ACTIVITY MEASUREMENT ALGORITHM IN SOLID RADIOACTIVE WASTE CLEARANCE PROCEDURE

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1. Waste characterization process basics

The radionuclides produced in a nuclear reactor are carried by the fluids that are flowing within the nuclear reactor plant systems. Some of the radionuclides are collected in radioactive waste such as ion exchange resins generated from reactor water purification systems, concentrated liquids produced by evaporating various kinds of process water from the plant systems, and some which adhere to the inner surfaces of the equipment of the plant systems, are present in dry active wastes generated when performing maintenance or when replacing equipment components. Such waste may include the component itself or various other materials, such as worker's protective clothing, that become contaminated through contact with the contaminated items.

Most of nuclear installations provide long-term regular and systematic monitoring of activity inventory and dispose with large database of measurement results. Activity of "difficult to measure radionuclides" (DTM) is in general determined using radiochemical analysis of waste streams samples. On the pre-decommissioning stage of shut down nuclear facilities exhaustive radiological analyses for radiological characterization are performed. Large databases of measuring values are used for nuclide activity correlation analysis for each waste stream. The correlation analysis outcome is "radionuclide vectors" (RNV) alternatively "key nuclide" (KN) determination and "scaling factors" (SF) calculation. The idea of the scaling factors method is that radionuclides originating from the same source and showing the same physicochemical behavior of their transport tothe waste should have a constant ratio of their activity concentrations. SF method is later used to determine activities of difficult-to-measure radionuclides from measured activity of key radionuclide, which is selected from among easy to measure radionuclides (ETM). Results of waste characterization are finally used in free release or repository acceptance criteria judge procedures.

2. Scaling factors method and radionuclide vectors method principle

Scaling factors method provide a mechanism for estimating the quantities of difficultto-measure nuclides in individual waste packages based on limited radiochemical analysis of samples from the bulk waste stream. DTM nuclidesare typically represented by beta emitting and alpha emitting nuclides, which exist in these radioactive wastes such as ¹⁴C, ⁶³Ni and ²⁴⁰Pu. In the reactor, key nuclides (gamma emitting nuclides such as ⁶⁰Co and ¹³⁷Cs, which are measurable from outside of the waste package usually by high resolution gamma spectrometry) are generated concurrently with DTM nuclides and transfer within the plant systems. In the SF method, the mass activity of DTM nuclides in radioactive wastes are evaluated by multiplying the mass activity of the key nuclide by the coefficients calculated based upon radionuclide data obtained by sampling and radiochemical analysis (i.e. the SFs, which are the ratios of mass activity between DTM nuclides and key nuclides). SFs can be calculated from measurements of radionuclides obtained through appropriate radiochemical analysis, through modeling code calculation or by a combination of both techniques. Principle of the Scaling factors method is given in ISO 21238:2007 and international experience with the scaling factor methodology is presented in IAEA NW-T-1.18. Flow chart of SF method application in characterization process shows figure 1.



Fig.1. Basic flow of SF method application in characterization process.

Radionuclides vectors method is dominantly used in Slovakian praxis. In general radionuclides vector is also some set of scaling factors, when we use instead of one key nuclideactivitythetotal activity of all radionuclides in waste batch. In this case the RNV components represent the relative contribution of each radionuclide to total activity. Total activity is calculated from easy to measure radionuclides response measured by gamma radiation measurement device.

RNV method has advantages in measurement stage of characterization process, because the non-spectrometric devices can be used and throughput of wastes is greater. On the other hand non-spectrometric device give null information about the RNV applicability. This is the essential attribute which restrain RNV method usage in confrontation with SF method. Spectrometric measurement can decrease uncertainties associated with ETM radionuclides estimation, which is in case of non-uniformity and instability of waste stream its crucial feature.

3. Free release measurement facility design

High throughput requirement to measurement facility and low free release criteria indicate needs of big size and high sensitivity. For high resolution gamma spectrometry system it means using of active and passive systems for background minimization(robust shielding, ventilation, collimators).



Fig.2. Radioactive waste free release measurement facility (MetroRWM experiment), general view and inner space with 4 HPGe detectors

In frame of European Metrology Research Project MetroRWM, was experimental measurement facility designed, constructed and tested, with the next properties:

- 4 HPGe detectors (50% rel. efficiency) with electric cooling
- 4 measurements in 3 position leads to 12 spectra what allow to test homogeneity
- Energy range 50 2000 keV
- Lead collimators
- Integrated weighing machine, range 0 500 kg,accuracy 0,2 kg
- Shielding built from very low-level activity bricks (without lead)
- Measuring container volume $0,33 \text{ m}^3$ (120x80x40), up to 700 kg
- Capacity 6 containers per hour
- Automatic measurement and analysis,
- SF method used for DTM radionuclides determination
- Protocol from measurement
- Results (including spectra files) are stored in database
- QA (stability and background tests)

Very low background level and high efficiency detectors results in low Minimum Detectable Activity (MDA) with short measurement time (T_m). In table 1. are results of MDA calculations for different measurement time for contaminated gravel with density 1,49 g.cm⁻³ in 0,33 m³ container.

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Radionuclide	MDA for T _m =100 s	MDA for T _m =300 s	Clearance limit
	$(Bq.g^{-1})$	$(Bq.kg^{-1})$	$(Bq.kg^{-1})$
Mn-54:	2	0,8	300
Co-57:	3	1,4	3000
Co-58:	1	0,9	300
Co-60:	2	0,7	300
Cs-137:	3	0,8	300
Eu-152:	6	3,1	300
Am-241:	20	6,6	300

Tab. 1. Minimum detectable activity calculations

4. Conclusion

Most of European metrology experts prefer in radiation wastes characterization process to use scaling factor method in combination with high definition gamma spectrometry rather than radionuclide vector method in combination with gross gamma activity measurement. The second method is actually used in Slovakian nuclear facilities. The international (IAEA) and Slovak national authorities (UVZ) recognize both approach but there is a fact, that high definition gamma spectrometry "sees" radionuclides more properly.

Acknowledgement

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5. References

- [1] ISO 21238:2007 Nuclear energy Nuclear fuel technology Scaling factor method to determine the radioactivity of low- and intermediate-level radioactive waste packages generated at nuclear power plants
- [2] ISO 14850-1:2004Nuclear energy Waste-packages activity measurement Part 1: High-resolution gamma spectrometry in integral mode with open geometry
- [3] IAEA NUCLEAR ENERGY SERIES No. NW-T-1.18 Determination and use of scaling factors for waste characterization in nuclear power plants. Vienna: International Atomic Energy Agency, 2009. p. ; 29 cm.