BIPM Activities in the Field of Ionizing Radiations*

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I. Introduction

The General Conference on Weights and Measures (CGPM**), which consists of delegates from all the Member States of the Metre Convention (at present 46 States), decided unanimously at its 11th General Conference (Paris, October 1960) to extend the activities of the International Bureau of Weights and Measures (BIPM) to the field of standards of measurement of ionizing radiations and to create at BIPM a corresponding section. Like all decisions taken by CGPM this one had been carefully prepared by the International Committee for Weights and Measures (CIPM), assisted in its task by the Consultative Committee for the Standards of Measurement of Ionizing Radiations (CCEMRI). This Committee was set up early in 1958 to initiate a programme of work for the BIPM in the field of ionizing radiations and to make plans for its implementation. It seems that the CCEMRI proved to be efficient since:

- its first session took place in April 1959,
- at its third session, in October 1961, it had already sponsored two international comparisons of radionuclides (^{32}P and ^{131}I) and it had received for consideration the first progress report of the BIPM ionizing radiation section which consisted at that time of two physicists, one electronic technician and one secretary,
- at its fifth session (September 1964) the inauguration of the new buildings of the ionizing radiation section took place; the section consisted then of four physicists, five high-level technicians, one machine-shop technician and one secretary. Nine international comparisons of radionuclides had been carried out, with analyses performed or in progress. A comparison of the emission rate of a neutron source was in progress and the results of an international comparison of six Hönigschmid radium standards had been analysed.

In view of the extensive work entrusted to CCEMRI, the CIPM decided in October 1969 to establish four sections: Section I (Measurement of X and γ rays, electrons), Section II (Measurement of radionuclides), Section III (Neutron measurements), Section IV (α-energy standards). In 1975 this last section was dissolved and Section II became responsible for its field of activity.

Since 1975 the three Sections of CCEMRI have met every two years. The chairman of CCEMRI meets also every two years with the chairmen of the three sections to analyse the work carried out and to present it to CIPM. Detailed information on the activity of CCEMRI can be found in a series of reports published periodically by BIPM.

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** For a glossary of acronyms see Appendix 1.

The impressive work performed by CCEMRI and the BIPM ionizing radiation section has been made possible thanks to a spirit of constant mutual understanding between these two bodies and also because the chairmen and the members of CCEMRI and their respective laboratories have always taken an active part in the work and have given excellent guidance to the staff of the BIPM ionizing radiation section. It should be mentioned also that the BIPM has taken advantage of the active long-term cooperation of high-level guest workers.

II. International coherence of measurements in the field of ionizing radiations

The joint work which has just been mentioned appears in the following tables:
Table I - International comparisons of X- and γ-ray measurements (page 4),
Table II - " " " radionuclides (page 5),
Table III - " " " neutron measurements (page 7).
These tables are self-explanatory, but a few short remarks may well be in order here.

X and γ rays

ICRU, which played an important role in the establishment of BIPM activities in the field of ionizing radiations, organized and analysed exposure comparisons before BIPM was able to do so. In the conventional region where free-air chambers are used, an agreement to within about ± 0.5% was reached [1]. Since then the situation has not changed drastically, but the analysis of the corrections has revealed errors (for example scattered photon contribution, humidity, etc.). Moreover, and this is a fundamental point, the results of the comparisons are now kept at BIPM, where any national laboratory can at any time join in the system and, what is still more important, national references which do not make possible an absolute determination of exposure can be incorporated in the system. This role of a "central point" for the X- and γ-ray measurements is of primary importance.

The conversion from exposure to absorbed dose in a given material and under specified conditions has been performed in the best possible way. The recent work of CCEMRI Section I has been analysed by Jennings [2].

Radionuclides

Between 1961 and 1967 eleven comparisons of radionuclide measurements have taken place. The aim of this work was to ensure a worldwide coherence of activity measurements, but it became rapidly clear that the comparisons could also be used to investigate specific technical problems [3]. However, a large scale comparison represents a considerable amount of work for the organizer as well as for the participants. For this reason, CCEMRI Section II decided to make them at longer intervals and to fill the gap between two comparisons as follows:

- to investigate particular problems and to publish monographs (Procedures for accurately diluting and dispensing radioactive solutions, The detection and estimation of spurious pulses, The application of liquid-scintillation counting to radionuclide metrology) [4, 5, 6],
- to organize small-scale comparisons in order to identify the problems and to prepare more efficient reporting forms,

- to implement at BIPM the "International reference system for measuring activity of gamma-ray emitting nuclides" (SIR) [7]. This system, which has been in operation since 1976, is becoming more and more useful (245 results so far); it enables each participating national laboratory to check rapidly the relative position of its measurements. The SIR is an excellent means of establishing what is sometimes called "traceability to BIPM".

The progress made in the radionuclide comparisons is shown in Table IV [8].

Table IV - Progress made in radionuclide comparisons

Radionuclide	Date	σ (%)	
^{60}Co	1967	0.31	increasing difficulty of measurement
^{139}Ce	1976	0.19	
^{134}Cs	1978	0.15	

Neutron measurements

There exists a great variety of neutron sources with very different energy spectra and the technical problems relative to their detection and measurement are complex and difficult to investigate. The national standardizing laboratories, as well as BIPM, spend a great deal of effort in investigating absolute methods to perform accurate measurements concerning the determination of neutron source emission rates, neutron fluence rates and (more recently) the relevant dosimetric quantities. On the other hand, thanks to a very successful cooperation under the auspices of CCEMRI Section III, they are also developing transfer instruments of adequate sensitivity and stability in order to organize international comparisons. Tables III and V summarize, for the field of neutron measurements, the international comparisons carried out or in progress; they show the complexity of the problems and the long-term joint task which is now well under way. It should be noted that the intercomparisons first dealt with isotopic-type radioactive neutron sources and thermal neutron sources, then with monoenergetic fast neutron beams using ion accelerators, and now they are extending to the "white" sources available from linear accelerators and filtered beams of nuclear reactors.

Table V - New fast neutron fluence rate intercomparison

Neutron energy (MeV)	Method (transfer instruments)				
	$^{115}In(n,n')^{115}In^m$	Nb/Zr	$^{115}In(n,\gamma)^{116}In^m$	Fission chamber ^{235}U	^{238}U
0.144			X	X	
0.565			X	X	
2.5	X			X	X
5.0	X			X	X
14.8	X	X		X	X
Coordinator	BCMN	NPL	NPL	AERE/NPL	

Table I - International comparisons of X- and γ-ray measurements

Quantity		Participants	Organization	Achievements
Exposure 10 to 50 kV X rays		15	- Direct comparisons made at BIPM between the BIPM standard and 8 national primary standards. - Calibrations at BIPM of ionization chambers used as national exposure references.	Standard deviation between standards: 0.2 to 0.3% according to radiation quality.
Exposure 100 to 250 kV X rays		17	- Indirect comparisons made by means of transfer instruments calibrated successively against the BIPM standard and the concerned national standard. - Calibrations at BIPM of ionization chambers used as national exposure references.	Standard deviation between standards: 0.2 to 0.35% according to radiation quality.
Exposure ^{60}Co gamma rays		19	- Direct comparisons made at BIPM between the BIPM standard and national primary standards. - Indirect comparisons by means of transfer chambers. - Calibration at BIPM of ionization chambers used as national exposure references.	Standard deviation between standards: 0.2%.
Absorbed dose ^{60}Co gamma rays	in graphite	6	- Comparison made at BIPM between four national calorimetric standards and the BIPM ionometric standard. - Calibration at BIPM of ionization chambers used as absorbed dose national references.	Agreement of 0.3% between calorimetric and ionometric standards.
	in water	8	- Comparison of Fricke dosimetry systems organized by NPL under the auspices of CCEMRI Section I: irradiations made at BIPM; the absorbed dose in water is calculated from the absorbed dose in graphite in the same beam.	- Standard deviation between Fricke systems: 0.3%. - Agreement between mean Fricke value and BIPM value: 0.4%.

Table II - International comparisons of radionuclides

Radionuclide	Date and ref.	Participants	Organization	Achievements
^{32}P pure β emitter	Jan 61 [9]	16	Solution supplied by LMRI. Results reported by BIPM. No analysis.	Sources of errors recognized: dilution, determination of absorption corrections.
^{131}I β-γ complex, short half life	Apr 61 [10]	19	Solution supplied and analysis performed by NRC.	Large scatter of results due to: dilution, aliquoting, amplifier performance, calculations. ^{60}Co would be more useful.
^{198}Au β-γ, short half life	Jan 62 [11]	25	Solution supplied and analysis performed by NPL. No reporting form. BIPM took part ($4\pi\beta$ only) for the first time.	Analysis difficult: not enough information.
^{60}Co	Jan 62 [12]	21	Solution supplied and analysis performed by IMM.	Large scatter. Errors due to source preparation. Suggestion: distribution of solid sources.
^{204}Tl pure β emitter, 2% electron capture (EC)	May 62 [13]	19	Solution supplied by NPL. Analysis performed by BIPM.	Very large scatter; errors due to: self and foil absorption, unknown efficiency to electron capture (no decay scheme given). Much information was obtained regarding source preparation.
^{35}S pure β emitter	Jun 62 [14]	15	Solution supplied by LMRI. Organization and analysis by BIPM (without participation). Reporting form distributed, further inquiry after end of measurements.	Large scatter. Bad LS results. Efficiency tracing may be best. Low energy, pure β emitter is difficult to standardize.
^{60}Co	Mar 63 [15]	22	Distribution by BCMN of 4 solid sources and 1 ampoule of solution. Solid sources checked (BCMN, NPL) before and after. 106 sources distributed (15 broken). Detailed reporting form and analysis by BIPM.	Detailed analysis: 0.5% difference in source-weighing procedures (evaporation/pycnometer). Results obtained with sources were good, those obtained with solutions were bad. Source preparation should improve.
^{241}Am α, γ	Jun 63 [16]	21	Detailed reporting form. Solution prepared and bottled by NBS, distributed by BIPM. Instructions given about uncertainty evaluation. Analysis by BIPM.	Many methods used. For the first time it is possible to calculate a meaningful mean result, with a standard deviation of 0.2%.

Table II (cont'd)

Radionuclide	Date and ref.	Participants	Organization	Achievements
$^{90}Sr+^{90}Y$ pure β decays	Feb 64 [17]	24	2 solutions with concentration ratio 1:15 distributed by PTB. Analysis performed by BIPM.	2 solutions prove to be useful, difference of the 2 weighing methods confirmed. Better determination of absorption corrections, better estimation of uncertainties. Details about efficient source treatment.
^{54}Mn simple EC, single γ ray	Apr 65 [18]	16	A more complicated case of coincidence counting. Solution supplied by LMRI. Analysis performed by PTB.	A 2% ^{35}S contamination of unknown origin makes analysis more difficult. Results rather poor, but a good value of ω_K is obtained. Repeating is not considered necessary.
^{60}Co simple β-γ decay	May 67 [19]	25	Two solutions in known concentration ratio issued by NPL. Analysis performed by BIPM.	Comparison of dilution and source preparation methods by means of ^{60}Co. Although participants did not have to dilute, the weak solution results were not much better than those of the strong one.
^{139}Ce simple EC, single γ ray and conversion electrons	Mar 76 [20]	22	Solution supplied by NAC/IAEA. Analysis performed by BIPM.	This EC nuclide has considerable int. conv. ($\alpha_t \approx 0.25$). Results satisfactory (no dilution required). Analysis of coincidence formulae used. Good value for α_t obtained.
^{134}Cs complex β-γ decay	Oct 78 [21]	24	Solution supplied by AECL/LMRI. Analysis performed by BIPM.	All participants sent results, total spread 0.68%, standard deviation 0.17. Best comparison since the program started, even though it involved counting with complex β-γ spectrum.
^{137}Cs metastable state of ^{137}Ba	May 82 [22]	19	Solution supplied by PTB. Analysis performed by BIPM.	Efficiency tracing by ^{134}Cs, tracer standardized on the grounds of SIR measurements. Analysis under way. Difficult technique, satisfactory results.

Table III - International comparisons of neutron measurements

Quantity	Participants	Organization	Achievements
Neutron emission rate	11	Circulation of the NRC Ra-Be (α,n) neutron source n° 200-1. BIPM organized the comparison, participated and performed analysis of results.	3% spread in results. The overall uncertainty estimated by participants ranges from 0.7 to 1.7% [23].
Thermal neutron fluence rate	11	Organized by Section III of CCEMRI. Gold foils were activated by nine laboratories in their respective thermal neutron flux assemblies. Each participant sent his activated foils to several other laboratories for absolute activity measurements. Analysis of results carried out separately by NPL and NBS using very different methods.	All individual adjusted values are within ± 2.6%, whereas their average standard deviation is about 1% [24, 25].
Monoenergetic fast neutron fluence rate. Neutron energies: 250 keV, 565 keV, 2.2 MeV, 2.5 MeV, 14.8 MeV	9	Transfer instruments carried from laboratory to laboratory and operated by BIPM physicist. BIPM participated at 2.5 and 14.8 MeV. Analysis performed by BIPM.	Agreement among the results is better than 5%, except at 250 keV with Bonner sphere, where the discrepancy ranges from 10 to 20%, due partly to the contribution of scattered neutrons. The overall uncertainty estimated by participants for absolute fluence rate measurements ranges from 2.0 to 3.5% [26].
^{252}Cf neutron emission rate	12	Organized by Section III of CCEMRI by circulating the NBS ^{252}Cf fission neutron source (10^7 s^{-1}). Analysis of results will be carried out by NPL. BIPM participated in the comparison.	Measurements are completed. Analysis of results in progress.

III - Some research work at BIPM

The work performed jointly by CCEMRI and BIPM has undoubtedly increased the coherence of measurements in the world, but one must not forget the importance of the accuracy with which the standards are realized. To reach this goal, it is necessary to study carefully the corrections applied to measurements, to use alternative measurement methods for the determination of the same physical quantity, or to invent and apply a new method of measurement. These three tracks have been followed by the staff of the BIPM ionizing radiation section within the frame of their research work. In view of the limited length of this abstract, I shall mention only some examples.

1. Work in the field of X- and γ-ray measurements

The research work in this field has developed in a way similar to the international comparisons. One should note the continuous and logical effort directed to a gradual transition from exposure to absorbed dose measured in strictly specified experimental conditions.

a) Free-air ionization chambers - The installation in the BIPM laboratory of very stable X-ray sources and of an automatic device for measuring ionization currents was necessary to perform comparisons of exposure standards by the substitution method. This instrumentation has been used to investigate carefully some corrections which are applied to free-air chambers. An original method of measurement using magnetic induction has been developed and enabled us to study the radial distribution of the ionization per unit mass produced by an X-ray pencil in air [27]. An experimental study of ion recombination in parallel-plate free-air ionization chambers has shown that both initial and volume recombinations may take place. A technique was set up to separate these two contributions and the extrapolation conditions have been analysed [28]. A very careful study of the influence of humidity on the ionization of air, performed at BIPM [29] (and later confirmed by other laboratories), has shown that the humidity correction as currently used in the sixties was in error. This can be explained by assuming that W for moist air does not obey the Bragg law for low values of relative humidity (Fig. 1). The results given on this figure have been used [30] to estimate by calculation the ionization in the case of a cavity chamber. The expectations are in agreement with measurements made in other laboratories [31, 32]. Within the frame of a critical analysis of various determinations of W in air, made while preparing an ICRU report [33], it has been possible, thanks to the above results, to correct published experimental values for which data were available on the humidity existing during measurements.

b) Cavity ionization chambers as exposure standards - A certain familiarity with the techniques described above has led us to a unified analysis of the principle of exposure measurements in a photon beam produced by ^{60}Co, with free-air ionization chambers and cavity ionization chambers. A first theoretical study [34] deals with the case of an ideal chamber with a very thin cavity; it was extended later [35] to the case of a cavity with finite dimensions, the influence of side walls being taken into account experimentally. These theoretical studies had a twofold influence: on the one hand, they have made possible a rational definition of the geometry of the standard cavity chamber of BIPM and, on the other hand, the analysis of

the mechanism of energy transfer by electrons has been very useful in the subsequent ionometric determination of absorbed dose in a phantom (perturbation correction).

The accuracy of exposure determinations is strongly affected by the uncertainty in $S_{C,a}$ the mean ratio of the mass collision stopping powers of graphite and air. For this reason any experiment giving information on this parameter is interesting. This was in particular the case for a comparison of activity, power and exposure measurements performed with a 37 GBq ^{60}Co source. A method of measuring the activity of such a source has been developed [36]. It is based on:

- the precise experimental determination, by gamma spectrometry, of the activity ratio of two sources which are very different in strength, but of identical geometry,
- the measurement of the activity of the weak source, after its dissolution, by the absolute $4\pi\beta-\gamma$ coincidence method.

The activity of the 37 GBq source was determined with an accuracy of 0.2%. The results obtained by this method were subsequently compared to those of the power measurement of a similar ^{60}Co source performed at NBS (agreement 0.02%) [37]. A comparison of the exposure rate at a point calculated from activity and the exposure rate measured at that point has shown deviations which can reach 0.8%. We shall have to wait for the recent estimations of stopping powers [38] to reduce the deviation to 0.23%, which is the indication of a better internal coherence of dosimetric measurements.

c) Ionometric determination of absorbed dose - The BIPM has chosen an ionometric method to determine the absorbed dose in a graphite phantom irradiated by a ^{60}Co beam. This choice is based on the following considerations:

- the ionometric method is flexible and precise, the stability in time of its various elements can be easily checked; therefore it is the method of choice "to maintain" at BIPM the various determinations made by the national laboratories,
- it is interesting to compare these results with the calorimetric determinations.

It is possible to determine the absorbed dose by using the Bragg-Gray relation, provided one is able to correct for the perturbations of photon fluence and electron fluence due to the real cavity. The photon fluence perturbation has been investigated by the Monte-Carlo method [39], whereas the mechanism of energy dissipation by electrons has been analysed by means of techniques previously developed for exposure standards [34, 35]. The results are given in figure 2. The good agreement (0.3%, assuming W = 33.85 eV) between ionometric measurements and the calorimetric measurements of national laboratories gives strong support to the reliability of the calculated correction. Moreover, one can use this comparison to derive from the experimental results a highly accurate value (for ^{60}Co gamma rays) of the product $S_{C,a}$ W with a standard deviation of only 0.1%. This is particularly important for the establishment of standards of air kerma, because the accuracy of such standards would be greater than the accuracy of the present exposure standards.

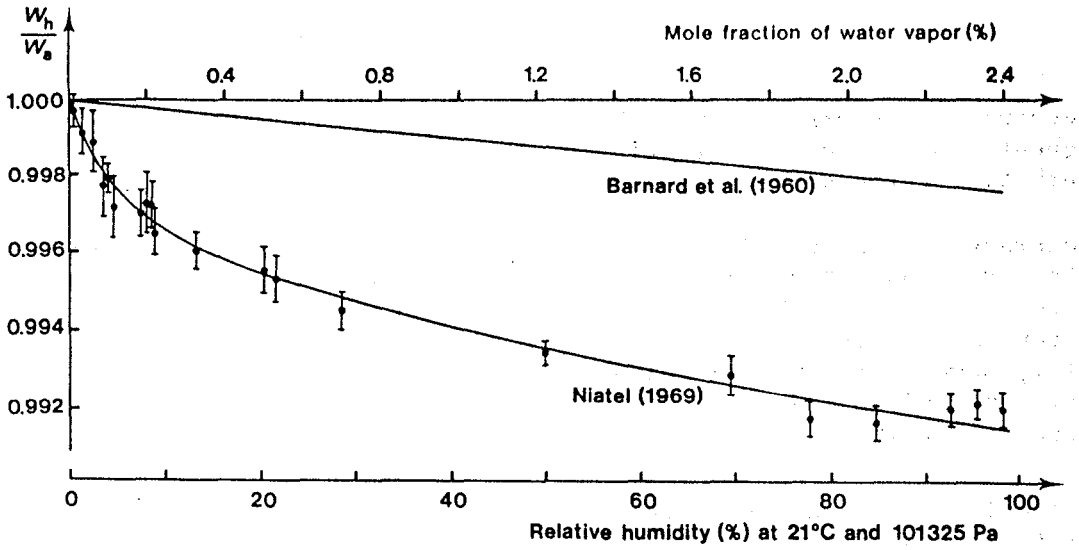

Fig. 1. Mean energy W_h expended in humid air per ion pair formed as a function of the mole fraction of water vapor. The ordinates are values relative to the energy W_a for dry air. While Barnard's curve [40] is theoretical (assuming independent contributions from dry air and water vapor), Niatel's curve [29] is based on measurements of ionization current in a free-air chamber. The errors correspond to a 95 percent confidence level.

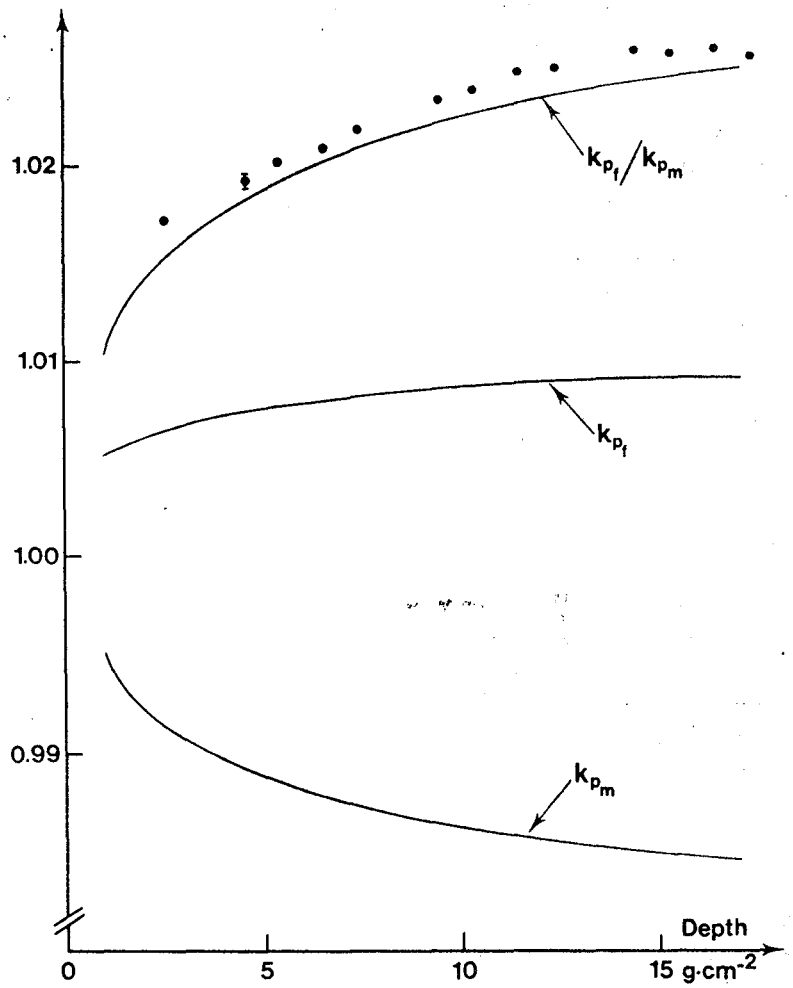

Fig. 2. Correction factors k_p [41] for the perturbation due to the presence of a flat cavity when determining the absorbed dose in a graphite phantom (^{60}Co photon radiation). The curves k_{p_m} and k_{p_f} give the theoretical variation [39] with depth of the factors k_p calculated for two positions of the cavity chamber with respect to the reference plane Π where one wishes to determine the absorbed dose: k_{p_m} and k_{p_f} correspond to the middle plane or to the front plane of the cavity at Π respectively. The upper part of the figure shows the ratios k_{p_f}/k_{p_m} (theoretical curve and experimental points). The uncertainties (estimated standard deviations) associated with the experimental points are given for illustration.

If the product $S_{C,a}$ W is used to calculate the W value which brings the calorimetric and ionometric absorbed dose measurements into complete agreement, the W value varies from 33.72 eV and 33.96 eV, depending on the stopping power data used [42] or [38]. Here, too, the coherence of measurements is increased by the new stopping power values.

2. Absolute measurements of α-particle energies

The knowledge of accurately determined alpha-particle energies is highly important, not only in alpha-ray spectroscopy, but also for recalibrating numerous reaction-energy (Q value) measurements as well as, still more directly, in atomic-mass systematics.

The BIPM spectrometer is unique because it has been built especially for absolute energy measurements [43]. The radii of the semicircular orbits vary from 365 mm to 475 mm. The magnetic induction B, produced by an electromagnet, (8.5 t; 40 kW) ranges from 0.6 T to 1.01 T and is stabilized by proton resonance to $\approx 1.5 \cdot 10^{-6}$. The induction is measured along an orbit and averaged according to Hartree's procedure. Until recently the α particles were detected by nuclear track plates which are now replaced by nitrocellulose films (Kodak LR 115).

The tracks are counted singly and the high-energy edge of the line histogram is extrapolated to background level (Fig. 3). The orbit diameters are determined to better than 0.01 mm. With the best sources it is thus possible to obtain a combined uncertainty of about 100 eV.

Twenty-six α-particle emitters have been measured: ^{253}Es, ^{244}Cm, ^{242}Cm, ^{241}Am, ^{240}Pu, ^{239}Pu, ^{238}Pu, ^{236}Pu, ^{232}U, ^{228}Th, ^{227}Th, ^{226}Ra, ^{224}Ra, ^{223}Ra, ^{222}Rn, ^{220}Rn, ^{219}Rn, ^{218}Po, ^{216}Po, ^{215}Po, ^{214}Po, ^{212}Po, ^{212}Bi, ^{211}Bi, ^{210}Po, ^{148}Gd.

These results were widely used in the latest atomic mass evaluation by Wapstra and Bos [44]. A new catalogue of recommended alpha-energy and intensity values has been published [45].

3. On some BIPM work relating to counting statistics

All the studies performed in this field have their origin in experimental problems arising during conduct of programs. In order to permit some kind of overview on the rather large number of reports dealing with seemingly quite different subjects, they are grouped here in three categories. For an extended summary up to 1972 see [46]; references till 1981 are assembled in [47].

a) Methods for measuring a dead time - Two of the early BIPM reports issued [48] contained the proposal of a new measuring method in which the two "natural" sources used in the traditional two-source method are replaced by two independent quartz oscillators. Simplicity, accuracy and speed of this approach have led to a rapid acceptance in national laboratories [49] where it is now part of the standard equipment. The method has also given rise to more mathematically-minded studies [50] where the practical implementation of the condition of an irrational frequency ratio is critically analyzed.

A possibly unique feature of the method lies in the fact that not only the value, but also the type of the dead time involved can be determined. Since P. Bréonce has constructed an automatic version of the measuring system [51], routine measurements with an accuracy of better than 10^{-3} take less than one minute. As for the alternate source-pulser method [52], a rigorous analysis [53] has revealed that the formula usually used is not an exact solution; its safe use is limited to relatively low pulser frequencies.

b) Effect of a dead time on interval densities and counting statistics - This is a very broad field and we have to restrict ourselves to some selected items. Whereas the interval density for an undisturbed Poisson process is described by an exponential, and insertion of a non-extended dead time results in a simple shift, the effect of an extended dead time is more complicated. An exact explicit expression for the modified distribution has first been derived in [54]. Much effort has been put in the evaluation of exact expressions which state the probability for observing exactly k counts in a given time interval. A complete set of such relations (which generalize the Poisson distribution) for a non-extended dead time has been assembled in [55]; analogous expressions for an extended dead time have been derived by Libert [56]. Of special interest are the corresponding moments of lowest order (mean, variance, etc.). For the situation where the time of observation is much longer than the dead time, the powerful methods of asymptotic integral transforms can be applied and they yield all the desired results [57]. Even the third central moments could be determined [58].

Several studies have been devoted to the series arrangement of two dead times, which is of great practical importance. The influence on the count rates for the four cases to be considered has already been given in [46]; on the other hand, the corresponding counting statistics are still unknown.

Another subject extensively treated concerns the deformation of the Poisson law due to the decay of the source during the time of measurement. A review of the results is given in [59]. They also lead to a new way of measuring half lives.

c) Methods for the absolute measurement of activity - During the past 30 years the coincidence method has occupied an unrivalled position among the few known approaches to measure the activity of a radionuclide which undergoes a beta-gamma decay. The principle of correlation counting, although known since about 1965, was not taken very seriously for several years by those engaged in activity measurements. A comparison performed in 1968 at the NPL confirmed the validity of the new approach, but did not reveal any real advantages compared to the conventional coincidence counting. BIPM studied the method from 1970 onwards, in particular the effect of a dead time on the correlation. It was then found that a particular form of correlation counting, namely modulo counting [60], could be used for distinguishing between single and multiple events and allowed thereby a new approach to the difficult problem of afterpulses [61, 5]. In the meantime it became also clear that correlation counting was applicable to nuclides with delayed states [62], a field where coincidence measurements run into serious trouble.

However, the technique is a difficult one and the various corrections needed are still not sufficiently well known [63]; it has therefore been little used, but the last word may not yet have been said. On the other hand, the principle of modulo-2 counting has recently gained renewed interest [64].

Meanwhile efforts continued for a better comprehension of the coincidence method. In particular, an exact and general solution was found for the case of extended dead times [65]; however, this was clearly overshadowed by the nearly simultaneous publication of Cox and Isham [66] on the same problem, but involving the more popular type of non-extended dead times. Smith's "translation" of this work into the language of physicists [67] was of great importance. This decisive advance, after some 20 years of "trial and error approaches", was generally considered as the definite and final answer to the problem of absolute activity measurements. However, in the meantime it had been overlooked that the measurement of coincidences, although very simple from the practical point of view, was in fact an unnecessary detour for what we actually need, namely the measurement of the efficiency of a counter. The real problem is to decide whether two beta and gamma pulses stem from the same disintegration or not; their coincidence in time is only a possible (but not necessary) consequence. The systematic pursuit of this idea, helped by the insight gained from the previous studies, led to an entirely different approach, called "selective sampling" [68], where all the difficult questions related to coincidences are avoided; there remain only problems with one channel left, and they are easy to treat. The practical implementation of this new approach is described in some more detail in the following section.

Selective sampling

The selective sampling method takes advantage of the fact that in a two-step decay the possibility to distinguish between pulses which do or do not have a partner (originating from the same disintegration) in the other channel is equivalent to a knowledge of the detector efficiency (for instance ε_β). Thus, any gamma pulse, measured in a given time interval T, is necessarily single or unpaired if no beta event is detected during T. Such "gaps" or regions of guaranteed absence of pulses can be experimentally "created" by an extended dead time. As for this type, by definition, each new pulse arrival starts a dead time T, the observation of a pulse at the output of such a circuit assures the absence of any other incoming pulse during the time interval T before its occurrence. This simple trick allows us to choose time zones which are free of betas (Fig. 4a). If we now observe simultaneously the events in the gamma channel, we can conclude that all gamma pulses arriving within such time regions have no partners, whereas outside these zones there is no such restriction. A comparison of the average arrival densities for the two zones, measured by the channel contents of a multichannel analyzer (Fig. 4b), leads readily to the simple relation

$$g/G = 1 - \varepsilon_\beta .$$

Since the beta count rate N_β can be measured directly, the source activity $N_o = N_\beta/\varepsilon_\beta$ is now readily obtained.

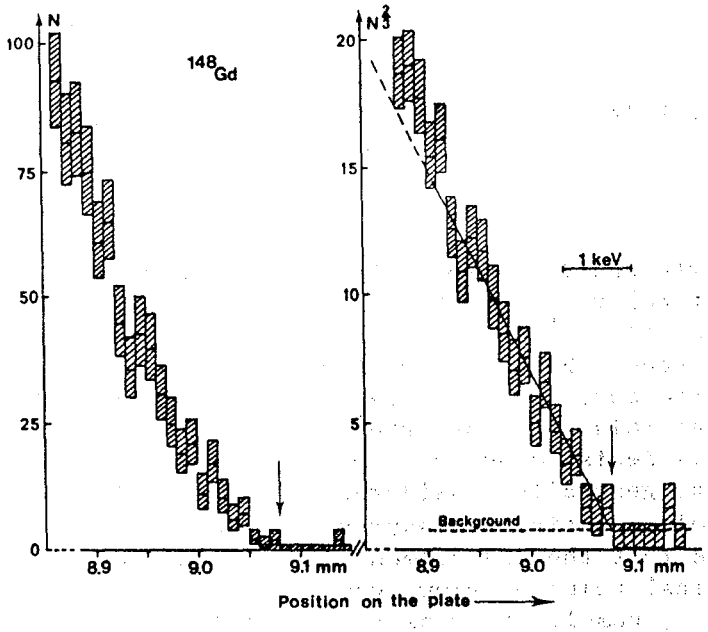

Fig. 3. Example of an α-particle spectrum. One section of the ^{148}Gd α-particle group, with increasing energy to the right. The ordinate on the left is the number N of α particles counted in a band width of 10 μm; it falls off proportional to $(X_o - X)^{3/2}$, where X is the position on the nuclear track plate and X_o the extrapolated value (shown by the arrow). On the right a straight line is fitted to the experimental points $N^{2/3}$. Its intersection with the background yields X_o.

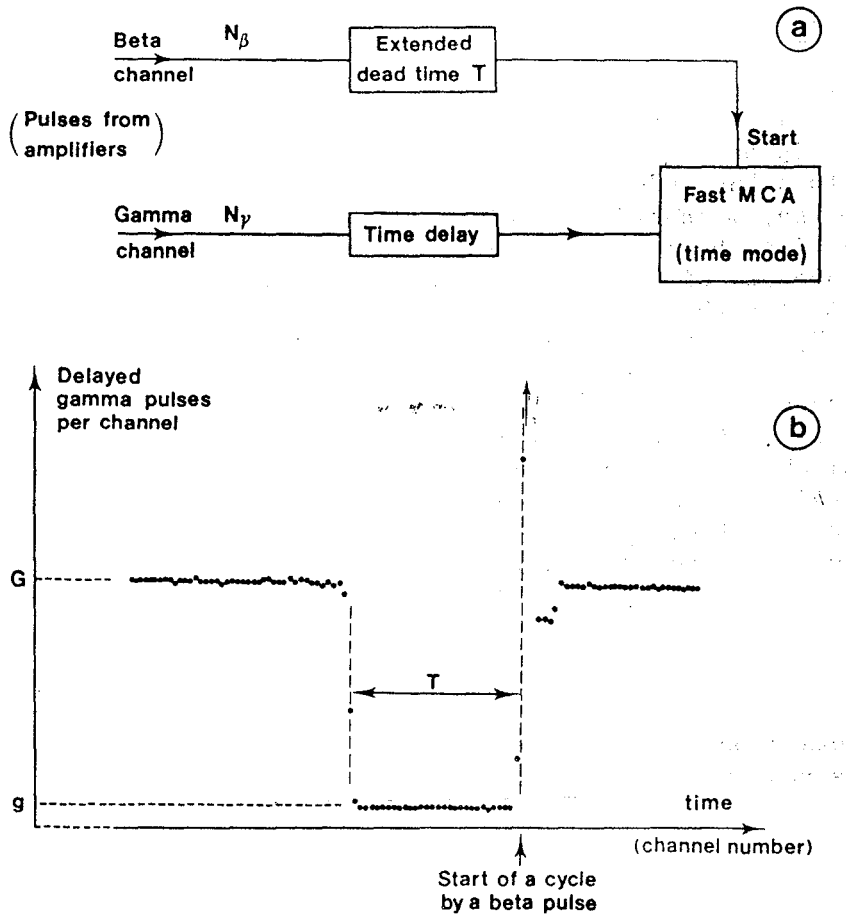

Fig. 4. Selective sampling. a) principle of the method
b) time spectrum of registered gamma pulses.

4. Work in the field of neutron measurements

The BIPM has installed a laboratory for measuring neutron sources and started by using the manganese sulphate bath method; in particular, (γ,n)-type neutron sources were measured by circulation of the $MnSO_4$ solution. The laboratory is also equipped with a 2.5 MeV and 14 MeV neutron generator. The fluence rates of these two beams are calibrated by the associated particle method. The neutron group is currently measuring dosimetric quantities in close cooperation with the X- and γ-ray group who had already developed similar techniques at BIPM in the photon field.

Neutron source emission rate determinations by the manganese bath method - Thanks to an improvement of the electronic equipment, the activity of manganese sulphate bath solutions can now be determined with a precision better than 0.1 %. In addition, the development of the circulation technique allows an accurate measurement of the ratio of thermal neutron absorption cross sections for hydrogen and manganese (σ_H/σ_{Mn}). This ratio is currently obtained with a standard deviation of 0.3 %, instead of the value of 1 % previously quoted in the literature, by using the cross sections of σ_H and σ_{Mn}. Consequently, an uncertainty of only 0.13 % in the neutron emission rate determinations is introduced, instead of the previous value of 0.45 %.

Neutron fluence rate measurements at 14.65 MeV - In collaboration with IRK a precise absolute measurement of the $^{27}Al(n,\alpha)^{24}Na$ cross section with 14.65 MeV neutrons has been performed. The associated particle method was used to determine the neutron fluence. Corrections were applied for scattered alpha particles, contamination of the alpha spectrum by protons from the $^2H(d,p)^3H$ reaction and contributions from (n,α) and (n,p) reactions in the silicon surface barrier detector used for the detection of the associated alpha particles. The absorption and scattering of neutrons by the target backing, the target holder, the beam line and the sample mounting system were taken into account. The result obtained is $(112.2 \pm 1.0)\ 10^{-3}$ barn. It should be noted that the $^{27}Al(n,\alpha)^{24}Na$ reaction is at present considered as a possible candidate for a neutron cross section standard, especially in the 14 MeV region, and it may be useful as a fluence monitor in fusion reactor studies.

Another example of research work carried out at BIPM in view of improving the accuracy of neutron fluence rate measurements is given below.

Production and measurement of 2.5 MeV neutrons - The BIPM has at its disposal a calibrated monoenergetic neutron beam: 2.5 MeV neutrons produced by the reaction $^2H(d,n)^3He$. The deuterons are accelerated by an electrostatic generator with a maximum energy of 150 keV. The absolute measurement of the neutron fluence rate is made using the associated particle method. The present accuracy of measurements is 1.5% (standard deviation). A stilbene scintillator with a neutron-gamma discrimination system, based on pulse shape, is also used to measure the neutron fluence rate directly; the efficiency of this neutron detector is determined by using the coincidence with the associated particles. The experimental set-up and the method of evaluation are indicated on figure 5.

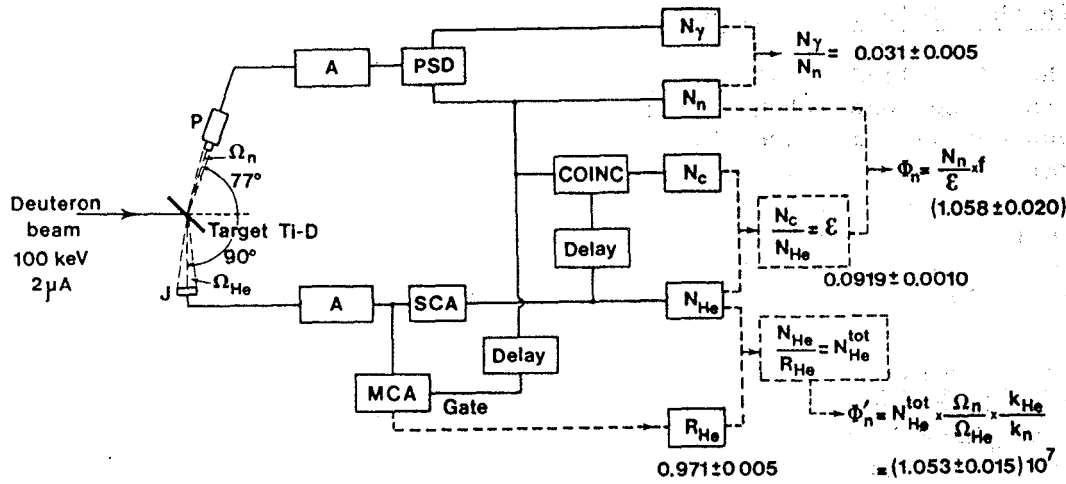

Fig. 5. Measurement of neutron fluence. J = silicon surface barrier detector; P = stilbene scintillator; A = amplifier; PSD = pulse shape neutron-gamma discriminator; SCA = single channel analyser; MCA = multichannel analyser; COINC = neutron-^{3}He coincidence system; N_{He}, N_n, N_γ and N_c = numbers of counts for ^{3}He (single channel), neutrons, gamma rays and neutron-^{3}He coincidence, respectively; $R_{He} = N_{He}/N_{He}^{tot}$, the ratio of the partial ^{3}He counting (corresponding to that from single-channel analyser) to the total ^{3}He counting; ε = efficiency of the stilbene scintillator; f = correction factor taking into account neutron scattering, geometry effect, anisotropy and secondary neutron source, etc.; Ω_n, Ω_{He} = solid angles for stilbene scintillator and silicon detector respectively; k_{He}, k_n = solid angle transformation factors from the centre-of-mass to laboratory system for ^{3}He and neutrons respectively; Φ_n = fluence measured by the stilbene scintillator; Φ'_n = fluence measured by the silicon detector (with the solid angle subtended by the stilbene scintillator as as reference). The numerical values given in the figure are obtained for a measuring time of 12 000 s.

The results of an intercomparison between these two methods show a very good agreement, so that increased confidence has been obtained for the absolute associated particle counting. Nevertheless, it should be pointed out that the measurement made with the stilbene scintillator is not a completely independent method and that the precision obtained is limited by the determination of the correction factor, f, specially due to the uncertainty in the evaluation of the scattered neutron and the gamma-ray contributions. Up to now, the associated particle method seems to have a better accuracy than the other methods for neutron fluence rate measurements. However, a critical point of this latter method should be mentioned: it concerns the determination of the solid angle transformation factors, k_{He} and k_n, which depend on the average energy of the incident deuterons producing the reaction. Fortunately, this dependence is less important for an observation angle of 90° for ^{3}He-particle detection. In this case, a variation of 5 keV in the deuteron average energy introduces a variation of 0.3% in the value of the ratio k_{He}/k_n.

Acknowledgments

Although this review is published under the name of a single author, it is clear that it has been prepared by a team consisting of the staff and the guest workers of the Ionizing Radiation Section of the BIPM. I wish to thank them all here. Mme Müller prepared the manuscript with the care that she brings to every task; she knows my gratitude.

APPENDIX 1

List of laboratories and other acronyms mentioned in the text

AERE : Atomic Energy Research Establishment, Harwell, United Kingdom
AECL : Atomic Energy of Canada Limited, Chalk River, Canada
BCMN : Bureau Central de Mesures Nucléaires, Euratom, Geel, Belgium
BIPM : Bureau International des Poids et Mesures, Sèvres, France
CCEMRI : Comité Consultatif pour les Etalons de Mesure des Rayonnements Ionisants
CIPM : Comité International des Poids et Mesures
CGPM : Conférence Générale des Poids et Mesures
IAEA : International Atomic Energy Agency, Vienna, Austria
ICRU : International Commission on Radiation Units and Measurements, Bethesda, USA
IMM : Institut de Métrologie D.I. Mendéléev, Leningrad, USSR
IRK : Institut für Radiumforschung und Kernphysik, Vienna, Austria
LMRI : Laboratoire de Métrologie des Rayonnements Ionisants, Saclay, France
NAC : National Accelerator Centre, Faure, South Africa
NBS : National Bureau of Standards, Washington, D.C., USA
NPL : National Physical Laboratory, Teddington, United Kingdom
NRC : Conseil National de Recherches du Canada, Ottawa, Canada
PTB : Physikalisch-Technische Bundesanstalt, Braunschweig, Federal Republic of Germany
SIR : International reference system for activity measurements of gamma-ray emitting nuclides

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