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Standardization of $^{68}\text{Ge}/^{68}\text{Ga}$ using the $4\pi\beta\text{--}\gamma$ coincidence method based on Cherenkov counting

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Keywords

Radionuclide metrology, Cherenkov effect, Liquid scintillation, $4\pi\beta\text{--}\gamma$ coincidence counting, Geant4 modeling.

Abstract

In the framework of an international BIPM comparison (Bureau International des Poids et Mesures), the activity standardization of ^{68}Ga in a solution of $^{68}\text{Ge}/^{68}\text{Ga}$ in equilibrium provided by NIST was carried out at LNHB. This exercise was organized to meet the growing interest of ^{68}Ga as a radiopharmaceutical in nuclear medicine services (e.g. as a surrogate of ^{18}F for PET imaging). Due to the volatility of ^{68}Ge , the activity standardization of ^{68}Ga was investigated at LNHB by means of $4\pi\beta\text{--}\gamma$ coincidence counting based on Cherenkov measurements. This technique was applied to take advantage of the Cherenkov threshold (~ 260 keV in aqueous solutions) in order to avoid the counting due to electron-capture events

associated to ^{68}Ge disintegrations. Cherenkov counting was performed using glass and polyethylene vials and the resulting activity concentrations were compared with $4\pi\beta\text{-}\gamma$ coincidence measurements based on liquid scintillation. The efficiency-extrapolation curve obtained with Cherenkov measurements in glass vials was compared to Monte Carlo simulations based on the Geant4 code.

1. Introduction

In clinics ^{68}Ga is obtained by elution from a $^{68}\text{Ge}/^{68}\text{Ga}$ generator system. As depicted in the simplified decay-scheme of ^{68}Ge - ^{68}Ga in Fig. 1, the parent radionuclide ^{68}Ge disintegrates by pure electron capture to the ground state of ^{68}Ga (half-life of 270.95 d) with radiation energies having maximum values lower than 10 keV (Bé et al., 2013). The daughter radionuclide ^{68}Ga mainly decays by β^+ emission (~ 88.9 (4) %; maximum energy of ~ 1899 keV; half-life of ~ 67.83 min) that makes it suitable for PET imaging as a surrogate of ^{18}F . ^{68}Ga also disintegrates through electron capture (~ 11.1 %) with maximum energies lower than 10 keV. Due to the growing interest of ^{68}Ga -based radiopharmaceuticals in molecular imaging and nuclear medicine, there was a need for activity standardizations at an international level (Zimmerman and Judge, 2007) under the auspices of the BIPM (Bureau International des Poids et Mesures). For that purpose, an international comparison was proposed and conducted by NIST in 2014. The chemical composition of the $^{68}\text{Ge}/^{68}\text{Ga}$ solution in equilibrium sent to national metrology institutes (NMIs) was optimized at NIST to prevent the potential volatility of germanium (Zimmerman et al., 2016).

Several methods were reported in the literature for the activity standardization of $^{68}\text{Ge}/^{68}\text{Ga}$ (Schönfeld et al., 1994; Grigorescu et al., 2004; Zimmerman et al., 2008; Kulkarni et al., 2017). Because of the volatility of germanium, primary methods based on liquid scintillation (LS) can be preferred to those using dried sources. When the $4\pi\beta\text{-}\gamma$ coincidence method is applied, countings that come from electron-capture events of both ^{68}Ge and ^{68}Ga , are generally avoided by adding thick absorbers on solid sources or by setting a low-level threshold in the β -channel (~ 20 keV). As an alternative to those techniques, the activity concentration of the $^{68}\text{Ge}/^{68}\text{Ga}$ solution provided by NIST was measured at LNHB by means of $4\pi\beta\text{-}\gamma$ coincidence counting directly in aqueous solutions in order to take advantage of the Cherenkov threshold for discrimination purpose.

For several years, Cherenkov counting has been investigated in radionuclide metrology for activity measurements by means of the Triple to Double Coincidence Ratio (TDCR) method. To this end, new models of light emission were developed in NMIs in order to take into account the physical properties characterizing the Cherenkov effect (Kossert, 2010; Bobin et al., 2010). The detection efficiency is directly related to the Cherenkov threshold which depends on the refractive index of the transparent medium where electrons or positrons pass through (~ 260 keV in aqueous solutions). Cherenkov light emission is anisotropic: it is produced from the surface of a cone at an angle with respect to the path of charged particles. In addition, this angle depends on velocity and it diminishes as the particle energy decreases due to Coulomb interactions and radiative losses in the medium. When using a three-photomultiplier system for TDCR measurements, the anisotropy of Cherenkov light emission is at the origin of geometrical dependence on coincidence counting between photomultipliers (PMTs). As depicted in Fig. 2 in the case of Cherenkov measurements with the $^{68}\text{Ge}/^{68}\text{Ga}$ solution, this geometrical phenomenon can be observed by comparing TDCR-Cherenkov counting using glass and polyethylene vials. A shift is clearly observed on the rates of double coincidences as a function of TDCR ratios. Higher detection efficiencies are also obtained with polyethylene vials due to their diffusive wall that modifies reflection and refraction processes occurring at the wall/air interface compared to glass vials. This problem of geometrical dependence in LS counting using diffusing vials was also discussed by Bobin et al. (2012) in the case of LS measurements of low-energy emitters.

Because the accuracy of $4\pi\beta\text{-}\gamma$ coincidence counting can also be affected by geometrical effects when the efficiency-extrapolation technique is applied (Baerg, 1966; 1973), $4\pi(\text{CH})\beta\text{-}\gamma$ coincidence counting based on Cherenkov (CH) measurements was carried out using both glass and polyethylene vials. The activity concentrations were also compared with $4\pi(\text{LS})\beta\text{-}\gamma$ coincidence measurements applied by setting a low-level threshold in the β -channel (~ 20 keV). Finally Monte Carlo simulations of the efficiency-extrapolation curve based on the Geant4 code are compared with the experimental results obtained with Cherenkov measurements in glass vials.

2. Standardization of $^{68}\text{Ge}/^{68}\text{Ga}$

2.1 The $4\pi\beta\text{-}\gamma$ coincidence system for LS and Cherenkov counting at LNHB

In the β -channel, LS counting is implemented using a three-PMT detection system first designed for TDCR measurements (Bobin et al., 2016). This detection apparatus is equipped with XP2020Q PMTs (Photonis) which are symmetrically arranged around an optical chamber containing the LS vial. For each PMT, the detection threshold is usually set below the single photoelectron signal as for TDCR measurements. For $4\pi\beta\text{-}\gamma$ coincidence counting based on LS and Cherenkov measurements, the three-PMT detector is coupled with a 3"x3" NaI(Tl) scintillator detector (76B76/3M, Scionic Holland) in the γ -channel.

As already described in previous studies (Bobin et al., 2007), $4\pi(\text{LS})\beta\text{-}\gamma$ coincidence measurements are usually carried out at LNHB by means of a live-timed anticoincidence instrumentation based on home-made electronics. Countings in both β - and γ -channels are performed using two MTR2 modules based on an electronic circuitry designed to implement the live-time technique using extendable dead times (Bouchard, 2000). In the β -channel, a counting event is triggered when at least a coincidence between two PMTs is obtained (resolving time ~ 80 ns). As usual in an anticoincidence system, the signals provided by a shaping amplifier in the γ -channel are first delayed (2 μs duration). Subsequently, the delayed pulses feed an analog-to-digital converter (ADC 3512, Lecroy) in order to register the coincident and non-coincident spectra. The amplitude analysis is monitored by a MTR2 module implementing the dead time in the γ -channel. The classification of the γ -photon amplitudes according to the two spectra is implemented using additional home-made modules (Bouchard, 2002). The amplitude of a delayed γ -pulse is classified in the coincident spectrum when occurring during a dead time in the β -channel. In the opposite case, the non-coincident spectrum is incremented. The two γ -spectra are used to calculate indirectly the coincidence rates according to anticoincidence counting (Bobin et al., 2007). Regarding the efficiency-extrapolation technique, the detection-efficiency variation in the LS counter is usually carried out by defocusing the PMTs.

2.2 Activity measurements based on LS counting

Six radioactive sources were prepared in standard 20 mL vial, each containing 10 mL of UG cocktail plus aliquots (masses ~ 10 mg) of the master solution of $^{68}\text{Ge}/^{68}\text{Ga}$ provided by NIST (Zimmerman et al., 2016). In order to prevent the counting related to electron-capture events, a detection threshold of about 20 keV was imposed in the β -channel and

controlled using a ^3H source (maximum energy: ~ 18.6 keV). The efficiency-extrapolation technique was implemented by PMT defocusing. Coincidence countings were calculated using a γ -window centered on the 511-keV annihilation yielding experimental $\frac{N_c}{N_\gamma}$ ratios ranging within 0.9 and 0.96 (see Fig. 3). These high-efficiency values correspond to positron interactions in the LS cocktail. The linear fitting of the experimental results was implemented using the conventional straight-line function:

$$\frac{N_\beta N_\gamma}{N_c} = A_{ext} \left[1 - \frac{(1 - N_c/N_\gamma)}{N_c/N_\gamma} \varepsilon_{\beta\gamma} \right] \quad (1).$$

The terms N_β , N_γ , and N_c correspond respectively to the β -, γ - and coincidence counting rates and A_{ext} is the extrapolated activity. As shown in Fig. 3, a low extrapolation slope ($\varepsilon_{\beta\gamma} \sim 0.1$ %) was obtained from the fitting procedure despite the high contribution of 511-keV annihilation photons to the detection efficiency in the β -channel (Bobin et al., 2016). This result can be explained by the counting of self-coincidences resulting from the two 511-keV annihilation photons emitted in cascade following a disintegration and detected in the β - and γ -channels respectively.

The uncertainty associated to the y-intercept (corresponding to the activity related to the β^+ branches) was estimated to 0.03 %. The analysis of the fitting residuals obtained with expression (1) did not reveal a problem of non-linearity. For conservative purpose, the uncertainty attached to the activity concentration was enlarged to 0.08 % in the uncertainty budget (see Tab. 1) in order to take into account the influence of 1077-keV photons in the measurements. The extrapolated y-intercept remains coherent within an uncertainty equal to 0.05 % when using a wider γ -window ranging between 200 keV and 650 keV. The decay-scheme correction (88.88 (41) %) used to determine the activity concentration was modified by the contribution of 1077-keV photon interactions (related to electron-capture disintegrations) in the β -channel estimated equal to 0.14 (2) % using the Geant4 simulation (Bobin et al., 2016).

2.3 Activity measurements based on Cherenkov counting

Two sets of 6 sources were prepared in glass and polyethylene vials filled with 15 mL of carrier solution (65 $\mu\text{g/g}$ of Ge^{4+} and Ga^{3+} in 0.5 M HCl) plus weighed aliquots (masses

~ 10 mg) of the master solution of $^{68}\text{Ge}/^{68}\text{Ga}$. The detection threshold in the β -channel was set below the single-photoelectron pulse as for TDCR measurements. Similarly to LS counting, the efficiency-extrapolation technique was carried out by PMT defocusing for both types of vials. The results displayed in Fig. 4 were obtained in the case of polyethylene vials using a γ -window centered on the 511-keV annihilation peak for coincidence counting. Using these settings, the experimental $\frac{N_c}{N_\gamma}$ ratios ranged between 0.56 and 0.72. The efficiency-extrapolation technique was applied using the same procedure for glass vials. In that case, the experimental $\frac{N_c}{N_\gamma}$ ratios were comprised between 0.53 and 0.68. As expected the maximum detection efficiency in the β -channel is higher for polyethylene vials.

For both types of vials, the linear fitting of experimental data was implemented using the classic expression (1) in a same way as for LS activity measurements. The results obtained in the case of polyethylene vials are presented in Fig. 4. For both glass and polyethylene vials, the extrapolation slopes are respectively $\varepsilon_{\beta\gamma} = 1,6$ (2) % and $\varepsilon_{\beta\gamma} = 2,2$ (2) %. No trend was observed on the shape of the fitting residuals. Compared to LS measurements, the higher values obtained for the slopes were not expected as well as the difference between both types of vials. The uncertainty related to the y -intercept was estimated to 0.1 %. The extrapolated activity remains coherent within this uncertainty value using a wider γ -window ranging between 200 keV and 650 keV. The activity concentrations at the reference date were obtained using a decay-scheme correction (0.8888 (41)) modified by the contribution of the 1077-keV photon interactions in the β -channel (0.10 (2) %) using the Geant4 simulation. The activity concentrations normalized with the LS measurement are compared in Fig. 5. The three values are in good agreement within the uncertainties (the component associated to the decay-scheme correction is not considered). The uncertainty budget in Tab. 2 corresponds to the Cherenkov result obtained with glass vials.

3. Efficiency-extrapolation calculations of $4\pi\beta\text{-}\gamma$ coincidence measurements based on Cherenkov counting in glass vials

3.1 Geant4 modeling

The Geant4 modeling implemented for the simulation of efficiency-extrapolation curves was already described in the case of $4\pi(\text{LS})\beta\text{-}\gamma$ coincidence measurements (Bobin et

al., 2016). The first development of this modeling was carried out to confirm the existence of Cherenkov light emission in a 3-PMT detection system due to electrons created in PMT windows by Compton scattering (Thiam et al., 2010). The Geant4 code was chosen for its capability to simulate the transport of ionization radiation as well as light emission produced by scintillation or Cherenkov effect (Agostinelli et al., 2003). In the present study, the version Geant4.10 was applied using the low-energy electromagnetic physics based on the PENELOPE models (Apostokalis et al., 1999). The geometry modeling of the detection system in both β - and γ -channels is similar to the description already given for $4\pi(\text{LS})\beta\text{-}\gamma$ measurements (Bobin et al., 2016). In the β -channel, optical properties are specified for each element in the optical chamber (aqueous solution, glass vial, PMT window) using the UNIFIED model (Levin and Moisan, 1996). Regarding refraction and reflection processes at medium boundaries, refractive indices and optical transmittances are defined according to photon wavelengths ranging in the spectral sensitivity of the XP2020Q PMT (160 nm-650 nm). The main optical parameters defined for the different elements constituting the optical chamber of the 3-PMT detector in the β -channel are listed hereafter:

- A standard 20-mL borosilicate vial is filled with 15 mL of carrier solution (65 $\mu\text{g/g}$ of Ge^{4+} and Ga^{3+} in 0.5 M HCl). The wavelength-dependent refractive indices were measured at LNHB with a multi-wavelength refractometer DSR- λ (Schmidt+Haensch) as described by Kossert (2013). The refractive indices of the carrier solution are slightly higher than for pure water ($n=1.345$ at 435 nm). The meniscus between the liquid surface and the inner vial wall is also included.
- The wavelength-dependent refractive indices ($n=1.53$ at 400 nm) and the optical transmittance ($\sim 50\%$ at 270 nm) are defined for the borosilicate vial (1-mm wall thickness).
- The vial is hung inside a Teflon spherical cavity (in which the PMT windows emerge) having a surface modeled with a Lambertian-type reflectivity of 95 %.
- The optical properties of the PMT window (fused silica) are specified ($n=1.47$ at 400 nm) as well as the bialkali photocathode ($n=2.7$ at 440 nm) for the calculation of the optical photons refracted at the interface between fused silica and the photoemissive layer. The physical properties of the bialkali photocathode are drawn from experimental data available in the literature: wavelength-dependent refractive indices (Harmer et al., 2006) and quantum efficiencies (Araújo et al., 1998).

In the present study on the standardization of ^{68}Ga with Cherenkov counting, each positron is randomly generated in the 15-mL volume of aqueous solution with energies distributed according to the β -spectra of ^{68}Ga (Mougeot et al., 2010). The transport of Cherenkov photons along the track of positrons is considered in each transparent element of the optical chamber in the β -channel. The number of Cherenkov photons in a spectral region is calculated according to a Poisson distribution determined from the Franck and Tamm theory (Frank and Tamm, 1937). The conversion into photoelectrons in each PMT is computed from the optical photons refracted at the interface between the PMT window and the bialkali photocathode. The calculation is implemented using binomial trials with quantum efficiencies (~ 0.24 in the 300-400 nm region) depending on photon wavelengths (Araújo et al., 1998).

In the γ -channel, the geometry modeling consists of the NaI(Tl) crystal and its housing (1.5 mm for the reflector and 0.5 mm of aluminium). The front of the γ -detector is placed at a distance of 7 cm from the center of the optical chamber of the β -channel.

For each disintegration, the Monte Carlo simulation takes into account the decay-scheme pathway of ^{68}Ga (Bé et al., 2013). For $4\pi\beta$ - γ coincidence calculations, the simulation results consist in the number of photoelectrons obtained in each PMT and the energy released in the γ -detector. The energy resolution of the NaI(Tl) scintillator (7 % at 662 keV) is taken into account by a convolution with a Gaussian distribution. The simulation of the efficiency-extrapolation technique by PMT defocusing is implemented using a second binomial trial applied on the photoelectrons reported for each PMT. This calculation determines the photoelectrons reaching the first dynode leading to a count in a PMT. At least two binomial successes are needed in two different PMTs to trigger a count in the β -channel.

3.2 Simulation results

The results obtained with the Geant4 modeling are based on the simulation of 10^7 events corresponding to disintegrations in the 15-mL volume of aqueous solution. The simulation of the efficiency-extrapolation technique with glass vials was implemented using the same γ -window applied on experimental data (centered on the 511-keV annihilation peak) and by decreasing the focusing parameter from 0.95 to 0.4 for the second binomial trial. The simulated slope ($\varepsilon_{\beta\gamma} = 0,2$ (1) %) estimated from the fitting of the calculated plots using

expression (1) is significantly lower than the experimental result ($\varepsilon_{\beta\gamma} = 1,6$ (2) %). The corresponding y-intercept is equal to 0.894 (1), that is higher than the value based on the decay-scheme parameters used to assess the activity concentration (0.890)

The simulated data were also used by only considering the results corresponding to the maximum experimental values in abscissa ($(1 - \frac{N_c}{N_\gamma}) / \frac{N_c}{N_\gamma} = 0.5$) yielding a normalized activity equal to 0.895 (1). In the case of experimental results, the normalized activity is equal to 0.897 (1) which is closer to the calculated value compared to the deviation of the simulated y-intercept mentioned above. Considering this result, the difference obtained between the calculated and experimental slopes can highlight a problem in the simulated variation of the detection-efficiencies in the transparent parts of the optical chamber when decreasing the calculated $\frac{N_c}{N_\gamma}$ ratios. Indeed, depending on the energies and the position of the charged particles, the emission of Cherenkov photons can occur according to two different Cherenkov thresholds (~ 260 keV for the aqueous solution and ~ 140 keV for the borosilicate glass vial and PMT windows). This difference in emission thresholds can be at the origin of geometrical effects in $4\pi\beta\text{-}\gamma$ coincidence counting yielding to variations in efficiency-extrapolation slopes as observed with LS and Cherenkov measurements. This issue is quite different from the geometrical effects described in the literature when measuring point sources with a proportional counter in the β -channel (Campion, 1959; Williams et al., 1968; Baerg, 1973).

4. Discussion

The activity standardization of ^{68}Ga in a solution of $^{68}\text{Ge}/^{68}\text{Ga}$ in equilibrium (provided by NIST) was performed at LNHB by means of the $4\pi\beta\text{-}\gamma$ coincidence method based on LS and Cherenkov measurements using glass and polyethylene vials. As depicted in Fig. 5, the activity concentrations are in good agreement. The efficiency-extrapolation technique was performed by PMT defocusing. No problem of non-linearity was found from the linear-fitting residuals. Nevertheless significant differences were observed in the extrapolation slopes obtained with the three counting techniques used in the β -channel (LS counting and Cherenkov measurements using glass and polyethylene vials).

The extrapolation slope was calculated using a Geant4-based simulation applied in the case of Cherenkov measurements with glass vials. The simulated slope is significantly lower

than the experimental value obtained by PMT defocusing. Because Cherenkov photons can be emitted according to two detection thresholds (~ 260 keV for the aqueous solution and ~ 140 keV for the borosilicate glass vial and PMT windows), this issue is interpreted as a problem in the simulated variation of the detection-efficiencies in the transparent parts of the optical chamber when decreasing the calculated $\frac{N_c}{N_\gamma}$ ratios. Further investigations on the Geant4-based simulation will be needed in the future in order to reduce the deviation of the calculated extrapolation slope.

The activity standardization based on $4\pi\beta\text{-}\gamma$ coincidence counting using Cherenkov effect is more complex than classical measurements by means of point sources in a proportional counter (Campion, 1959; William et al., 1968; Baerg, 1973). Coincidences between the β - and γ -channels can be affected by geometrical phenomena due to the physical properties of Cherenkov effect: optical photons emitted according a cone, detection thresholds depending on refractive indices of transparent materials. To reinforce this observation, preliminary studies of ^{56}Mn reveal significant differences in extrapolated activities depending on the γ -window used to calculate experimental $\frac{N_c}{N_\gamma}$ ratios. ^{56}Mn is a high-energy multi-branch β - emitter ($\beta_{0,1}, E_{max} = 2849$ keV (56.6 %); $\beta_{0,3}, E_{max} = 1038$ keV (27.5 %); $\beta_{0,4}, E_{max} = 736$ keV (14.5 %)). The deexcitation of ^{56}Fe yields three main γ -photon energies (Bé et al., 2004): 847 keV (~ 98 %), 1811 keV (26.9 %) and 2849 keV (14.2 %). A difference of about 7 % has been obtained when comparing the extrapolated activities given by two different γ -windows (150 keV-1150 keV and 150 keV-3500 keV). This problem could result from the large difference of detection efficiencies between β -branches due to Cherenkov threshold. Usually this situation is not encountered when measuring β - emitters deposited on point sources with a proportional counter. Clearly, this issue shows the need of an accurate Monte Carlo modeling to simulate extrapolation curves and to avoid activity biases in the case of $4\pi\beta\text{-}\gamma$ coincidence measurements based on Cherenkov counting (Fitzgerald, 2016; Bobin et al., 2016).

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Captions

Figure 1: Simplified decay scheme of ^{68}Ge - ^{68}Ga

Figure 2: Comparison of TDCR-Cherenkov counting between glass and polyethylene vials of the $^{68}\text{Ge}/^{68}\text{Ga}$ solution provided by NIST. Because of geometrical effect due to their diffusive wall, better detection efficiencies are obtained with polyethylene vials.

Figure 3: Experimental plots obtained by PMT defocusing for the efficiency-extrapolation technique in the case of $4\pi(\text{LS})\beta\text{-}\gamma$ coincidence measurements. The dashed line represents the fitting with a straight-line function.

Figure 4: Experimental plots obtained by PMT defocusing for the efficiency-extrapolation technique in the case of $4\pi(\text{CH})\beta\text{-}\gamma$ coincidence measurements using polyethylene vials. The dashed line represents the fitting with a straight-line function.

Figure 5: Comparison of activity concentrations obtained by means of $4\pi\beta\text{-}\gamma$ coincidence measurements using LS and Cherenkov counting. The results are normalized with the value given by LS coincidence measurements.

Table 1: Uncertainty budget of activity standardization of ^{68}Ga based on $4\pi(\text{LS})\beta\text{-}\gamma$ coincidence measurements.

Table 2: Uncertainty budget of activity standardization of ^{68}Ga based on $4\pi(\text{CH})\beta\text{-}\gamma$ coincidence measurements using glass vials.

Figure 1

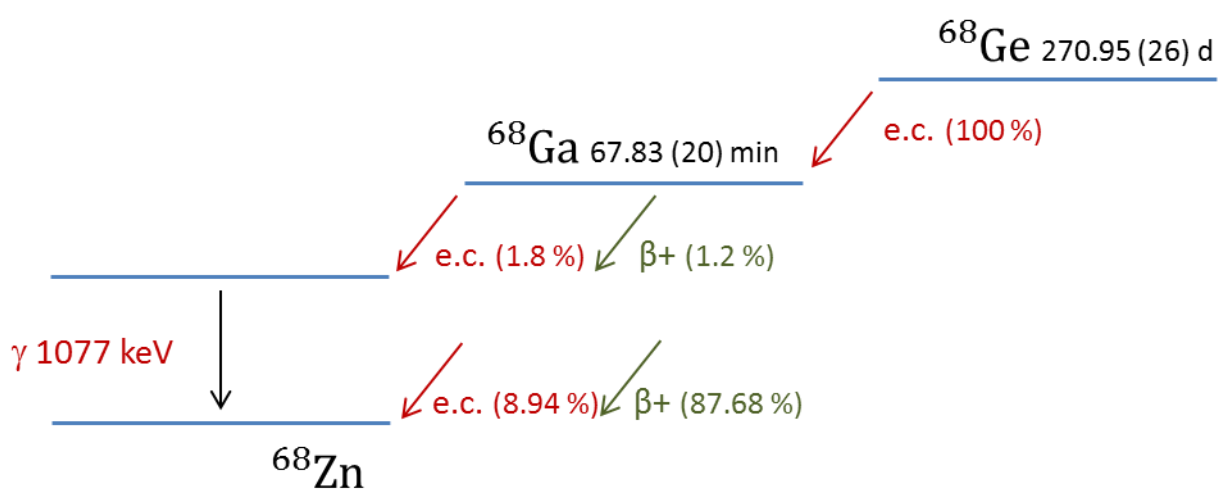


Figure 2

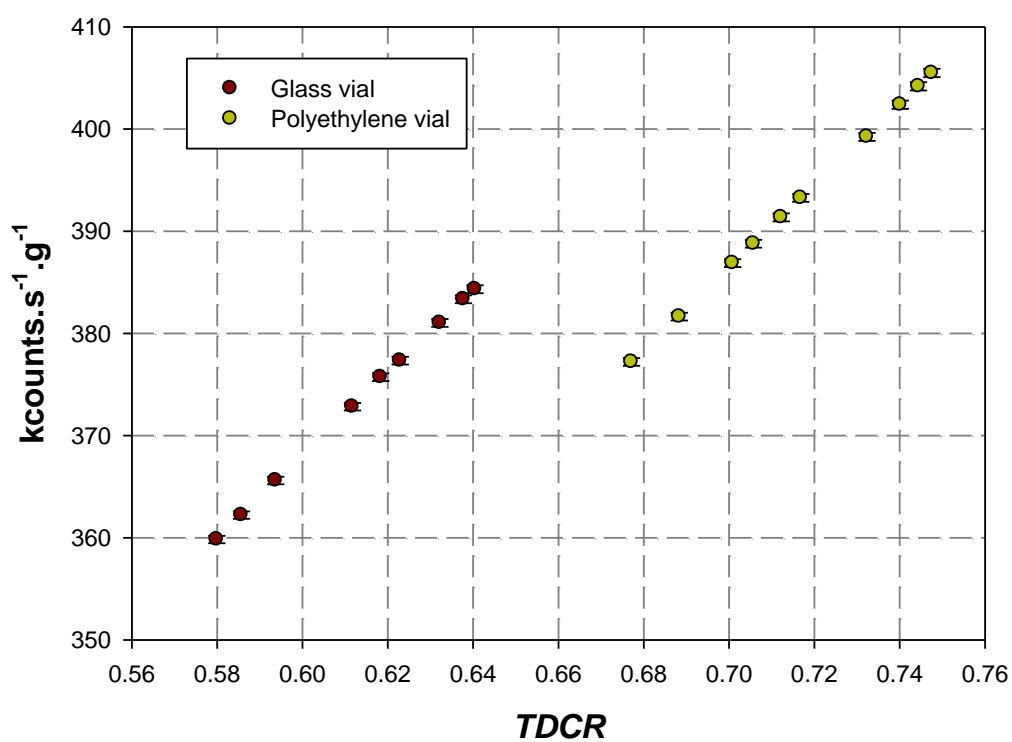


Figure 3

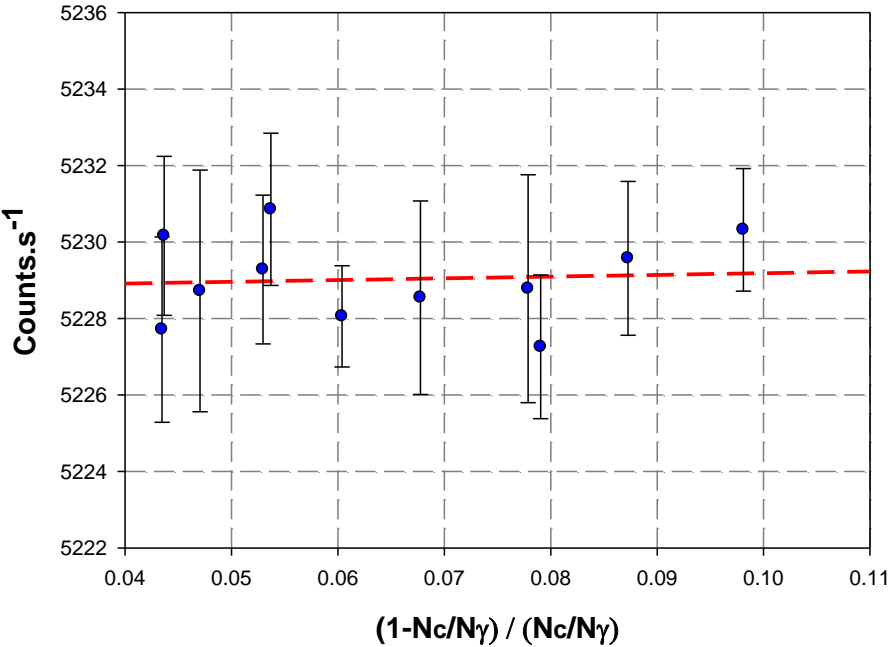


Figure 4

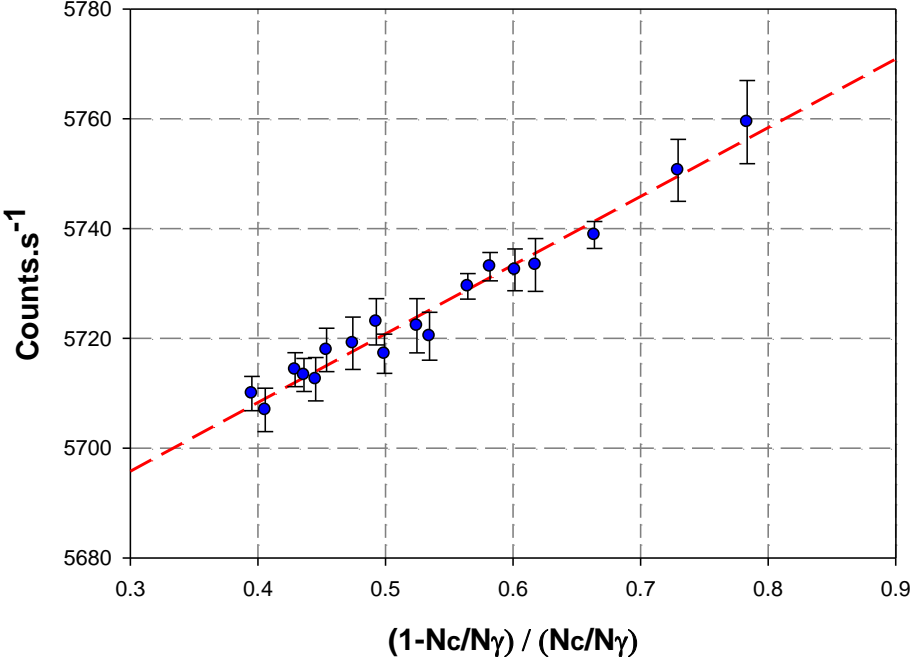


Figure 5

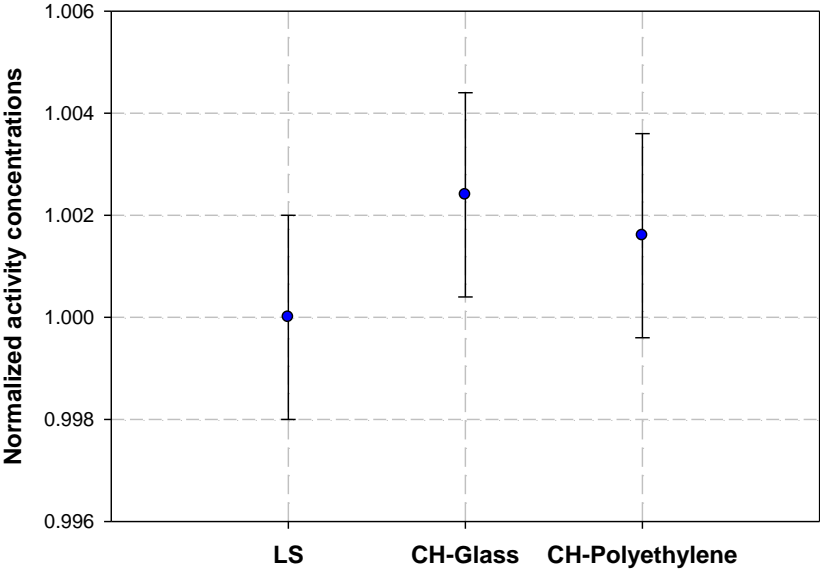


Table 1

Uncertainty component	Comments	<i>u</i> (%)
Measurement variability	Standard deviation of 6 sources measured	0.15
Weighing	Gravimetric measurements (pycnometer method)	0.1
Live time	1-MHz frequency clock used for the live-time	0.01
Decay correction	Half-life of ⁶⁸ Ge: 270.95 (26) d	0.02
Extrapolation technique	PMT defocusing	0.08
Decay-scheme correction	Parameters corresponding to β+ branches	0.46
Background		0.05
Relative combined standard uncertainty		0.5

Table 2

Uncertainty component	Comments	<i>u</i> (%)
Measurement variability	Standard deviation of 6 sources measured	0.12
Weighing	Gravimetric measurements (pycnometer method)	0.1
Live time	1-MHz frequency clock used for the live-time	0.01
Decay correction	Half-life of ⁶⁸ Ge: 270.95 (26) d	0.02
Extrapolation technique	PMT defocusing	0.1
Decay-scheme correction	Parameters corresponding to β+ branches	0.46
Background		0.05
Relative combined standard uncertainty		0.5