Radiological characterization of metallic waste on decommissioning by comparing real and simulated spectrum

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Abstract

One aim of decommissioning planning, is to obtain a radiological understanding of the involved installation. The characterization could be carried out by: (a) neutron activation calculations; (b) dose rate measurements; (c) in-situ gamma spectrometry; (d) sampling for determination of the scaling factors (SF).

Neutron activation calculations contains several uncertainties based on: (a) input data - such as material data (composition and impurities); (b) neutron flux and energy; (c) nuclear data libraries; (d) the methodology of the process and the simulation codes.

This work is focused on the development of a technique for validation of neutron calculations.

A non-destructive gamma spectrometry technique by using MCNP6.1 simulations is under development for interpretation of the resulting gamma-ray spectra of the radionuclides in activated components. In particular, a spectrum will be produced, based on the activities of the main radionuclides in the activated component and the results of MCNP6.1 simulations. This spectrum will be compared with the experimental spectrum.

Background

- Many of the important long lived radionuclides in radioactive waste are difficult to measure (DTM) by non-destructive techniques.
- Identification of these DTM nuclides requires radiochemical techniques to separate the various radionuclides for measurement and determination of the SF.
- SF are used for determination of the activities of DTM radionuclides, based on the correlation between easily measurable gamma emitting nuclides (key nuclides) and DTM nuclides.
- The key radionuclides are ^{60}Co which is an activation product and ^{137}Cs - a main fission product - which due to its high water solubility is easily transported and settled as surface contamination.
- The spectrum of these two radionuclides as well as of ^{90}Sr - a fission product and a pure β^- emitter - is generated by the MCNP6.1 code and compared with the corresponding experimental one.

Materials & Methods

MATERIALS

- Bicron Monoline Scintillation detector NaI(Tl) (Model 3M3/3) with a 3'' x 3'' crystal
- GenieTM 2000 spectroscopy software for spectrum acquisition
- MCNP6 simulation code

METHODS

- Point source geometry for ^{60}Co, ^{137}Cs and ^{90}Sr placed at 15 cm distance in front of the scintillation detector NaI(Tl), on the main axis of symmetry.

- Regarding the simulated spectra, F8 pulse-height tally combined with the "FT8 GEB card" of MCNP6 code is used.

- The Gaussian Energy Broadening (GEB) option defines the energy broadening according to the Gaussian formula:

$$f(E) = \frac{1}{\sigma\sqrt{2\pi}} e^{-\frac{(E-E0)^2}{2\sigma^2}}$$

σ: standard deviation depends on $FWHM = 2\sqrt{2 \cdot ln2} \cdot \sigma \approx 2{,}35482 \cdot \sigma$

- FWHM is defined experimentally from $FWHM = a + b\sqrt{E + cE^2}$

A non-linear function adjusted by least-squares procedure is applied to determine the values of a, b and c coefficients (Fig. 1).

The adjustment coefficients of the resolution curve for the Na(Tl) detector were obtained by the method of least squares and the values are α= -0.0055 MeV, b=0.06214 MeV$^{1/2}$ and c=0 MeV^{-1}, using data from four radioactive sources of ^{60}Co, ^{137}Cs, ^{133}Ba, ^{241}Am (Table 1).

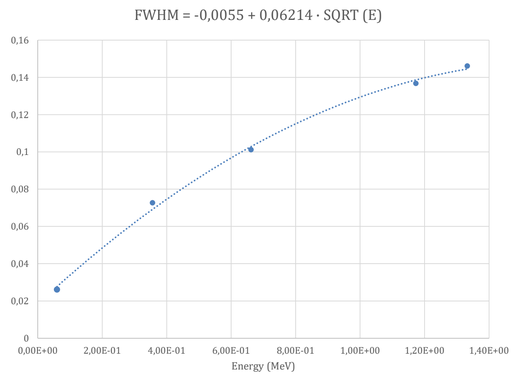


Fig. 1. *Function of FWHM for Bicron Monoline scintillation detector NaI(Tl) (Model 3M3/3) with a 3'' x 3'' crystal.*

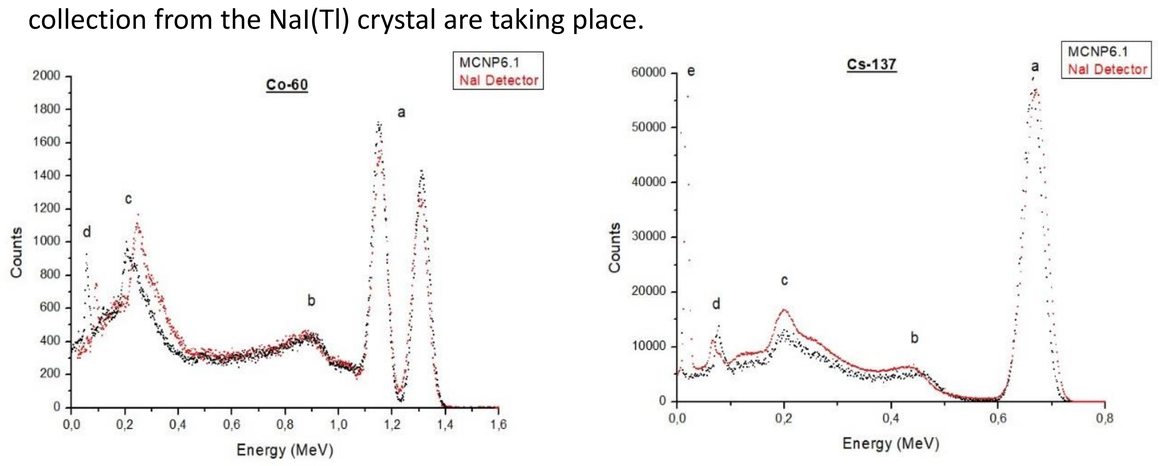
Source	Energy (MeV)	FWHM
Am-241	5.95E-02	0.026139
Ba-133	3.56E-01	0.072639
Cs-137	6.62E-01	0.101197
Co-60	1.17E+00	0.136747
	1.33E+00	0.14613

Table 1. *Energy and resolution calibration of NaI scintillation detector*

Results

The comparison of simulated and the experimental spectrum of ^{60}Co, ^{137}Cs and ^{90}Sr sources shows that they are in perfect agreement (Fig. 2).

The experimental spectrum at low energy in both the ^{60}Co and ^{137}Cs spectrum has the same form leading us to the conclusion that the background as well as additional effects of photon collection from the NaI(Tl) crystal are taking place.



peak a : Photoelectric effect, **peak b** : Compton edge ($E_e = h\nu \cdot \frac{1-cos\theta}{\frac{m_e c^2}{h\nu}+1-cos\theta}$ for θ=π), **peak c** : Backscatter peak from the Compton scattering of the gamma rays in the walls of the lead shield surrounding the detector ($\Delta\lambda = \lambda' - \lambda_0 = \frac{h\cdot(1-cos\theta)}{mc}$, for θ=π), **peak d**: Kα1 shell X-ray (0,75keV) arises from the lead excitation of the shielding

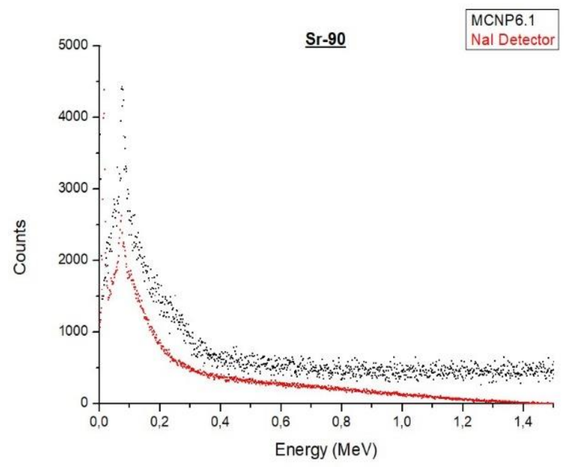


Fig. 2. *Comparison between simulated and experimental pulse height distribution to Co-60, Cs-137 and Sr-90 sources.*

- MCNP6.1 uses the temperature of 2,53·10^{-8} MeV (equal to 20^{o}C) as a default value. Scintillators are affected by the existing temperature, but MCNP code does not take into account the differential of a small temperature rate of 10 or 20 degrees.
- No uncertainty about source activities has been taken into account.
- There are no detailed data for the ^{90}Sr source used → geometry errors

Conclusion

- The qualitative agreement between the simulated and experimental spectrums justifies the validity of the simulation results for the radioactive sources.
- The simulated spectrums presented in this work show that not only the gamma peaks are characteristic but also the continuum are representative for each of the radionuclide.
- The technique will be used for radiological characterization of nuclear reactor components. The characterization of the components which appeared with surface contamination, is essential for the decision making process during decommissioning. The cutting techniques in order to reduce the production of secondary waste and limit the doses to personnel and the selection of decontamination techniques should be based on accurate determination of the radionuclides inside the material and/ or in the surface contamination.

The next steps includes the comparison of simulated real spectrums in case of complex geometries of sources with multiple radionuclides and the use of HPGe detector.

International Conference on Radiation Applications RAP 2023, Hellenic Center of Marine Research, Anavyssos, Attica, GREECE, 29 May – 2 June 2023