In the present work, a comparative study is made among nuclear methods for $^{235}$U/$^{238}$U ratios determination: activation analysis followed by high-resolution gamma-ray spectrometry, delayed neutron counting, passive gamma-ray and alpha spectrometry. Activation analysis followed by high-resolution gamma-ray spectrometry yielded a relative standard deviation down to 0.1% and a relative error of about 1% for standards of uranium enriched to 14%. Passive gamma-ray spectrometry using Ge(Li) detectors yielded a relative error down to 0.1% for enriched uranium and values even lower for the standard deviation. Passive gamma-ray spectrometry using Low Energy Photon Detector (LEPD) yielded a precision of 0.2% and a still better accuracy for enriched standards. In the case of alpha spectrometry, a relative error down to 0.5% and a precision of about 1% were obtained, also for enriched uranium standards. Delayed neutron counting allowed a relative standard deviation of about 7% and a relative error of about 2%, for standards of depleted uranium.

Introduction

The determination of $^{235}$U/$^{238}$U isotopic ratios is of great importance in nuclear technology. It is normally assumed that precise and accurate isotopic analysis of uranium has to be carried out by mass spectrometry. In nuclear laboratories, on the other hand, it can be more convenient and less expensive to use other available methods of analysis.

In the present work, a comparative study is made among some nuclear methods for $^{235}$U/$^{238}$U ratios determination, such as: activation analysis followed by high-resolution gamma-ray spectrometry, delayed-neutron counting, passive gamma-ray and alpha spectrometry.

The methods were studied by using uranium isotopic standards from the Compagnie Generale de Matieres Nucleaires (COGEMA) from Pierrelatte, France.

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The precision (in terms of the standard deviation) and the accuracy (in terms of the relative error) of the four methods studied are compared, as well as their range of applicability. Also a comparison is made with the results obtained by Mantel et al. /1/ and by John et al. /2/, who also worked with neutron activation analysis followed by high-resolution gamma-ray spectrometry.

Materials and methods

1. Thermal Neutron Activation Analysis Followed by High Resolution Gamma-ray Spectrometry

As was described by Mantel et al and by other authors /1-6/, thermal neutron activation analysis followed by high resolution gamma-ray spectrometry can be used successfully for the determination of $^{235}\text{U}/^{238}\text{U}$ isotopic ratios, by calculating the ratios between the areas of the peaks corresponding to the fission products of $^{235}\text{U}$ and to $^{239}\text{Np}$, formed by activation of $^{238}\text{U}$.

The precision of the method can be greatly improved by using the average of several peak ratios obtained in the gamma spectra. Lima et al /6/ have applied this procedure to the determination of $^{235}\text{U}/^{238}\text{U}$ ratios in rocks, in search for an "Oklo Phenomenon" in Brazil.

In the present work, the calibration curves constructed by linear regression from the peak ratios and from the values of enrichment of the standards were established by using uranium isotopic standards from the Compagnie Generale de Matieres Nucleaires, COGEMA (France).

The uranyl nitrate solutions of these $\text{U}_3\text{O}_8$ standards were prepared by dissolution with 1:1 HNO$_3$ and dilution with water, in order to obtain a final concentration from about 2 to 10 mg U/mL.