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A number of research programs have addressed development of analytical techniques for the determination of difficult-to-measure (DTM) radionuclides for the comprehensive characterization of radioactive wastes. DTM nuclides are of low-abundance, long half-life, they emit either α particles, or electrons (β, Auger) or X-rays that all suffer significant self-absorption. Liquid scintillation is an ideal tool to avoid absorption and assure high detection efficiency, but its use is limited by the poor energy resolution. The biggest challenge in the analyses is the identification of the small amounts of DTM nuclides and confirmation of the absence of interferences/impurities. In order to obtain correct results i) high requirements towards the selectivity of the separation have to be met, ii) isotopic interferences have to be taken into account, iii) the methods have to be carefully validated. For the latter purpose, various techniques such as measurement of reference materials, comparison with other methods, inter-laboratory comparisons, systematic assessment of the influencing factors, assessment based on scientific understanding of the principles can be used and have to be used according to ISO 17025[1].

RADANAL has been involved in the development of methods for the determination of a couple of β-decaying DTM nuclides, such as ^{90}Sr, ^{63}Ni, ^{93}Zr, ^{151}Sm. The methods are based on radiochemical separations followed by LS spectrometry. Extraction chromatography (EC) as a major tool is generally adequate to obtain high selectivity. The procedures for Sr, Ni, Zr, and Sm separations - developed at our laboratory - are based on EC using Sr resin, DMG resin, UTEVA resin and DGA resin[2], respectively in combination with further purification procedures. Chemical recoveries using properly selected tracers (e.g., ^{95}Zr, ^{140}La) or carriers (Sr, Ni, Zr, La) and decontamination factors for the major interferences have been determined.

Isotopic interferences in ^{90}Sr, ^{63}Ni, ^{93}Zr, ^{151}Sm determination (^{89}Sr, ^{91}Sr, ^{92}Sr, ^{59}Ni, ^{95}Zr, ^{154}Eu, ^{155}Eu, ^{147}Pm, respectively) have been taken into correction by careful spectrum evaluation, and performing further purification.

For method validation, the *use of reference materials* and participation in *interlaboratory exercises* are realistic choices only in case of a few well-known radionuclides (^{90}Sr, occasionally ^{63}Ni). For the more "exotic" nuclides method validation can be based on the application of an *independent analytical technique*. Inductively coupled plasma mass spectrometry (ICP-MS) is an excellent tool

for detection of relatively long-lived radionuclides, such as ^{93}Zr and there is a chance to use it for radionuclides that have half-lives down to around 100 years (^{151}Sm). *Standard addition* is a good technique for "quench independent" efficiency calibration (^{63}Ni). *Isotope dilution technique* is appropriate for calculating the accurate quantity (activity) of the analyte (^{93}Zr) when validation is done via ICP-MS measurement. But neither of the latter two techniques is able to reveal possible contamination in the source. *Scientific understanding of the principles* can help improve the performance of the results. As such principle, the law of radioactive decay is used to check the half-life of the analyte (identify ^{90}Sr via the ingrowth of its ^{90}Y progeny). Another example is the comparison of measured activity/mass ratios with theoretical values. We compared the measured ^{93}Zr/stable Zr mass ratio with the calculated ones in case of Zr fuel cladding in order to confirm the trueness of the analytical results.

In the presentation, the methods developed for the determination of ^{90}Sr, ^{63}Ni, ^{93}Zr, ^{151}Sm in radioactive wastes will be briefly discussed and emphasis will be given to the various aspects of method validation. Examples for good and bad practices will reveal the need for competent method validation.

References:

1. General requirements for the competence of testing and calibration laboratories, ISO/IEC 17025, 2005.
2. Homepage of TRISKEM International: http://www.triskem.com/full_technical%20information%20resins.asp