METHOD AND SET-UP FOR MEASUREMENTS OF TRACE LEVEL CONTENT OF HEAVY FISSIONABLE ELEMENTS BASED ON DELAYED NEUTRON COUNTING

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Abstract

Methods and set-up for measurements of trace level content of fissionable nuclides based on the delayed neutron counting technique are presented. It is shown that the electrostatic accelerator based method using the $^9\text{Be}(d,n)^{10}\text{B}$ reaction as a neutron source allows to determine the content of fissionable nuclides in ultra trace level (nano gram). New method for the determination of isotopic content of the sample is proposed. This method is based on the new systematics of the average half life of delayed neutron precursors for different fissioning systems.

Introduction

Availability of the method for the identification of ultra trace levels of fissionable elements (actinides) in samples of varied origins is of great importance for many area of applications. The commonly used methods of elemental analysis are the neutron activation analysis (NAA), neutron-induced prompt gamma-ray analysis (PGA), proton-induced X-ray analysis. The minimum detectable amounts are a function of many factors such as the source strengths, detector efficiency, geometry, sample quality, interfering reactions and other factors related to specific experiments. For instance reactor-based PGA method has the highest sensitivity that allows to obtain for thorium and uranium elements the detection limit approximately of ~ 1 mg/g [1]. At the present time the alpha spectrometry method with preliminary chemical separation of actinides is routinely used for the identification of these elements in trace levels but it is time consuming.

The method of elemental analysis based on the delayed neutron (DN) activity counting was estimated as having for thorium and uranium elements the minimal detectable amount ~50 $\mu$g [2]. However the developments of more reliable data base for DN parameters and utilization of high strength neutron sources make it possible to extend the DN counting technique to quantitative and qualitative analysis of fissionable elements in ultra trace level.

Experimental method and set-up for content determination of fissionable elements in samples

Experimental set-up primarily was designed and successfully used for the investigations of the delayed neutron yields from neutron induced fission of heavy nuclei [3]. The set-up was installed at the electrostatic accelerator KG-2.5 and has the following main parameters: ion (proton and deuteron) current - up to 500 $\mu$A, pneumatic sample delivery system - 150 ms and 1 s for 'fall down' sample delivery system, high voltage - up to 2 MV, neutron flux monitor - calibrated fission chamber, neutron detector - 30 boron counters embedded in the polyethylene moderator. Neutron detector efficiency is 0.084 with very low sensitivity to gamma-ray...
background of the sample under investigation. The intensity of the neutron background during delayed neutron counting period is about 0.008 counts/s per 1 µA of deuteron current in case of the (d, n) neutron production reactions.

The general equation for elemental analysis on the basis of the delayed neutron counting can be expressed as the following:

\[
N(t_k) = A \cdot \sum_{i=1}^{m} F_i \cdot \frac{a_i}{\lambda_i} \cdot (1 - \exp(-\lambda_i \Delta t_k)) \cdot \exp(-\lambda_i t_k) + B \cdot \Delta t_k , \tag{1}
\]

\[
F_i = (1 - \exp(-\lambda_i t_{ir})) \left( \frac{n}{1 - \exp(-\lambda_i T)} - \exp(-\lambda_i T) \left( \frac{1 - \exp(-n\lambda_i T)}{(1 - \exp(-\lambda_i T))^2} \right) \right) ,
\]

\[A = \varepsilon \sigma_f \varphi N_f \nu_d ,\]

where \(N(t_k)\) - the number of counts registered by the neutron detector in the time-channel \(t_k\) with time-channel width \(\Delta t_k\), \(\nu_f\) - the total delayed neutron yield per one fission, \(B\) - the intensity of neutron background, \(\lambda_i\) and \(a_i\) - the decay constant and relative abundance of \(i\)-th group of DN, \(n\) - the number of cycles, \(m\) - the number of DN groups, \(T\) - the duration of one cycle of measurements, which includes the irradiation and the delayed neutron counting time, \(t_{ir}\) - irradiation time, \(\varepsilon\) - efficiency of neutron detector, \(\varphi\) - the neutron flux, \(\sigma_f\) - fission cross section, \(N_f\) - the number of atoms of fissionable element (nuclide) under investigation.

Equation (1) and the value of parameters of the set-up allow to estimate the (detectable concentration) detection limit of fissionable elements (as well as minimal detectable amount) in the samples for the neutron source based on the \(^{9}\text{Be}(d,n)^{10}\text{B}\) reaction and deuteron ion current of 500 µA [4]. It was assumed that for the reliable analysis one needs to register one hundred delayed neutron counts above the background. The result of the estimation for thorium, uranium and plutonium elements are presented in the Table 1.

Table 1

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Minimal detectable amount *, g</th>
<th>Detectable concentration *, g/g</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Fast neutron flux</td>
<td>Thermal neutron flux</td>
</tr>
<tr>
<td>(^{235}\text{U})</td>
<td>6.3•10^{-6}</td>
<td>1.5•10^{-6}</td>
</tr>
<tr>
<td>(^{238}\text{U})</td>
<td>1•10^{-5}</td>
<td>2.6•10^{-6}</td>
</tr>
<tr>
<td>(^{239}\text{Pu})</td>
<td>1.7•10^{-5}</td>
<td>1.9•10^{-8}</td>
</tr>
<tr>
<td>(^{232}\text{Th})</td>
<td>1.7•10^{-5}</td>
<td>1.9•10^{-8}</td>
</tr>
</tbody>
</table>

*) Amounts which were obtained at the experimental conditions indicated in the text

**) Degradation of the neutron flux in the neutron slowing down process was taken into account.
Ten cycles of irradiation and delayed neutron counting were taken into consideration. The sample irradiation time was 100 s and the delayed neutron counting time was 25 s starting at 1 s after the end of irradiation. The total time spent for analysis was 1260 s. The estimation was made both for the fast neutron flux from the $^{9}$Be(d,n)$^{10}$B reaction at 2 MeV deuteron energy and for the thermal neutron flux which can be easily obtained by slowing down the neutrons from the neutron target. Degradation of the neutron flux during the neutron slowing down process was accounted for.

It is seen from the Table 1 that the set-up under discussion affords to determine the fissionable elements containing in the sample in trace level. Utilization of many cycles measurements leads to increasing the sensitivities of the analysis. Moreover in contrast to the gamma rays and alpha particles analysis methods the delayed neutron counting method has no restriction on the weight of the sample under investigation that also leads to increasing the sensitivity of the analysis based on the delayed neutron counting. The detectable concentration of fissionable nuclides was estimated for 500 g sample.

The combination of the fast neutron flux and the thermal neutron flux analysis allows to make the identification of the isotopic content of the sample.

**Method of identification of isotopic content of sample**

Until now the identification of isotopic content of the sample in the frame of the DN counting technique was based on the difference between the values of relative abundances of the definite DN group for different nuclides [5]. This method requires a high statistical accuracy of DN decay curve [6] and reliable data base for the DN group parameters (decay constants and relative abundances). The first condition is difficult to reach because of small amount of fissionable elements in the sample.

We propose another approach for the identification of isotopic content of sample which is based on the new systematic of the delayed neutron parameters [7]. According to this systematic the average half-life of the delayed neutron precursors for the isotopes of thorium, uranium, plutonium and americium elements can be presented by the following expression

$$\langle T_i \rangle = a_{1i} \cdot \exp \left[ (a_{2i} \cdot (-A_c - 3Z) \cdot A_c / Z) \right],$$

(2)

where index $i$ is related to the certain fissioning systems (thorium, uranium, etc.), $A_c$ and $Z$ - the mass number and atomic number of the fissioning nuclei respectively. The experimental data on the average half life parameters were obtained using the formula

$$\langle T \rangle = \sum_{i=1}^{6} a_i \cdot t_i,$$

where $a_i$ and $t_i$ are the relative abundance and period of the $i$-th DN group. The above expression (2) was presented in the form

$$\ln \langle T_i \rangle = a_{3i} + a_{2i} \cdot \left( (-A_c - 3Z) \cdot A_c / Z \right),$$

$$a_{3i} = \ln a_{1i},$$

(3)
and the appropriate delayed neutron data were analyzed for obtaining the values $a_{3i}$ and $a_{2i}$ on the basis of the least-square procedure. The results of the fitting procedure (solid lines) are shown in Fig.1. The obtained $a_{3i}$ and $a_{2i}$ values for each of the considered element are presented in Table 2. Thus all of the known isotopes can uniquely be identified by only one parameter - the average half life of the delayed neutron precursors. Therefore for the identification of the isotopic content of the sample one needs to make measurements and the least squares analysis of DN decay curve with the purpose to obtain the value of average half life parameter (for the mixture of nuclides). As compared with the six group parameters analysis [6] such analysis can be done using decay curves with much less statistical accuracy.

Table 2

<table>
<thead>
<tr>
<th>Element</th>
<th>$a_{3}$</th>
<th>$a_{2i}$  ($\times 10^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Th</td>
<td>-10.44 ± 0.23</td>
<td>12.94 ± 0.24</td>
</tr>
<tr>
<td>U</td>
<td>- 5.69 ± 0.21</td>
<td>7.65 ± 0.20</td>
</tr>
<tr>
<td>Pu</td>
<td>- 4.21 ± 0.60</td>
<td>6.10 ± 0.57</td>
</tr>
<tr>
<td>Am</td>
<td>0.38 ± 1.26</td>
<td>1.81 ± 1.17</td>
</tr>
</tbody>
</table>

Fig. 1. Systematics of the average half life of delayed neutron precursors
In case of presence of two nuclides in the sample the obtained value \( <T_{1,2}> \) is connected to unknown value of the fractional amount of the number of atoms of nuclides 1 and 2 by the following expressions

\[
<T_{1,2}> = \frac{(v_1 \sigma_1 \varphi_1 m_1 <T_1> + v_2 \sigma_2 \varphi_2 m_2 <T_2>)/(v_1 \sigma_1 \varphi_1 m_1 + v_2 \sigma_2 \varphi_2 m_2),}{m_1 + m_2 = 1},
\]

where \( v_1, v_2 \) - the total delayed neutron yields related to nuclide 1 and 2, \( \sigma_1, \sigma_2 \) - the fission cross section of nuclides 1 and 2, \( <T_1>, <T_2> \) - the average half life of DN precursors of nuclide 1 and 2, \( m_1, m_2 \) - the fractional amount of the number of atoms of nuclide 1 and 2 respectively, \( \varphi \) - the neutron flux through the sample.

In case of three nuclides in the sample with two of them which are fissionable by thermal neutrons (for example \(^{235}\text{U}, ^{238}\text{U}, ^{239}\text{Pu} \)) the combination of the fast neutron and thermal neutron flux analysis will give respectively the average half life values \( <T_{12}> \) and \( <T_{23}> \) for the mixture of nuclides which are connected to the fractional amount of the number of atoms of nuclides \( m_1, m_2, m_3 \) in the sample by the following expression

\[
<T_{1,2,3}>=\frac{(v_1 \sigma_{1f} \varphi_{1f} m_1 <T_1> + v_2 \sigma_{2f} \varphi_{2f} m_2 <T_2> + v_3 \sigma_{3f} \varphi_{3f} m_3 <T_3>)/(v_1 \sigma_{1f} \varphi_{1f} m_1 + v_2 \sigma_{2f} \varphi_{2f} m_2 + v_3 \sigma_{3f} \varphi_{3f} m_3),}{m_1 + m_2 + m_3 = 1},
\]

where \( \sigma_{1f}, \sigma_{2f}, \sigma_{3f} \) and \( \varphi_{1f}, \varphi_{2f}, \varphi_{3f} \) - are the fission cross sections and the neutron fluxes for thermal and fast neutrons respectively. \( <T> \) values for thermal and fast neutron induced fission of all fissioning system were assumed to be equal.

Conclusion

The DN counting technique coupled with the electrostatic accelerator based neutron source \(^9\text{Be(d,n)}^{10}\text{B}\) is a powerful instrument in performing the analysis of trace level content of fissionable elements in the samples of varied origins. The combination of thermal and fast neutron measurements and the analysis of the appropriate aggregate decay curves with the purpose to obtain the average half life parameters affords to extend the possibilities of the techniques to the identification of the isotopic abundances in the sample under investigation.

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References


