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ABSOLUTE MEASUREMENT OF THE ^{125}I DESINTEGRATION RATE*

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ABSTRACT

The procedure followed by the Laboratório de Metrologia Nuclear at the IPEN (Instituto de Pesquisas Energéticas e Nucleares), São Paulo - Brazil, for the absolute determination of the ^{125}I desintegration rate by means of the X-(X,γ) Coincidence and Sum-Peak methods is described. The results were submitted to the BIPM (Bureau International des Poids et Mesures), France, for an International Comparison of this radionuclide.

MEDIDA ABSOLUTA DA TAXA DE DESINTEGRAÇÃO DO ^{125}I

RESUMO

O presente trabalho descreve o método adotado pelo Laboratório de Metrologia Nuclear do IPEN, em São paulo, para a determinação da taxa de desintegração do ^{125}I, aplicando os métodos de Coincidência X-(X,γ) e Pico-Soma. Estes resultados foram submetidos ao BIPM (Bureau International des Poids et Mesures) da França, para a participação em uma Comparação Internacional deste radionuclídeo.

INTRODUCTION

The Laboratório de Metrologia Nuclear (LMN) at IPEN (Instituto de Pesquisas Energéticas e Nucleares), São Paulo - Brazil, has participated in several international comparisons sponsored by the BIPM (Bureau International des Poids et Mesures), France, for the standardization of radionuclides, during the past twenty years. The main goal of these comparisons is to verify the performance of the methods and procedures followed by different laboratories throughout the world, seeking the best possible accuracy. The choice of the radionuclide is made among

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those which have some difficulty in the standardization because of their complex decay schemes or some experimental problems.

The most recent radionuclide comparison sponsored by the BIPM was the ^{125}I (May, 1988). This radionuclide is of great interest in nuclear medicine but it is difficult to be standardized by means of ionization chambers due to its low energy gamma-rays.

The present report gives the details on the procedure followed by the LMN at IPEN for the determination of the ^{125}I desintegration rate, using the X-(X,γ) Coincidence and Sum-Peak methods.

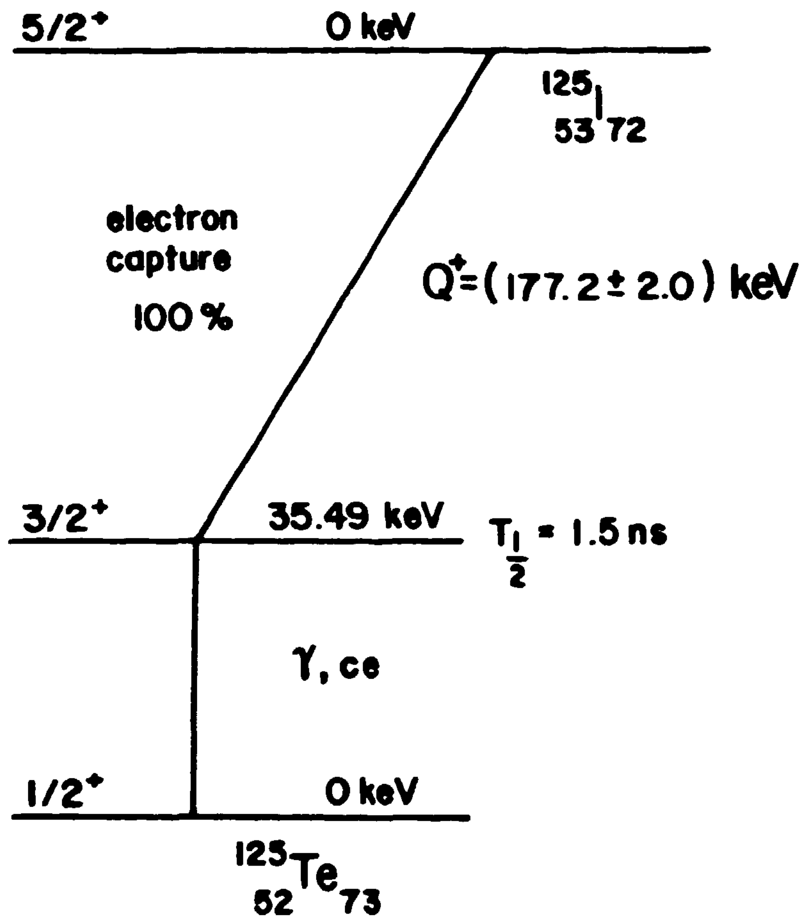
The ^{125}I solution was prepared by OMH (Országos Mérésügyi Hivatal), Budapest, Hungary and sent to each participant laboratory. The LMN at IPEN took part in this comparison together with the IRD (Instituto de Radioproteção e Dosimetria) - Rio de Janeiro, Brazil. The measurements done by LMN and IRD were completely independent and uncorrelated.

The decay scheme of ^{125}I is shown in Figure 1. The nuclide decays 100% by electron capture followed by 35 keV gamma-ray emission. The K X-rays (27.4 and 31 keV) emitted after the electron capture events in the ^{125}I are coincident with the photons emitted in the decay of the 35 keV excited state of ^{125}Te. These photons are either the K X-rays following the internal conversion of the gamma ray or the unconverted gamma-ray itself. The adopted half-life for ^{125}I was (59.5 ± 0.4) d [7]. The 35 keV gamma-ray transition has a half-life of 1.47 ns and a high internal conversion probability (93.4%)[3].

THEORY OF THE METHODS

X-(X,γ) Coincidence Method

This method makes use of two scintillation counters. The K X-rays emitted after the ^{125}I electron capture events are detected by one of the scintillators whereas the other scintillator detects the 35 keV gamma-ray or the K X-rays following the internal conversion process.



$$P_1 = 0{,}7038 \pm 0{,}0223^{(8)}$$

$$P_2 = 0{,}7704 \pm 0{,}0102^{(8)}$$

Figure 1 - Decay scheme of ^{125}I (3).

The measurement can be performed in two different ways: the first sets the descrimination window to count only singles from the K X-rays or gamma-rays emission (window 1); the second sets the descrimination window to count singles plus the coincidence pulses between K X-rays (capture) and K X-rays (conversion) or K X-rays (capture)and the 35 keV gamma-rays, detected by the same scintillation counter (window 2), see figure 3.

The formula for the activity (desintegration rate) in the window 1 case is given by [9] :

$$No = \frac{4K}{(1+K)^2} \cdot [N_{11} + \frac{N_{c1}(1-N_{c1}/N_{21})}{1-N_{c1}/N_{21}}] \cdot [N_{21} + \frac{N_{c1}(1-N_{c1}/N_{11})}{1-N_{c1}/N_{21}}] \cdot \frac{1}{2N_{c1}} \quad (1)$$

where

N_{11} and N_{21} are the counting rate at window 1 for detectors 1 and 2 respectively,

N_{c1} is the coincidence rate, and

$K = P_2/P_1$ where

$P_1 = P_K W_K$ is the X-rays emission probability for the electron capture events, and

$P_2 = (1+\alpha_K W_K)/(1+\alpha_T)$ is the K X-rays emission probability for the internal conversion events plus the 35 keV gamma-rays emission probability.

For the window 2 case, the activity is given by [9]:

$$No = \frac{4K}{(1+K)^2} \cdot [N_{12} + \frac{N_{c2}(1-N_{c2}/2N_{22})}{2(1-N_{c2}/2N_{12})}] \cdot [N_{22} + \frac{N_{c2}(1-N_{c2}/2N_{12})}{2(1-N_{c2}/2N_{22})}] \cdot \frac{1}{2N_{c2}} \quad (2)$$

The parameters have the same meaning as in equation (1), except that N_{12} , N_{22} and N_{c2} are measured at window 2.

The observed counting rates N_{ij} were corrected for background and dead time in the usual way. The dead time was measured by the source-pulser method[1]. The coincidence rates N_{cj} were corrected for dead time and accidental coincidences by means of the Cox-Isham formulae[4].

Sum-Peak Method

This method makes use of only one scintillation counter. In the case of ^{125}I two peaks are observed in the pulse height spectrum (see figure 3). The first peak with lower pulse height and higher intensity corresponds to pulses arising from K X-rays events overlapping the 35 keV gamma-rays, due to the poor resolution of the scintillator. The second peak is produced by the sum of pulses of coincident events arising from K X-rays from capture and K X-rays from conversion or gamma-rays from the 35 keV transition. The latter is called Sum-Peak.

The activity is given by[6]:

$$No = \frac{P_1+P_2}{(P_1+P_2)^2} \cdot \frac{(A_1 + 2A_2)^2}{A_2} \qquad (3)$$

where

A_1 and A_2 are the counting rates corresponding to the first and Sum-Peaks, respectively, corrected for background and dead time in the usual way.

A_1 correspond to N_{i1} in equation (1) and

$A_2 = (N_{i2}-N_{i1})$ from equations (1) and (2), for each detector.

P_1 and P_2 were defined previously.

Dependence of Observed Activity on Source Counting Rate

For the X-(X,γ) Coincidence method it is not expected any dependence of No on the counting rate since the N_{ij} and N_{cj} parameters were properly corrected for dead time and resolving time. The effect of pile-up in this case should not be significant because losses in the single rates of each detector give rise to losses in the coincidence rates and the expected change in the activity should be negligible. However, measurements were made at different source-detector distances and with different source masses to verify these assumptions. There was no change in the calculated activity as discussed later on.

For the Sum-Peak method it is expected that accidental summing of pulses produced by X-rays or the 35 keV gamma-rays can change the values of A_1 and A_2 given in equation 3.

The expected change in A_2 is approximately given by:

$$A_2 = A_2^o + 2\tau_r A_1^2 \qquad (4)$$

where

τ_r is the effective resolving time of the pulses contributing to the accidental summing and

A_2^o is the A_2 value extrapolated to zero counting rate.

From equations (3) and (4) it can be deduced that

$$No \cong No' \left(1 + \frac{B}{A} A_1\right) \qquad (5)$$

where

No' is the observed activity,

$B = 2\tau_r \left[\left(\frac{A_1}{A_2^o}\right) - 4 \right]$ and

$A = \left(\frac{A_1}{A_2^o} + \frac{4 A_2^o}{A_1} + 2 \right)$

A more detailed description of this correction will be given in a separate publication[5].

COUNTING EQUIPMENT

The counting rates needed for the ^{125}I desintegration rate by the two methods: X-(X,Y) Coincidence and Sum-Peak. were measured simultaneously by means of the electronic set up shown in figure 2. Two 3" x 3" NaI(Tl) scintillation detectors, type BICRON MODEL 3M3 were used, with a measured resolution FWHM (Full Width at Half Maximum) of 22.2 % for detector 1 and 24.8 % for detector 2. at the 28 keV photon energy corresponding to the ^{125}I K X-rays (see figure 3).

The set up allows the variation of the source-detector distance symmetrically with respect to the source. For most measurements this distance was fixed around 10 mm.

In figure 2, the components labeled 1 refer to window 1 and those labeled 2 refer to window 2. The energy range covered by window 1 was between 16 and 45 keV, and for window 2 it was between 16 and 72 keV, approximately. Windows 1 and 2 corresponds to the first peak and (first + sum) peaks, respectively, as shown in figure 3. The positions of the lower and upper discrimination levels are also shown in figure 3.

SAMPLE PREPARATION

The radioactive sources were prepared by dropping known aliquots of the ^{125}I solution on a 30 $\mu g/cm^2$ thick COLLODIOM substrate. The picnometer technique was used (2) and the aliquot masses were determined by means of a METTLER M5SA microbalance. The estimated mass uncertainty was around $\pm$ 15μg.

The diluited solutions were prepared from the ^{125}I master solution, having diluition factors of 7.82120 and 5.90248, respectively. A total of 20 sources were prepared, 8 from the master solution and 6 from each of the diluited solutions. To avoid volatilization of radioactive material, an aliquot (around 10 mg) of an aqueous solution containing

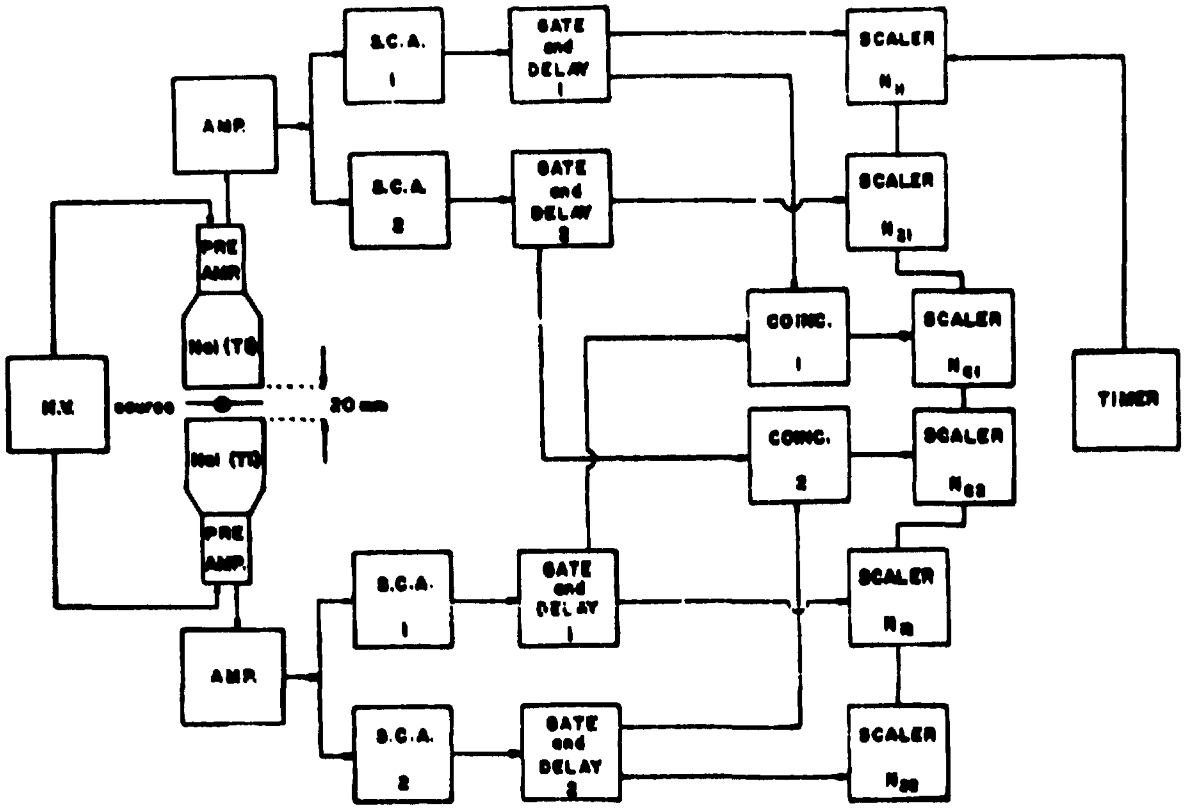


Figure 2 - Block diagram of the coincidence system.

160 μg of $AgNO_3$ per gram of solution was dropped on the ^{125}I source before drying in a dessecator atmosphere.

EXPERIMENTAL RESULTS

A typical ^{125}I pulse height spectrum obtained by the upper NaI(Tl) detector is shown in Figure 3. The first peak is produced by singles from X or gamma-rays and the second peak is produced by coincidence events adding up in the same detector as described in the section "Sum-Peak Method". The ratio between the counts in the valley between the two peaks and the centroid of the second peak was around 0.023. This allowed the separation between the counts of the two peaks to be performed with good accuracy.

The typical counting rates, measured dead time and resolving time for both detectors are shown in table 1. The highest rates correspond to sources prepared from the ^{125}I master solution and the lowest to sources from the diluited solutions.

TABLE 1

Typical counting rates, measured dead time (τ) and resolving time (τ_r) for the X-(X,γ) Coincidence method

Parameter	Window	
	J = 1	J = 2
N_{1j}	400 - 2700 cps	370 - 2400 cps
N_{2j}	420 - 2900 cps	390 - 2500 cps
Bg_{ij}	5.7 cps	8.2 cps
τ	(3.100 ± 0.040) μs	(3.172 ± 0.038) μs
τ_r	(0.0529 ± 0.0009) μs	(0.9806 ± 0.0010) μs

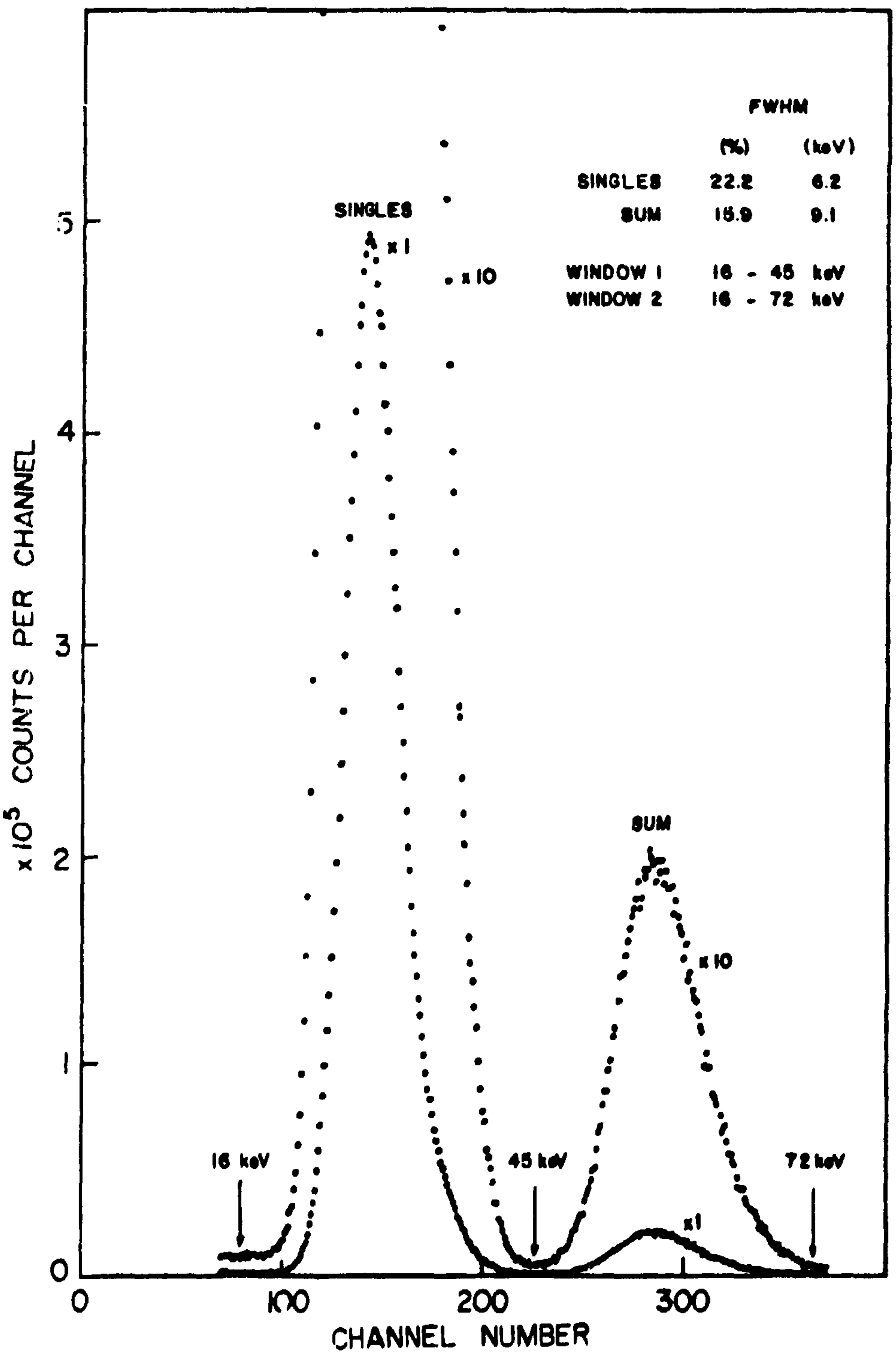


Figure 3 - A typical ^{125}I pulse height spectrum.

The counting rates for the Sum-Peak method are not shown since they are taken from N_{1j} and N_{2j}, as given by equation (3).

The dependence of the ^{125}I activity by the X-(X,γ) Coincidence method, as a function of the source-detector distance was verified from measurements at five different distances. The results agreed within the statistical uncertainty of $\lesssim 0.1\%$. There was also agreement in the case of the Sum-Peak method, but with a higher statistical uncertainty (~0.5%) due to the reduced number of counts A_2 (equation 3) at larger distances.

The parameters envolved in the extrapolation to zero counting rate for the Sum-Peak method were obtained by a least square fitting using 29 measured points. The results are shown in table 2. The value of A_2^o/A_1 for detector 2 is slightly lower than for detector 1 because the latter was a little closer to the source. The values of τ_r for both detectors are in excellent agreement. This should be the case since the electronic conditions for both detectors are the same. The ratio B/A indicates a correction around 1.3% for a counting rate around 1000 cps.

TABLE 2

Parameters envolved in the extrapolation to zero counting rate for the Sum-Peak method

Parameter	Value	
	Detector 1	Detector 2
A_2^o/A_1	$(6.083 \pm 0.026) \times 10^{-2}$	$(5.362 \pm 0.019) \times 10^{-2}$
τ_r	(0.427 ± 0.068) μs	(0.444 ± 0.056) μs
A	(18.682 ± 0.068)	(20.865 ± 0.066)
B	$(-2.276 \pm 0.34) \times 10^{-4}$	$(-3.054 \pm 0.36) \times 10^{-4}$
B/A	$(-1.218 \pm 0.18) \times 10^{-5}$	$(-1.464 \pm 0.17) \times 10^{-5}$

Table 3 shows the final results of the ^{125}I activity for the master and diluited solutions. The results for each window (17 to 45keV and 17 to 72keV) are shown for the X-(X,γ) Coincidence and Sum-Peak methods, together with their statistical uncertainty. The mean activity for the two methods agree well with each other for the three different solutions. The statistical uncertainty of the mean activity among the three solutions resulted in 0.20% for the X-(X,γ) Coincidence method and 0.23% for the Sum-Peak method.

TABLE 3

Final results for the ^{125}I activity (x 10^6 Bq/g), together with the statistical uncertainty (one standard deviation)

Condition		Master Solution Activity	σ(%)	Diluition 1 Activity	σ(%)	Diluition 2 Activity	σ(%)
Coinc	Window 1	1.4373	0.21	1.4282	0.17	1.4390	0.22
	Window 2	1.4350	0.21	1.4269	0.17	1.4379	0.23
Sum	Window 1	1.4375	0.16	1.4203	0.16	1.4386	0.29
	Window 2	1.4437	0.28	1.4300	0.27	1.4364	0.23
Coinc Mean		1.4362	0.21	1.4276	0.17	1.4386	0.22
Sum Mean		1.4406	0.23	1.4252	0.23	1.4375	0.26

Table 4 shows the source of errors envolved in the determination of the activity for the two methods used (mean of the three solutions). The predominant systematic errors are in the B/A value for the Sum-Peak method and in the K value for the X-(X,γ) Coincidence method. The overall uncertainty in the activity was 0.41% and 0.28% for the Sum-Peak and the X-(X,γ) Coincidence methods, respectively.

TABLE 4

Uncertainties in the ^{125}I activity measurements (in %)

Source	Sum-Peak	X-(X,γ) Coincidence
Statistics	0.23	0.20
Weighing	0.10	0.10
Dead Time	0.006	0.002
Background	0.005	0.008
Timing	0.005	0.005
$P_1P_2/(P_1+P_2)^2$	0.16	-
$4K/(1+K)^2$	-	0.16
Extrapolation (B/A)	0.28	-
Decay	0.06	0.06
Total	0.41	0.28

CONCLUSIONS

The consistency between the Sum-Peak and X-(X, γ) Coincidence methods was quite satisfactory within the estimated uncertainties. The extrapolation technique to zero counting rate developed for the Sum-Peak method showed good results and is a promising alternative for simple measurements of standardization performed with a single detector.

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