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# Application of "wet" extrapolation method for activity standardisation of electron capture radionuclides

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**Abstract**

The activity of electron capture radionuclides is usually determined by 4π(proportional counter, PC)–γ coincidence counting. The corrections necessary for the final activity value calculation are obtained by an extrapolation method. Variation of the PC detection efficiency can be achieved through different methods, e.g. by changing the self-absorption of the source using absorbing foils or by adding carriers. Another possibility is a "wet" extrapolation method, which utilises an absorption change during the drying of a water droplet added onto the source surface. In this paper, slopes of extrapolation curves and resulting activity values obtained by different methods are compared for several radionuclides ($^{54}$Mn, $^{139}$Ce, $^{88}$Y, $^{57}$Co). In some cases a digital coincidence system was used for the analysis of measured data. The "wet" extrapolation, due to its very simple procedure, seems to be convenient for routine measurement and its accuracy is similar to the other methods.


*Keywords:* 4πβ–γ coincidence counting; Efficiency extrapolation; $^{54}$Mn standardisation; $^{139}$Ce standardisation; $^{88}$Y standardisation; $^{57}$Co standardisation

## 1. Introduction

4π(Proportional counter, PC)–γ coincidence counting is a commonly used measurement method for the activity of radionuclide solutions. The corrections necessary for the final activity value calculation are usually obtained from an extrapolation, in which the detection efficiency in the PC channel is varied. Variable PC detection efficiency can be achieved through different methods, e.g. by variation of the electronic threshold level (this is not generally applicable for a PC operating in atmospheric pressure), addition of absorber foils, or altering the self-absorption of the source by addition of conductive seeding agents (e.g. aquadag).

In the case of electron capture (EC) nuclides, the energy of electrons detected in the PC channel is low; therefore the change of source self-absorption using foil addition etc. is too large. A simple efficiency extrapolation method—"wet" extrapolation—usable in 4π(PC)–γ coincidence counting was proposed by Zajíc (1987). It utilises an absorption change during drying of a water droplet added onto the source surface. A "wet" source is inserted into the PC and measured while the detection efficiency in the PC increases as the water evaporates. This method enables a smooth decrease of source self-absorption and can be reproduced using the same source. This paper describes the use of this method for the activity measurement of four EC decaying radionuclides, namely $^{54}$Mn, $^{139}$Ce, $^{88}$Y and $^{57}$Co.

## 2. Measurement

The correct γ-window setting, which is derived from the coincidence equation, is crucial for proper determination of the disintegration scheme correction. It is important especially for nuclides with competing EC and $\beta^+$ branches or if some of the decay branches are highly converted.

For EC nuclides with complex decay scheme, the observed counting rates for PC, gamma and coincidence channels can be written as:

$$N_{PC} = N_0 \sum_i a_i \left[ \varepsilon_{AXi} + (1 - \varepsilon_{AXi}) \frac{\varepsilon_{PC\gamma i} + \alpha_{Ti} \varepsilon_{CEi}}{1 + \alpha_{Ti}} \right], \quad (1)$$

$$N_\gamma = N_0 \sum_i \frac{1}{1 + \alpha_{Ti}} \varepsilon_{\gamma i}, \quad (2)$$

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$$N_C = N_0 \sum_i \frac{1}{1+\alpha_{Ti}} [\varepsilon_{XAi}\varepsilon_{\gamma i} + (1-\varepsilon_{XAi})\varepsilon_{Ci}], \quad (3)$$

where $N_{PC}$, $N_\gamma$, $N_C$ are the count rates in the PC, $\gamma$ and coincidence channel, corrected for background, dead time and accidental coincidences, respectively, $N_0$ is the activity of the source, $a_i$ the branching ratio, $\varepsilon_{XAi}$ the PC detection efficiency for EC X-rays or Auger electrons associated with the $i$th EC branch, $\varepsilon_{PC\gamma i}$ the PC detection efficiency for $\gamma$-rays associated with the $i$th branch, $\alpha_{Ti}$ the total internal conversion coefficient of $\gamma_i$; $\varepsilon_{CEi}$ the PC detection efficiency for conversion electrons associated with the $i$th branch and $\varepsilon_{Ci}$ the $\gamma$-detector efficiency for $\gamma$-rays associated with the $i$th branch, $\varepsilon_{\mathrm{Ci}}$ is the probability of coincidence detection of the $\gamma$-rays associated with the $i$th branch and detected in the $\gamma$-detector that were already detected in PC, In the case of the $\beta^+$ branch, the $j$th term in $N_{PC}$ is

$$a_j \left[ \varepsilon_{\beta j+} + (1-\varepsilon_{\beta j+}) \left( \frac{\varepsilon_{PC\gamma i} + \alpha_{Ti}\varepsilon_{CEi}}{1+\alpha_{Ti}} + \varepsilon_{PC\gamma(\beta+)} \right) \right]$$

and similarly in $N_C$ it is

$$\frac{a_j}{1+\alpha_{Tj}} [\varepsilon_{\beta j+}\varepsilon_{\gamma j} + (1-\varepsilon_{\beta j+})\varepsilon_{Cj}],$$

where $\varepsilon_{\beta j+}$ is the PC detection efficiency for $\beta^+$-particles and $\varepsilon_{PC\gamma(\beta+)}$ the PC detection efficiency for $\gamma$-rays associated with $\beta^+$-decay; $N_{PC}$ can be written as a linear function of the PC detection efficiency estimate $\varepsilon_{PC} = N_C/N_\gamma$:

$$N_{PC} = N_0 \left[ 1 - K \left( 1 - \frac{N_C}{N_\gamma} \right) \right]$$

or

$$\frac{N_{PC}N_\gamma}{N_C} = N_0 \left[ 1 + (1-K) \frac{1 - N_C/N_\gamma}{N_C/N_\gamma} \right]. \quad (4)$$

The function is linear only if $K$ does not change with $N_C/N_\gamma$. Thus the linearity conditions (Grigorescu, 1983) have to be fulfilled:

$$\varepsilon_{\gamma 1} \frac{\varepsilon'_{\gamma 1}(1-\varepsilon_{C1}/\varepsilon'_{\gamma 1})}{1-((\varepsilon_{PC\gamma 1}+\alpha_{Ti}\varepsilon_{CE1})/(1+\alpha_{T1}))} = \ldots \frac{\varepsilon'_{\gamma i}(1-\varepsilon_{Ci}/\varepsilon'_{\gamma i})}{1-((\varepsilon_{PC\gamma i}+\alpha_{Ti}\varepsilon_{CEi})/(1+\alpha_{Ti}))} = \ldots = E,$$

where $\varepsilon = \varepsilon_{\gamma i}/(1+\alpha_{Ti})$.

These conditions result in a less rigid condition: for a given $\gamma$-channel setting, the detection efficiency of photons corresponding to all branches must be constant. Then the coincidence equation can be written as follows:

$$\frac{N_{PC}N_\gamma}{N_C} \approx N_0(1+sk_\varepsilon), \quad (5)$$

where

$$s = \frac{1-(N_C/N_\gamma)}{N_C/N_\gamma}$$

and

$$k_\varepsilon = \sum_i a_i \left( \frac{\varepsilon_{PC\gamma i} + \alpha_{Ti}\varepsilon_{CEi}}{1+\alpha_{Ti}} - \varepsilon_{Ci}/E \right).$$

If the linearity conditions are met, the slope of the extrapolation curve depends on the $\gamma$-detection efficiency of the PC, the detection efficiency for conversion electrons and the total internal conversion coefficient of $\gamma$. The slope may not change with $N_C/N_\gamma$, so the detection efficiency of the conversion electrons may not be altered. Fig. 1 shows the transmission of electrons through a water layer for different electron energies. It is evident that the water layer that fully absorbs 6–14 keV electrons has no influence on the electrons with energy above 30 keV. The water layer in which all the 6 keV electrons are absorbed is very thin. This might increase the inhomogeneity of the source during the final phase of extrapolation, as it may have already partially dried out at that point. This inhomogeneity can result in an incorrect value of $N_{PC}N_\gamma/N_C$. However, this is rarely observed in sources prepared on Ludox- and insulin-treated foils. A typical "wrong" process of the final phase of extrapolation is shown in Fig. 2.

The detection efficiency of EC nuclides in a PC is usually low due to source self-absorption. The "wet" extrapolation makes it possible to obtain results in a sufficient range of $N_C/N_\gamma$ with a fine efficiency decrease. Since the measuring time of one point should be about 20 s, the use of a high-activity source is recommended. The problem posed by low statistical accuracy is crucial, especially in the case of $\gamma$-window setting, which reduces the count rate in the coincidence channel. That was the case during the $^{54}$Mn and $^{88}$Y measurements.

The $4\pi$(PC)–$\gamma$ coincidence system consists of a stainless-steel cylindrical PC with dimensions of $2 \times (18\,\mathrm{mm} \times \varnothing 64\,\mathrm{mm}$, using methane at atmospheric pressure in a gas flow arrangement and the $\gamma$-ray detection assembly (two opposing NaI(Tl) detectors mounted close to the PC).

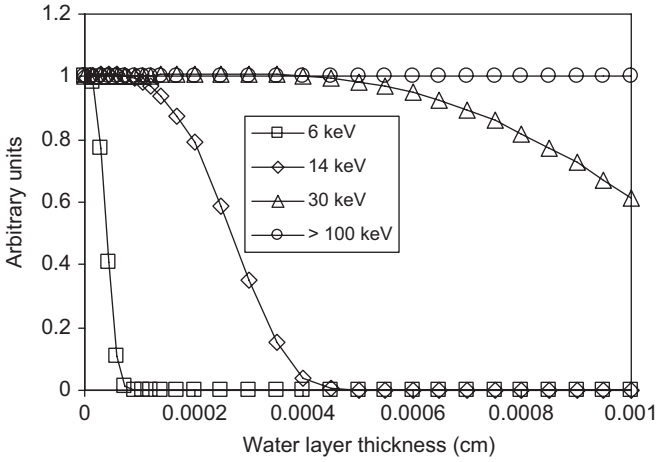


Fig. 1. The transmission of electrons through a flat water layer as a function of thickness for different electron energies. The ratio of electrons that passed through the layer to the original electrons ($y$-axis) was calculated using MCNPX 2.6 code.

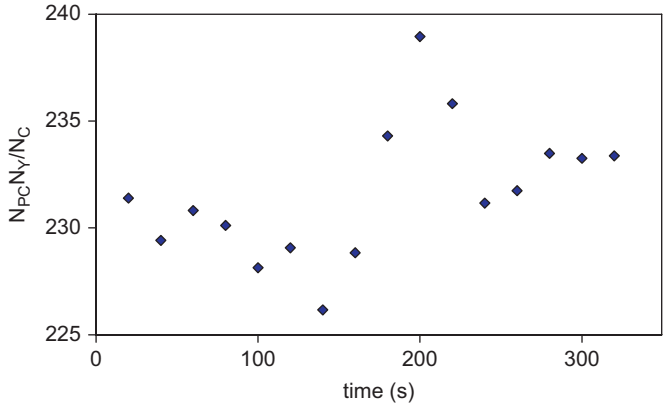


Fig. 2. The final phase of "wet" extrapolation with an inhomogeneous source. (The inhomogeneity was caused by the fact that in the final phase a part of the source was dry, whereas the rest of the source remained wet.)

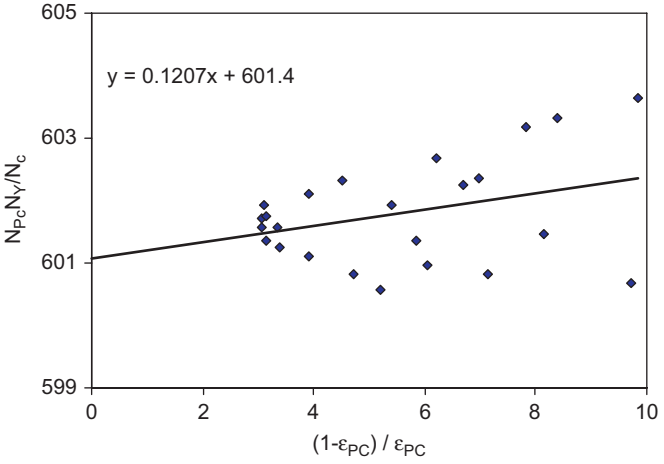


Fig. 3. Typical "wet" extrapolation curve for $^{54}$Mn, combining data of two sources.

The sources were prepared by deposition of 20–50 mg aliquots of active solution onto a conducting foil (gold-coated VYNS foils ~40 μg g$^{-1}$), treated with Ludox and insulin. The detection efficiency was changed either by the "wet" method or by adding extra mass to the source. Some of the sources were treated in an $NH_3$ atmosphere to achieve higher PC efficiencies.

### 2.1. Measurements of $^{54}$Mn

$^{54}$Mn nuclide has a very simple decay scheme, it decays by EC to the first excited state of $^{54}$Cr. The $K$ and $L$ probabilities for EC are about $P_K = 0.89$, $P_L = 0.1$, $\omega_K = 0.289$, the total internal conversion coefficient of $\gamma$ is about 0.0002. The Energy of Auger electrons is 0.4–0.7 keV ($e_{AL}$) and 4.5–5.99 keV ($e_{AK}$); (Bé et al., 2004). The coincidence equation can be easily derived from Eqs. (1)–(3) in the way described below and according to (4) and (5)

$$\frac{N_{PC}N_\gamma}{N_C} \approx N_0(1 + sk_\varepsilon)$$

and

$$k_\varepsilon \approx \varepsilon_{PC\gamma} - \frac{\varepsilon_C}{\varepsilon_\gamma} = \varepsilon_{PC\gamma}\left(1 - \frac{\varepsilon'_\gamma}{\varepsilon_\gamma}\right),$$

where $\varepsilon'_\gamma$ is the $\gamma$-detector efficiency for $\gamma$-rays already detected in the PC.

It is obvious that the value of the normalised extrapolation slope depends on the $\gamma$-window setting. For the "wide" window (LLD~100 keV) $k_\varepsilon$ is about zero ($k_\varepsilon \approx 0$), for an energy window encompassing the 834.8 keV peak, $k_\varepsilon \approx \varepsilon_{PC\gamma}$. Since the detection probability in the PC for 5 keV electrons is low due to absorption within the source, it is convenient to achieve a low value of $k_\varepsilon$.

Five sources were measured by the "wet" method, and the results were compared with those obtained by repeated addition of a drop of $Al(OH)_3$ into the source. The difference between the activity value resulting from the "wet" extrapolation and from the addition of extra mass was 0.1%. This is not significant considering that the relative standard extrapolation uncertainty was 0.1% for extra mass and 0.2% for "wet" extrapolation.

A typical extrapolation curve obtained from measurement of two "wet" sources is shown in Fig. 3.

### 2.2. The measurements of $^{139}$Ce

$^{139}$Ce decays by EC to the first excited state of $^{139}$La, which continues with a 165.85 keV partly converted $\gamma$-transition. The K and L probabilities for EC are about $P_K = 0.7$, $P_L = 0.23$, $\omega_K = 0.905$, $\omega_L = 0.117$ and the total internal conversion coefficient of $\gamma$ is 0.2516. The energy of Auger electrons is 2.7–6.2 keV ($e_{AL}$) and 26–39 keV ($e_{AK}$) and the energy of conversion electrons is 126–165 keV (Bé et al., 2004).

If the window in the $\gamma$-channel is set around the $\gamma$-peak so that X-rays are excluded, the coincidence equation is similar to the one derived for $^{54}$Mn:

$$\frac{N_X N_\gamma}{N_C} \approx N_0(1 + sk_\varepsilon)$$

and

$$k_\varepsilon \approx \varepsilon_{PC\gamma T} - \frac{\varepsilon_C}{\varepsilon_\gamma},$$

where $\varepsilon_{PC\gamma T}$ is the probability of a $\gamma$-transition detection in the PC:

$$\varepsilon_{PC\gamma T} \approx \frac{\varepsilon_{PC\gamma}}{1 + \alpha_T} + \frac{\alpha_T}{1 + \alpha_T}\varepsilon_{CE}.$$

The high value of the conversion coefficient makes the normalised slope $k_\varepsilon$ significant. It is obvious that linearity is achieved if the detection efficiency of the conversion electrons does not vary with change of $\varepsilon_{XA}$ during the extrapolation measurement. Fig. 4 shows a spectrum taken with the PC. The source was measured before and after the reduction of efficiency by adding a drop of aquadag; while the $L$-peak was decreased by 50%, the residual part of

spectra—corresponding to the (126–165) keV conversion electrons and X-rays—was reduced only by 1%.

A set of 15 sources was measured immediately after drying, then treated in an $NH_3$ atmosphere and measured, and finally, aquadag was added into five of the sources. The efficiency range was 0.2–0.48.

Another five sources treated with $NH_3$ were measured using the "wet" extrapolation method. The efficiency range was 0.15–0.49. The efficiency extrapolation curve is shown in Fig. 5. Results of both extrapolation measurements are shown in Table 1.

For $^{139}$Ce the result of "wet" extrapolation, which requires less sources and is very simple, is comparable with other methods.

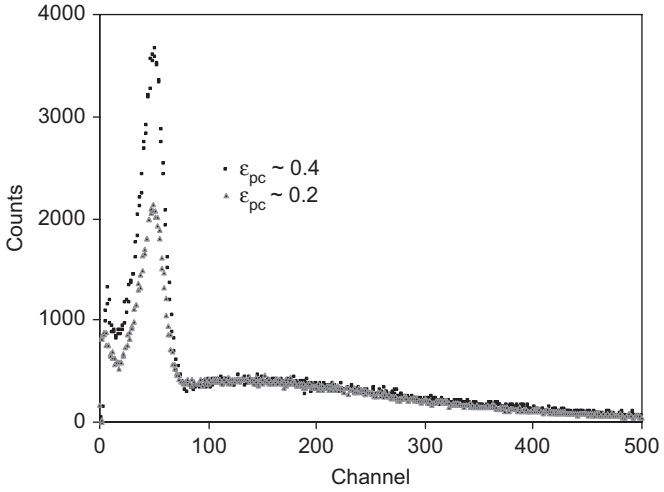


Fig. 4. Pulse height spectrum of $^{139}$Ce in a PC. The peaks correspond to L-Auger electrons.

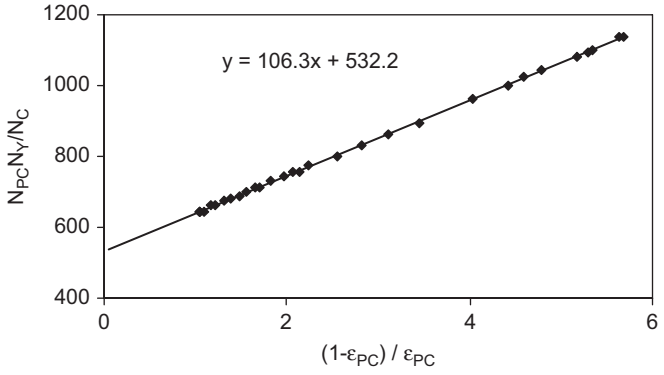


Fig. 5. Efficiency extrapolation curve of $^{139}$Ce.

### 2.3. The measurements of $^{88}Y$

As shown in Fig. 6, $^{88}$Y, decays 99.8% by EC and 0.2% by $\beta^+$ emission towards excited levels of $^{88}$Sr. Internal conversion coefficients of all listed $\gamma$-transitions are negligible ($\sim 10^{-4}$).

Considering that all branches pass through the 1836 keV level, it is useful to set the $\gamma$-window on $\gamma_3$. The observed count rates in individual channels according to Eqs. (1)–(3) are as follows:

$$\begin{aligned}
N_{PC} &= N_0\{a_1[p_1(\varepsilon_{XA} + (1-\varepsilon_{XA})(\varepsilon_{PC\gamma 1} + \varepsilon_{PC\gamma 3} \\
&\quad - \varepsilon_{PC\gamma 1}\varepsilon_{PC\gamma 3})) + p_2(\varepsilon_{XA} + (1-\varepsilon_{XA})\varepsilon_{PC\gamma 2})] \\
&\quad + a_2[\varepsilon_{XA} + (1-\varepsilon_{XA})\varepsilon_{PC\gamma 3}] + a_3[\varepsilon_{PC\beta+} \\
&\quad + (1-\varepsilon_{PC\beta+})(\varepsilon_{PC\gamma 3} + \varepsilon_{PC\gamma(\beta+)} \\
&\quad - \varepsilon_{PC\gamma 3}\varepsilon_{PC\gamma(\beta+)})]\}, \\
N_\gamma &= N_0(a_1 p_1 + a_2 + a_3)\varepsilon_{\gamma 3} = N_0(1 - a_1 p_2), \\
N_C &= N_0\{a_1[p_1(\varepsilon_{XA}\varepsilon_{\gamma 3+}(1-\varepsilon_{XA})(\varepsilon_{pC\gamma 1}\varepsilon_{\gamma 3} + \varepsilon_{C1}] \\
&\quad + a_2[\varepsilon_{XA}\varepsilon_{\gamma 3+}(1-\varepsilon_{XA})\varepsilon_{C2}] + a_3[\varepsilon_{PC\beta+}\varepsilon_{\gamma 3} \\
&\quad + (1-\varepsilon_{PC\beta+})\varepsilon_{C3})]\},
\end{aligned}$$

where $\varepsilon_{C3}$ is the probability for annihilation photons or $\gamma_3$-rays to be detected in the PC detector when they are also detected in the photon detector. The coincidence

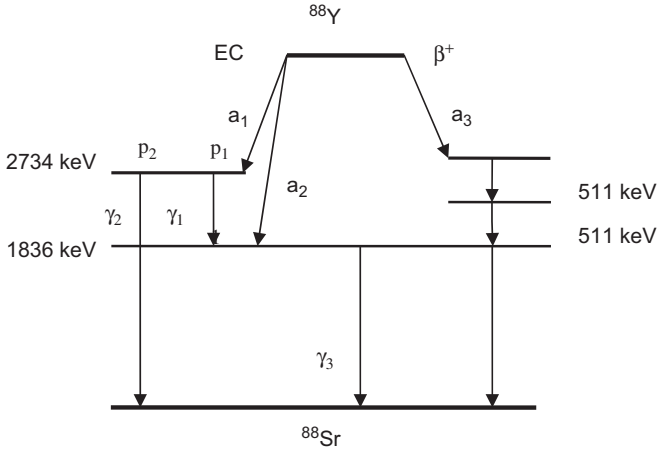


Fig. 6. Simplified decay scheme of $^{88}$Y. The branching ratios are $a_1 = 0.945$, $a_2 = 0.052$ and $a_3 = 0.002$. The $K$ and $L$ probabilities for EC are about $P_K = 0.72$, $P_L = 0.23$, $\omega_K = 0.696$, $\omega_L = 0.03$ and the total internal conversion coefficient of $\gamma_3$ is 0.001. The energy of Auger electrons is 1.2–2.1 keV ($e_{AL}$) and 11.6–16 keV ($e_{AK}$; Bé et al., 2004).

Table 1
Results of extrapolations–$^{139}$Ce

| Extrapolation method | Number of sources measured | Slope $k_\varepsilon$ | Activity (kBq/g) |
|---|---|---|---|
| Source treatment with $NH_3$ and aquadag | 15 + 12 + 5 | 0.2001 ± 0.0005 | 531.56 |
| Wet extrapolation | 5 | 0.1998 ± 0.0005 | 532.18 |
| Approximate calculation from coincidence formula | | 0.1989 ± 0.0015 | |

equation is

$$\frac{N_{PC}N_\gamma}{N_C} \approx N_0\left\{1 + s_x\overline{k_\varepsilon} + \underbrace{a_3\frac{1-\varepsilon_{\beta+}}{\varepsilon_{XA}}\left(\varepsilon_{PC\gamma 3} - \frac{\varepsilon_{C3}}{\varepsilon_{\gamma 3}}\right)}_{B}\right\},$$

where

$$s_x = (1-\varepsilon_{XA})/\varepsilon_{XA},$$

$$\overline{k_\varepsilon} = \left(\overline{\varepsilon_{PC\gamma}} - \frac{\overline{\varepsilon_C}}{\varepsilon_{\gamma 3}}\right)\frac{1}{1+a_1p_2} \quad \text{and}$$

$$\overline{\varepsilon_C} = a_1p_1(\varepsilon_{C1} + \varepsilon_{PC\gamma 1}\varepsilon_{\gamma 3}) + a_2\varepsilon_{C2}.$$

For positron detection efficiency $\to 1$, the term $B$ can be neglected and the coincidence formula can be, according to (5), written in the usual form.

The correct implementation of the extrapolation measurement requires that the change of $\varepsilon_{PC}$ has a negligible influence on positron detection efficiency. Calculations were performed to determine that the water layer, which absorbs all Auger electrons, virtually does not affect the positrons (Fig. 1). The influence of the summing effect in the $a_1$ branch was estimated by comparison of two measurements, the first with one NaI(Tl) detector in the $\gamma$-channel, and the second with two detectors. The summing effect in the first case was considerably lower, but the resulting activity was changed only negligibly.

Measurements of five "wet" sources were performed and compared with extra mass addition measurements. A typical progress of efficiency increase during the water droplet drying is shown in Fig. 7.

The occurrence of L-Auger electrons may cause problems in the final phase of the drying process. This can be prevented by setting the LLD in the PC channel high enough to cut off L-electrons, but this reduces the PC detection efficiency by approximately 10%. If the L-electrons are being detected, it is necessary to make sure that a linearity of efficiency extrapolation is achieved for points measured during the final phase of drying.

The results of "wet" and conventional (five sources, extra mass added three times) extrapolation were compared. The results did not differ notably, as the difference

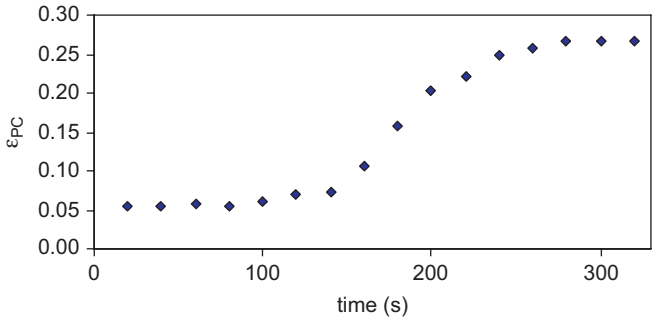


Fig. 7. Typical progress of efficiency increase during the drying of a water drop on a $^{88}$Y source.

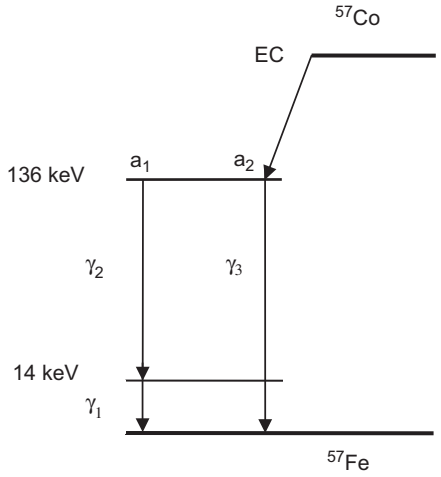


Fig. 8. Simplified decay scheme of $^{57}$Co. The branching ratios for $\gamma_2$ (122 keV) and $\gamma_3$ (136 keV) branches are $a_1 = 0.876$ and $a_2 = 0.123$. The $K$ and $L$ probabilities for EC are about $P_K = 0.888$, $P_L = 0.096$, $\omega_K = 0.352$, $\omega_L = 0.006$ and the total internal conversion coefficients $\alpha_T$ are 0.0236 (122 keV) and 0.148 (136 keV). The energy of Auger electrons is 0.6–0.7 keV ($e_{AL}$) and 5.37–7.1 keV ($e_{AK}$) and energy of conversion electrons is 7.3–136 keV (Bé et al., 2004).

between the resulting activities was 0.15%, while the relative standard extrapolation uncertainty was 0.2% and 0.4% for conventional and "wet" extrapolation, respectively.

### 2.4. The measurements of $^{57}$Co

As mentioned in Havelka and Sochorová (2007), the activity standardisation of $^{57}$Co does not present any difficulties as long as the $\gamma$-window setting is adjusted for identical detection efficiencies of $a_1$ and $a_2$ branches (Fig. 8). The coincidence equation may be written as

$$\frac{N_{PC}N_\gamma}{N_C} \approx N_0\left(1 + \left(\frac{1-\varepsilon_{PC}}{\varepsilon_{PC}}\right)k\right),$$

where the normalised extrapolation slope $k$ is approximated as

$$k \approx P_{ce} + (1-P_{ce})\varepsilon_{PC\gamma 122}\left(1 - \frac{\varepsilon'_{\gamma 122}}{\varepsilon_{\gamma 122}}\right),$$

$$P_{ce} \cong \frac{\alpha_1\alpha_{T122}}{1+\alpha_{T122}} + \frac{\alpha_2\alpha_{T136}}{1+\alpha_{T136}} = 0.0360,$$

$$\varepsilon_{PC} = N_C/N_\gamma,$$

where $\varepsilon_{\gamma 122}$, $\varepsilon'_{\gamma 122}$ are the $\gamma$-detector efficiencies for 122 keV $\gamma$-rays and for 122 keV $\gamma$-rays already detected in the PC, respectively, $\varepsilon_{PC\gamma 122}$ is the detection efficiency of the PC for 122 keV $\gamma$-rays, $\alpha_{T122}$, $\alpha_{T136}$ are the total internal conversion coefficients for 122 and 136 keV $\gamma$-rays, respectively, and $a_1$, $a_2$ are the branching ratios.

It is clear that the slope value is mainly determined by the term $P_{ce}$. The maximum PC detection efficiency of sources treated with $NH_3$ is approximately 80%, hence for

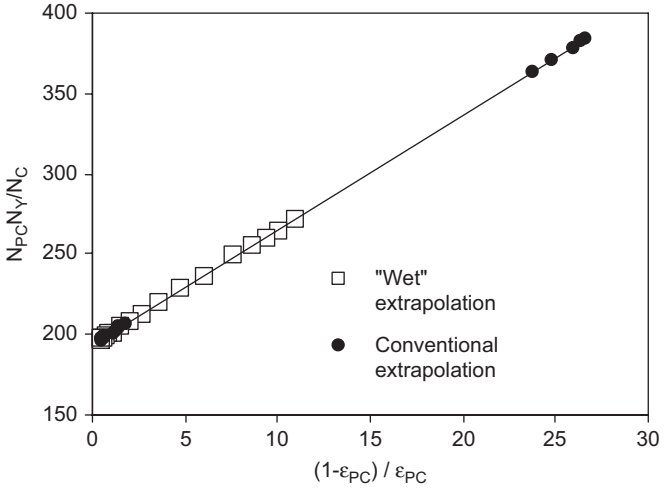


Fig. 9. Efficiency extrapolation curve for $^{57}$Co, using "wet" and conventional variation techniques.

such sources the term $((1-\varepsilon_{PC})/\varepsilon_{PC})k$ should be in the order of 0.1%.

In Fig. 9, two methods of efficiency extrapolation are compared. The conventional extrapolation was performed by measuring five sources directly after drying, after their treatment in $NH_3$ and after the addition of a drop of carrier (20 mg $CoCl_3$/l + 4 g HCl/l). The "wet" extrapolation represents a measurement of one source. The difference between the results of the two methods is negligible. The relative standard extrapolation uncertainty was 0.1% for conventional and 0.15% for "wet" extrapolation.

## 3. Conclusions

Standardisations of four nuclides decaying by EC using the $4\pi$(PC)–$\gamma$ coincidence counting efficiency extrapolation were presented. The "wet" extrapolation method was tested and compared with the conventional methods of source self-absorption variation. For $^{139}$Ce and $^{57}$Co the "wet" method is quite comparable with other methods. For $^{54}$Mn and $^{88}$Y the differences between resulting activity values were lower than corresponding uncertainties, but the relative standard extrapolation uncertainty was significantly higher for the "wet" method (approximately twice the uncertainty of the conventional method). Due to its simplicity, the method should be convenient for routine measurement and for checking the proper extrapolation slope. An indispensable advantage of the method is the possibility to measure a single source repeatedly.

This simple and convenient method of detection efficiency variation has proved to be suitable for activity measurement of above-stated EC nuclides. It can also be used for various other radionuclides, but one has to ensure a sufficient count rate in the coincidence channel and take into account the effect of water on chemical conditions of the source.

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