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Disintegration rate and gamma ray emission probability per decay measurement of $^{123}\mathrm{I}$

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ABSTRACT

A series of ¹²³I measurements have been carried out in a $4\pi(e_A,X)-\gamma$ coincidence system. The experimental extrapolation curve was determined and compared to Monte Carlo simulation, performed by code ESQUEMA. From the slope of the experimental curve, the total conversion coefficient for the 159 keV total gamma transition, α_{159} , was determined. All radioactive sources were also measured in an HPGe spectrometry system, in order to determine the gamma-ray emission probability per decay for several gamma transitions. All uncertainties involved and their correlations were analyzed applying the covariance matrix methodology and the measured parameters were compared with those from the literature.

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1. Introduction

One of the research areas under development by the Nuclear Metrology Laboratory (Laboratório de Metrologia Nuclear—LMN) at the Nuclear and Energy Research Institute (IPEN) in São Paulo, Brazil, is the primary standardization of radionuclides applied to Nuclear Medicine. In this context the radionuclide ¹²³I plays an important role because it is routinely supplied by IPEN to Nuclear Medicine Services for diagnosis of cardiac metabolism and functional and morphological thyroid studies. Moreover, it is being produced routinely at the IPEN Cyclone-30 cyclotron, by means of ¹²⁴Xe (p, 2n) ¹²³Cs reaction, followed by ¹²³Cs \rightarrow ¹²³I decay. For this reason, there was a need to develop its standardization by a primary method.

The radionuclide ¹²³I decays solely by electron capture process, with a half-life of 13.2234(37) h, mainly to the first excited state of ¹²³Te (97.18%) followed by a 158.99 keV gamma transition (Chisté and Bé, 2004a), as shown in Fig. 1. The intensity of this transition amounts to 99.22(30)% and it is partly converted with α_{159} equal to 0.1918(18). Several low yield gamma transitions are also present comprising the energy interval from 174 keV to 1068 keV and with probabilities per decay in the range from 0.0006% to 1.3%.

The LMN has two $4\pi\beta$ - γ coincidence systems composed of gas-flow or pressurized 4π proportional counters coupled to a single or a pair of NaI(Tl) scintillation counters. The latter may be replaced by an HPGe detector for high resolution measurements. A series of ¹²³I measurements have been carried out with one of

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these systems. The experimental extrapolation curve was determined and compared to Monte Carlo simulation, performed by code ESQUEMA (Takeda et al., 2005; Dias, et al., 2006). From the slope of the experimental curve, the total conversion coefficient for the 159 keV gamma transition was determined.

All radioactive sources were also measured in an HPGe spectrometry system. The ¹²³I radioactive solutions were checked for the presence of impurities by means of this system and a series of measurements were performed in order to obtain the emission probability per decay for several gamma-rays. All uncertainties involved and their correlations were analyzed applying the covariance matrix methodology (Smith, 1991) and the measured parameters were compared with those from the literature.

2. Methodology

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2.1. Coincidence equations

Full description of the coincidence equations can be found elsewhere (E.G. Campion, 1959; Baerg, 1966, 1967, 1973). In the case of ¹²³I, the equations were already described in the literature (Reher et al., 1984). Neglecting low intensity gamma transitions, and selecting the gamma window to cover only the total energy absorption peak at 159 keV, the formulae can be given by:

$$N_{4\pi} = N_0 \left\{ \varepsilon_{EC} + (1 - \varepsilon_{EC}) \left[\frac{\alpha_{159}}{1 + \alpha_{159}} \varepsilon_{ec} + \frac{1}{1 + \alpha_{159}} (\varepsilon_{4\pi})_{\gamma} \right] \right\}$$
(1)

$$N_{\gamma} = N_0 \varepsilon_{\gamma} \tag{2}$$

$$I_c = N_0 \varepsilon_{EC} \varepsilon_{\gamma} \tag{3}$$

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Fig. 1. Decay scheme of ¹²³I (Chisté and Bé, 2004a). The numbers on the right are the energy levels, in keV. The numbers close to the arrows correspond to electron capture decay probability for the main transitions, in percent.

where N_0 is the source disintegration rate; $N_{4\pi}$ is the 4π (PC) counting rate; N_{γ} and N_c are the gamma and coincidence counting rates, respectively; ε_{EC} is the PC total efficiency including X-rays and Auger electrons; ε_{γ} is the 159 keV gamma-ray peak efficiency; α_{159} is the total conversion coefficient, ε_{ec} is the conversion electron efficiency and ($\varepsilon_{4\pi}$)_{γ} is the PC gamma-ray efficiency for 159-keV transition. Combining Eqs. (1)–(3), the final expression is given by:

$$\frac{N_{4\pi}N_{\gamma}}{N_{c}} = N_{0} \left\{ 1 + \frac{(1 - \varepsilon_{EC})}{\varepsilon_{EC}} \left[\frac{\alpha_{159}}{1 + \alpha_{159}} \varepsilon_{ec} + \frac{1}{1 + \alpha_{159}} (\varepsilon_{4\pi})_{\gamma} \right] \right\}$$
(4)

In the extrapolation limit where $(1 - \varepsilon_{EC}) \rightarrow 0$, the value of N_0 can be determined. From the slope of the experimental extrapolation curve, it is possible to obtain α_{159} . For this purpose, the value of $(\varepsilon_{4\pi})_{\gamma}$ has been calculated by Monte Carlo code MCNPX (ORNL, 2006), as explained in Section 2.4. The calculated value of ε_{ec} by MCNPX resulted close to unity, as expected. Possible contribution from Compton scattered photons with higher energies might affect the slope of the extrapolation curve. However, this effect was estimated to be negligible by Monte Carlo simulation, as described in Section 2.4. Corrections for dead time and accidental coincidences were applied according to formulae taken from the literature (Smith, 1978).

2.2. Gamma-ray emission probability per decay

In the case of ¹²³I, the total gamma transition from the first excited state of ¹²³Te to ground state (159 keV) has probability per decay close to 99%. Therefore, if the probabilities per decay of the other transitions that reach the ground state are known, the value for 159 keV can be obtained by subtraction from 100%. The gamma-ray emission probability per decay for this transition will depend on the total gamma transition probability and on the value of the internal conversion coefficient α_{159} . On the other hand, this latter parameter can be obtained from the slope of the

extrapolation curve, as explained in Section 2.1. Therefore, in the present paper, the gamma emission probability per decay for the 159 keV transition was obtained from the gamma transition probabilities per decay of other low yield transitions, combined with the experimental value of α_{159} , as follows:

$$I_{\gamma}(159\,\text{keV}) = \frac{1}{1 + \alpha_{159}} \left(1 - \sum_{i=1}^{n} P_{\gamma i} \right)$$
(6)

where $P_{\gamma i}$ is the gamma transition probability per decay of the *i*-th gamma transition which reaches the ¹²³Te ground state; n is the total number of these gamma transitions. From Fig. 1, it can be observed that the corresponding gamma-ray energies are 440.02, 505.34, 599.69, 687.95, 783.62, 894.8, 1036.64 and 1068.18 keV. All of these transition probabilities were determined in the present paper, considering the gamma-ray emission probabilities per decay determined experimentally and the conversion coefficients taken from the literature (Chisté and Bé, 2004a). The theoretical internal conversion coefficients were calculated by ICC Computer code (program Icc99v3a-GETICC) (Chisté and Bé, 2004b). The validity of Eq. (6) depends on the assumption that there is no direct feeding of the Te ground state during the ¹²³I electron capture decay. This assumption was considered on the basis of decay scheme information from the literature (Chisté and Bé, 2004a). The gamma-ray emission probability per decay for the 159 keV transition obtained directly from the activity value and HPGe measurements has not been included in the present results because its uncertainty turned out to be 0.89% which is much higher than if Eq. (6) is used (0.27%).

In order to obtain the gamma-ray emission probability per decay for the low yield transitions, a series of measurements were carried out in a gamma-ray spectrometry system consisting of a thin Be window HPGe detector with 20% relative efficiency. The radioactive sources were positioned in a well-defined geometry, approximately 18 cm away from the detector front face. At this distance, the corrections for cascade summing are expected to be small. In spite of this, they were taken into account applying a Monte Carlo methodology developed at LMN (Dias et al., 2002). This system was calibrated by means of MCNPX (ORNL, 2006) calculations and compared with experimental peak efficiencies obtained with ¹⁵²Eu, ¹³⁷Cs, ⁶⁰Co and ¹³³Ba standards supplied by the IAEA. Monte Carlo efficiency calculation by means of MCNPX code was used in order to compensate for the lack of experimental points around 159 keV. The uncertainties in the calculated efficiency values were estimated by the average residuals between experimental values and calculation, which resulted 1.6%. This procedure has been considered more accurate than polynomial interpolation because the presence of Pb filter in front of HPGe detector changes the efficiency behavior and it shows a maximum around 158 keV, falling down below this energy, making it difficult to interpolate with a limited number of experimental points.

For ¹²³I, high yield X-rays and the 159 keV gamma-ray line are mixed together with several very low yield gamma-rays, for which the emission probability per decay was intended to be measured. At high counting rates, necessary to detect low yield gamma-rays, pile-up problems may arise and interfere with small peaks present in the HPGe spectrum. For this reason a 0.15 mm Pb filter has been placed between the radioactive source and the detector, in order to cut off the ¹²³I X-rays. Nevertheless, the 159 keV pile up events were present but did not interfere strongly with the low yield measurements.

2.3. Standardization setup

The extrapolation technique was employed to determine the activity of the solution by means of the $4\pi(e_AX) - \gamma$ coincidence system, consisting of a 4π gas-flow proportional counter (PC) coupled to a pair of 76×51 mm NaI(Tl) crystals. The PC counter was filled with 0.1 MPa P-10 gas mixture and operated at +2050 V. The ¹²³I radioactive sources were prepared by depositing known aliquots of solution with approximately 30 mg on a Collodion substrate 30 μ g cm⁻² thick, coated on both sides with 10 μ g cm⁻² thick gold layers, and applying the pycnometer technique (Campion, 1975). Aliquots of AgNO₃ were dropped onto the sources to prevent iodine evaporation. The beta efficiency was changed by applying external absorbers made of Collodion films $20 \,\mu g \, cm^{-2}$ thick placed over and under the ¹²³I sources. The extrapolation curve was obtained by measuring four ¹²³I sources and applying several different absorbers on each source. All sources were plotted together in order to obtain a single extrapolation curve.

The measurements were performed selecting a gamma-ray window at the total absorption peak of 159 keV in the NaI(Tl) and the events from the coincidence system were registered by a method developed at the Nuclear Metrology Laboratory (LMN) which makes use of a Time to Amplitude Converter (TAC) associated with a Multichannel Analyzer (Baccarelli et al., 2008).

All sources were measured in the HPGe gamma-ray spectrometer for impurity checks and for gamma-ray emission probability per decay determination. The results were obtained as the average of six measurements, covering a period of 58,000 s.

2.4. Monte Carlo simulation

The theoretical response functions of all detectors from the $4\pi(e_A,X) - \gamma$ coincidence system have been calculated using MCNPX Monte Carlo code (ORNL, 2006). More details on the experimental setup and calculation procedure can be found elsewhere (Takeda et al., 2005 and Dias et al., 2006). The value of ($\varepsilon_{4\pi}$)_{γ} to be applied in Eq. 4 has also been calculated by this code. In addition, MCNPX has been used for calculating all HPGe peak efficiencies involved in the gamma spectrometry calibration, including all standard sources and ¹²³I gamma-ray energies. Monte Carlo code ESQUEMA Version 2.0 (Takeda et al., 2005 and Dias et al., 2006), developed at LMN, has been used for calculating the extrapolation curve in the $4\pi(e_{A,}X) - \gamma$ coincidence experiment. All transitions from the precursor radionuclide down to the ground state of the daughter radionuclide have been taken into account. As a result, the whole coincidence experiment can be simulated and Eq. (4) could be reproduced theoretically as a function of the $4\pi(PC)$ detection efficiency, for the selected gamma-ray window. This version includes all Auger electrons that may be detected in the 4π detector.

The activity value (N_0) using the Monte Carlo procedure was obtained by minimizing the following chi-square:

$$\chi^2 = (\vec{y}_{exp} - N_0 \vec{y}_{MC})^T V^{-1} (\vec{y}_{exp} - N_0 \vec{y}_{MC})$$
⁽⁷⁾

where \vec{y}_{exp} is the experimental vector of $N_{4\pi} N_{\gamma}/N_c$; \vec{y}_{MC} is the $N_{4\pi} N/N_c$ vector calculated by Monte Carlo for unitary activity; N_0 is the activity of the radioactive source; V is the total covariance matrix, including both experimental and calculated uncertainties, and T stands for matrix transposition.

A series of simulated values were calculated for a wide range of beta efficiency parameter in small bin intervals. The \vec{y}_{MC} values used in Eq. (7) correspond to the same efficiency obtained experimentally.

The slope in the extrapolation curve in the $4\pi(e_A,X) - \gamma$ experiment is directly related to the value of the 159 keV transition internal conversion coefficient, as shown by Eq. (4). This value of 0.1918(18), derived by Chisté and Bé, (2004a) from theoretical tables, was used here to compare the calculated slope to the experimental one. The slope obtained in the Monte Carlo simulation turned out to be very close to the expected value given by Eq. (4). This result indicates that the contribution from higher energy photons below the 159 keV total absorption peak may be considered negligible. There is a systematic error in the Monte Carlo extrapolation curve due to the uncertainty affecting the theoretical α_{159} internal conversion coefficient, which is about 0.9%.

3. Results and discussion

Impurity checks detected < 0.01% contribution of 121 Te in the 123 I solution, at the reference date, and this contribution has been considered negligible. Fig. 2 shows the theoretical gamma-ray spectrum calculated by code ESQUEMA for 123 I, in comparison



Fig. 2. Gamma-ray spectra obtained for ¹²³I using Nal(TI) crystals from the $4\pi(e_A X) - \gamma$ system. The black marks correspond to the experimental data and white marks to the Monte Carlo calculation using code ESQUEMA (Takeda et al., 2005 and Dias et al., 2006).

with the experiment, using the pair of Nal(Tl) crystals at the $4\pi(e_AX) - \gamma$ coincidence system. The observed peak corresponds to 159 keV gamma-rays. The other low yield gamma-rays are barely visible. A reasonable good agreement between experimental and calculated spectra can be observed.

Fig. 3 shows the experimental extrapolation curve obtained by changing the absorber thickness, for a set of four radioactive sources. This curve was fitted by a straight line using covariance methodology. The intercept of this line yields the N_0 value, which turned out to be 544.5(25) kBq g⁻¹, as shown in Table 1. The residuals between experimental data and fitted curve are shown in Fig. 4. The error bars represent the overall error in the ordinate for each point. The distribution of points is quite uniform and does not show any noticeable bias. The uncertainty budget of activity determination is shown in Table 2. The main contribution to the overall uncertainty comes from the extrapolation curve fitting parameters.

The value of $(\varepsilon_{4\pi})_{\gamma}$ at 159 keV was calculated by MCNPX and resulted 0.0019(4). The uncertainty in this value comes from



Fig. 3. Extrapolation curves obtained for ¹²³I. The black marks are experimental points and the white marks are the Monte Carlo calculation. The straight line is the linear least square fitting.

Table 1

Extrapolation parameters and corresponding covariance matrix obtained by least square fit of extrapolation curve shown in Fig. 3. The intercept corresponds to the ¹²³I activity value. On the right is the total internal coefficient of 158.99 keV transition obtained experimentally.

Extrapolation parameters $(\times 10^3)$		Covariance matrix (× 10 ⁶)		α ₁₅₉	
a	544.5 ± 2.5	6.14473		0.1905 ± 0.0032	
b	$\textbf{87.95} \pm \textbf{1.20}$	-2.35576	1.43703		



Fig. 4. Percent residuals in the extrapolation curve least square fitting. The error bars are the standard deviation in the experimental points.

Table 2

Partial uncertainties involved in activity determination by $4\pi(e_A,X)-\gamma$ coincidence measurements, in percent (u = 1).

Component	Value (%) Remarks	
Weighing	0.09	Total uncertainty
Dead time	0.10	Pulser method
Background	0.10	Counting statistics
Decay	0.01	-
Resolving time	0.10	Accidental coincidences
Extrapolation curve	0.46	Statistics in curve fitting
Combined uncertainty	0.49	



Fig. 5. Percent residuals between the HPGe experimental peak efficiencies and the calculated values obtained by MCNPX code.

statistics in the calculation, which is the dominant contribution. The slope of the experimental extrapolation curve, after subtraction of $(\varepsilon_{4\pi})_{\gamma}$, as indicated by Eq. (4), yielded the total conversion coefficient α_{159} for the 159 keV transition, which resulted 0.1905(32), slightly lower but still in good agreement with 0.1918(18), taken from the literature (Chisté and Bé, 2004a).

From Eq. (7) the value for N_0 was 546.7(14) kBq g⁻¹, obtained by combining Monte Carlo calculation and experimental data, and agrees well with the linear fit result given in Table 1. An alternative procedure for obtaining the best fit between calculated and experimental extrapolation curves could be leaving the conversion coefficient value as a free parameter in the Monte Carlo simulation. In cases where the slope is not only affected by conversion coefficients but also includes other effects, such as contribution from other scattered gamma-rays below the selected gamma window, this procedure would not be adequate, because the curve behavior is nonlinear (see for instance the case of ¹³³Ba, described in Dias et al. (2002)). In the case of ¹²³I, these effects turned out to be negligible; therefore this procedure could be adopted. However, it is not justifiable because it is very time consuming and the expected slope yielded the same value as the experimental one, within their uncertainties.

Fig. 5 shows the residuals between the HPGe experimental peak efficiencies and the calculated values obtained with MCNPX code. The trend is uniform with no bias. Since there was a lack of experimental points between 121 and 244 keV, it was decided to use the Monte Carlo calculated efficiency instead of the experimental data in order to obtain peak efficiencies for ¹²³I. The error in the calculated peak efficiency was estimated from the average residual, resulting in 1.6%.

The experimental results for the gamma-ray emission probability (I_{γ}) per decay are presented in Table 3. The uncertainty

Table 3

Gamma-ray emission probability per decay of ¹²³I. The number inside the parenthesis corresponds to the uncertainty in the last digits (one standard deviation). The values from the literature correspond to Chisté and Bé, (2004a).

Transition	E (keV)	Literature ^a	This work	Transition	E (keV)	Literature ^a	This work
γ _{1,0} (Te)	158.99	83.25(21)	83.36(22)	γ _{10,2} (Te)	454.76	0.00412(22)	0.0033(2)
γ _{13,10} (Te)	174.20	0.00083(25)	0.00098(47)	γ _{4,0} (Te)	505.33	0.266(42)	0.281(5)
$\gamma_{6,4}$ (Te)	182.61	0.018(5)	0.015(6)	$\gamma\gamma_{6,1}$ (Te)	528.96	1.28(12)	1.245(24)
γ _{7,4} (Te)	192.17*	0.0199(7)	0.0162(7)	γ _{7,1} (Te)	538.54*	0.3788(43)	0.343(6)
γ _{10,7} (Te)	197.22	0.00383(72)	0.0027(41) ^b	γ _{11,2} (Te)	556.05	0.0029(3)	0.0024(2)
γ _{6,3} (Te)	198.23	0.0035(7)		γ _{13,4} (Te)	562.79	0.00115(7)	0.00088(13)
γ _{10,6} (Te)	206.79	0.00442(86)	0.0038(4) ^c	γ _{13,3} (Te)	578.26	0.00126(17)	0.0020(8)
γ _{7,3} (Te)	207.80	0.00112(32)		γ _{5,0} (Te)	599.69	0.00266(17)	0.0026(3)
γ _{6,2} (Te)	247.96	0.0698(23)	0.0687(15)	γ _{8,1} (Te)	610.05*	0.0011(3)	< 0.0001
γ _{7,2} (Te)	257.51*	0.0016(2)	0.00016(4)	γ _{9,1} (Te)	624.57	0.0798(2)	0.0759(14) ^e
γ _{9,4} (Te)	278.36	0.0023(4)		γ _{13,2} (Te)	628.26	0.00164(14)	
γ _{2,1} (Te)	281.03*	0.0789(9)	0.0701(13)	γ _{6,0} (Te)	687.95	0.0269(6)	0.0268(7)
γ _{10,5} (Te)	295.17*	0.001582(4)	< 0.0007	γ _{10,1} (Te)	735.78*	0.0616(8)	0.0494(9)
γ _{8,2} (Te)	329.38	0.0026(6)		γ _{9,0} (Te)	783.59*	0.0591(11)	0.0520(10)
γ _{3,1} (Te)	330.70	0.01164(33)		γ _{11,1} (Te)	837.10	0.000582(8)	0.00041(11)
		0.01424(7)	$0.0124(5)^{d}$	γ _{12,1} (Te)	877.52	0.00083(7)	0.00071(9)
γ _{9,2} (Te)	343.73	0.0044(3)		γ _{10,0} (Te)	894.80*	0.00101(7)	0.00068(7)
$\gamma_{4,1}$ (Te)	346.35	0.1257(9)	0.1183(21)	γ _{13,1} (Te)	909.12	0.00141(8)	0.00128(9)
γ _{10,3} (Te)	405.02	0.00298(23)	0.0026(3)	γ _{12,0} (Te)	1036.63	0.00097(7)	0.00076(6)
γ _{12,5} (Te)	437.50	0.0007(7)		γ _{13,0} (Te)	1068.12*	0.00142(7)	0.00100(8)
γ _{2,0} (Te)	440.02*	0.4229(43)	0.387(7)				

^a Chisté and Bé, 2004a.

^b Sum of (197.22+198.23).

^c Sum of (206,79+207,80).

^d Sum of (329,38+330,70).

^e Sum of (624.57+628.26).

Table 4

Uncertainty budget for the gamma-ray emission probability per decay measurements (u=1).

Component	Value (%)	Remarks
Monte Carlo HPGe peak efficiency	1.6	Estimated from average residual between calculation and experimental results
Counting statistics	0.1-36	Standard deviation of the mean
Cascade summing	0.03-0.20	Monte Carlo code
Doublet correction factor	0.01-0.40	For 182.61 keV, 528.96 keV, 538.54 keV and 578.26 keV gamma-ray peaks
HPGe Dead time correction	0.33	Pulser method
Combined uncertainty	1.7-48	Propagation

budget for these measurements is shown in Table 4. The value 83.36(22) for the main gamma-ray transition was obtained by means of Eq. (6), combined with the experimental value of α_{159} , given in Table 1. This result is in good agreement with the literature: 83.25(21) (Chisté and Bé, 2004a). Some transitions were too close in energy to be separated and their values given in Table 3 correspond to the sum of neighbor values, as indicated in the table footnote. Most of the other I_{ν} values obtained in the present work are in agreement with the literature (Chisté and Bé, 2004a) within three standard deviations. Some other I_{γ} values are not far from the literature but their values are not in agreement within the imposed deviation limit. These values are indicated by an asterisk at the right side of the energy value. The gamma-rays corresponding to 295.17 and 610.05 were not observed, and Table 3 gives an upper limit for their emission probabilities. The experimental value at 257.51 keV was double checked, but the value from the literature is exactly one order of magnitude higher, which is a quite odd result. The reasons for discrepancies in the remaining eleven values may be large background subtraction and spectrum distortions due to pile up.

The main contribution to the overall uncertainty in I_{γ} comes from background subtraction and counting statistics. Since the values in Table 3 come from relative measurements, most of corrections cancel out and do not contribute to the overall uncertainty.

4. Conclusion

The experimental result of the ¹²³I extrapolation curve, for the selected gamma window, agrees with the Monte Carlo simulation within the experimental uncertainty, indicating that the theoretical prediction can be used in a reliable way. The specific activity of the ¹²³I radioactive solution has been obtained with good accuracy.

The total conversion coefficient for the main transition (159 keV) was obtained experimentally with good accuracy and agrees with the literature. The gamma-ray probability per decay for this transition agrees with the literature within the experimental uncertainty.

Twenty-one of the low I_{γ} values agree with the literature within three standard deviations. Two gamma-rays were not observed and upper limits for their intensities were given. The other eleven experimental I_{γ} values do not agree with the literature, suggesting that new measurements may be necessary to confirm the present results.

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