The Half-life of ^{129}I

Eduardo García-Toraño[*1], Timotheos Altzitzoglou[2], Pavel Auerbach[3], Marie-Martine Bé[4], Christophe Bobin[4], Philippe Cassette[4], Frédéric Chartier[5], Rainer Dersch[6], Marta Fernández[7], Hélène Isnard[5], Karsten Kossert[6],Valérie Lourenço[4], Ole Nähle[6], Anthony Nonell[8],Virginia Peyrés[1], Stefaan Pommé[2], Andrej Rozkov[2], Anabel Sánchez-Cabezudo[1], Jana Sochorová[3]

[1] Laboratorio de Metrología de Radiaciones Ionizantes (CIEMAT), Avda. Complutense 40, 28040 Madrid, Spain
[2] European Commission, Joint Research Centre (JRC), Directorate for Nuclear Safety and Security, Retieseweg 111, B-2440 Geel, Belgium
[3] Czech Metrological Institute (CMI), Radiova 1, CZ 10200 Praha 10, Czech Republic
[4] CEA-LIST, Laboratoire National Henri Becquerel (LNE-LNHB) CE-Saclay, 91191 Gif sur Yvette Cedex, France
[5] Den – Département de Physico-Chimie, CEA, Université Paris-Saclay, F-91191 Gif sur Yvette Cedex, France
[6] Physikalisch-Technische Bundesanstalt (PTB), Bundesallee 100, 38116 Braunschweig, Germany
[7] Unidad de Espectroscopía de Masas y Apl. Geoquimicas (CIEMAT), Avda. Complutense 40, 28040 Madrid, Spain
[8]Den – Service d'Etudes Analytiques et de Réactivité des Surfaces (SEARS), CEA, Université Paris-Saclay, F-91191 Gif sur Yvette Cedex, France

* Corresponding author. Tel.: +34 91 346 6225; fax: +34 91 346 6442.
E-mail address: e.garciatorano@ciemat.es

Abstract

The radionuclide ^{129}I is a long-lived fission product that decays to ^{129}Xe by beta-particle emission. It is an important tracer in geological and biological processes and is considered one of the most important radionuclides to be assessed in studies of global circulation. It is also one of the major contributors to radiation dose from nuclear waste in a deep geological repository. Its half-life has been obtained by a combination of activity and mass concentration measurements in the frame of a cooperation of 6 European metrology institutes. The value obtained for the half-life of ^{129}I is 16.14 (12) × 10^6 a, in good agreement with recommended data but with a significant improvement in the uncertainty.

Keywords ^{129}I; half-life; standardization; mass spectrometry; activity measurements

1 Introduction

The radionuclide ^{129}I, a long-lived fission product, decays to ^{129}Xe via two beta branches, one containing 99.5% of all disintegrations to an excited level at 39.578 keV in the daughter nuclide and a second with 0.5% to the ground level. The gamma transition depopulating the excited level is converted, with a total conversion coefficient α_T=12.41(13) [1]. Its decay scheme is presented in Figure 1. Small amounts of this nuclide are produced in nature, but natural levels have been altered in the past by nuclear weapons testing that released large amounts into the atmosphere. At present, it is mainly produced by the nuclear fission of ^{235}U and ^{239}Pu and released as a consequence of the operation of nuclear power plants and fuel reprocessing centres [2].

Iodine-129 is an important tracer in geological and biological processes and is considered to be one of the most important radionuclides to be assessed in studies of global circulation [3]. It is also one of the major contributors to the radiation dose from nuclear waste in a deep geological repository. Its transmutation has been suggested as a possible method to destroy it, as its immobilization in repositories poses difficult problems [4].

FIGURE 1

The recommended value[1] for the half-life of ^{129}I is 16.1 (7) × 10^6 a. It was obtained by Chechev and Sergeev in 2004 [1] as the weighted mean of a set of 4 results with a significant dispersion (from 1.56 (6) × 10^7 a to 1.97 (14) × 10^7 a). For this reason, it was selected for half-life determination in the frame of the coordinated research project "ENV09 /Metrology for Radioactive Waste Management" of the European Metrology Research Programme [5] with the aim of reducing its uncertainty.

Given its long half-life, the determination by following the decay rate had to be discarded and its value has been obtained using the relationship:

$$T_{1/2} = \frac{\ln(2).N}{A} \qquad (1)$$

where A is the activity and N is the number of atoms in a source of a nuclide whose half-life is $T_{1/2}$. For this determination, a combination of measurements of activity and mass concentrations of the same solution was carried out by the participants. Five European Metrology laboratories (CIEMAT, CMI, JRC, LNE-LNHB, PTB) performed activity concentration measurements whereas mass spectrometry measurements were carried out at CEA-LANIE (Laboratory of Analytical development for isotopic and elemental analysis) and CIEMAT.

2. Material procurement and initial measurements

The ^{129}I solution used for the comparison, with an approximate massic activity of 30 kBq·g^{-1}, was purchased by CIEMAT from CERCA-LEA. Its chemical composition was

[1] Unless otherwise indicated, uncertainties stated in this work are given as standard uncertainties (k=1)

NaI-NH_4OH-$Na_2S_2O_3$. Upon reception, the solution was tested at CIEMAT by gamma spectrometry to assess the purity and was found to be free of detectable radioactive contaminants. Preliminary mass spectrometry measurements indicated a $^{127}I/^{129}I$ mass ratio higher than 5.

Aliquots of the solution were dispensed into 5 mL penicillin-type vials previously treated with a carrier solution of similar composition to that of the radioactive solution. Since the original solution already contained a certain (unknown) amount of ^{127}I, a possible addition of that isotope coming from lixiviation of the vial wall cannot be a problem for mass determination, given that it would be included in the $^{127}I/^{129}I$ measured isotopic ratio. Furthermore, no difference was found between the isotopic ratio measured in a sample contained in a vial saturated with the carrier solution (CEA-LANIE) and another contained in a vial not treated in this way (CIEMAT). Therefore, the incorporation of ^{127}I from the walls can be considered negligible or not detectable by ICP-MS.

An amount of 1.5 g of the solution was dispatched to each of the laboratories participating only in activity measurements: CMI, JRC and PTB; LNE-LNHB received 2 grams of solution, as this laboratory, through other CEA units, was also involved in mass spectrometry measurements. Given the low energy of the gamma emissions from this radionuclide, homogeneity tests could not be performed either by ionization chamber measurements or by gamma-ray spectrometry. In the latter case, differences in the structure of the vials combined with the gamma-ray attenuation would make the analysis unreliable. The hypothesis of homogeneity of the material was adopted based on the fact that all vials were treated before dispensing the material using the same procedure (vial saturation with a carrier solution) and dispensing was done within a short period of time.

3. Massic activity measurements

The massic activity of the solution was determined using the methods presented in Table I. Since there was no previous comparison of massic activity for this nuclide in the frame of EURAMET or the Consultative Committee for Ionizing Radiation, Section II: Measurement of radionuclides, CCRI(II), an intercomparison was agreed; after acceptance it was registered as EURAMET.RI(II)- S6.I-129 Supplementary Comparison. The technical protocol for the comparison was agreed between partners; it included the recommendation of using nuclear data from the NUCLEIDE database [1]. According to it, the half-life value is $T_{1/2} = 16.1\ (7) \times 10^6$ a. This value was used by all participants, excluding PTB that used its own reference with $T_{1/2} = 15.7\ (5) \times 10^6$ a. Neither the absolute value, nor the difference between both references had significant influence on the activity measurements. The reference date was established as 1st July 2013, 0:00 UTC. Complete details about the methods used by participating laboratories as well as detailed uncertainty budgets and inter-comparison results are presented elsewhere [6].

Table I. Participanting laboratories and techniques used for the determination of the massic activity of the ^{129}I solution

	Liquid Scintillation Counting	4πγ counting	Coincidence counting
CIEMAT	CIEMAT/NIST	NaI well detector	
CMI			4πβ-γ coincidence counting with proportional counter
JRC	TDCR, CIEMAT/NIST		
LNE-LNHB	TDCR		4πβ-γ anti-coincidence counting with TDCR
PTB	TDCR, CIEMAT/NIST		

3.1 Impurities

Radionuclidic impurity assessment was done by gamma-ray spectrometry by all participants. The nuclide ^{129}I is a fission product and the potential contaminants of interest are all iodine isotopes. HPGe detectors of various types (n-type coaxial with Be window, extended-range p-type coaxial with carbon-epoxy window and planar) were used with good counting efficiency at low energies. No evidence of gamma-ray emitting contaminants was found in the measurements of any of the participating laboratories.

Adsorption tests made by JRC reported negligible values. All participants measured the original solution without dilution. The pycnometer method was adopted for all source preparations.

3.2 Measurements by Liquid Scintillation Counting

Two LSC-based methods were used to determine the activity per unit mass: The CIEMAT/NIST method [7, 8] and the Triple to Double Coincidence Ratio method (TDCR) [9,10].

The CIEMAT/NIST efficiency tracing method was used by CIEMAT, JRC and PTB. All measurements were done using commercial counters with two photomultiplier tubes (PMT). Tritium activity standards required by the method were from different origin: CIEMAT used a ^{3}H standard from NIST whereas JRC and PTB used their own standards. All participants used samples with Ultima Gold as scintillation cocktail, and some participants used in addition samples with Instagel Plus or Hisafe 3. Samples were prepared in glass vials. At JRC and PTB, about 1 mL water was added to some of the samples. Low-level counting options (i.e. guard mode), when available, were disconnected because photon emission from the source can interact with the guard detector, thus making the application of this method impossible. Typical counting efficiencies for ^{129}I samples were higher than 94% in all counters.

The TDCR method was applied at JRC, PTB and LNE-LNHB. In all cases, participants used their custom-built counters with 3 PMTs. Ultima Gold was selected as scintillation cocktail by PTB and JRC (that also used samples with Instagel Plus). LNE-LNHB preferred Hionic Fluor. All participants used glass vials. PTB also prepared some samples with polyethylene (PE) vials. As for CIEMAT/NIST measurements, JRC and PTB added about 1 mL water to some samples (0.5 mL for PTB to samples in PE vials). Typical counting efficiencies were also high, in the order of 95%.

Results for each laboratory and technique are given in Table II and typical uncertainty budgets from both methods are presented in Table III. Additional details are given in ref. [6].

3.3 Measurements by coincidence and anti-coincidence counting

Coincidence and anti-coincidence counting was performed at CMI and LNE-LNHB. At CMI, the 4πβ-γ coincidence counting method was used with a proportional counter and two gamma-ray detectors. Samples were prepared on Mylar foils with thickness from 350 $\mu g \cdot cm^{-2}$ to 25 $\mu g \cdot cm^{-2}$ attached to metallic rings. They were covered by Al foils (2 $\mu g \cdot cm^{-2}$) on both sides and placed in a pill-box-type 4π proportional counter at atmospheric pressure in gas-flow mode using methane as counting gas. The discrimination threshold was set at 0.7 keV. A non-extending dead time of 5.995(5) μs was used. The gamma channel comprises two NaI crystals with 76.2 mm diameter and a height of 76.2 mm each. Non-extending dead times of 6.031(5) μs and 6.113(5) μs, respectively were used in the NaI detectors. In order to implement the efficiency extrapolation procedure, the counting efficiency was varied by adding Al foils to the sources. The highest counting efficiency reached in the proportional counter was 88 %.

The LNHB applied the 4πβ-γ anti-coincidence counting using liquid scintillation sources prepared with Hionic Fluor. A TDCR counter based on 3 PMTs was used in the β−channel and a HPGe semiconductor detector in the γ−channel. The extrapolation method was carried out by PMT defocusing. The maximum detection efficiency in the β-channel was 94%. Dead time control was carried out by a live-time technique based on home-made modules.

Results and uncertainties from both laboratories are presented in Tables II and III respectively.

3.4 Measurements by 4π gamma counting

This method, also known as integral gamma counting was used only at CIEMAT. The basis of the method has been described by Winkler and Pavlik [11]. Its application to the standardization of some nuclides with complex decay schemes is described by García-Toraño et al. [12], and an overview of uncertainty calculation has been discussed by Thiam et al. [13]. The detection efficiency for this specific nuclide was close to 58%. It was calculated by Monte Carlo simulation with the PENELOPE package [14]. Results are given in Table II.

3.5 Results of the massic activity measurements

Table II shows the results of the measurements obtained by the laboratories. Only one final value was reported for each laboratory, in most cases obtained by combination of two methods. Final results from each laboratory are presented in graphical form in Figure 2.

The final value was obtained as the power-moderated mean [15] of 4 final laboratory results, excluding the CIEMAT contribution as outlier. The reason for the discrepancy is, for the moment, unknown. The final value is 33.10 (5) $kBq \cdot g^{-1}$. The stated uncertainty corresponds to the standard deviation of the 4 results.

FIGURE 2

Table II

Final results for the massic activity *A*, and its uncertainty *u*, as reported by the participants in the EURAMET.RI(II)- S6.I- 129 Supplementary Comparison.

Laboratory	A kBq·g^{-}	$u(A)$ kBq·g^{-1}	Method to obtain the final value	Results from each measurement technique
CIEMAT	32.14	0.11	Mean value of results obtained with two techniques. Uncertainty obtained combining both uncertainties with the expression $1/u^2 = 1/u_1^2 + 1/u_2^2$	32.15 (13) CIEMAT/NIST 32.13 (23) Gamma Counting
CMI	33.10	0.57	Only one result	33.10 (57) coincidence
JRC	33.0	0.1	Mean value of results obtained with two techniques. The uncertainty obtained with a single method is kept as more realistic	33.0 (5) CIEMAT/NIST 33.0 (1) TDCR
LNE- LNHB	33.16	0.06	Result from anticoincidence counting	33.1 (11) TDCR 33.16 (6) Anticoincidence
PTB	33.003	0.099	Weighted mean of the results obtained with two techniques.	32.90 (50) CIEMAT/NIST 33.007 (99) TDCR

Table III. Typical uncertainty budgets associated to each technique. Only major components are indicated. Since uncertainties declared by participants for a given technique were similar, only results from one laboratory are given. Additional details can be found in ref. [6].

Laboratory	JRC	PTB	CIEMAT	CMI	LNE
Measurement technique	CIEMAT/NIST	TDCR	$4\pi-\gamma$ counting	Coincidence Counting	Anti-coincidence Counting
Contribution due to	***u*(*a*)/*a* in %**				
Counting statistics	0.04	0.04	0.2	0.2	0.05
Weighing	0.12	0.03	0.1	0.01	0.05
Background	0.01	0.03	0.02	0.02	0.1
Dead time		0.03			
Resolving time	-			0.01	
Decay data	0.13	0.22	0.2		
Half-life		0.01			
Quenching	0.1				
Interpolation/ extrapolation of efficiency curve	0.02		0.1	0.50	0.13
Impurities		0.03	0.01	0.1	
Adsorption	0.04	0.05	0.01	0.1	
PMT asymmetry		0.02			
Counting time		0.01			
Ionization quenching and *kB*	0.02	0.11			
LS spectrometer dependence	0.2				
Calculation code dependence	0.16				
Shape factor dependence	0.15				
TDCR value		0.13			
Numerical model			0.2		
Monte Carlo statistics			0.2		
Escape from sample				1.6	
Combined relative standard uncertainty	**0.38**	**0.30**	**0.43**	**1.7**	**0.18**

4. Mass concentration measurements

The number of ^{129}I atoms per gram of solution was determined at CEA-LANIE and CIEMAT by mass spectrometric measurements combined with the isotope dilution (ID) technique.

4.1 Analytical strategy

Isotope dilution is a method of determining the quantity of chemical substances and is regarded among the chemistry measurement methods of the highest metrological standing. This method [16] was associated to mass spectrometric measurements in order to obtain high accuracy on mass concentration and has been used in the past for half-life determinations [17-19].

The ^{129}I mass concentration was determined by isotope dilution using certified ^{127}I solutions according to the ID equation (2):

$$C_{^{129}I} = \left|^{129}I\right|_S \times C_{Sp} \times \frac{m_{Sp}}{m_S} \times \frac{M_S}{M_{Sp}} \times \frac{1}{(^{127}I)_S} \times \frac{\left(\frac{^{129}I}{^{127}I}\right)_{Mix}}{\left(\frac{^{129}I}{^{127}I}\right)_S - \left(\frac{^{129}I}{^{127}I}\right)_{Mix}} \qquad \textbf{(2)}$$

where *Mix*, *S* and *Sp* stand for mixture, sample and spike, respectively; m_{Sp} and m_S are the masses used to prepare the blends (in g), M_{Sp} and M_S are the respective atomic weights (in g·mol^{-1}) for the spike and the sample, $(^{127}I)_S$ is the ^{127}I abundance (in atom%) in the sample, and $|^{129}I|_S$ is the ^{129}I abundance (w%) in the sample.

The two laboratories have used two different mass spectrometers and two different spike solutions. Details are given below.

4.1.1 Mass spectrometry instrumentation

The main difference between the work carried out by LANIE and CIEMAT refers to the instrumentation, a Multicollector Inductively Coupled Plasma Mass Spectrometer (MC-ICPMS) and a Quadrupole based Inductively Coupled Plasma Mass Spectrometer (Q-ICPMS), respectively. Regarding the operation mode this implies: (i) the mass analyzer, a magnetic/electrostatic sector (flat-topped peaks) in the case of the MC-ICPMS equipment and a quadrupole (Gaussian peaks) in the Q-ICPMS instrument and (ii) the detection system, multiple high stability Faraday cup detectors to simultaneously measure several isotopes (MC-ICPMS) and single electron-multiplier detector that measures the ions (Q-ICPMS).

Technical information regarding the measurements performed at LANIE by MC-ICPMS is presented in a recent work [20]. The mass bias corrections related to the

enhanced extraction and transmission of the heaviest ions [21] were performed by both laboratories applying the exponential law fractionation [22] and using tellurium as an internal standard. The instruments used by both laboratories are presented in Table IV together with other relevant details as the media of analysis and the ratios used for normalization.

Table IV. Instrumentation, media of analysis and strategy for mass bias correction performed by CIEMAT and LANIE laboratories in charge of mass spectrometric measurements.

	Instrument	Media of analysis	Mass bias correction	Ratio(s) used for mass bias correction [23]
CIEMAT	ICPMS Q (iCap Q, Thermo Instruments)	NH_4OH 3.4M	Internal correction (tellurium)	$^{126}Te/^{130}Te$ and $^{128}Te/^{130}Te$
LANIE	MC-ICPMS (Isoprobe, GV Instrument)	Dilution of the sample in HNO_3%	Internal correction (tellurium)	$^{126}Te/^{130}Te$

4.1.2 ^{127}I Spike

Laboratories have used independent spikes. LANIE has used a ^{127}I spike provided by the National Institute of Standards and Technology (NIST SRM 3180). The presence of other elements is indicated in the certificate and no trace of ^{129}I is mentioned. This was confirmed by qualitative Q-ICPMS measurements. It was sent in five independent closed vials in Na_2SO_3 and NaOH media to ensure the stability and conservation of these solutions. The concentration for each vial was certified at 1.0006 (12) $mg \cdot g^{-1}$.

CIEMAT has used a spike that was gravimetrically prepared from potassium iodide (99.995% minimum purity, Merck Suprapur), assuming that 100% of the iodine was ^{127}I. The concentration of the solution used as spike was 1.5155 (25) $\mu g \cdot g^{-1}$. The uncertainty of the spike was calculated taking into account all components of weighing and purity.

4.2 Results of the mass concentration measurements

Table V presents the values of the ^{129}I mass concentration obtained by both laboratories. Each one has carried out the determinations on a set of mixtures, prepared independently from the sample and spike.

Table V. Final results for the mass concentration, as reported by the participants.

Laboratory	[^{129}I] atom·g^{-1}	u([^{129}I]) atom·g^{-1}	Method to obtain the final value
CIEMAT	2.450×10^{19}	0.018×10^{19}	Mean of five sample-spike mixtures
LANIE	2.415×10^{19}	0.017×10^{19}	Mean of six sample-spike mixtures

The uncertainty was calculated in accordance with the ISO Guidelines for the Expression of Uncertainty in Measurement [24]. For uncertainty propagation calculations, the numerical method of Kragten [25, 26] was applied. Uncertainty contributions of the major component of the ID equation evaluated by the two laboratories are indicated in Table VI. The (^{129}I/^{127}I) Mix and (^{129}I/^{127}I)$_S$ ratios were corrected for mass bias by tellurium isotope ratios and their uncertainties have been taken into account in the uncertainty budget. Figure 3 presents graphically the main sources of uncertainty in the measurements of the two laboratories.

Table VI. Typical uncertainty budgets associated to each technique by CIEMAT and LANIE

Laboratory	CIEMAT	LANIE
Measurement technique	Isotope dilution associated to ICPMS Q measurements	Isotope dilution associated to MC-ICPMS measurements
Uncertainty component	**Uncertainty contribution in %**	
(^{127}I/^{129}I)$_S$	0.22	0.02
[^{127}I]$_{Sp}$	0.12	0.12
m_{Sp}	0.08	0.09
m_S	0.1	0.08
^{126}Te/^{130}Te ^{128}Te/^{130}Te	0.66	0.67
(^{127}I/^{129}I)$_{Mix}$	0.15	0.12
Combined relative standard uncertainty	0.73	0.70

As can be seen in Table VI, the main sources of uncertainty in the CIEMAT measurements come from the concentration of the spike solution and the measured isotope ratio of sample and mixtures. The uncertainty of the absolute ^{126}Te/^{130}Te and ^{128}Te/^{130}Te isotope ratios used for mass bias correction has already been considered in the calculation of the ^{127}I/^{129}I ratios involved in the equation. Note that the weighing of sample and spike has a very small contribution. The major source of uncertainty for CIEMAT and LANIE comes from the ^{126}Te/^{130}Te and ^{128}Te/^{130}Te IUPAC ratios [23] used for instrumental mass bias correction. The IUPAC uncertainty associated with this representative tellurium isotopic composition includes the overall range of variations that are likely to be encountered in both natural materials and chemicals. Since this component dominates the uncertainty budgets from the two mass determinations, the

final relative uncertainty of the whole process was conservatively estimated as equal to that of CIEMAT (0.73 %).

FIGURE 3

The final value of the mass concentration was obtained as the average of values obtained by CEA and CIEMAT which is:

$2.432\ (18)\times10^{19}$ atoms·g^{-1}.

5. The half-life of ^{129}I

As indicated in Section 1, the half-life of ^{129}I is obtained from Eq. 1, using the values determined in this work.

$T_{1/2}$ (^{129}I) = 16.14 (12) × 10^6 a

Its relative standard uncertainty was evaluated using the expression:

$$u_r\left(T_{\frac{1}{2}}\right) = \sqrt{\left(\frac{u_A}{A}\right)^2 + \left(\frac{u_N}{N}\right)^2} = 7.\times 10^{-3}$$

from which the mass determination is the major component.

The result is in good agreement with the evaluated value of 16.1 (7) × 10^6 a [1], but carrying an uncertainty that is more than 5 times lower.

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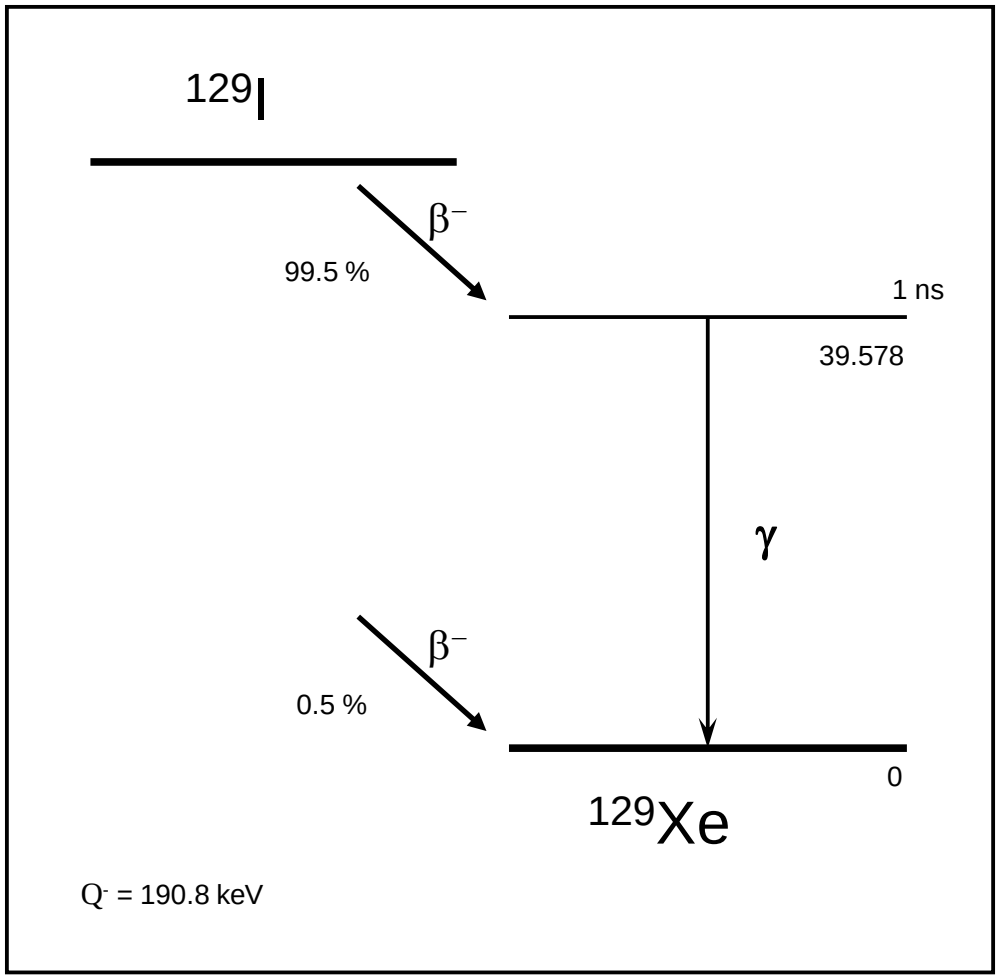


Figure 1. Simplified decay scheme of ^{129}I. Data have been taken from [1].

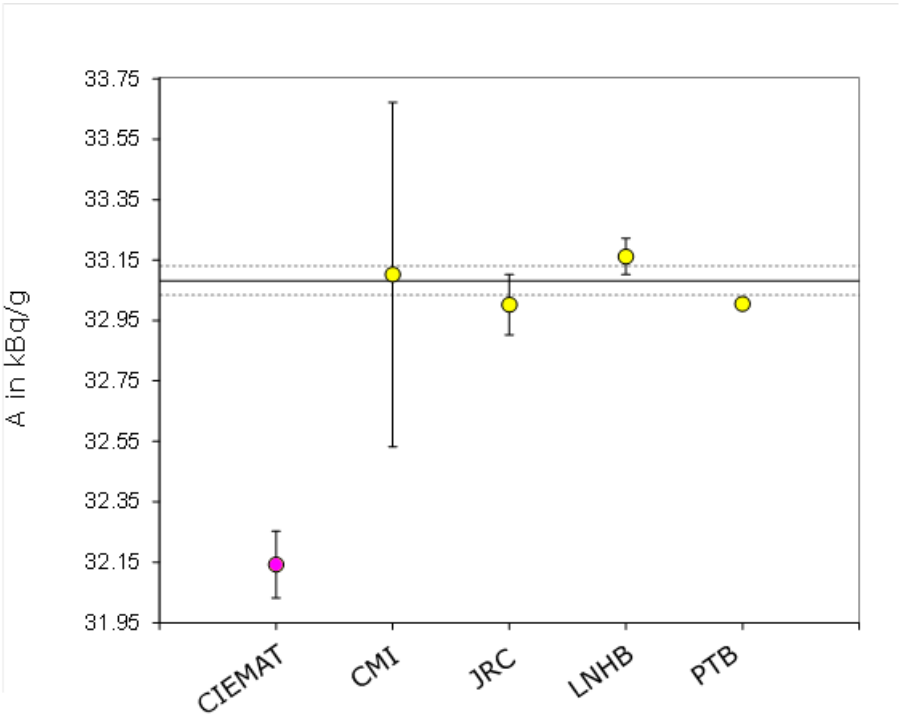
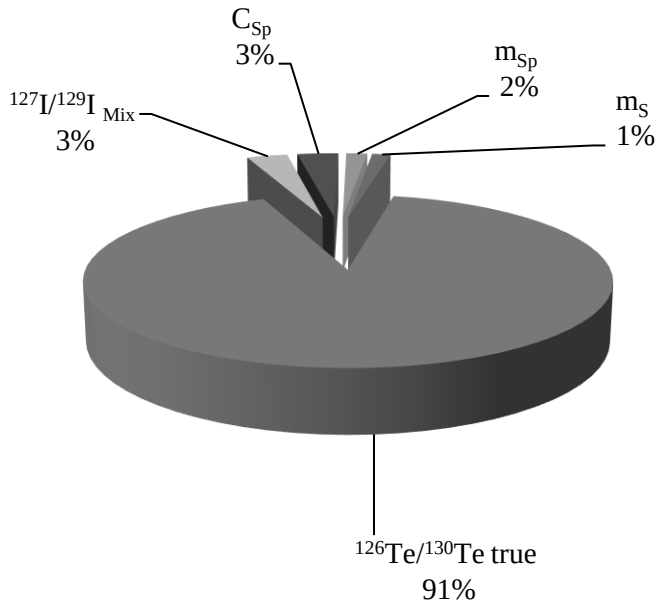
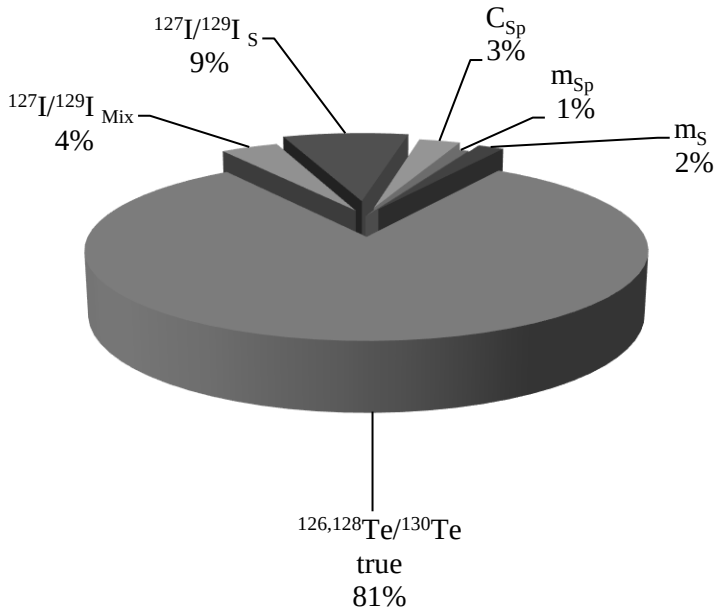


Figure 2

Final laboratory results for the massic activity of aliquots of the same ^{129}I solution. The horizontal line corresponds to the value of the power-moderated mean of the 4 consistent values.



1



2

3 **Figure 3: Relative contribution of the major sources of uncertainty in the**
4 **determination of the ^{129}I concentration at LANIE (upper) and CIEMAT.**