French National Atomic Energy Agency (CEA)

French National Metrology Office (LNE)

Laboratoire National Henri Becquerel

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Laboratoire National Henri Becquerel (LNHB) has been designated as the "Primary Standards laboratory for Ionizing Radiation Metrology"

Conservation of the national standards

• Takes part in the international comparisons

(Bureau International des Poids et Mesures, BIPM)

(Euromet organisation)

Research and Developments

•Measurement methods

•Nuclear Data measurements

• sources preparation

• simulation calculations

•Atomic and Nuclear data Evaluation

LNHB has the responsibility for the preparation and improvement of the French national primary standards for the units in radioactivity (becquerel) and ionizing radiation dosimetry (gray)

Traceability chain

•Calibration

•Transfer device

Tasks of a National Metrology Institute

Provide users with the metrological standards they need through a strictly established traceability

R & D

International traceability

Transfer to users

Maintenance and upgrading of standards

Coherence and excellence of the national metrology chain

Teaching

From NMI to final user

Primary standard:

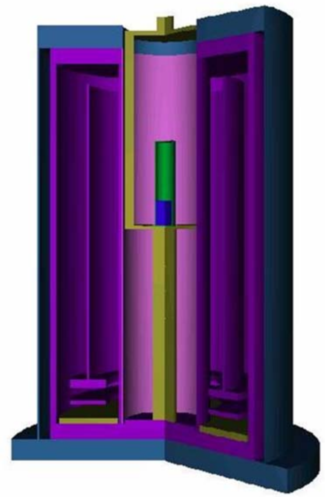
Liquid Scintillation Counting Technique

TDCR method

u_c=0.5 %

Secondary standard:

Ionization chamber



u_c=1.5 %

Users: dose calibrator

(activimeters)

u_c=3 to 4 %

Traceability chain of activity measurement : ^{18}F example

A good knowledge of the decay scheme data is required at each part of the metrology chain.

1) Which ones ?

The data describing the radioactive disintegration : **half-life, emission energy and intensity of the various radiations, etc.**

2) For which needs ?

- metrology : **detector calibration, simulation calculations, etc.**
- medical uses **: diagnostic (Tc-99m, Tl-201, F-18, …), therapy (Ir-192, I-131, Y-90, …)**
- Nuclear fuel cycle **: residual power in the reactor, waste management , control of the nuclear matter, etc.**

Our job :

As primary standard laboratory, we must determine data which will be recommended data.

1) Data Evaluation**, is done from :**

- **the published measured values**
- **theoretical calculations**
- **the consistency of the decay scheme**

2) To make available the recommended data

- **« Nucleide », a computerized database, on CD rom**
- **publication of articles and Tables of data on "paper"**
- **publication of a website :** www.nucleide.org
- **participation to working groups : RNVL, IAEA, etc.**

- The evaluation of data is time consuming
- An international working group has been formed in 1995 (DDEP):

↓

LNHB (France), PTB (Germany), INEEL and LBNL (USA), KRI (Russia)

The current members of DDEP are :

Marie-Martine Bé (LNHB, France); *Editor-In-Chief*
Filip G. Kondev (ANL, United States); *Coordinator, Editor*
Valery P. Chechev (KRI, Russia)
Christophe Dulieu, Vanessa Chisté, Xavier Mougeot (LNHB, France)
Mark Kellett, Alan L. Nichols (IAEA, Austria)
Edgardo Browne (LBNL, United States)
Tibor Kibédi (ANU, Australia)
Aurelian Luca (IFIN, Romania)
Monica Galan (Ciemat, Spain)
Andy Pearce, Arzu Arinc (NPL, UK)
Huang Xiaolong (CIAE, China)

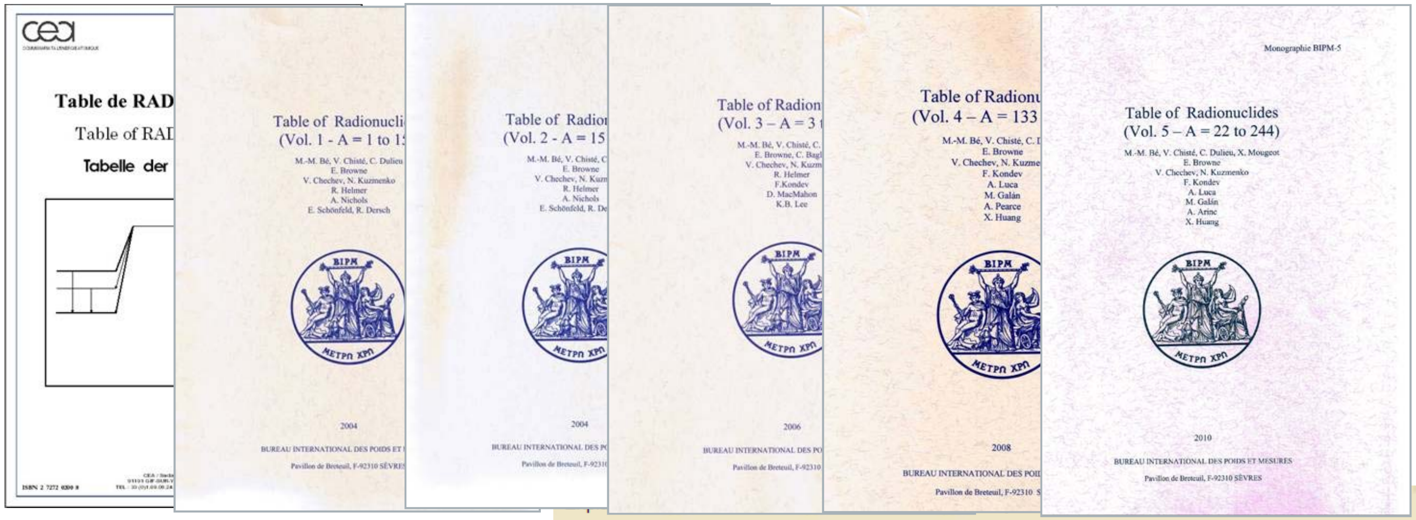
In this group we :

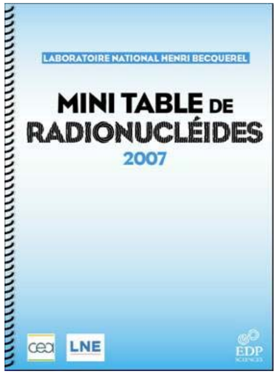
- use the same methodology in the evaluations,
- use the same set of specific data, like Q-values or Internal Conversion Coefficients, established by specialists in this field,
- we provide written documentation of all data used and all decisions and calculations done during the evaluation process,
- the review of each new evaluation is done by other members of the group.

Then, the results of these DDEP evaluations are compiled and edited by the LNHB as a Monographie published under the auspice of the Bureau International des Poids et Mesures (BIPM).

Moreover the *International Committee for Radionuclide Metrology* (ICRM) formally approved the recommendation made by the Nuclear Data Working Group of using the DDEP evaluated decay data in all future nuclear data studies.

Publications





Tables of evaluated data and comments on evaluation
Pages updated by the Laboratoire National Henri Becquerel
All questions about the data must be sent to the authors. See chapter Addresses.

updated: 3rd March 2011
latest entries: Bi-213, Pa-231, Ra-223, Rn-219
latest updates: Cu-64, Po-216, Ra-224, Rn-220
(181 nuclides in table, sorted by alphabetical order / atomic number / mass number / edition date

Subscribe to DDEP RSS feed

(Type of updates since last revision: 1 - update in comments only ; 2 - minor update in table ; 3 - major update in table)

Nuclide		Tables	Comments	ENSDF	UpDate	Type
Ac-225	^{225}Ac	table	comments	ensdf	26/08/2009	3
Ac-227	^{227}Ac	table	comments	ensdf	16/02/2009	2
Ac-228	^{228}Ac	table	comments	ensdf	22/01/2010	3
Ag-108	^{108}Ag	table	comments	ensdf	04/09/2006	2
Ag-108m	^{108}Ag^{m}	table	comments	ensdf	24/01/2008	1
Ag-110	^{110}Ag	table	comments	ensdf	12/03/2004	1
Ag-110m	^{110}Ag^{m}	table	comments	ensdf	24/03/2004	1

Nuclide		Tables	Comments	ENSDF	UpDate	Type
Np-236m	^{236}Np^{m}	table	comments	ensdf	20/09/2006	3
Np-237	^{237}Np	table	comments	ensdf	07/01/2010	2
Np-238	^{238}Np	table	comments	ensdf	16/02/2009	2
Np-239	^{239}Np	table	comments	ensdf	16/02/2009	2
O-15	^{15}O	table	comments	ensdf	01/06/2004	1
P-32	^{32}P	table	comments	ensdf	08/04/2004	1
P-33	^{33}P	table	comments	ensdf	08/04/2004	1

Web site :
www.nucleide.org

Emerging radionuclides

Radio pharmaceutical cited by order of importance :

1) **Cu-67** (2,6 d; β-) **;** **At-211** (7,2 h; α -> Bi-207, ε -> Po-211 then α -> Pb-207) **;** **Lu-177** (6,7 d; β-)
2) **Sc-47** (3,3 d; β-) **; Sn-117m** (13,8 d; IT) **; Ge – Ga-68** (288 d, 1,1 h ; ε, β+),
3) **I-124** (4,2 d; β+, ε) **; Cu-64** (12,7 h; β-, ε , β+) **; Y-86** (14,7 h; ε, β+) **; Sc-44** (3,9 h; β+, ε) and **Sc-44m** (58,6 h; IT)**; Sr – Rb-82** (25,5 d; ε and 1,3 min β+)
4) **Fe-52** (8,3 h; β+, ε)
5) **Zr-89** (78,4 h; β+, ε) **; Co-55** (17,5 h; β+, ε) **; Ac-225** (10 d; α)**; Bi-213** (45,6 min; β-, α)

BIPM Inter comparison

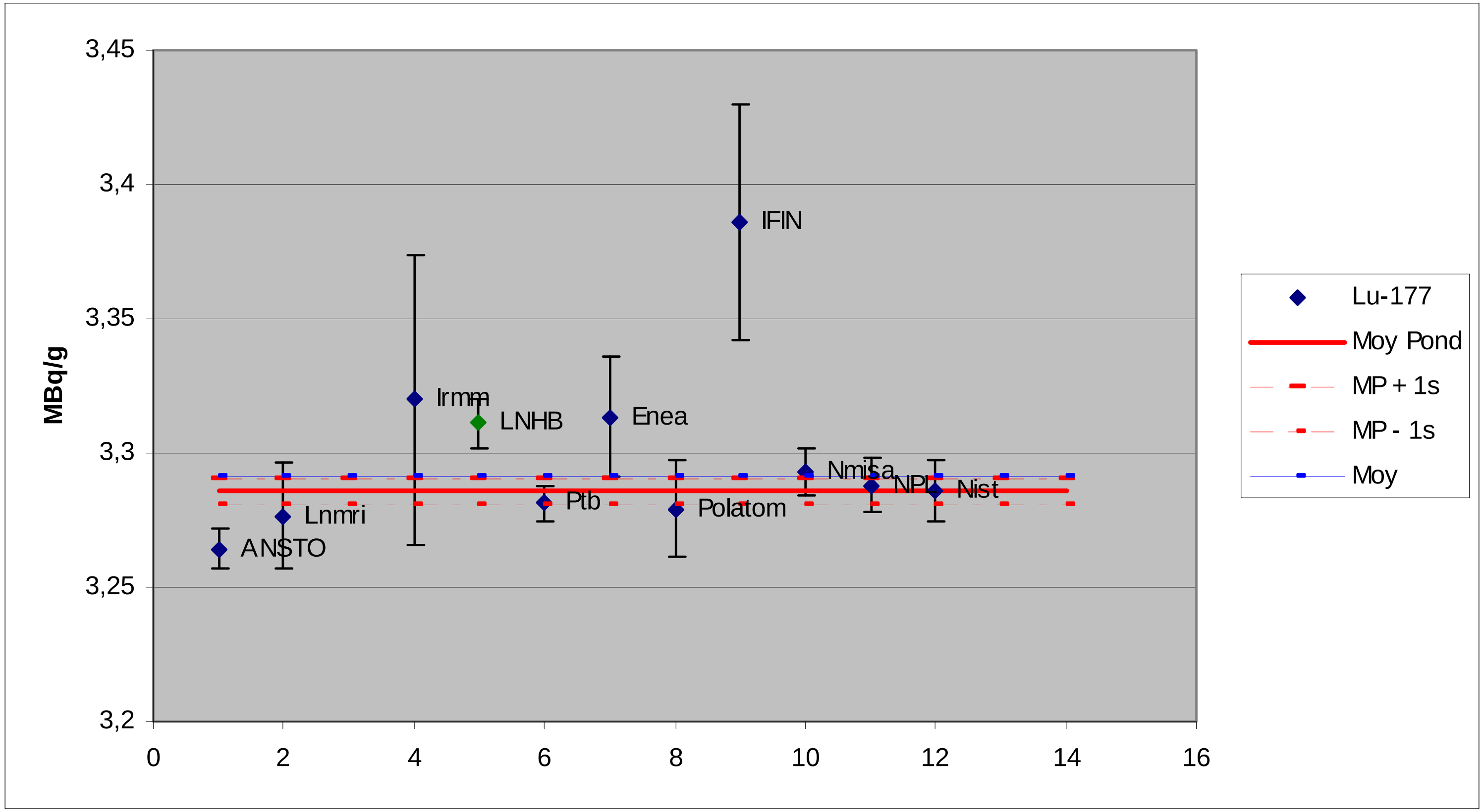
Quoted in IAEA-INDC-0501- Nuclear Data for the production of Therapeutic Radionuclides

First Remarks on this list

1) For all of these nuclides, there are no primary standards for activity measurements, then there is no traceability in the chain of measurements.
2) This list comes from ARRONAX, in other countries other nuclides are considered as potentially useful, e.g. our colleagues in NPL (GB) are studying Cu-61.
3) There is a lack of knowledge in the decay data for most of the so-called “Emerging nuclides”.
4) At present, most of these nuclides are under study and not yet currently produced, they are not available for measurements in laboratories, so it is difficult to initiate new studies.

Recent international exercises have been conducted on Lu-177 and Cu-64:

For **Lu-177**, 12 laboratories measured the activity of the same solution, results have been sent for insertion to the SIR (BIPM). Then, a Key Comparison Data Reference value will be created.



^{64}Cu international exercise

An Euramet project 1085 was proposed with the following aims:

- to create national activity standards of ^{64}Cu which can be disseminated to practitioners in the medical field through secondary standards;
- to establish an international metrological infrastructure bringing significant input to the key comparison data base (KCDB) at the *Bureau International des Poids et Mesures* (BIPM);
- to determine nuclear decay data such as the β+/ β- branching ratio, absolute emission intensities of x- rays, the annihilation and gamma photons, and the half-life with high accuracy;
- to evaluate and publish an updated decay scheme, based on previously published results as well as on those coming from this project.

Participants in this Euramet project are:

LNE-LNHB (coordinator, France),

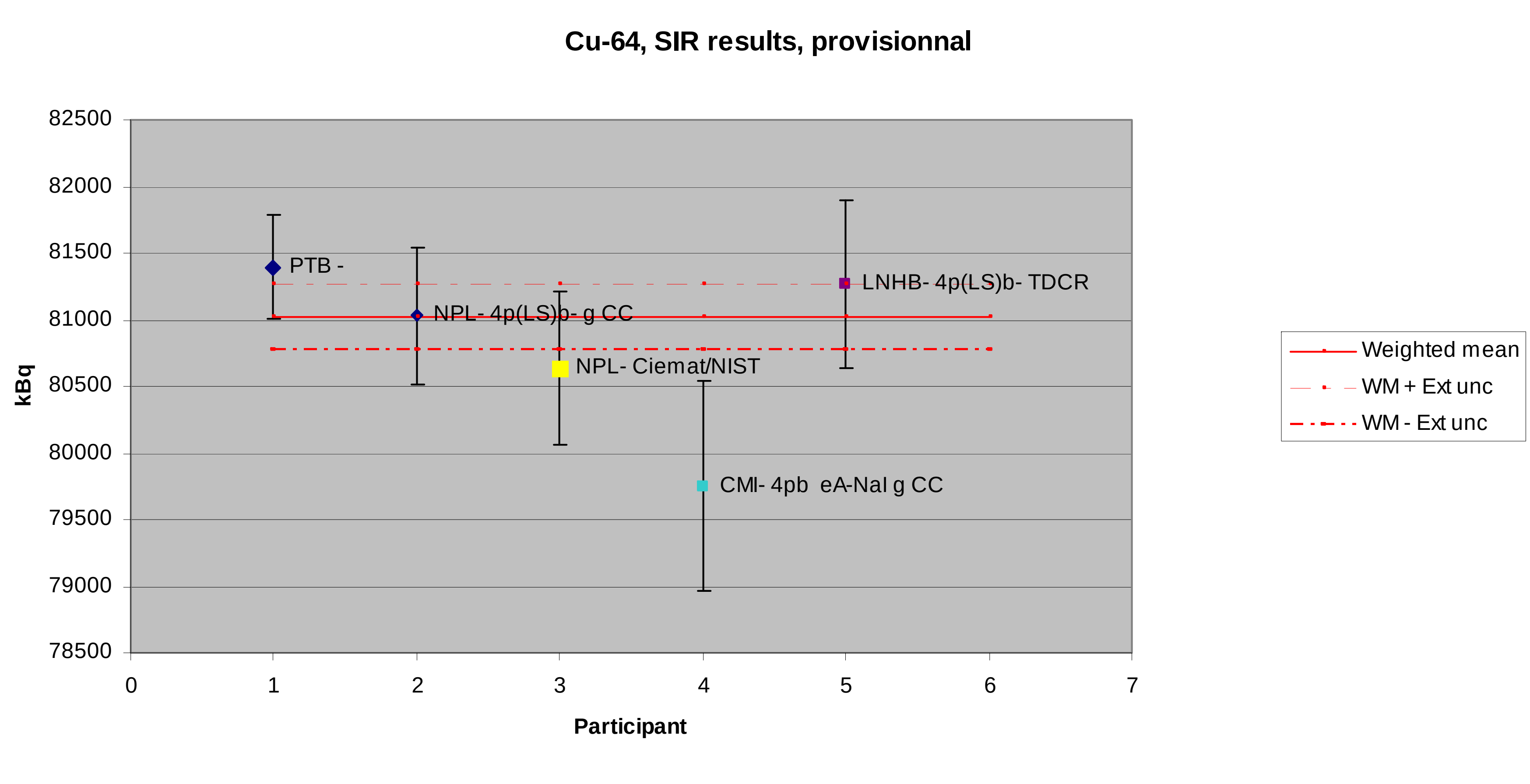
PTB (Germany),

CMI (Czech Republic),

NPL (Great Britain),

IFIN-HH (Romania).

^{64}Cu international exercise – Activity measurement results



From these results a Key Comparison Reference value will be established by the BIPM

Reference	$P_{\beta+}$ (%)	P_{EC0} (%)	P_{β^-} (%)	$P_{EC}/P_{(\beta+ + \beta-)}$
Helmer (Bé *et al.*, 2004)	17.86 (14)	42.6 (5)	39.0 (3)	0.758 (10)
This work	17.52 (15)	43.53 (20)	38.48 (28)	0.786 (10)
Δ_{Rel}	- 1.9 %	+ 2.1 %	1.3 %	3.6 %

Note:

The LSC- TDCR method is very sensitive to the EC/(β^- + β^+) ratio, The activity determination by means of the triple-to-double coincidence ratio (TDCR) method failed when using evaluated data as published in 2009 or before. For some LS samples the experimental TDCR value was lower than computed ratios of the triple and double efficiencies. The situation changes when using the new decay data.

The activity results obtained with this method are in good agreement with the results of other methods.

Two laboratories used this method and did the same remark.

Needs for new measurements ….

Looking at the list, it appears that most of them have not a well established decay scheme.

Example: Cu-67

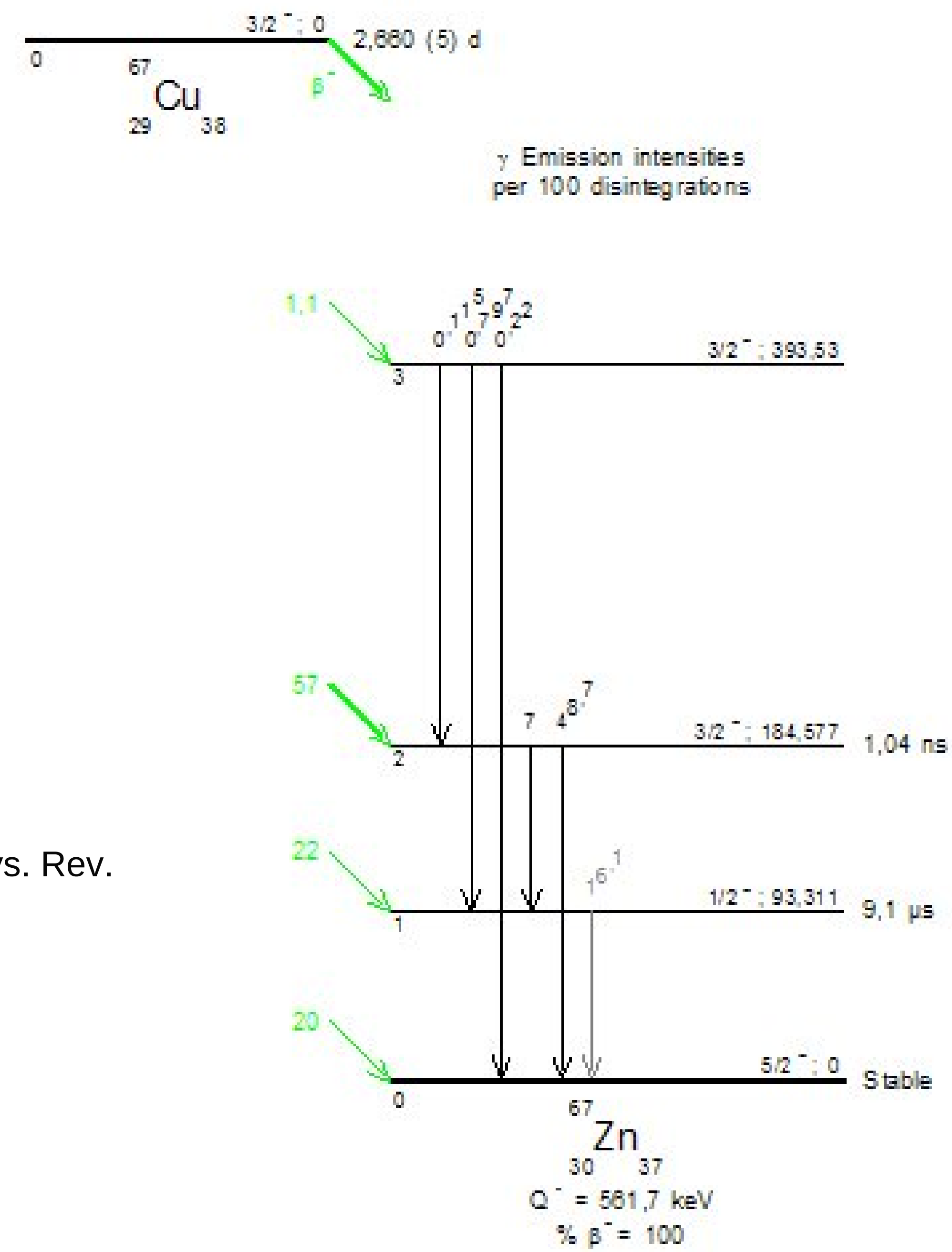
Copper 67 is a new possible therapeutic nuclide to be used in cancer treatment, its production is planned next year in the Cyclotron ARRONAX (Nantes).

BUT:

Decay scheme based on :

- Iβ to the ground state ~ 20% (1953Easterday, Phys. Rev. 91,3,p 653)
- Iγ relative values (1978Me10)
- Theoretical internal conversion coefficients

⇨ Rough decay scheme, <u>all data required</u>



Nuclides selection

- ✓ Numbers of nuclides potentially useful for medicine purposes have been cited in various publications, however only a small number of them are planned to be really used.
  - ➢ A first action, would be to cross the information and demands coming from various countries/institutes in order to establish a reduce list of nuclides of major or urgent interest.

Impurities determination

- ✓ Depending on the route of production, other nuclides are produced in addition to the nuclide of interest.
  - ➢ It is of importance to determine which are these impurities and in which quantity they are present in a solution.

For example, an impurity of 1% in Lu-177m in a solution of Lu-177 leads to a correction of ~13 % when determining the activity in Lu-177 of this solution by using a ionization chamber.

Standards

- ✓ A real production and the use of nuclides require to have standards.

  At present, for most of these nuclides, these standards don't exist !

  because most of these nuclides are under study and not yet currently produced, they are not available for measurements in laboratories.

- ➢ The production, in sufficient quantity, of the selected nuclides should be encouraged in order to make possible the measurements of activity and decay data

- Current activity measurements using “secondary instruments” such as ionization chambers or gamma spectrometry require a good knowledge of the decay data.

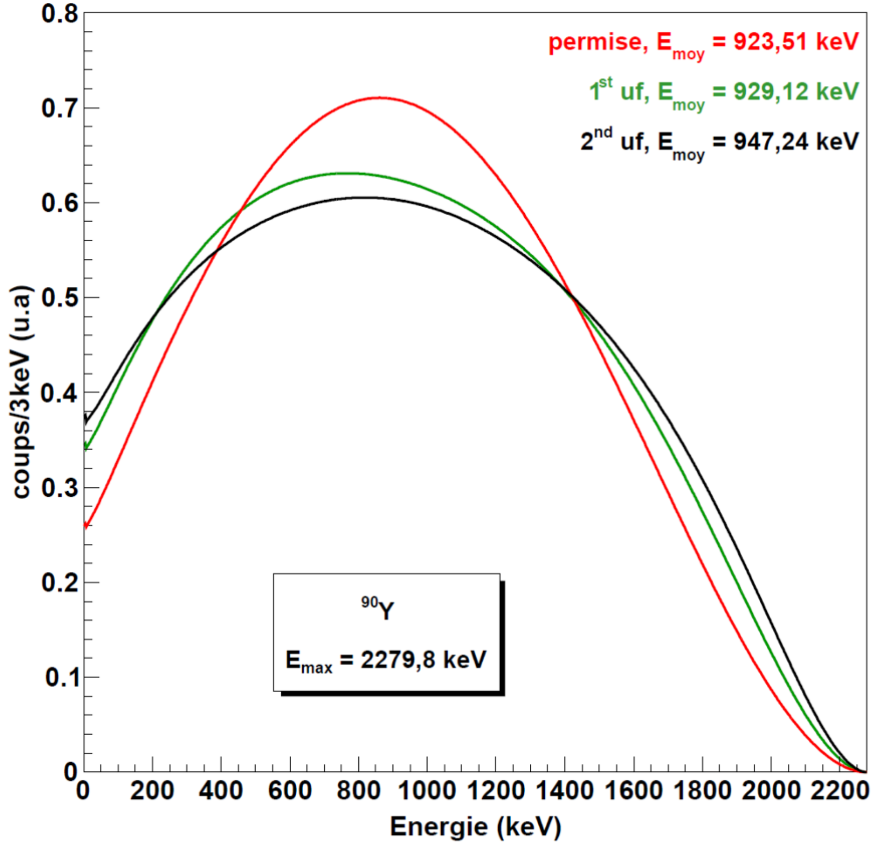
  However, most of the potentially “medical” nuclides are not well-known in term of decay data.

  - A programme of measurements and evaluations of decay data should be initiated

Some of the interesting nuclides are beta minus emitters, the range, in tissue, of such particles is of the order 1 – 100 mm depending on the energy.

For the new emerging nuclides the beta spectra have not been measured so, the spectra are calculated following theoretical models.

The shapes of the beta spectra depend on the nature of the transition (allowed, unique forbidden, non unique forbidden).



Beta spectra

But the existing computer programs can calculate the spectra for allowed and unique forbidden transitions only.

In any treatment using ionizing radiations, the knowledge of the dose delivered within the tumor and the rest of the body is essential to ensure the efficacy and the safety of the treatment. Among the problems, one is related to the calculation of the dose delivered in the volumes of interest according to the quantity of radionuclide which is present there. The dose assessment requires to know the energy spectrum of the β transition.

- In order to improve the existing calculation codes of β spectra and to add the calculation of non unique forbidden transition, new measurements of β spectra are required. This will help to derive experimental shape factors from which reductions of the theoretical calculations will be made possible.

To conclude

The actions to initiate would be :

1) To prioritize the nuclides to be studied.
2) To create actions/supports to encourage the production of these nuclides in enough quantity to make possible activity and decay data measurements, as soon as possible.
3) To determine and study the impurities as well.
4) To create actions/supports to encourage the measurements of beta spectra for beta emitting nuclides.
5) With new measurement results to evaluate the decay scheme data.