

to point out that, most of the time, solutions for measurement of trace are very dilute, this fact avoiding problems due to quenching and inner filter effect.

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## E15

### Neutron Activation Analysis: a Powerful Tool for Rare-earth Elements Assay in Terrestrial Materials

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The ability of rare-earth elements to act as indicators of physico-chemical processes in the environment has drawn a great interest on the knowledge of their abundance and distribution. This interest is growing with the availability of reliable data on rare-earth element concentrations.

Neutron activation analysis [1–4] is the most used analytical technique for the determination of the rare-earth elements in environmental matrices on account of its high sensitivity, of the possibility of determining all the elements and of the undisputed accuracy and reliability of its results. Both instrumental and destructive techniques may be applied but when the content of all the rare-earth elements is required, the use of destructive methods is compulsory.

Neutron activation analysis methods for rare-earth elements determination in different matrices have been developed at the University of Pavia using the 250 Kw TRIGA Mark II reactor [5]. In the present paper a critical review of both instrumental and destructive methods is presented as well as the indication of the best working conditions for irradiation, counting and radiochemical separations.

The 'optimized' procedures were then utilized in the determination of rare-earth element in standard reference materials of both mineral and biological origin to emphasize a widespread application of the method. The chosen materials were: SRM-278 'Obsidian rock', SRM-1632 'Coal', SRM-1633 'Fly Ash', SRM-1571 'Orchard Leaves' from U.S. National Bureau of Standard and Standard rock NIM-G (South Africa).

All these matrices were submitted to instrumental and radiochemical activation analysis for rare-earth elements determination.

The choice of the nuclear reaction, irradiation and decay times, and of the proper gamma radiation in counting are presented and discussed. The adopted radiochemical procedure is based on the separation of the rare-earth element group by fluoride precipitation, which allows the removal of  $^{46}\text{Sc}$  and alkaline, alkaline-earth radionuclides interfering in the high resolution gamma-ray spectrometry.

Results, given as the average of four or more independent determination and relative standard deviation, are reported and discussed. Precision of the methods can be deduced from the reproducibility of data, whereas accuracy is evaluated by comparison with certified information and literature values. Sensitivity limits under the described operational conditions are also reported. In some cases it is the first time that a rare-earth element content is reported, as for instance Pr, Dy, Ho and Er content in NBS-SRM 278 'Obsidian rock', Pr, Nd, Gd, Er and Tm content in NBS-SRM 1633 'Fly Ash' and Er content in NBS-SRM 1571 'Orchard Leaves'.

Trends and correlations among data are discussed as well.

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## E16

### Effect of Lanthanum(III) on Noninvasive Tumour Imaging with Gallium-67

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Gallium-67 citrate is the most widely used non-invasive, positive tumour imaging radiopharmaceutical [1]. Healthy organ uptake of the radionuclide in

addition to that in the tumour with commercial gallium-67 is however its serious drawback [2]. We have succeeded in avoiding this inconvenience by injecting gallium-67 made free just before its administration [3]. In this way we could image malignancies even in the liver which has not been reported earlier due to high concentration of the radionuclide in the healthy organ [1–3]. But, the preparation of free or loosely bound gallium-67 from commercial gallium-67 citrate is not easy because the composition of these solutions is not always the same [4]. Literature reports the administration of iron or iron complexes for improving the quality of tumour image with gallium-67 [5]. Although a more rapid tumour to blood radioactivity ratio is obtained by the administration of these substances, they invariably lead to reduction in the radionuclide uptake in the tumour and thus to low quality image. We have examined the effect of administration of low doses of lanthanum on the rate of elimination and on the tumour affinity for gallium-67.

Aqueous solutions of different salts of lanthanum(III) were administered in Morris hepatoma-3924A-bearing rats after the injection of commercial gallium-67 citrate solutions. Whole body scintigram of the rat was taken at different intervals of time after the administration of the radionuclide. After the animal died, the radioactivity in different organs was measured with a well-type gamma counter.

Lanthanum administration although favours the elimination of the radioactivity from the blood it does not improve the quality of the tumour image. The distribution of gallium-67 in lanthanum(III)-treated animals and those which did not receive lanthanum will be discussed.

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## E17

### Separation and Sequential Determination of Americium and Plutonium in Urine Samples

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Simultaneous determination of low levels of Americium and Plutonium in complex matrices has

always been difficult to carry out owing to the chemical behavior differences of the two actinides.

An analytical extraction and separation technique of the two actinides has been realized utilizing Dibutyl, N,N-diethyl carbamylphosphonate (DDCP) as a reagent. The possibility has been ascertained of performing a separation through partition chromatography techniques and through liquid–liquid extraction. In particular, such technique has been applied to the determination of the two actinides in urine samples from personnel exposed to the risk of internal contaminations due to emitter alpha radionuclides. In this case the liquid–liquid extraction technique has been adopted for reasons connected with the operation rapidity and ease.

This method involves: wet way sample mineralization by HNO<sub>3</sub> and evaporation concentration; liquid–liquid extraction of interesting actinides by DDCP from a 12 M solution in HNO<sub>3</sub>; sequential stripping of Am by HNO<sub>3</sub> 2 M and of Pu by HCl 3 M and HN<sub>4</sub>I 0.1 M; electrodeposition on a steel plate and final measurement through alpha spectrometry with surface barrier detector.

Average final yields obtained through this analytical method have been: 80% for Americium, 66% for Plutonium. Activity values found in 'urine whites' are comparable with the full-scale value. Min. detectable values at a 300 min count time have been the following: Americium  $1.3 \times 10^{-3}$  Bq/l (0.03 pCi/l), Plutonium  $1.6 \times 10^{-3}$  Bq/l (0.04 pCi/l). At present, experiments are carried out for applying this analytical method on other matrices as well.

## E18

### Actinide Behavior and Radiation Damage Produced by $\alpha$ -Decay in Materials to Solidify Nuclear Waste

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Solidification of High Level (radioactive) Waste, HLW, from reprocessing of nuclear reactor fuels in a suitable matrix and subsequent burying in a suitable repository are the envisaged final steps of the nuclear fuel cycle. Because of the long half-lives of some of the actinides (mainly Np, Pu, Am and Cm) and because of their decay by emission of an  $\alpha$ -particle (~5 MeV energy) and a heavy recoil atom (e.g. U from Pu-decay, ~100 keV energy), the actinide behavior in the candidate materials for waste solidification deserves particular attention. The materials for solidification are glasses or certain minerals, or a