On-site measurement of ^{11}C and ^{18}F half-life by TDCR counting

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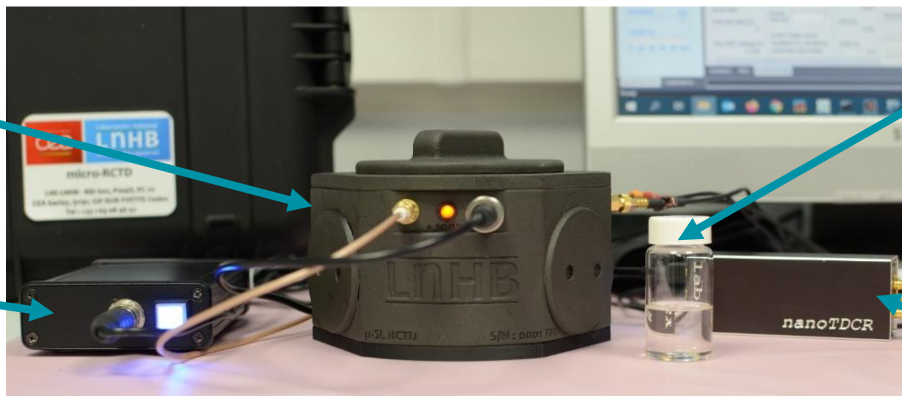
The triple to double coincidence ratio (TDCR) liquid scintillation measurement technique is commonly used in national metrology institutes (NMIs) to perform standardization of pure beta emitters. LNHB has developed two new portable TDCR devices [1]. Such portable instrumentation gives end-users access to a reference measurement method that can be used for a large number of radionuclides. It addresses a wide range of industrial and medical applications for radionuclide metrology such as calibrating solutions with short-lived radionuclides, preventing radioactive source transportation, and performing on-site comparisons to promote radionuclide metrology harmonization. The linearity of response of such an instrument allows measurements at very high count rates, close to one million pulses per second and at very low count rates, a few pulses per second. Such a property is highly suitable for the measurement of short-lived radionuclides such as ^{18}F and ^{11}C radiopharmaceuticals that are presented in this work.

µ-TDCR module, including optical chamber, high voltage divider and PMTs

High voltage power supply



Measurement geometry: Liquid scintillation vial with 10 mL of UltimaGold scintillator

LabZY nanoTDCR acquisition system [2] connected to a quartz clock @ 10 MHz and a computer with NTP server from CEA

Photography of µ-TDCR without shield protection at Orsay hospital.
The setup was similar for experiments with mini-TDCR.

Experimental setup

- Two portable TDCR devices; µ-TDCR and mini-TDCR
- Reference quartz clock @ 10 MHz
- NTP time server from CEA network
- Samples composition:
  - 10 mL of UltimaGold® in glass Vial and PE-PTFE vial
  - 100 µL of radiopharmaceutical solution [^{11}C]PIB or [^{18}F]FDG in saline solution with 10% of ethanol
- 5 cm of lead shielding surrounding the device; ventilated and stable temperature in the laboratory

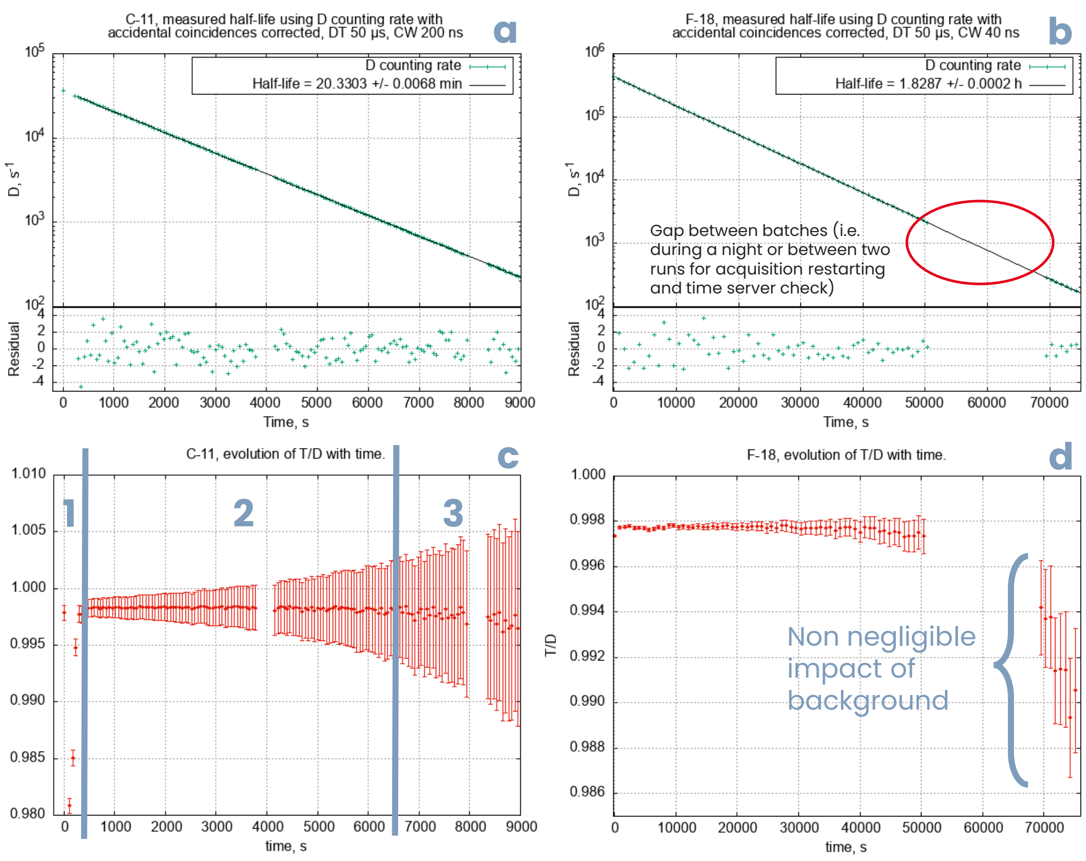
Analysis procedure

- Fitting of T and D counting rate corrected from accidental coincidences [3] with equation:

$$f(x) = D_0 e^{\left(\frac{ln(2)t}{T_{1/2}}\right)} + D_{Blk}$$

- D_{Blk}: background counting rate, D_0: counting rate at the beginning of the experiment, t: time, $T_{1/2}$: measured half-life
- χ^2 minimization using the non-linear least squares Levenberg–Marquardt algorithm implemented in Gnuplot with weighted value using counting uncertainty
- Uncertainty evaluation according to previous detailed work [4]

Typical results for 2 measurements with both nuclides



- D counting rate is used to derive half-life (fig. a and b);
- T/D analysis for linearity; i.e. stable counter and negligible background dependency (two extreme case presented in fig. c and d):
  1. relaxation of PMTs or cocktail after sample placement;
  2. very good stability, part of interest to be analyzed;
  3. background influence, larger result spreading.

Typical uncertainty budget for the different measurements performed in this study

Component	^{18}F Rel. Std. Unc. (%)	^{11}C Rel. Std. Unc. (%)
Background	0.01	0.01
Dead time and counter linearity (T/D variation)	0.01 to 0.03	0.01
Coincidence Window and accidental correction	0.01	0.01
Fit (contains part of model uncertainty)	0.006 to 0.019	0.02 to 0.03
Impurities (none detected)	< 0.001	< 0.001
Square root of the sum of quadratic components	0.018 to 0.026	0.026 to 0.035

Summary of fitting results from different experiments

Experiments (dead time; coincidence window)	$T_{1/2}(^{18}F)$	$T_{1/2}(^{11}C)$
Mini-TDCR (10 µs; 40 ns)	1.82825 (33) h	20.327 (6) min
Mini-TDCR (10 µs; 200 ns)	1.82835 (33) h	20.328 (6) min
Mini-TDCR (50 µs; 40 ns)	1.82873 (33) h	20.327 (6) min
Mini-TDCR (50 µs; 200 ns)	1.82873 (33) h	20.329 (6) min
µ-TDCR (10 µs; 40 ns)	1.82877 (33) h	20.330 (5) min
µ-TDCR (10 µs; 200 ns)	1.82877 (33) h	20.330 (7) min
µ-TDCR (50 µs; 40 ns)	1.82900 (48) h	20.333 (7) min
µ-TDCR (50 µs; 200 ns)	1.82900 (48) h	20.332 (7) min

Final results

- Gamma spectrometry carried out at the SHFJ did not reveal the presence of impurities. Blank measurement of one night (µTDCR, ^{11}C; D = 7.22 (7) s^{-1} and T/D = 0.906 (7) s^{-1}) were compared with measurement of the sample after total decay of the radionuclide, (µTDCR, ^{11}C; D = 7.33 (6) s^{-1} and T/D = 0.920 (3) s^{-1}). No relatively long half-life impurities could be identified.
- Using all the measurements the calculated half-life according to the evaluation method from [5] for ^{11}C is **20.3292 (46) min** and for ^{18}F is **1.82863 (33) h.** These results are consistent with DDEP values **20.361 (23) min** for ^{11}C and **1.82890 (23) h** for ^{18}F. We were not able to improve the measurement uncertainty for ^{18}F.

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[1] B. Sabot *et al.*, 2022, NIM-A 1034. [2] V. Jordanov *et al.*, 2020, NIM-A 954. [3] C. Dutsov *et al.*, 2020, NIM-A 977.
[4] S. Pommé *et al.*, 2020, ARI 158. [5] Bé *et al.*, 2011, Table of Radionuclides – Introduction.