NON-CONVENTIONAL MEASUREMENT TECHNIQUES
FOR THE DETERMINATION OF SOME
LONG-LIVED RADIONUCLIDES PRODUCED IN NUCLEAR FUEL
A LITERATURE SURVEY

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The results of a literature survey on non-radiometric analytical techniques for the determination of long-lived radionuclides are described. The methods which have been considered are accelerator mass spectrometry, inductively coupled plasma mass spectrometry, thermal ionization mass spectrometry, resonance ionization spectrometry, resonance ionization mas spectrometry and neutron activation analysis. Neutron activation analysis has been commonly used for the determination of $^{129}$I and $^{237}$Np in environmental samples. Inductively coupled mass spectrometry seems likely to become the method of choice for the determination of $^{99}$Tc, $^{237}$Np and Pu-isotopes. The methods are discussed and the chemical separation methods described.

In assessing the long-term environmental effects of nuclear power production, the disposal of radioactive waste in general and spent fuel in particular, a number of long-lived radionuclides have to be considered. As an example of the nuclides to be considered, Table 1 shows the most important long-lived radionuclides produced in nuclear fuel. Critical nuclides in most nuclear fuel disposal scenarios are one or several of the following of these nuclides: $^{79}$Se, $^{99}$Tc, $^{126}$Sn, $^{129}$I, $^{135}$Cs, $^{226}$Ra, $^{229}$Th, $^{237}$Np and $^{242}$Pu. It should be important to know the concentrations of these long-lived nuclides in spent nuclear fuel and other waste as well as in the environment.

In practice the interest in different nuclides varies considerably. $^{226}$Ra, $^{229}$Th, $^{230}$Th and $^{233}$U are only daughters of shorter-lived nuclides, their relative activities during the first 1000 years being small. The specific activities of $^{235}$U and $^{238}$U are low, their concentration in nuclear fuel high, and they are present everywhere in the environment. The amount of $^{234}$U in nuclear fuel can be calculated and its behaviour is the same as that of the element. One could imagine that it may be interesting to determine $^{79}$Se, $^{93}$Zr, $^{107}$Pd, $^{126}$Sn and $^{135}$Cs, but that seems not to be the case. For the above reasons this paper will mainly deal with methods for the determination of $^{99}$Tc, $^{129}$I, $^{237}$Np and the different Pu isotopes. $^{135}$Cs is also discussed because two references to its determination have been found.
The classical method for the determination of radionuclides involves measuring the radiation emitted by the nuclides in connection with their spontaneous decay. However, because of the low specific activity, and in some cases absence of α- or γ-radiation, other analytical methods may be more sensitive and specific. In this report non-radiometric methods for the determination of the above nuclides are described and compared with radiometric methods. The report is based on a literature study and some simple calculations.

### Determination of radionuclides

#### General

As already mentioned, radionuclides are traditionally determined by measuring the radiation they emit during disintegration. For short-lived nuclides this is the most sensitive method to use owing to their high specific activity, which means that the obtained signal per unit weight material is very high. This can be illustrated by the following examples. One Bq corresponds to 0.3 pg of $^{137}$Cs (half-life 30 years), 27 pg of $^{226}$Ra and 152 ng of $^{129}$I. Therefore, non-radiometric methods have not been considered for the determination of important radionuclides like $^{90}$Sr, $^{137}$Cs, $^{241}$Am and the different Cm-isotopes. For very long lived nuclides the specific activity may be so low that the detection limits are too poor for certain types of samples. When the mass (or number of atoms per unit weight) becomes significant compared to the intensity of the emitted radiation per unit weight of the radionuclide, other mass-sensitive detection methods may be more favorable. An example of this is given by TOOLE et. al. While the α-spectrometric detection limit is 0.1 mBq for all actinides, the detection limit for inductively coupled plasma mass spectrometry (ICP-MS) changes from 0.004 μBq...