

Report on a trial comparison
of activity measurements of a solution of ^{109}Cd

by A. Szörényi

Országos Mérésügyi Hivatal, Budapest, Hungary

Abstract

Activity measurements of a ^{109}Cd solution have been carried out in view of a forthcoming large-scale international comparison. Details on the widely differing methods and detectors used by the six participants are reported. The measured activity-concentration values have a spread of 1.7 % and a standard deviation of the mean of 0.53 %. Some results of the γ -ray-emission rate have also been produced.

1. Introduction

This trial comparison was carried through by the Working Group for advising on future comparisons on behalf of Section II (Mesure des Radionucléides) of the Comité Consultatif pour les Etalons de Mesure des Rayonnements Ionisants (CCEMRI).

As decided during the 1983 meeting of CCEMRI Section II, OMH diluted and bottled the ^{109}Cd solution supplied by the National Accelerator Centre (NAC, South Africa) for the trial comparison.

The ampoules were dispatched from the OMH on 26 November 1984. Each participant (Table 1) received one flame-sealed NBS-type ampoule containing about 2 g of solution. The exact mass was indicated. In addition, BIPM received one ampoule, containing 3.6 g of the undiluted solution, for the International Reference System (SIR).

Table 1 - List of participants

AECL	Atomic Energy of Canada Limited, Chalk River,	Canada
BCMN	Bureau Central de Mesures Nucléaires, Euratom, Geel,	Belgium
LMRI	Laboratoire de Métrologie des Rayonnements Ionisants, Saclay,	France
NPL	National Physical Laboratory, Teddington,	United Kingdom
OMH	Országos Mérésügyi Hivatal, Budapest,	Hungary
PTB	Physikalisch-Technische Bundesanstalt, Braunschweig	Federal Republic of Germany

The ^{109}Cd activity concentration was about 1.2 MBq g^{-1} in an aqueous solution of HCl (0.1 mol per dm^3) with $20 \text{ }\mu\text{g}$ of CdCl_2 per gram of solution.

Purity checks by OMH disclosed no gamma-ray-emitting impurities at the level of about 10^{-6} of the ^{109}Cd activity. For the half life it was proposed to use the value $(462.6 \pm 0.4) \text{ d}$ [1].

A reporting form had been set up and was distributed with the ampoules. The dead line for submitting the results to OMH was 28 February 1985. The results arrived at OMH between 4 March and 3 May 1985.

2. Mass of solution contained in the ampoules, activity concentration from ionization-chamber measurements, adsorption tests

The participants were asked to measure the activity concentration by a calibrated ionization chamber and the mass of solution contained in the ampoules as well as to carry out adsorption tests. The results of these measurements can be seen in Table 2.

3. Source preparation

A summary of technical details reported in the forms is presented in Table 3.

Dilutions were made by AECL, BCMN, LMRI and NPL.

AECL: Dilution was needed for both the ^{109}Cd and the ^{109}Pd solution because of the large difference between the rate of detectable radiations and the differences in detection efficiency for the PC and the $\text{Ge}(\text{Li})$ counting.

NPL: It was necessary to dilute the solution in order to reduce the correction for x-x summing.

4. Activity and gamma-ray emission-rate measurements

A summary of technical details reported in the forms is presented in Tables 4, 5, 6 and 7.

BCMN, LMRI, NPL, OMH and PTB applied 4π proportional detectors for counting the conversion electrons. BCMN, NPL and OMH calculated the activity from the count rate of conversion electrons. Typical spectra are presented on Figure 1. In addition to the measurements of conversion electrons, LMRI and PTB measured also the γ -ray emission rate. AECL used the following method: a solution of $^{109}\text{Pd}(+^{109}\text{Ag}^{\text{m}})$ prepared from irradiated palladium was standardized by $4\pi(\text{PC})$ counting. Sources

prepared from the ^{109}Cd solution distributed by OMH were compared as to the 88 keV γ -ray-emission rate with sources from the standardized ^{109}Pd solution. Both ^{109}Pd and ^{109}Cd decay via the 88 keV level of $^{109}\text{Ag}^m$ by γ -ray emission. This comparison was made with both a Ge(Li) detector and a pair of NaI crystals. Despite the poor statistics of the NaI γ counting, the agreement with the Ge(Li) counting was satisfactory.

In addition to the 4π (PPC) measurement BCMN used also two other methods:

- 4π LS counting of the rate of conversion electrons,
- 4π CsI(Tl) scintillation counting of the rate of conversion electrons and γ photons.

5. Uncertainties

The combined uncertainties and their components are summarized in Table 8.

6. Final results

The results are presented in Table 8 and Figure 2.

BCMN used three different methods for the activity measurement, but the results were not combined into a single result. The three BCMN results have therefore been taken as three independent values, in the calculation of the mean.

The weighted mean of 8 results is $(1\ 187.6 \pm 2.4)$ kBq g^{-1} ,
and the unweighted mean $(1\ 187.9 \pm 6.3)$ kBq g^{-1} ,
at the reference date (1984-12-15, 00 h UT).

The results of this trial comparison show a total spread of 20 kBq g^{-1} (1.7 %).

The deviation of the lowest value from the mean is -1.2 %,
that of the highest value is +0.5 %.

Table 2 - Mass measurements, ionization-chamber measurements and adsorption tests

	AECL 1)	BCM	IMRI	NPL	OMH	PTB
Ampoule number	6991	6992	6993	6994	6990, 6998	6995
Mass of solution (g)	2.0049	2.0018	2.0023	2.0023	2.0009, 2.0024	2.0030
- indicated by OMH						
- determined by laboratory	1.997 ± 0.020	-	-	1.999 51	1.9969, 1.9984	2.0029 ± 0.0002
Activity concentration (kBq g ⁻¹)	1 196 ± 14 2) o	-	1 260 3) o	1 186		
at reference date	1 201 ± 23 t		1 179 t	(1976 cal.) o 1 210 (1979 cal.) o	the chamber was not calibrated	activity too low for IC measurements
Activity remaining in the "empty" ampoule after 2 rinsings with distilled water (Bq)	14 ± 13	-	-	400 4)	200 ± 20	5

- 1) The uncertainty of measurements made with a calibrated ionization chamber includes an allowance for the calibration ($\approx 1\%$) of the ionization chamber. The large uncertainty in the total mass of solution reflects the fact that the amount of solution remaining in the pycnometer was only estimated.
- 2) o - original ampoule
t - transfer to another ampoule
- 3) Geometry conditions different from those used for calibration.
- 4) Three rinsings with carrier solution.

Table 3 - Source preparation

	AECL	BCMN	IMRI	NPL	OMH	PTB
<u>Dilution</u>						
Diluent: μg of CdCl_2 per g of solution	18	20	10	20	3	-
mol of HCl per dm^3 of solution	0.1	0.1	1	0.1	0.1	-
Number of dilutions	2	2	1	2	-	-
Dilution factors	10.100	9.7	73.388 5	72.584	-	-
	47.618	102	$\pm 0.002 5$	57.018		
<u>Source preparation for proportional counting</u>						
Source backing: substrate	VYNS	VYNS	Cellulose	VYNS	VYNS	VYNS
Metal coating	Au + Pd	Au	Au	Au	Au + Pd	Au + Pd
Number of films	1 or 2	1	1	1	1	1
Metal layers: above	0	1	1	1	1	1
below	1 or 2	1	1	1	1	1
Total mass ($\mu\text{g cm}^{-2}$)	15	26	20	50	30 ± 5	55
Drops dispensed onto metal.	No	Yes	Yes	Yes	Yes	Yes
Wetting or seeding agent	Catanac SN	Catanac 1)	-	Catanac 2)	Ludox + Teepol	Ludox SM 3)
Drying	Dry air $50-60^\circ\text{C}$	Air stream	Air	Air	IR lamp	Air
Special treatment	-	-	4), 9)	-	-	5)
Range of source mass (mg)	6)	10 to 15	19 to 26	30 to 80	8 to 12	9 to 11
<u>Source preparation for γ and/or x-ray count.</u>						
Substrate	Polyester tape	VYNS	Mylar	-	-	Polyethylene
	sandwich	Polyimide	(sandwich)			
Ring diameter (outer/inner) (mm)	38/25	64	10			20
" thickness (mm)	0.5	20 to 50 μgcm^{-2}	0.04			0.015
Diameter of active area (mm)	8	4	5			4 to 5
	6.3 mg cm^{-2} 10)	8)				11)

Remarks

- 1) 7 mg per g of solution.⁷ 2) 50 μg per g of solution. 3) 10^{-4} on 4 sources. 4) Electro-deposition of resin (2 sources).
 5) Electro-deposition (4 sources), electro-sprayed resin (8 sources). 6) ^{109}Pd : 0.42 to 18.14 mg, ^{109}Cd : 0.31 to 3.39 mg
 (drop masses/dilution factor). 7) A spot of about 10 mm diameter was wetted with a droplet of Catanac and removed immediately
 thereafter. This left a very thin film sufficient to spread the activity droplet. 8) No metal coating; wetting agent: Catanac
 (7 mg per g); drying in air stream; mass range: 10 to 15 mg. 9) AlCl_3 and drying in NH_3 atmosphere (1 source).
 10) Tape thickness. 11) Precipitated in H_2S atmosphere.

Source preparation for liquid-scintillation counting (BCMN): about 15 mg of solution in 12 cm^3 of scintillator Aqualuma Plus (Lumac)
 pre-loaded with 0.5 μg of CdCl_2 per g of solution.

Table 4 - Equipments for counting

	AECL	BCMN	IMRI	NPL	OMH	PTB
<u>4π proportional counter</u>						
Wall material	stainless steel	Al	Plexi + Au	steel	Al	Al
Height of each half (mm)	21	40	22	120	20	20
Anode						
- Nature	stainless steel	stainless steel	stainless steel	gold-coated W	stainless steel	gold-coated Mo
- Wire diameter (mm)	0.013	0.021	0.02	0.05	0.021	0.1
- Wire length (mm)	36	150	160	250	40	40
- Distance from source (mm)	10	20	20	70	10	10
- Voltage applied (kV)	2.4	2.0 to 2.6	3.5	4.6	3.65	7.3
Gas						
- Nature	CH ₄	Ar + CH ₄				
- Pressure (MPa)	0.1	0.3 to 0.5	1.0	0.5	1.1	1.1
Discrimination level (keV)	≈ 0.1	3.5	≈ 40	31.0	37.0	30.0
<u>Scintillation detector</u>						
		LS	CsI	NaI	-	-
Number of crystals: ordinary	2 NaI	-	2 (sandwich)	1		
well type	None	-	None	Yes		
Diameter (mm)	76	32	52.4	125		
Height (mm)	76	15	25.4	100		
Well diameter (mm)	-	-	-	14.50		
depth (mm)	-	-	-	48.42		
Year of purchase	1976/77	-	1975	1970		
Phototube(s)	75 BOL (SRC)	RCA 8850, EMI 9658R	?	?	-	-
Resolution at 662 keV (FWHM*) (%)	7.2	- 20 (at 60 keV)	?	?	-	-
Solid angle (sr/4 π)	0.7	1	1	0.993	-	-

* full width at half maximum

Table 4 (cont'd)

	AECL	BCMN	IMRI	NPL	OMH	PTB
<u>Semiconductor detector</u>						
Nature	Ge(Li)	-	1) Ge(Li) 2) high-purity Ge	-	-	high-purity Ge*
Type (geometry)	coaxial		1) coaxial 2) planar	-	-	planar
Volume or relative efficiency	nominal 10 %		1) 100 cm ³ 2) 2 cm ³	-	-	length 25 mm diameter 22 mm
Energy resolution FWHM (keV)/ref. energy (keV)	1.4/88		1) 1.2/122 2) 0.54/122	-	-	0.38/5.9 0.570/122
Material and thickness of window (mm)	Al, 0.5		1) Al, 0.5 2) Be, 0.06	-	-	Be, 0.2
<u>Dead times (μs)</u>						
- for PC channel	2.018 ± 0.015**	10.0 ± 0.1	cumulative	15.0 ± 0.2	3.026 ± 0.005	5.000 ± 0.002
- for γ and/or X-ray channel	2.015 ± 0.014 (NaI)	10.50 ± 0.05*** (CsI)	-	-	-	-

* Other detectors: Ge(Li), true coaxial, 5 % efficiency,
Ge(Li), one end open, 11 % efficiency.

** Another channel of electronics was set up by AECL to gate off the γ rays in coincidence with ¹⁰⁹Pd β rays in order to inhibit the counting of bremsstrahlung by the γ-ray channel. The dead time of this channel was the same as that for the PC channel.

*** The measurements by 4πLS counting were carried out with MCA live time.

Table 5 - Counting data for the different methods

	Counting channel window limits (keV)	Typical count rates (s ⁻¹)	Background rates (s ⁻¹)	Number of sources measured	Number of data points, time per point (s)	Time of the measurements
<u>AECL</u>						
PC channel	-	830 to 32 000	0.4 to 1.4	6 ¹⁰⁹ Pd	19 600	
NaI channel	70 to 108	4 to 530	1 to 3	6 ¹⁰⁹ Cd	19 600	85-01-15 to
NaI (anti-coincidence)	70 to 108	4 to 350	1 to 3	6 ¹⁰⁹ Pd	19 600	85-02-08
Ge(Li) for ¹⁰⁹ Pd	85 to 91	0.2 to 580	0.02 to 0.04	6 tape sources		
" for ¹⁰⁹ Cd		6.6 to 14		6 " "	9 6 000	
<u>BCMNI</u>						
LS channel, original sol.	-	12 000	1.2	2	2 2 000	85-03-25 to
" dilution	-	1 200	1.2	3	6 20 000	85-03-30
CsI channel	-	120 to 1 500	37	4	13	85-04-01 to 04-20
PPC channel	3.5 integral	140 to 1 300	3.4	3	15 3 000 to 10 000	85-03-28 to 04-17
<u>LMRI</u>						
PPC channel	40 integral	300	0.8	5	5 5 000	
4π channel	50 to 120	6 to 26	2.014	7	7 60 000	85-02
<u>NPL</u>						
PPC channel	31.0 integral	400	12	7	3 per source 4 000	85-03-05 to 03-15
<u>OMH</u>						
PPC channel	37.0 integral	9 600	0.2	28	26 (2 to 3)×200	84-12-10 to 12-21
<u>PTB</u>						
PPC channel	30.0 integral	9 000	0.6	10	10 1 000	85-02-06
Ge channel	88	50 (integral)	-	4	20 000	85-01-28

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Table 6 - Formulae used for calculating the resultsAECL

$$N_o(^{109}\text{Pd}) = \frac{N(^{109}\text{Pd} + ^{109}\text{Ag}^m)}{\text{EF}},$$

where N is corrected for dead time, background, impurities (^{65}Zn , ^{111}Ag) and decay,

$$\text{EF} = 4\pi(\text{PC}) \text{ efficiency for counting } ^{109}\text{Pd} + ^{109}\text{Ag}^m$$

$$= (1.9444 \pm 0.0095),$$

$$T_{1/2}(^{109}\text{Pd}) = (13.402 \pm 0.006) \text{ h.}$$

$$N_o(^{109}\text{Cd}) = \frac{N_o(^{109}\text{Pd}) \cdot 88 \text{ keV rate (Cd sources)}}{88 \text{ keV rate (Pd sources)}},$$

where the count rates are corrected for losses and decay.

BCMNI

$$N_o = N_{ce} C_{ce} C_{sp} (4\pi\text{LS}),$$

$$N_o = N_{ce+\gamma} C_{sp} (4\pi\text{CsI}),$$

$$N_o = N_{ce} C_{ce} C_{sp} (4\pi\text{PPC}),$$

where $N_{ce+\gamma}$ and N_{ce} are corrected for background and decay (live-time measurements were applied in all cases).

LMRI

$$N_o = N_{ce} + N_\gamma,$$

where N_{ce} is corrected for background and decay (cumulative dead times were applied), and extrapolated exponentially to 0 keV,

N_γ = result of $4\pi\gamma$ counting.

NPL

$$N_o = N_{ce} C_{ce} C_{sp} C_{sum} C_s,$$

where N_{ce} is corrected for dead time, background and decay.

OMH

$$N_o = N_{ce} C_{ce} C_{sp} C_g C_s.$$

where N_{ce} is corrected for dead time, background and decay.

PTB

$$N_o = N_{ce} + N_\gamma,$$

where N_{ce} is corrected for dead time, background and decay and N_γ = result of measurement of γ rate.

Table 7 - Corrections applied for calculating the results

(in parentheses uncertainty in units of the last digit)

	AECL	BCMN	IMRI	NPL	OMH	PTB
<u>Corrections applied</u>						
$C_{ce} = (1 + \alpha_t)/(\alpha_t + \epsilon_\gamma)$		1.0381(5) (LS,PPC) [2]		1.0376 (8)	1.0379 (7)	
$(\alpha_t = 26.4 \pm 0.5)$ [1]		$\epsilon_\gamma = 0.0025$ (25)		$\epsilon_\gamma = 0.008$ (2) at 0.5 MPa	$\epsilon_\gamma = 0.00$ % (5)	
for spectrum extrapolation, C_{sp} below threshold electrons above threshold x rays		1.012 (2) (LS) 1.007 (2) (CsI) 1.001 (1) (PPC)	0.1 to 0.3 %	0.20 % (20)	1.002 (2)	
for source self-absorption (C_s)				0.15 % (15)	1.0003 (3)	
for geometry loss (C_g)					1.0016 (5)	
for x-ray summing, above threshold (C_{sum})				- 0.01 % (1)		
for Ge(Li) losses (dead time, pile up)	1.0185 (21) (max.)					
for 4π counting, linear extrapolation to zero mass						
- for the spectrum between 38 and 50 keV (pile-up suppression) [3]			0.95 % (10)			
- and for the spectrum between 0 and 38 keV [3]			1.2 % (4)			

Table 8 - Uncertainty components of the final result (in %)

Components due to	AECL	BCMN			LMRI		NPL	OMH	PTB
		LS	CsI	PPC	PPC	4πγ			
Counting statistics	0.19	0.1	0.1	0.1	0.08	0.08	0.06	0.06	0.05
Weighing	0.02	0.1	0.1	0.1	0.017	0.01	0.05	0.01	0.02
Dead time	0.01 (NaI/PC)	0.1	0.1	0.1	-	-	0.02	0.005	0.02
Resolving time	-	-	-	-	-	-	-	-	-
Delay mismatch	-	-	-	-	-	-	-	-	-
Pile up	0.01 (NaI)	0.02	0.1	0.1	-	-	0.05	-	-
Background	included in counting statistics	0.02	0.2	0.2	0.004	0.05	0.1	0.01	-
Timing	0.05	-	-	-	0.003	0.01	0.005	0.005	-
Adsorption	0.01	0.05	-	-	-	-	0.02	0.01	-
Impurities	0.14 (in the ¹⁰⁹ Pd sol.)	-	-	-	-	-	0.05	2·10 ⁻⁴	-
Efficiency for PC counting of ¹⁰⁹ Pd + ¹⁰⁹ Ag ^m	0.49	-	-	-	-	-	-	-	-
Source self-absorption	-	-	-	-	-	-	0.15	0.03	-
Uncertainty in C _{Ce}	-	0.05	-	0.1	-	-	0.08	0.07	-
Correction for below-threshold electrons and above-threshold x rays	-	0.2	0.2	0.3	0.23	-	0.2	0.2	0.15
Foil absorption	-	-	0.1	-	-	-	-	-	-
Threshold setting	-	-	-	-	-	-	0.05	0.1	-
Correction for geometry loss	-	-	-	-	-	-	-	0.05	-
Measurement of γ rate by Ge(Li)	-	-	-	-	-	-	-	-	0.06
Half life	-	-	-	-	0.01	0.01	-	0.001	-
Dilution	-	0.1	-	0.1	0.003	0.003	-	-	-
Extrapolation for 4πγ counting (0 to 50 keV)	-	-	-	-	-	0.41	-	-	-
Efficiency for 4πγ counting	-	-	-	-	-	0.02	-	-	-
Combined uncertainty	0.55	0.29	0.4	0.4	0.25	0.42	0.31	0.25	0.17

Table 8 (cont'd)

	AECL	BQMN			LMRI	NPL	CMH	PTB
		LS	CsI	PPC				
FINAL RESULT								
Radioactivity concentration (kBq g ⁻¹) at the reference date (1984-12-15, 00 h UT)	1 184.5	1 194	1 193	1 189.2	1 174	1 190.0	1 189.0	1 189.7
Combined uncertainty (kBq g ⁻¹)	6.5	3.5	5	4.8	3	3.7	3.0	2.2
γ-ray emission rate (s ⁻¹ g ⁻¹) at the reference date					43.73 · 10 ³ (4 π) 43.6 · 10 ^{3*} 43.4 · 10 ^{3*}			43.73 · 10 ³
Combined uncertainty (s ⁻¹ g ⁻¹)					0.18 · 10 ³ (4 π) 0.3 · 10 ^{3*} 0.4 · 10 ^{3*}			0.85 · 10 ³

* Results by γ-ray spectrometry (Ge(Li) and Ge Hp).

Weighted mean of 8 results: (1 187.6 ± 2.4) kBq g⁻¹, at the reference date (1984-12-15, 00 h UT)

Unweighted mean " : (1 187.9 ± 6.3) kBq g⁻¹, " " " "

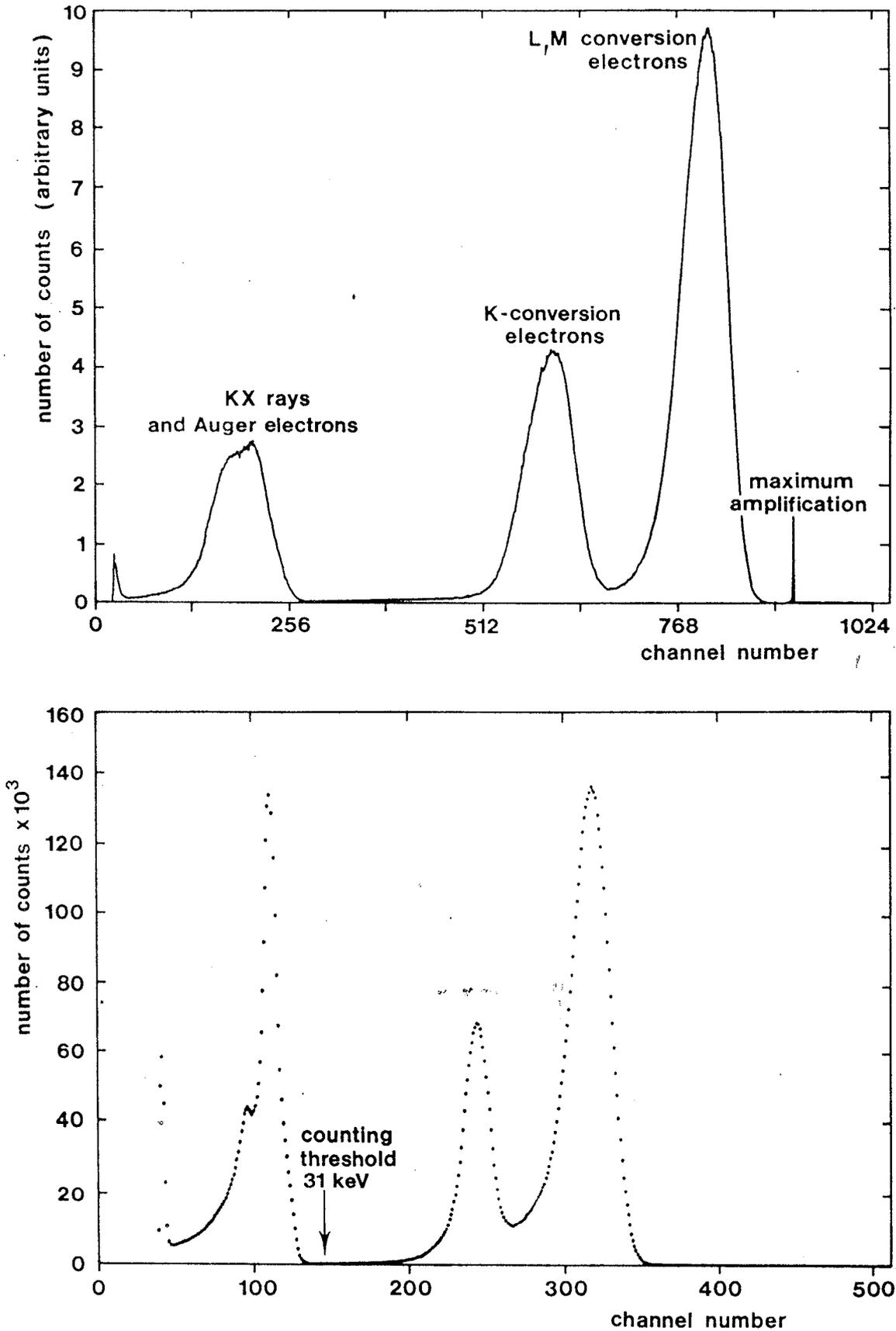


Figure 1 - Typical spectra observed with pressurized 4π proportional counters using a 9 to 1 mixture of Ar-CH₄ at a pressure of 0.3 MPa (BCMn, top) and of 0.5 MPa (NPL, bottom).

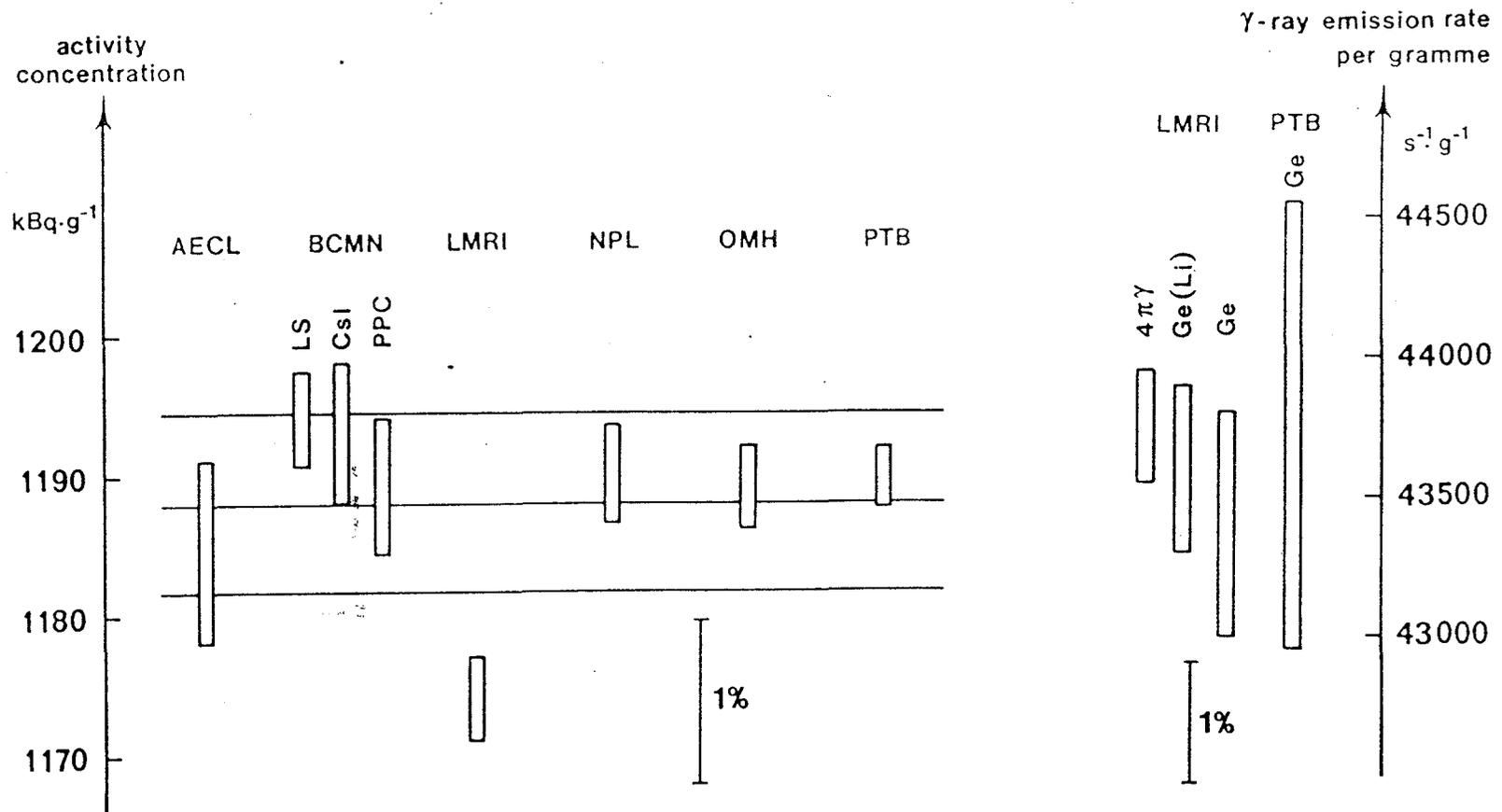


Figure 2 - Graphical representation of the results

References

- [1] Table de Radionucléides, CEA-BNM, LMRI, 1982.
- [2] $P_{\gamma} = 1/(1 + \alpha_t) = 0.0368$ (5), in R. Vaninbroukx, "Emission probabilities of selected gamma rays for radionuclides used as detector calibration standards", paper P52, IAEA Advisory Group Meeting, Nuclear Standard Reference Data, Geel, November 1984.
- [3] J.M.R. Hutchinson and S.B. Garfinkel, "Standardization of cadmium-109 sources for γ -ray emission rate", Int. J. Appl. Radiat. Isot. 22, (1971) p. 405-414.

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