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TRIAL COMPARISON OF ACTIVITY MEASUREMENTS OF A SOLUTION OF ^{75}Se

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Trial comparison of activity measurements of a solution of ^{75}Se

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Abstract

Activity measurements of a ^{75}Se solution supplied by the Physikalisch-Technische Bundesanstalt were performed by five laboratories in the frame of a trial comparison. Different methods were used. Details on source preparation, detectors and counting data are summarized in the report. The measured activity concentration values (based on six results) show a total spread of 8,8 % and a standard deviation of the mean of 1,3 %. The weighted mean value is (0,417 3 ± 0,005 6) MBq g^{-1}.

1. Introduction

At the 1987 meeting of Section II (Mesure des radionucléides) of the Comité Consultatif pour les Etalons de Mesure des Rayonnements Ionisants (CCEMRI), the Working Group advising on future comparisons chose ^{75}Se as a new nuclide to be the subject of a trial comparison. This radionuclide has already been measured with a good precision (apart from one measurement, the precision is better than 0,5 %) using the ionization chambers of the International Reference System for the measurement of gamma-ray emitting nuclides (SIR) (Fig. 1): it is of importance because it has applications in the domains of medicine, industry and agriculture [1]. The numerous and well-defined gamma lines emitted from this radionuclide are often used, for example for the calibration of Ge-Li detectors [2], but the measurement of the absolute activity of ^{75}Se presents serious difficulties. These are due mostly to the presence of a metastable intermediate state of about 17 ms half life in the decay scheme ([3] and Fig. 2), the feeding of which is not well established. Section II recommended [4] that special attention be paid to the contribution of this long-lived intermediate state and that the activity be determined, if possible, by a direct measurement or, after obtaining more reliable coefficients to describe the feeding of this state, by a conventional coincidence measurement.

The PTB kindly took care of the preparation, bottling and distribution of the solution of ^{75}Se required for the comparison. Unfortunately, the time schedule was too tight to allow Amersham-Buchler to supply the necessary amount of activity to schedule. As a result, the reference date of the measurements had to be postponed by about two months. The ampoules were dispatched from the PTB on 24 April 1989. Each participant (Table 1) received a flame-sealed ampoule filled with about 3,6 g of solution.

The ^{75}Se activity concentration was approximately 0,55 MBq g^{-1} in an aqueous and slightly alkaline (pH = 8) solution of Na_2SeO_3 (sodium selenite) at 50 mg l^{-1}. No radionuclide contaminant was detected at the PTB during purity checks. The detection limit for the photons, the emission probability of which is higher than 0,1, was 0,5 %. The recommended half life of (119,8 ± 0,1) d, taken from ref. [3], was adopted by all laboratories. Figure 2 shows the simplified decay scheme of ^{75}Se.

A new version of the reporting form incorporating suggested improvements was mailed to participants on 21 February 1989. The dead line for the submission of the results was originally fixed as 15 April 1989, but problems with the production of the radionuclide made it necessary to postpone it to 25 July 1989. The reference date was therefore chosen as 15 June 1989. Only one result arrived in time at the BIPM: three followed during the summer and the last two reached the BIPM in October. As the arrival of the results was later than expected, no conclusion concerning the trial comparison could be drawn during the 1989 meeting of Section II of CCEMRI.

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

The activity concentration of the ^{75}Se solution was measured by means of a calibrated ionization chamber in three laboratories (LMRI, OMH and PTB). The PTB chamber was calibrated using the efficiency results of the X-Auger (4πPPC)-γ(NaI(Tl)) coincidence measurements obtained by the extrapolation method in 1976 [5]. The AECL ionization chamber (IC) was not calibrated for ^{75}Se, so the activity was calculated from the known IC response as a function of energy. For the emission probability P_γ a recently recommended value [7] was adopted. The total mass of solution in the ampoules was also determined and adsorption tests were performed as required. Data concerning these items can be found in Table 2.

3. Source preparation

Most laboratories used an undiluted solution for the preparation of sources. The AECL used also a diluted solution, as did the PTB, for those sources for which the conventional coincidence method was used. For the NaI(Tl) high-efficiency integral counting, the PTB prepared sources from the undiluted solution. The AECL and the PTB brought the solution to pH 8 by adding appropriate amounts of LiOH and NaOH, respectively. All laboratories used gold-coated foils (Au-Pd coated in the case of the AECL). The BIPM made their sources in sandwich form. The OMH used only one foil

gold-coated on both sides. Three participants (AECL, BIPM and OMH) used $BaCl_2$ and Na_2SeO_3 in order to form barium selenite ($BaSeO_3$). The PTB added hydrazine to precipitate Se in metallic form. Further details on source preparation are assembled in Table 3.

4. Activity measurements

Measurements of the activity concentration of the ^{75}Se solution were performed by two independent methods. Each laboratory used the well-known coincidence method, for which the photons detected in an appropriate window of the γ channel, the events in the β channel (for ^{75}Se mainly delayed conversion electrons of 12,61 keV, prompt Auger electrons K of 9,11 keV and prompt K-X rays of 10,66 keV) and the coincidence between these two channels are registered. This method implies the use of two corrections: the first consists in an extrapolation to efficiency 1, the second reduces the count by allowing for delayed events which have not been detected in coincidence with the prompt ones. This point will be discussed in section 6. In order to perform the efficiency variation required by this method, four laboratories used a pressurized proportional counter, but with different techniques: the BIPM and the PTB varied the pressure between 0,4 and 1,5 MPa and 0,1 to 3,1 MPa, respectively; the OMH worked at 1,2 MPa and obtained the efficiency variation by adding gold-coated foils to the sources; the LMRI used a pressure of 0,91 MPa, with a threshold variation as extrapolation method. The AECL, as is their custom, maintained atmospheric pressure in the counter but changed the voltage over the range 1,8 to 2,6 kV [10].

Events in the γ-ray channel were detected at the AECL, BIPM and OMH by means of conventional NaI(Tl) detectors, whereas the PTB used a well-type NaI(Tl) detector.

The PTB also used the $4\pi\gamma$[NaI(Tl)] high-efficiency integral counting described in [8, 9]. A sketch of the experimental set-up is shown in Fig. 3. In this method, when the ^{75}Se radionuclide detection efficiency is calculated from decay-scheme data and detector efficiency, the isomeric state is treated as a separate nuclide. The activity is then determined from the relation $A = N/\varepsilon$, where N is the integral count rate measured and ε is the average detection efficiency for a decay of ^{75}Se. The efficiency was evaluated using the calibration curve of the detector in terms of the γ-ray energy and decay scheme data taken from [3]. Decays via the isomeric state are also taken into account. Detection efficiencies for the main decay paths - prompt or delayed - remain close to 1 and as a consequence, to a first approximation, the percentage of EC decays to the

isomeric state does not have to be known. However, two important assumptions have to be made in computing the activity value:

1. There is no electron capture directly to the ground state,
2. The feeding of the isomeric state by the 97 keV transition is (6,60 ± 0,15) % and its γ-ray emission probability is (3,48 ± 0,07) %.

Further details on the counting equipment and the counting data are listed in Tables 4 and 5.

5. Corrections

All participants corrected the observed rates for dead time, background, mass of the source and accidental coincidences. In addition, the AECL corrected the measured PC-γ coincidence rates for delay mismatch. The PTB indicated that the results obtained by means of the 4πβ-γ coincidence method depend on the slope of the fitted linear function. This effect is due to self absorption in the source reducing the efficiency of 13,5 keV conversion electrons from the decay of the metastable state. To estimate the self absorption of conversion electrons, the efficiency for K-Auger electrons has been measured via the capture efficiency from which the X-ray efficiency is subtracted as $P_K\omega_K$ for infinitely high gas pressure in the proportional counter. Both the K-Auger electron efficiency and the measured activity values show a linear dependence on the number of VYNS-foils placed on the source to vary the efficiency. This allows to extrapolate the function of activity values to the (negative) number of VYNS-foils corresponding to the case of efficiency equal to one suggesting to lower the result of the discriminator method by 1,2 %. This should be considered as a lower limit for the activity value, since on the average the conversion electrons from the decay of the metastable state have energies 50 % higher than the K-Auger electrons and may undergo less absorption than assumed.

6. Correction for the number of delayed events per decay

To obtain the true value for the activity of the radioactive ^{75}Se solution, it is necessary to correct the extrapolated values N_0 for the influence of the delayed events which have been registered in the β channel with the prompt ones. Some years ago Lewis *et al.* [11], by a correlation method, determined a value of (5,40 ± 0,35) % for this correction. This is in good agreement with the value (5,5 ± 0,5) % deduced from the nuclear decay

scheme data given in [12, 13, 14]. However, more recent decay-scheme evaluations, e.g. [3], suggest values as high as 7,4 %.

- The AECL recalculated the number of delayed conversion electrons on the basis of the nuclear data listed in [3] for two windows set on the γ channel. For the range 15 to 340 keV they found a value of (8,1 ± 1,5) %, whereas for the range 340 to 635 keV the value was (7,4 ± 1,5) %.

- The BIPM used for this correction the experimental value of 5,4 % taken from [11].

- The LMRI performed similar calculations taking the values P_K = 0,875 ± 0,003, and from [3] the values α_K = 167 ± 5 and α_t = 211 ± 7 for γ_1 = 24 keV. The γ emission probability $I_{\gamma 1}$ = 0,000 30 ± 0,000 06 was taken from an updated evaluation of A. Nichols for γ-ray standards [15]. This yields a value of (5,9 ± 1,2) % for the correction under the assumption that the detection efficiency of the conversion electrons is negligible. In order to approach this condition the sources were sandwiched and the assumption was partly tested using sources covered with additional absorbing foils.

- The OMH established the contribution of the delayed fraction to the total count rate in the EC channel by measuring weak ^{75}Se sources of 55 and 110 Bq, respectively, with increasing dead times of the extending type, the values of which were close to the half life of the isomeric state.

- The PTB applied the correlation method of Lewis *et al.* [4] to determine the fraction of delayed events in the proportional counter caused by transitions from the 304 keV isomeric level of ^{75}As. The experimental set-up is shown in Fig. 4. The experimental data leading to the determination of the fraction of delayed events are presented in Fig. 5. On this figure the ratio $(N_{pr}+N_{del})/N_{pr}$ is plotted versus $(1-\varepsilon)/\varepsilon$, where N_{pr} and N_{del} are the numbers of the prompt and the delayed events, respectively. The experimental data could be fitted by a straight line in the range $0 < (1-\varepsilon)/\varepsilon < 2$ and an extrapolation to 0 gave a value of (6,5 ± 0,2) % for the required parameter. Assuming the extrapolation function being curved this value is a lower limit for the fraction of detectable delayed events.

As may be seen, the estimates of the fraction of delayed events detected with the prompt ones range from (5,40 ± 0,35) % to (8,1 ± 1,5) %. The uncertainties of assessment differ greatly from one laboratory to another because the crucial datum, the intensity of the 24 keV transition, has not been well measured directly. As a consequence, it is

necessary to compare the values of the activity before and after applying the correction so as to have a suitable basis for judging the quality of the measurements.

7. Uncertainties

The combined uncertainties and their components on the final result are given in Table 7. The uncertainties in the experimental extrapolated value N_0, including the delayed events, are also listed. The total combined uncertainties refer to the value of the activity concentration and include the contribution due to the decay scheme parameters.

The following tentative conclusions can be drawn:

1. In the case of the β-γ coincidence method, the uncertainties due to the extrapolations and to the corrections for the delayed events give the main contributions to the total uncertainties for the AECL, the BIPM and the OMH. The PTB found that the main contribution to the uncertainty stemmed from self-absorption in the source. The LMRI did not report a value for the component due to the extrapolation.

2. In the case of the high γ-efficiency method, counting statistics and decay-scheme parameters are the main sources of uncertainty.

The indicated total uncertainties are in most cases larger than or equal to 1 %. Only the OMH found a lower value (0,5 %). This seems to indicate that the laboratories are not very confident of their results. Moreover, as noted earlier, the use of the value for the fraction of delayed events significantly increases the overall uncertainty. This confirms the PTB suggestion that the coincidence method will not be the best for determining the activity concentration of ^{75}Se until the value for the fraction of delayed events is more accurately known.

8. Final results

To obtain the activity concentration of the solution of ^{75}Se, different extrapolation methods were used. The AECL performed an extrapolation to efficiency $\varepsilon_x = 1$ by using polynomials of the first and the second order for each source and for two different windows. They then calculated a weighted mean value for each case. The BIPM made an extrapolation for all sources to efficiency 1. The LMRI performed an extrapolation for each source and evaluated an unweighted mean of the results obtained from their five

sources. The OMH performed a second-order extrapolation for each source and then calculated an unweighted mean from the individual results. The PTB used the coincidence method with just one source for the evaluation of the activity concentration. For the high γ-efficiency method, the final result is the unweighted mean of eight runs (each consisting of ten individual measurements of 100 s) with four sources.

The results are presented in Table 8 and in Figure 6. They are given before and after applying the correction for the fraction of delayed events detected with the prompt ones. All laboratories - with the exception of the BIPM - found similar activity concentrations and the agreement between their results is not affected by the correction for the delayed events. Measurements with the high γ-efficiency technique give results compatible with those obtained at the AECL, the LMRI, the OMH and the PTB after correction for the delayed events.

The complete set of results may be combined to provide weighted and unweighted means for the reference date of 1989-06-15, 0 h UT. Both means are listed in Table 9. The value used for the AECL in this computation is the weighted mean value of the two individual results which have been obtained with a second-order least squares fit and seem to be the most reliable ones. Table 10 shows the same quantities, without the BIPM results. In this case, the agreement is much better and, as expected, the uncertainties are smaller (up to three times). The reasons for the discrepancy between the BIPM results and those of all other laboratories have not been explained yet and further measurements are necessary.

The results of this trial comparison cover a total range of 44,7 kBq g^{-1} (10,1 %) and the deviations of the lowest and the highest values from the weighted mean are - 7 % and + 3,1 %, respectively, when no correction for the delayed events is applied. It follows that for the activity measurements the total range is 36,9 kBq g^{-1} (8,8 %) and the greatest deviations from the weighted mean are - 6,1 % and + 2,7 %.

Table 1 - List of the participants and names of the persons who carried out the measurements

AECL Atomic Energy of Canada Limited, Chalk River, Canada
(R.H. Martin)

BIPM Bureau International des Poids et Mesures, Sèvres, France
(P. Bréonce, C. Colas, G. Ratel and C. Veyradier)

LMRI Laboratoire de Métrologie des Rayonnements Ionisants, Saclay, France
(P. Blanchis, J. Bouchard and B. Chauvenet)

OMH Országos Mérésügyi Hivatal, Budapest, Hungary
(M. Csikós, Gy. Horváth, Á. Szörényi and A. Zsinka)

PTB Physikalisch-Technische Bundesanstalt, Braunschweig, Federal Republic of Germany
(U. Schötzig, $4\pi\gamma$ high efficiency counting;
E. Funck and R. Klein, coincidence method;
H. Janssen, correlation method)

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

Laboratory	AECL	BIPM	LMRI	OMH	PTB
Ampoule number	89-391	89-387	89-388	89-390	89-386
Mass of solution (g)					
- indicated by PTB	3,600 9	3,605 7	3,596 6	3,600 6	3,654 5
- determined by laboratory	3,581 9	3,598 9 (1) 3,595 5 (2)	3,596 0	3,587 1	3,654 5
Ionization chamber measurements					
Activity concentration (3) ($kBq\ g^{-1}$) at ref. date (1989-06-15, 0 h UT)	438 ± 15 (4)		418,0	426,2	426,27 (5)
Date of measurement	(1989-05-08)		(1989-07-27)	(1989-05-02)	(1989-04-14 to 1989-04-21)
Activity concentration (6) ($kBq\ g^{-1}$) at ref. date (1989-06-15, 0 h UT)			417,7	426,5	426,33
Date of measurement			(1989-08-02)	(1989-05-02)	(1989-04-24)
Adsorption tests					
Activity remaining in the "empty" original ampoule after 2 rinsings with 5 cm^3 of distilled water (Bq)	22 ± 4	0	144	42 ± 11	
Additional rinsings	0	0	2	2 (7)	
Final residual activity (Bq)			44	37 ± 9	
Date of test	(1989-05-11)	(1989-05-12)	(1989-08-03)	(1989-05-03)	
Measuring instrument used	Ge spectrometer	ionization chamber	Ge-Li spectrometer	calibrated Ge-Li spectrometer	

Table 2 (continued)

(1) With ampoule.

(2) With pycnometer.

(3) Before opening the original ampoule.

(4) The ionization chamber was not calibrated for this nuclide.

(5) The ionization chamber efficiency results from X-Auger (4πPPC)-γ(NaI(Tl)) coincidence measurements in 1976 [6]. The fraction of the detected conversion electrons following the isomeric 304 keV state was assumed to be 5,4 % [7] and subtracted from the result of the coincidence measurement. The results of the present measurement were not taken into account for this determination.

(6) After transfer to another ampoule.

(7) With diluent (50 µg Na_2SeO_3 per g of distilled water).

Table 3 - Source preparation

Laboratory	AECL*	BIPM	LMRI	OMH	PTB**
Dilution					
- Diluent	distilled H_2O + LiOH pH = 8			50 µg Na_2SeO_3/g of distilled water	45 µg ml^{-1} Na_2SeO_3 in aqueous solution adjusted to pH 8 with NaOH
- Number of dilutions	1				1
- Dilution factors	1,0 2,600 4	1,0		1,0	1,0 10,19
Source preparation			(2)		
- Source backing (substrate)	VYNS	VYNS (1)		VYNS (≈ 20 µg cm^{-2})	
- Metal coating	Au-Pd	Au		Au	Ag-Pd (≈ 15 µg cm^{-2})
- Number of films	1	2		2 (3)	1
- Metal layers					
above		1		1	1
below	1	1		1	1
- Precipitation	$BaCl_2$ 90 µg ml^{-1}, 1 to 3 drops	$BaCl_2$ 100 µg g^{-1}			(5)
- Disposed mass of solution (mg)	15 to 53 16 to 50	20 to 100	50	15	11 8
- Drying	dessicator or laboratory air at T = 25 °C laboratory air	air		dry silicagel at room temperature	air drying
- Type of balance used	M5 Mettler	Sartorius 4504MP8	M5 Mettler	M5 Mettler SA	ME22 Mettler
- Number of sources prepared	9 5	24	10	26 (4)	6 (6) 4 (7)

* When there are two values in this column the figure on the left-hand side correspond to the dilution factor 1 and those on the right-hand side to the dilution factor 2,600 4.

** When there are two values in this column the figures on the left-hand side correspond to the high γ-efficiency method and those on the right-hand side to the coincidence method.

Table 3 (continued)

(1) The sources were sandwiched.

(2) Two sets of 5 sources were prepared.
a) 2 Au-coated mylar (250 μg cm^{-2} mylar + 50 μg cm^{-2} Au coating).
b) 2 Au-coated thin films (40 μg cm^{-2} collodion + 40 μg cm^{-2} Au coating).
Only the first set was used to evaluate the activity because the coatings of these sources are assumed to cut the Auger and conversion electrons from the 24 keV transition and therefore to allow one to perform a linear extrapolation. The other set was used in order to look at non-linearities which are due to the variation of detection efficiency for conversion electrons.

(3) For the efficiency variation further foils were added.

(4) - 16 sources were prepared following the recommended method 1 [8] applying 60 μg $BaCl_2$/g of distilled water.
- 10 sources were prepared using an ion exchange resin as basis for the radioactive material. The procedure was as follows: anion- and cation-exchanging resins were deposited onto the usual VYNS foils; radioactive material was dropped onto the resin and dried by silicagel in a desiccator; distilled water was dropped onto the dried material to help it to spread uniformly over the active spot; sources were again dried.

(5) Aqueous solution of 60 μg ml^{-1} hydrazine ($N_2H_4 \cdot H_2SO_4$), one drop of about 10 mg per source.

(6) Prepared on 1989-05-05.

(7) Prepared on 1989-07-12.

Table 4 - Equipment for electron counting

Laboratory		AECL	BIPM	LMRI	OMH	PTB
Proportional counter						
Wall material		stainless steel	stainless steel	perspex	aluminium	aluminium (3)
Height of each half	(mm)	21	20	25	20	20
Anode						
- Nature		W(Au-coated)	stainless steel	stainless steel	stainless steel	stainless steel
- Wire diameter	(mm)	0,013	0,1	0,020	0,025	0,05
- Wire length	(mm)	36	36	170	40	40
- Distance from source	(mm)	10	10	12,5	10	10
- Voltage applied	(kV)	1,8 to 2,6 (1)	4,5 to 9,22	3,2	4,69	1,5 to 10,2
- Solid angle		4π	4π	4π	4π	4π
Gas						
- Nature		CH_4	Ar/CH_4 (9:1)	Ar/CH_4 (9:1)	Ar/CH_4 (9:1)	Ar/CH_4 (9:1)
- Pressure	(MPa)	0,1	0,4 to 1,5	0,91	1,2	0,1 to 3,1
- Discrimination level	(keV)	0,1 to 5	(2)	2 to 10	> 1	0,5 to 10,0

(1) The voltage was varied to produce an effective change in discrimination level.

(2) A window was set in order to count events with energy around 10 keV, e.g. prompt K-X rays (E_{XK} = 10,66 keV), prompt Auger electrons (E_{eAK} = 9,11 keV) and delayed conversion electrons ce_{1K} (E_{1K} = 12,61 keV).

(3) Of cylindrical form.

Table 5 - Counting equipment for X and γ rays

Laboratory		AECL	BIPM	LMRI	OMH	PTB (1)	PTB (3)
Scintillation detector (channel 1)							
Crystal material		NaI(Tl)	NaI(Tl)	NaI(Tl)	NaI(Tl)	NaI(Tl)	NaI(Tl)
Number of crystals							
- ordinary		2	2	1	1		
- well type						2	1
Diameter	(mm)	76	76	76	76	152 (2)	152
Height	(mm)	76	76	76	76	102	152
Well diameter	(mm)					51	50
" depth	(mm)					19	100
Resolution (FWHM*)	(%)	7,3	7	8,2	7,8 and 9,4	8,9	
	(keV)	48	46	46	54	51,6 and 62	59
at ...	(keV)	662	662	662	662	662	662
Solid angle	(sr/4π)	0,425	0,688			1,0	0,7
Distance from photon counter to source	(mm)		25	≈ 30	22		25

* Full width at half maximum.

(1) Equipment for 4πγ high-efficiency integral counting [8].

(2) A sketch of the detector arrangement is shown in Fig. 3.

(3) Equipment for the coincidence method.

Table 6 - Counting data for the different methods

Laboratory		Counting channel window limits /(keV)	Typical count rates /(s^{-1})	Background rates /(s^{-1})	No of sources measured	Typical time for one measurement /(s)	Dead times /(μs)		Coincidence resolving time /(μs)	
AECL										
- channel 1	(1)	Lo (2) 15 to 340	3 700	5,0 ± 0,3	14	400	2,50 ± 0,04	(4)	0,703 ± 0,005	(4)
		Up (3) 340 to 635	340	1,5 ± 0,2	14	400	2,49 ± 0,04	(4)	0,700 ± 0,005	(4)
- channel 2	(5)	0,1 to 5	1 250	0,4 ± 0,1	14	400	2,49 ± 0,05	(4)	0,703 ± 0,005	(4)
							2,48 ± 0,05	(4)	0,700 ± 0,005	(4)
BIPM										
- channel 1	(1)	15 to 340	360 to 2 100	2,2	15	500 to 1 000	10	(8)	0,900	(9)
- channel 2	(5)	(6)	3 900 to 15 200 (7)	1,1	15	500 to 1 000	10	(8)		
LMRI										
- channel 1	(1)	366 to 438	170	3,9	5	2 000	25	(10)	1,50 ± 0,01	
- channel 2	(5)	2 to 10	4 600	3,7	5	2 000	25	(10)		
OMH										
- channel 1	(1)	340 to 450	200	1,6	26	2 x 1 000	3,002 ± 0,005	(11)	1,078 ± 0,010	
- channel 2	(5)	≈ 1	4 800	0,7	26	2 x 1 000	3,004 ± 0,005	(11)		
PTB										
- method 1	(12)	12 (13)	2 000	35	4	100	3			
- method 2	(14)									
- channel 1	(1)	380	2 347	27,6	1	4 000	5,000 ± 0,015	(15)	1,000 ± 0,003	(9) (16)
- channel 2	(5)	0,5 to 10,0	4 000	0,55	1	4 000	5,000 ± 0,015	(15)		
- method 3	(17)	350 to 460	5	< 0,7	4	9 000	5,12 ± 0,03	(15)	1,64 ± 0,02	(9)
		2 to 15 (18)	100	< 1	4	9 000	5,06 ± 0,03	(15)		

Table 6 (continued)

(1) Measurement with a scintillation detector.

(2) Lo = lower window.

(3) Up = upper window.

(4) The source-pulse method was used for this measurement.

(5) Measurements performed with a proportional counter.

(6) A window was set so as to count every event with energies of about 10 keV as explained in the text.

(7) Values at pressure P = 1,0 MPa for the least and the most active sources respectively.

(8) Non-extended numerical dead time measured by means of the two-oscillator method.

(9) Measured by means of the two-oscillator method.

(10) Extended dead time common to channels 1 and 2.

(11) The dead times were measured by means of a random coincidence simulator.

(12) $4\pi\gamma$ high-efficiency counting.

(13) Lower level.

(14) Measurements performed with sources obtained with dilution 2.

(15) The dead times were determined by the two-oscillator method.

(16) A pulse synchronizer was used to match delays between channels 1 and 2.

(17) Correlation method.

(18) The discrimination level varied during the measurements.

Table 7 - Uncertainty components of the final result (in %)

Components due to	AECL	BIPM	LMRI	OMH	PTB	
					high γ effic. method	coincidence method
counting statistics	0,05 (1)	0,03	0,07	0,05	0,1	0,02
weighing	0,02 (2)	0,03	0,003	0,005	0,05	0,02
dead time	0,01 (3)		$< 10^{-5}$	0,005	0,05	0,014
background	(4)	(4)	(13)	0,005	0,02	0,1
pile-up	(5)					
timing	0,01 (6)		0,003	0,005		
adsorption	< 0,01 (7)			< 0,01	0,001	
impurities	< 0,01 (8)			< 0,005		
extrapolation uncertainty	1,05 (9)	0,5 (11)		0,2	0,01 (14)	
decay scheme parameter	1,40 (10)	0,33	1,2	0,47	0,3 (15)	
$T_{1/2}$	0,02	0,02				
resolving time				0,01		
delay mismatch				0,04		
self absorption						1,0
combined* uncertainty	1,05	0,5 (12)	0,07	0,21	0,32	1,0
total combined uncertainty	1,75	0,60	1,2	0,52	0,32	1,0

* Without uncertainty on decay scheme parameters.

Table 7 (continued)

(1) Error in intercept from fit of 210 individual data.

(2) Balance calibration.

(3) Calculated effect of 1σ variation in dead time.

(4) Included in counting statistics.

(5) Included in extrapolation.

(6) Manufacturer's specification.

(7) Relative γ counting with a Ge detector.

(8) From Ge counting and supplied data.

(9) Difference between first and second order fits.

(10) Uncertainty in delayed state correction.

(11) For a first order extrapolation.

(12) The uncertainty on P_e = 0,054 0 ± 0,003 5 [10] is not included.

(13) Included in the fitting procedure.

(14) Extrapolation of count rate from 12 keV threshold to zero energy.

(15) Influence of decay scheme uncertainties on the radionuclide detection efficiency.

Table 8 - Final results

	AECL (3)	BIPM	LMRI	OMH	PTB method 1	PTB method 2
Date of measurement	(1989-05-14)	(1989-05-24 to 1989-07-27)	(1989-08-08)	(1989-05-04 to 1989-05-25)	(1989-07-31)	(1989-05)
Activity concentration (1) ($MBq\ g^{-1}$) at ref. date (1989-06-15, 0 h UT)	0,473 5 (4)	0,413	0,443 7	0,452 3	N/A	0,445 2
Combined uncertainty (1) ($MBq\ g^{-1}$)	0,000 9	0,002	0,000 3	0,000 9	N/A	0,004 4
Correction for delayed events	1,074 ± 0,015 (5)	1,054 0 ± 0,003 5	1,059 ± 0,012	1,055 ± 0,005		1,065 ± 0,002
Maximal achieved efficiency	0,26	0,47 (7)	0,40	0,71		0,73
Number of degrees of freedom	207 (Lo) (4) 221 (Up) (4)		29	7		6
Order of extrapolation	2	1	1	2		
Method used for extrapolation	voltage variation	pressure variation	threshold variation	sources were covered with successive VYNS foils		discrimination variation
Activity concentration (2) ($MBq\ g^{-1}$) at ref. date (1989-06-15, 0 h UT)	0,440 9 (6)	0,391 8 (8)	0,419 (9)	0,428 7 (10)	0,421 6 (11)	0,418 0
Combined uncertainty (2) ($MBq\ g^{-1}$)	0,006 2	0,002 3	0,005	0,002 2	0,001 4	0,004 2

Table 8 (continued)

(1) Values not corrected for the delayed events.

(2) Values corrected for the delayed events.

(3) All these results were obtained with the diluted solution.

(4) Value obtained for a second-order fit with the upper window.

The other results are as follows:

- lower window	2nd order extrapolation	(0,456 7 ± 0,000 3) MBq g^{-1}
- upper window	1st order extrapolation	(0,491 2 ± 0,001 0) MBq g^{-1}
- lower window	1st order extrapolation	(0,461 4 ± 0,000 3) MBq g^{-1}.

The mean value of the AECL results for a second-order fit using type A uncertainties for weighting and combining the other uncertainties quadratically with the external error is (0,457 7 ± 0,004 1) MBq g^{-1}.

The mean value of the AECL results for a first-order fit using type A uncertainties for weighting and combining the other uncertainties quadratically with the external error is (0,463 2 ± 0,007 1) MBq g^{-1}.

(5) For the upper window (340 to 635 keV).
For the lower window (15 to 340 keV) the value is (1,081 ± 0,015).

(6) Value obtained for a second-order fit with the upper window.

The other results are:

- lower window	2nd order extrapolation	(0,422 4 ± 0,005 9) MBq g^{-1}
- upper window	1st order extrapolation	(0,457 3 ± 0,006 5) MBq g^{-1}
- lower window	1st order extrapolation	(0,426 8 ± 0,005 9) MBq g^{-1}.

The mean value of the AECL results for a second-order fit using type A uncertainties for weighting and combining the other uncertainties with the external error is (0,423 8 ± 0,007 6) MBq g^{-1}.

The mean value of the AECL results for a first-order fit using type A uncertainties for weighting and combining the other uncertainties with the external error is (0,429 0 ± 0,009 9) MBq g^{-1}.

(7) Efficiency spread between 0,27 and 0,47.

(8) Weighted mean of 92 measurements.

Table 8 (continued)

(9) Unweighted mean of all the results obtained with 5 different sources.

(10) Unweighted mean value calculated from the individual results.

(11) Unweighted mean of 8 runs (each consisting of 10 individual measurements of 100 s) with 4 sources.

Table 9 - Weighted and unweighted mean values of the activity concentration of ^{75}Se

	Activity concentration/(MBq g^{-1}) (1)	
	Weighted mean	Unweighted mean
Before correction for the delayed events	0,444 0 ± 0,002 6	0,442 4 ± 0,007 8
After correction for the delayed events (2)	0,417 3 ± 0,005 6	0,417 2 ± 0,005 3

(1) The weighted mean of the two AECL results obtained for a second-order fit was used as the AECL value.

(2) These values include the result obtained by means of the high γ-efficiency method.

Table 10 - Weighted and unweighted mean values of the activity concentration of ^{75}Se, without the BIPM results

	Activity concentration/(MBq g^{-1}) (1)	
	Weighted mean	Unweighted mean
Before correction for the delayed events	0,444 6 ± 0,001 6	0,449 7 ± 0,003 3
After correction for the delayed events (2)	0,423 0 ± 0,001 7	0,422 2 ± 0,001 9

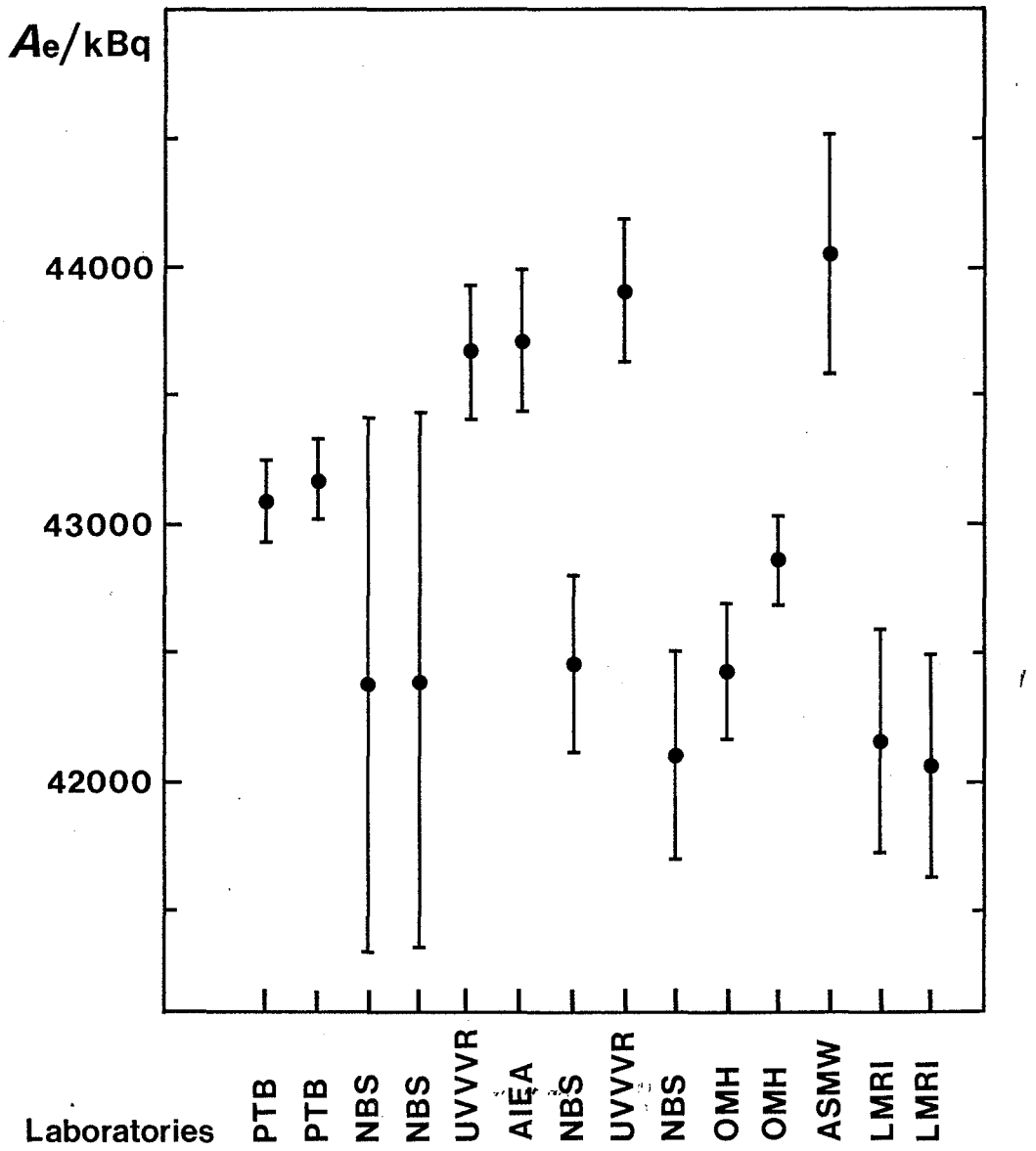

Fig. 1 - Results obtained for ^{75}Se in the frame of the International Reference System.

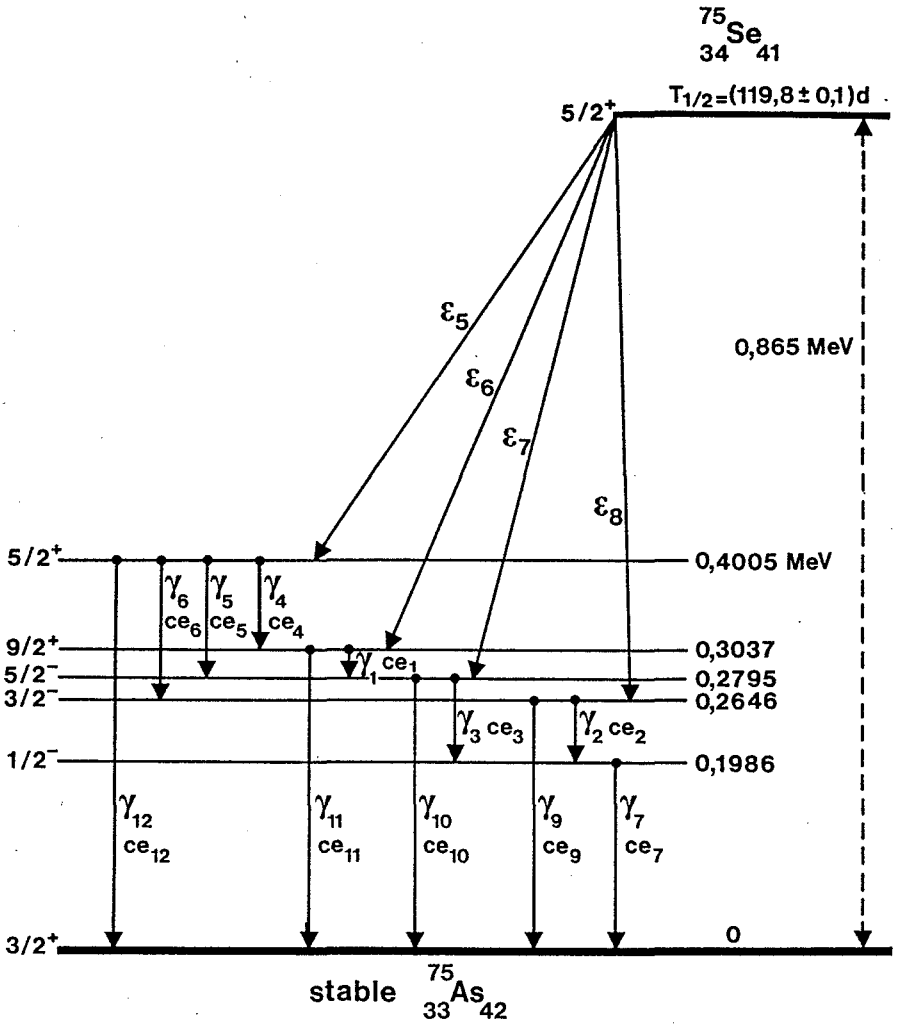

Fig. 2 - Decay scheme of ^{75}Se (from [3]).

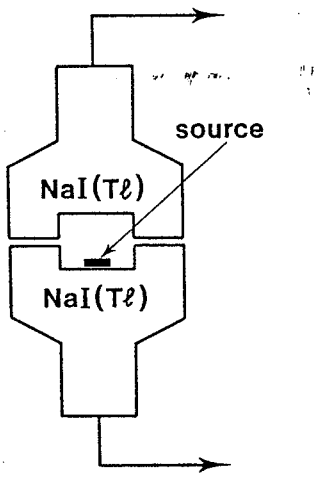

Fig. 3 - Experimental set-up for the $4\pi\gamma$(NaI(Tl)) high-efficiency integral counting.

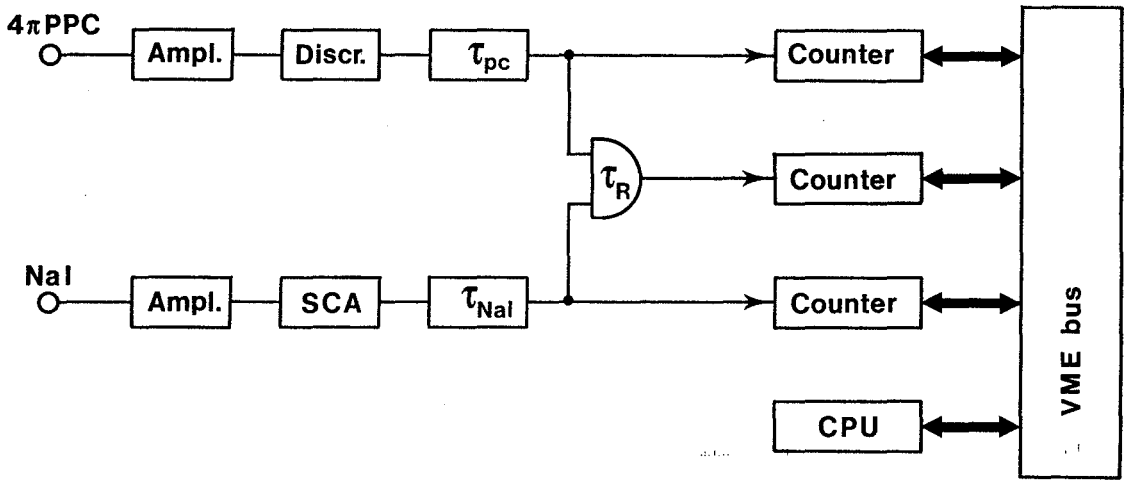

Fig. 4 - Experimental set-up used at the PTB for the correlation method.

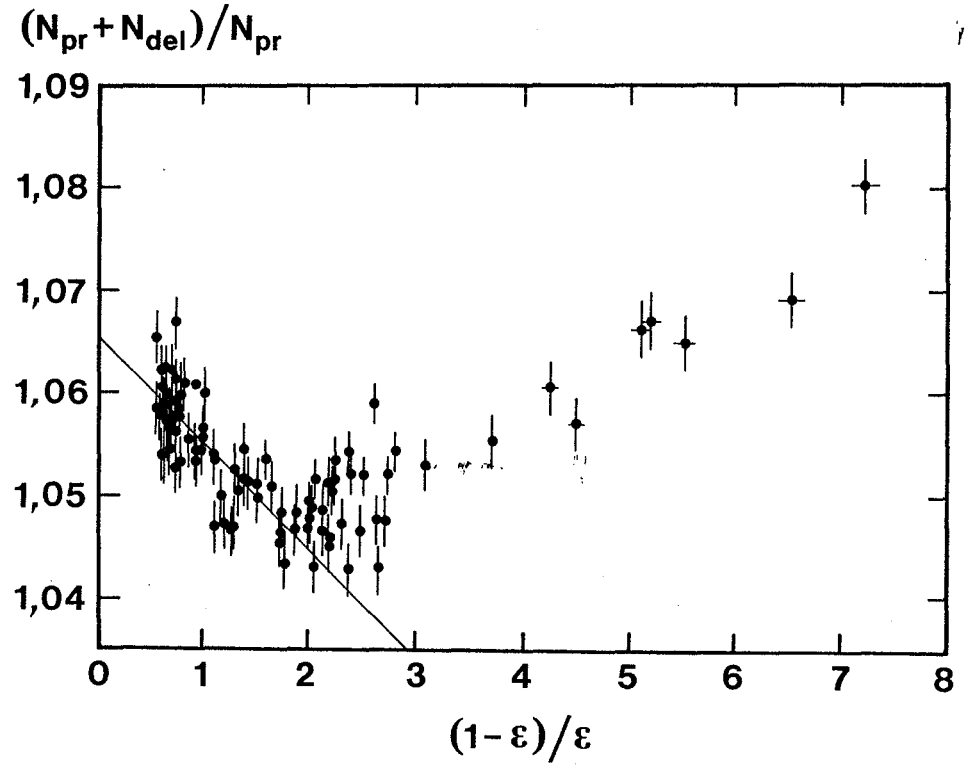

Fig. 5 - Experimental data obtained at the PTB, using the correlation method to determine the fraction of delayed events. N_{pr} and N_{del} are the numbers of prompt and delayed events, respectively, and ε is the efficiency.

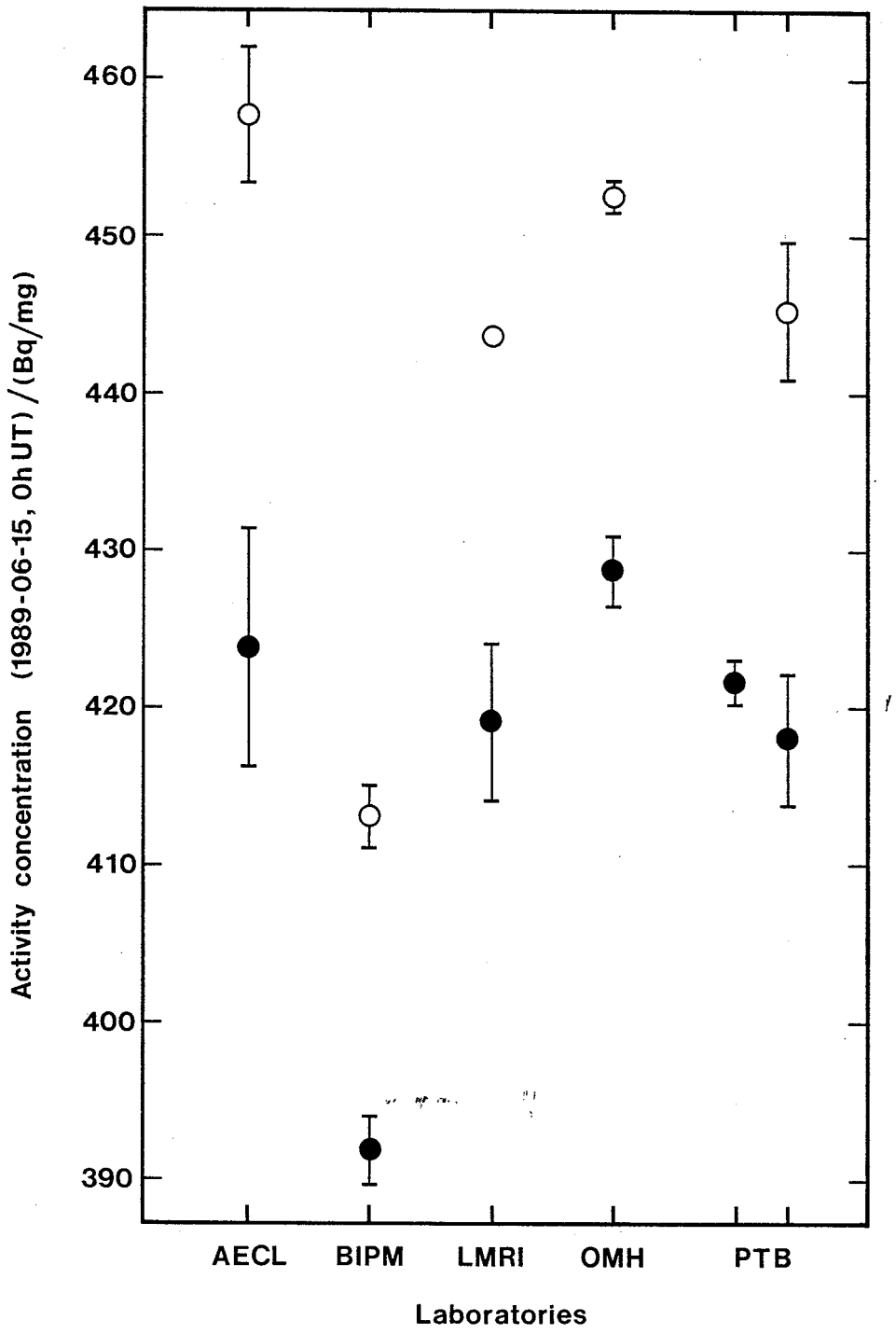

Fig. 6 - Results of the ^{75}Se trial comparison obtained before (○) and after (●) correction for delayed events.

Acknowledgments

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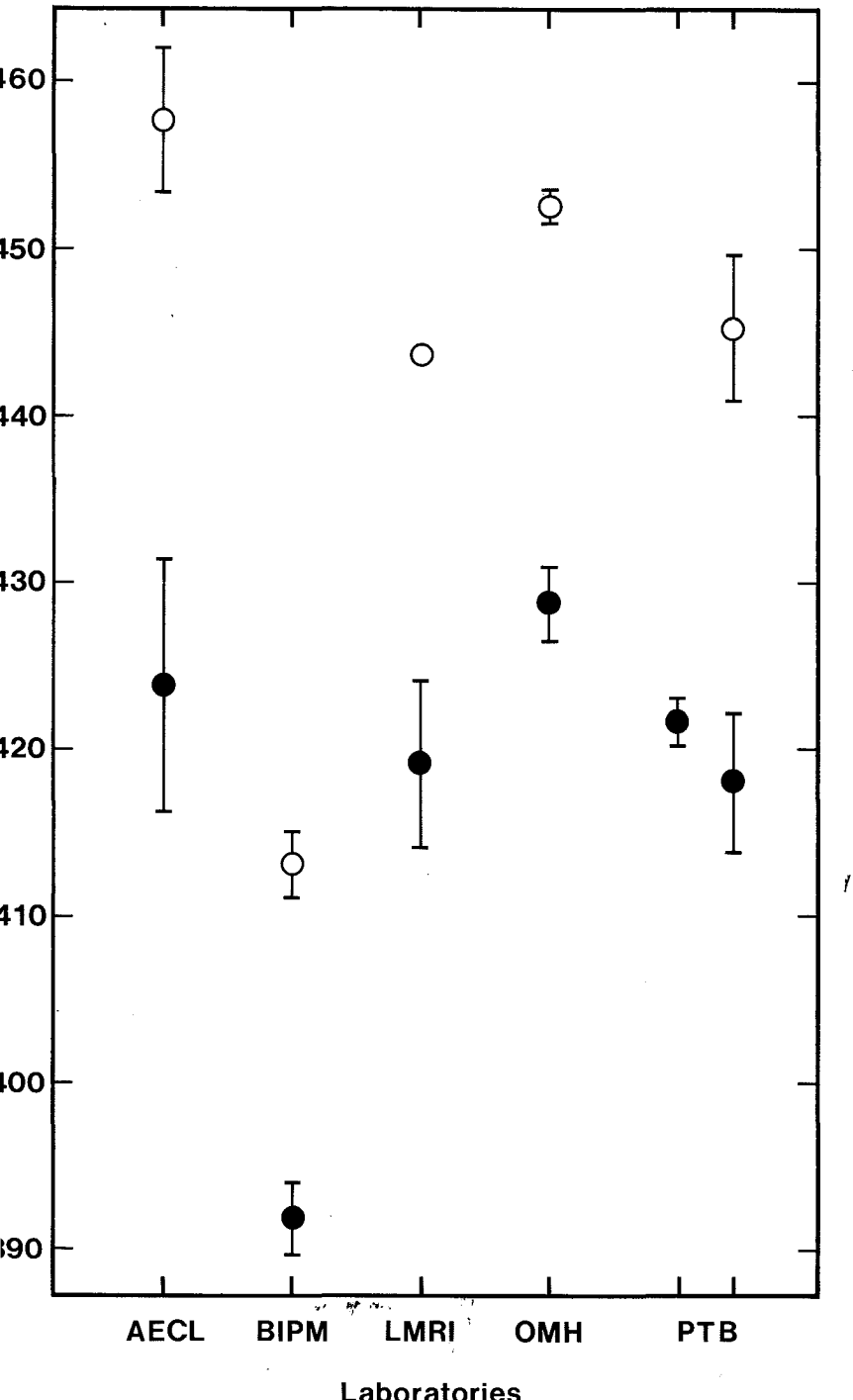
460
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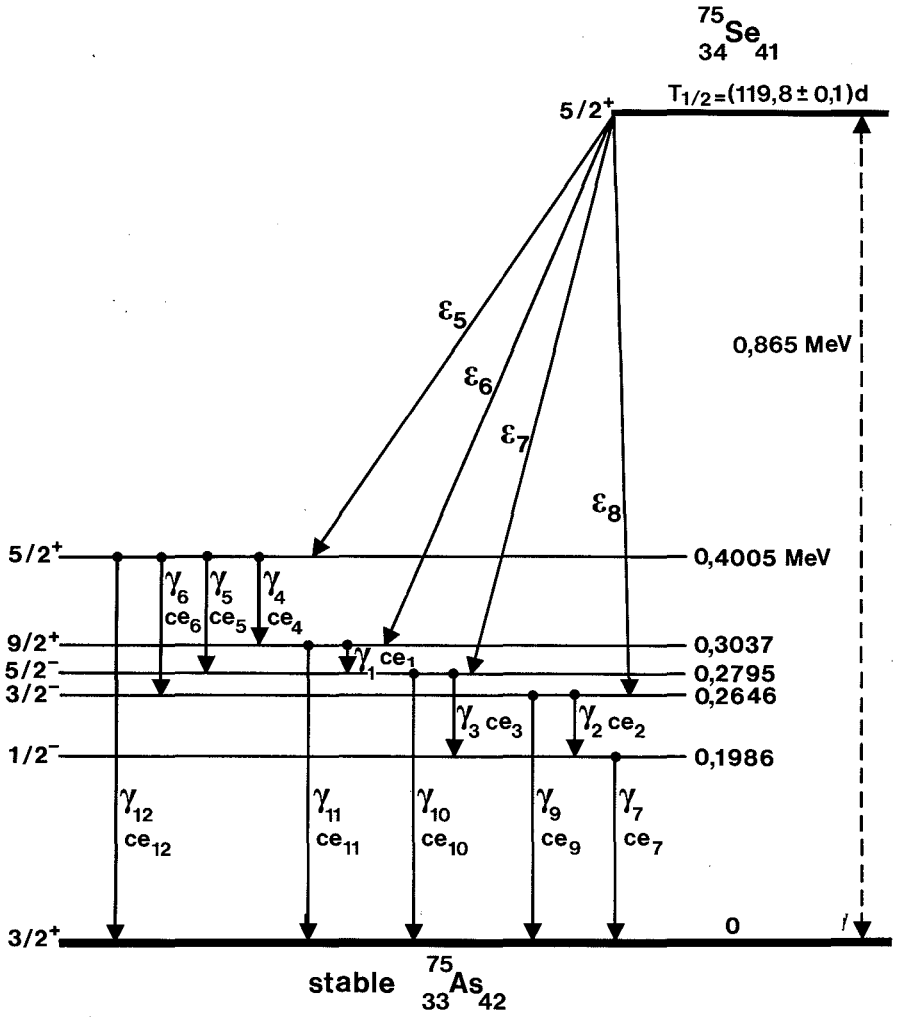
AECL
BIPM
LMRI
OMH
PTB
Laboratories


$^{75}_{34}Se_{41}$
$T_{1/2}=(119,8\pm0,1)d$
$5/2^+$
ε_5
ε_6
ε_7
ε_8
0,865 MeV
$5/2^+$
0,4005 MeV
γ_6 ce_6
γ_5 ce_5
γ_4 ce_4
$9/2^+$
0,3037
γ_1 ce_1
$5/2^-$
0,2795
$3/2^-$
0,2646
γ_3 ce_3
γ_2 ce_2
$1/2^-$
0,1986
γ_{12} ce_{12}
γ_{11} ce_{11}
γ_{10} ce_{10}
γ_9 ce_9
γ_7 ce_7
$3/2^+$
0
stable
$^{75}_{33}As_{42}$