Combined Photopeak and Compton Analysis for Radiological Characterization of Activated Steel in Nuclear Reactor Decommissioning

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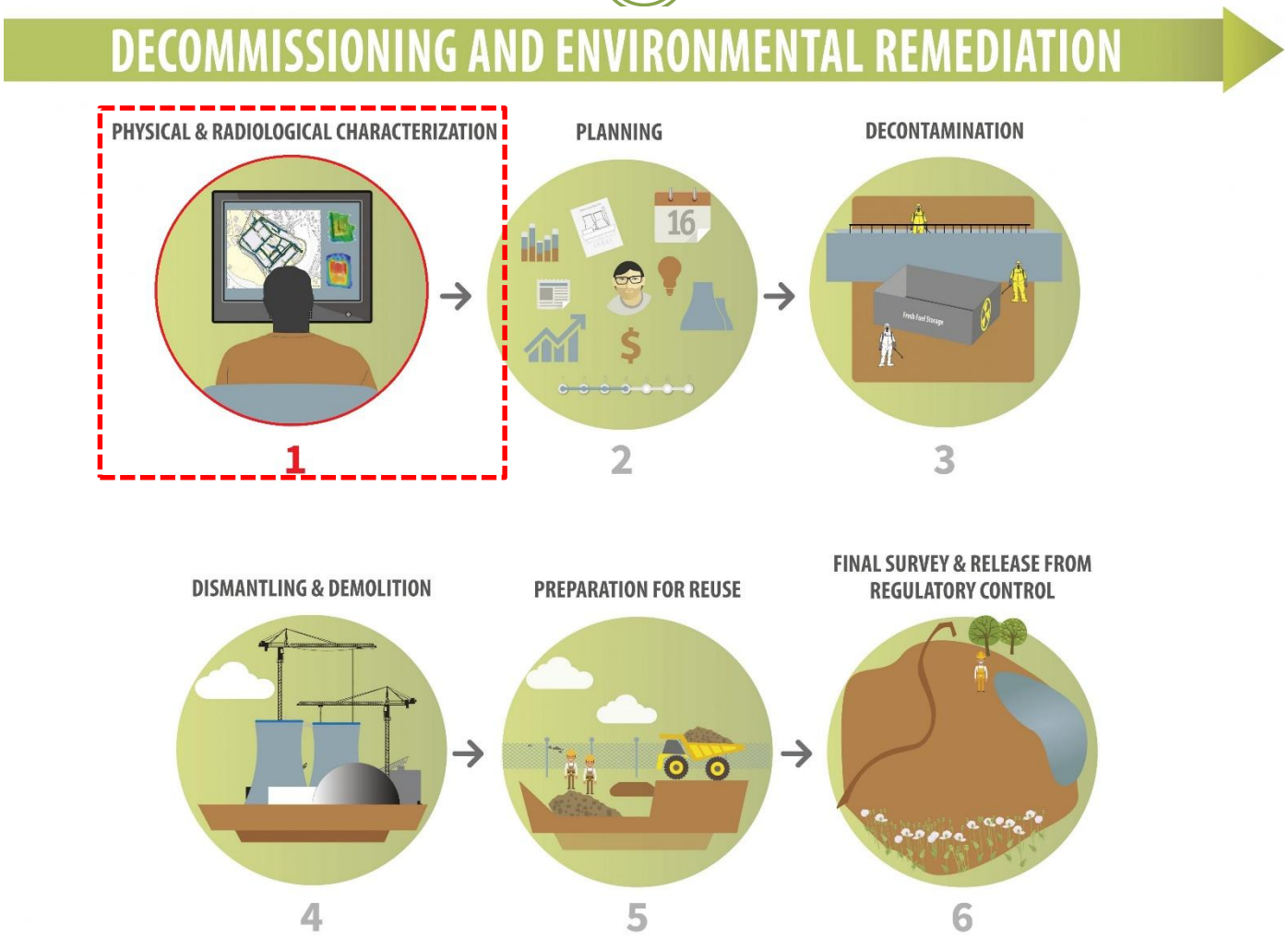
Introduction



Scope of this work

Radiological characterization of activated components which frequently exhibit surface contamination is essential for the decision making process regarding their management during decommissioning.

- Cutting techniques
- Decontamination techniques
- Clearance procedures

Characterization on decommissioning

The characterization could be carried out by means of :

(a) neutron activation & radionuclide transport calculations;

(a) dose rate measurements;

(b) in-situ gamma spectrometry;

(c) sampling for determination of scaling factors (SF).

Neutron Activation Calculation Uncertainties

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ISO16966:2013

Material Data
- Material composition
- Impurities (even in ppm-range they have a significant influence on results [Li, N, Co, U])

Nuclear Data Libraries
- Nuclide library with reliable cross sections (response lines related different neutron energies)
- Consideration of all relevant nuclides for the purpose

Modelling
- Level of detail of the model
- Position of the material in the reactor

Neutron fluence rates
- Neutron flux
- Energy spectrum

Neutron activation calculation

Irradiation history
- integrated exposure
- decay time

Results: Nuclide Activity

Software
- Numeric calculation: stability, convergence
- Different calculation methods (Point method, Range method)

Scaling Factors (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.
- In the SF method, the concentrations of DTM nuclides in radioactive wastes are evaluated by multiplying the concentrations of the key nuclide by the coefficients calculated based upon radionuclide data obtained by sampling and radiochemical analysis.
- 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.
- The key radionuclides are Co-60 which is mainly an activation product and Cs-137 - a main fission product - predominantly found as surface contamination and only rarely within the material volume.

Objective

This work is focused on the development of an **innovative technique** for:

- the **concurrent quantitative determination** of volume activity (due to activation) and surface activity (due to the surface contamination) in steel components, using **one single measurement**
- the **validation** of neutron calculations
- **limiting the sampling** for radiochemical analysis
- significantly improve the accuracy of radiological characterization and SF determination for metallic waste classified as Low Level Waste (LLW), Very Low Level Waste (VLLW), and Exempt Waste (EW).
- efficient decontamination for **clearance and recycling**, disposal volumes and associated **costs reduction**, and repository **capacity preservation**.

Method

A non-destructive gamma spectrometry technique by using MCNP6.1 simulations for interpretation of the resulting gamma-ray spectra of the radionuclides in activated and/ or contaminated components.

In particular, simulated spectra were produced and compared with the corresponding experimental spectra. These comparisons validated the simulation results and allowed the use of simulated spectra for interpretation of experimental γ-spectra.

Spectra produced using MCNP6.1

1. Use of F8 pulse-height tally (*energy distribution of the pulses created within the cell*) combined with the "FT8 GEB card" i.e FT8 GEB *α b c*
2. The *α*, *b* and *c* values are functions of the type of detector and the size of the detector with units of MeV, $MeV^{1/2}$ and MeV^{-1} respectively. These parameters specified the Full Width at Half Maximum

$$FWHM = a + b\sqrt{E + cE^2}$$ where *E* is the Energy of the tallied particle

3. The Gaussian Energy Broadening (GEB) option defines the energy broadening according to the Gaussian formula

$$f(E) = \frac{1}{\sigma\sqrt{2\pi}} e^{-\frac{(E-E_0)^2}{2\sigma^2}}$$

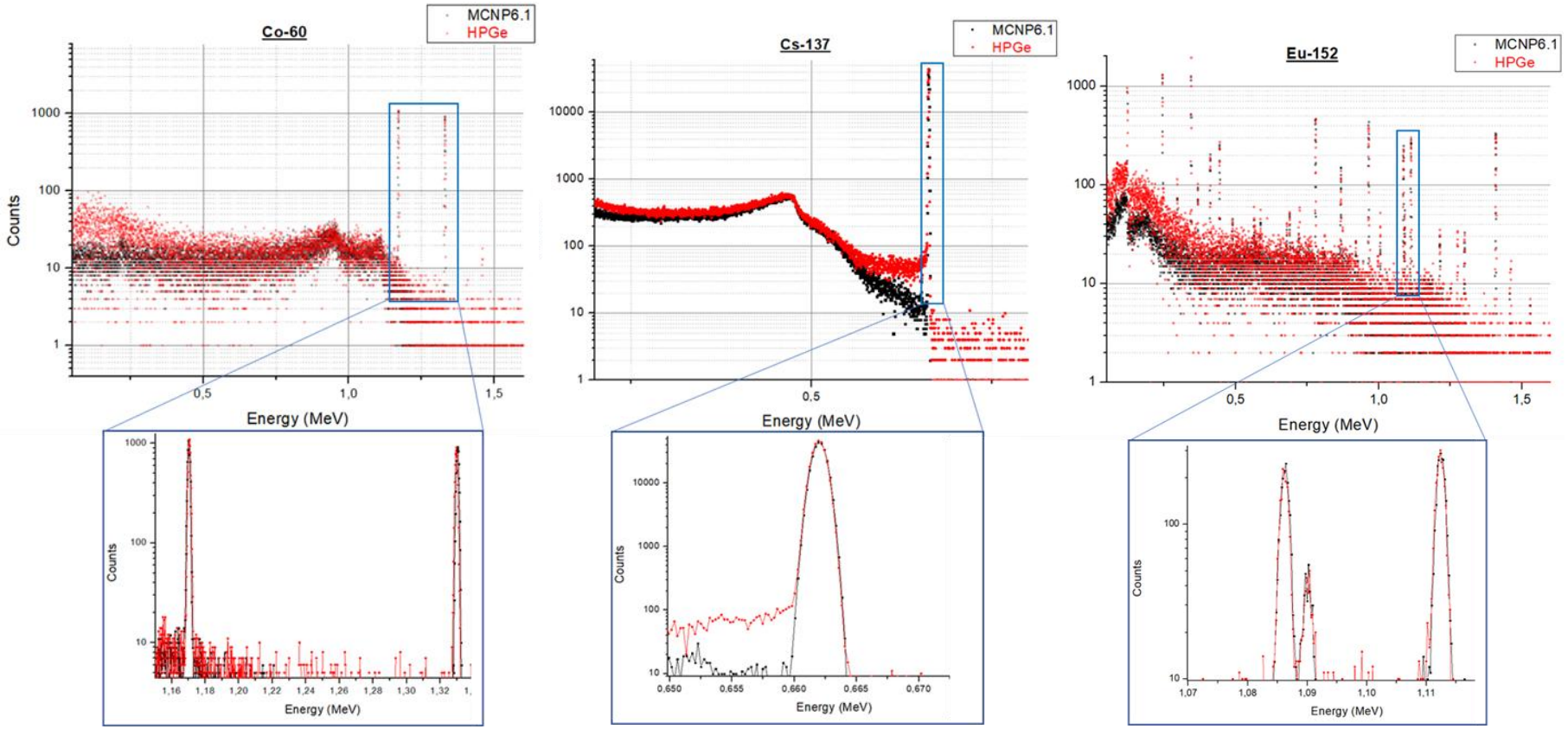
E: *broadened energy;*

E_o*: unbroadened energy;*

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

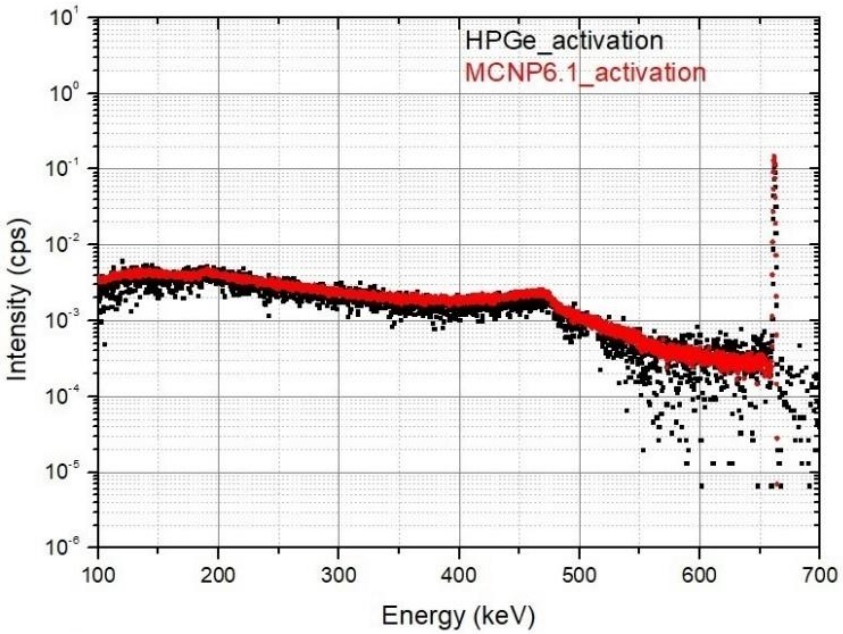
4. Energy bins in MCNP6.1 input file should be equal to the 8192 channels of the HPGe detector.

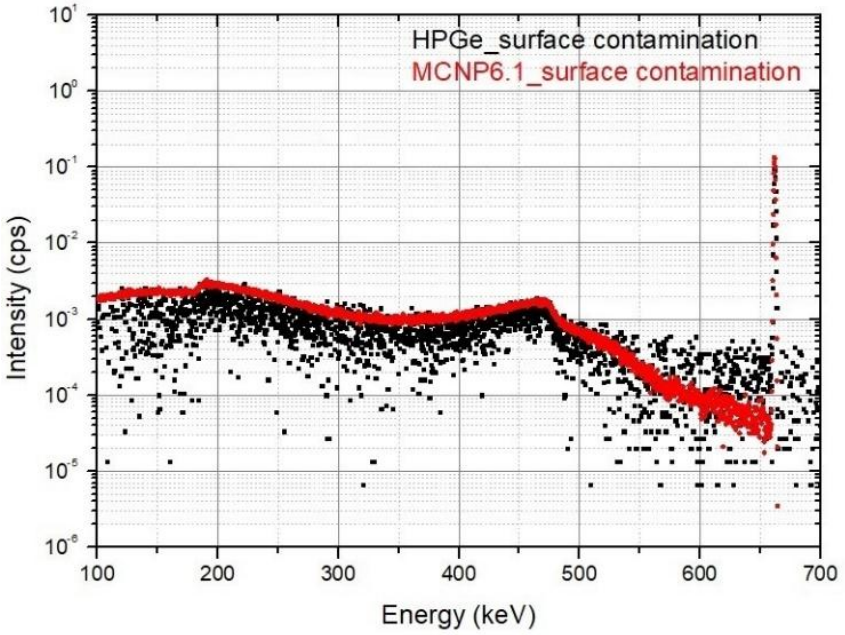
Quantitative comparison of real & simulated spectra – Point Sources



Co-60, Cs-137 and Eu-152 point sources spectra

Quantitative comparison of real & simulated spectra – Volume Sources





Nominal volume (16 sheets of contaminated with ^{137}Cs filter papers)

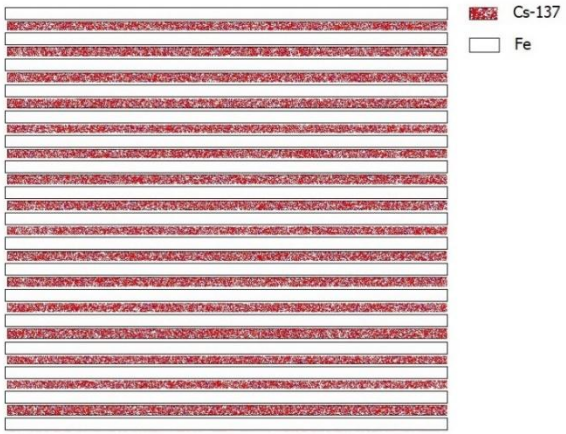
$$A_V = 4960 \pm 500\ Bq$$

Nominal surface (5 sheets of contaminated with ^{137}Cs filter papers)

$$A_S = 1550 \pm 160\ Bq$$

Volume sources used

- Volume source representing the activated and the surface contaminated metallic slab both with ^{137}Cs were prepared at the Radioactive Waste and Material Laboratory in the NCSRD.
- Filter paper was subdivided into squares of 9 cm^2 each and 0.05ml of ^{137}Cs acid solution (2M HNO_3) of 170 ± 17 Bq/ ml in the centre of each square. with ^{137}Cs filter paper was 5940 cm^2. The total surface was subdivided into 18 sheets of 33 × 10 cm^2. Hence, the activity for each one of the 18 contaminated sheets was 304 ± 30 Bq. (0.92 Bq/cm^2)



Volume source representing the activated slab for validation of MCNP6.1 models (VisedX_24E)

Volume source representing the surface contamination on a slab for validation of MCNP6.1 models (VisedX_24E)

First Method

The first method is based on the net counts of the detector in the photopeak and the Compton edge region arising from activities distributed both within the volume and on the surface of steel samples.

$$C'_{ph} = \lambda_1 \times A'_V + \lambda_3 \times A'_S$$

$$C'_{ce} = \lambda_2 \times A'_V + \lambda_4 \times A'_S$$

$$\lambda_1 = \frac{C_{phV}}{A_V} \qquad \lambda_2 = \frac{C_{ceV}}{A_V} \qquad \lambda_3 = \frac{C_{phS}}{A_S} \qquad \lambda_4 = \frac{C_{ceS}}{A_S}$$

C_{phV}: counts of the photo-peak for "Nominal Volume" with activity A_V

C_{ceV}: counts of the Compton-edge for "Nominal Volume" with activity A_V

C_{phS}: counts of the photo-peak for "Nominal Surface" with activity A_S

C_{ceS}: counts of the Compton-edge for "Nominal Surface" with activity A_S

C'_{ph}: counts of the photo-peak in the experimental spectrum for each of the three arrays exhibiting both surface (A'_S) and volume activity (A'_V)

C'_{ce}: counts of the Compton-edge in the experimental spectrum for each of the three arrays exhibiting both surface (A'_S) and volume activity (A'_V)

Second Method

The second method relies on the correlation between the Compton-to-photopeak ratio (C/P) and the absolute photopeak efficiency, as derived from simulated gamma-ray spectra.

The graphical representation of C/P as a function of the absolute photo-peak efficiency is used to determine the total absolute photo-peak efficiency as well as the relative contribution of volume and surface activity to the photo-peak counts.

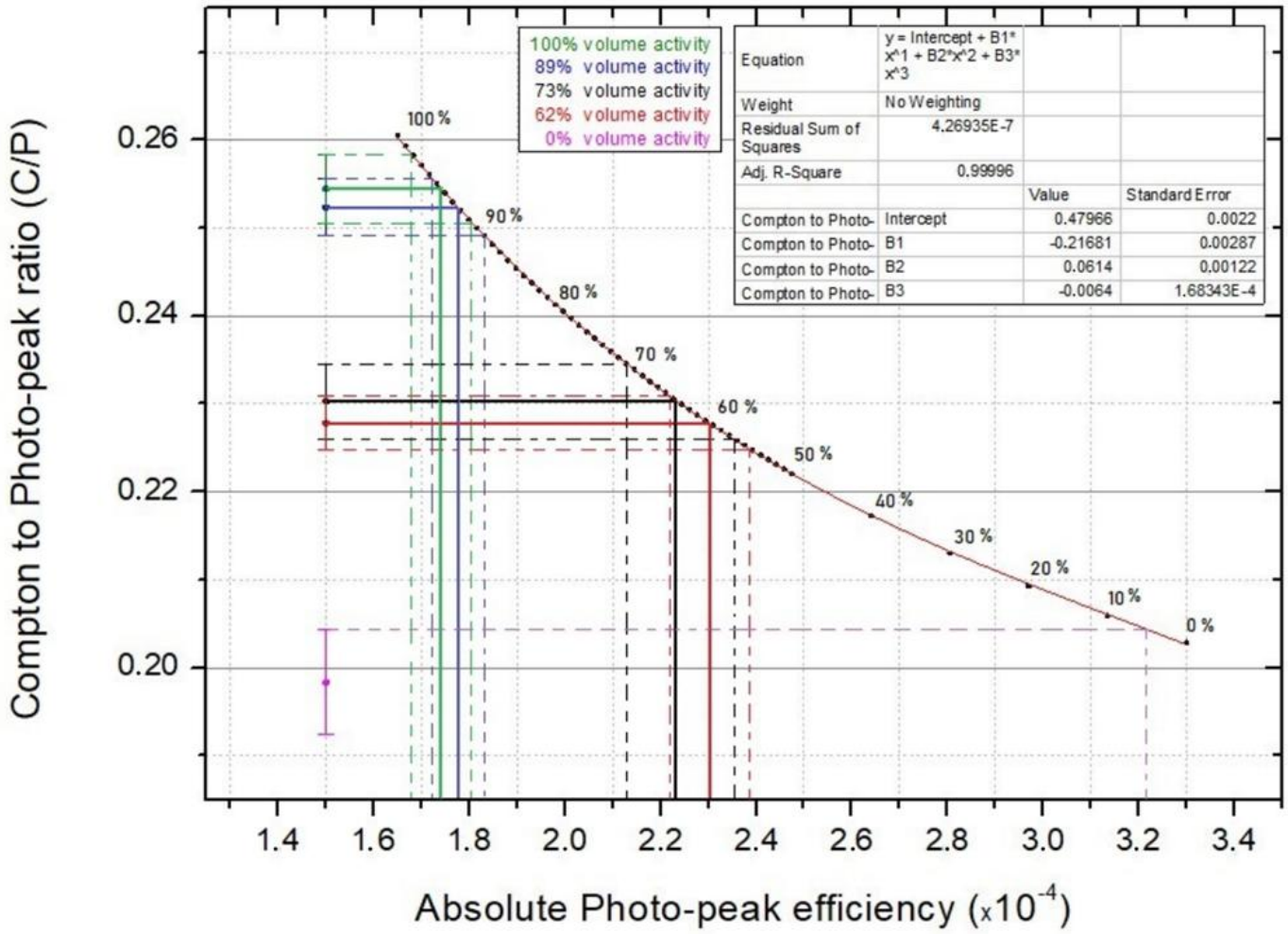
$$total\ efficiency = (a \times efficiency\ volume) + (\beta \times efficiency\ surface)$$

$$vol.effic.(\%) = (a \times efficiency\ volume) / total\ efficiency$$

$$surf.effic.(\%) = (\beta \times efficiency\ surface) / total\ efficiency$$

$$a + \beta = 1$$

Second Method



Results

Case	α vol. act. (%)	vol. effic. (%)	β surf. act. (%)	surf. effic. (%)	Counts from volume ($\times 10^3$)	Volume activity-A_V^i (Bq)	Counts from surface ($\times 10^3$)	Surface activity-A_S^i (Bq)	Total HPGe counts ($\times 10^3$)
Nominal Surface						**0**		**1550 ± 155**	
1st method						**110 ± 100**		**1200 ± 100**	
2nd method	0^{+5}_{0}*	$0^{+0.03}_{0}$	100^{0}_{-5}*	$1^{0}_{-0.03}$	$0^{+1.59}_{0}$	$\mathbf{0^{+60}_{0}}$	$61.16^{0}_{-1.59}$	$\mathbf{1240^{0}_{-30}}$	61.16 ± 0.24
Nominal Volume						**4960 ± 496**		**0**	
1st method						**3530 ± 250**		**210 ± 120**	
2nd method	$94.5^{+3.5}_{-4.0}$*	$89.6^{+6.7}_{-6.6}$	$5.5^{+4.0}_{-3.5}$*	$10.4^{+6.6}_{-6.7}$	$87.69^{+6.56}_{-6.46}$	$\mathbf{3540^{+270}_{-260}}$	$10.18^{+6.46}_{-6.56}$	$\mathbf{210^{+130}_{130}}$	97.87 ± 0.31
Nominal 1						**4960 ± 496**		**620 ± 62**	
1st method						**4100 ± 250**		**340 ± 120**	
2nd method	$92.0^{+3.5}_{-3}$*	$85.3^{+6.3}_{-5.0}$	$8.0^{+3}_{-3.5}$*	$14.7^{+5.0}_{-6.3}$	$100.56^{+7.43}_{-5.89}$	$\mathbf{4060^{+300}_{-240}}$	$17.33^{+5.89}_{-7.43}$	$\mathbf{350^{+120}_{150}}$	117.89 ± 0.3
Nominal 2						**2480 ± 248**		**1550 ± 155**	
1st method						**1930 ± 200**		**1270 ± 100**	
2nd method	$60.5^{+4.5}_{-5.5}$*	$43.4^{+5.1}_{-5.4}$	$39.5^{+5.5}_{-4.5}$*	$56.6^{+5.41}_{-5.1}$	$48.03^{+5.64}_{-5.98}$	$\mathbf{1940^{+230}_{-240}}$	$62.64^{+5.97}_{-5.64}$	$\mathbf{1270^{+120}_{-110}}$	110.67 ± 0.3
Nominal 3						**2480 ± 248**		**930 ± 93**	
1st method						**1670 ± 200**		**920 ± 100**	
2nd method	65^{+6}_{-8}*	$48.1^{+6.94}_{-8.1}$	35^{+8}_{-6}*	$51.9^{+8.1}_{-6.98}$	$41.86^{+6.0}_{-7.0}$	$\mathbf{1690^{+240}_{-260}}$	$45.16^{+7.10}_{-6.00}$	$\mathbf{910^{+140}_{-120}}$	87.02 ± 0.3

Outcomes

- Two equivalent methodologies were developed and validated for concurrent determination of homogeneous surface and volume activities in mixed-activity steel samples with specific activities at the level of specific clearance for melting.
- Both methodologies produced consistent results and the agreement between the nominal activities and the experimentally determined ones were within the 1σ to the 2σ uncertainty level, confirmed their reliability for the concurrent determination of surface and volume activities in steel using a single experimental gamma-ray spectrum of an unknown sample.

Conclusions

This technique is essential for:

- the decision making process on cutting techniques, decontamination techniques and clearance procedures
- neutron calculations validation
- SF determination via sampling
- safe and efficient waste management

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Thank you for your attention