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New concepts and instruments for ^{14}C and ^{36}Cl measurements in i-graphite

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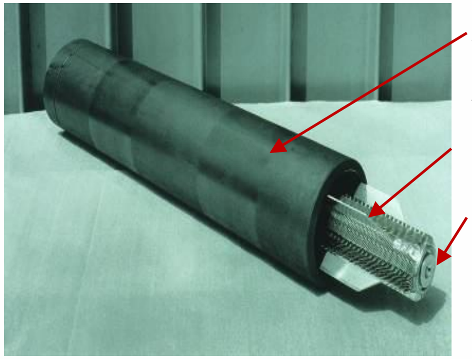
Outlook

- Intro : The need of field measurement capabilities for the characterization of the i-graphite

- What can be done for ^{36}Cl measurements ?
  - Conventional measurements
  - Using photon coincidence detection
  - Estimated performances

- What can be done for ^{14}C measurements ?
  - Calorimetry : new means
  - Gamma response
  - Estimated performances

- Conclusion : next steps



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Intro

- ^{14}C and ^{36}Cl are key nuclides for irradiated graphite waste management
  - Key for designing methods, tools and decommissioning means
  - Key for treatment and packaging
  - Key for means and ways of storage…
- Unfortunately they are difficult to measure. Need of sampling and laboratory
- Complementary to lab measurements, field measurement means would and could be very useful : no sampling, real time measurement
- Our goal, in this first step, is to propose and assess technical means apt to make this job
- Next steps will be then first to confirm experimentally on sites then, if relevant, to early design and evaluate how future processes would take benefit of such kind of instruments

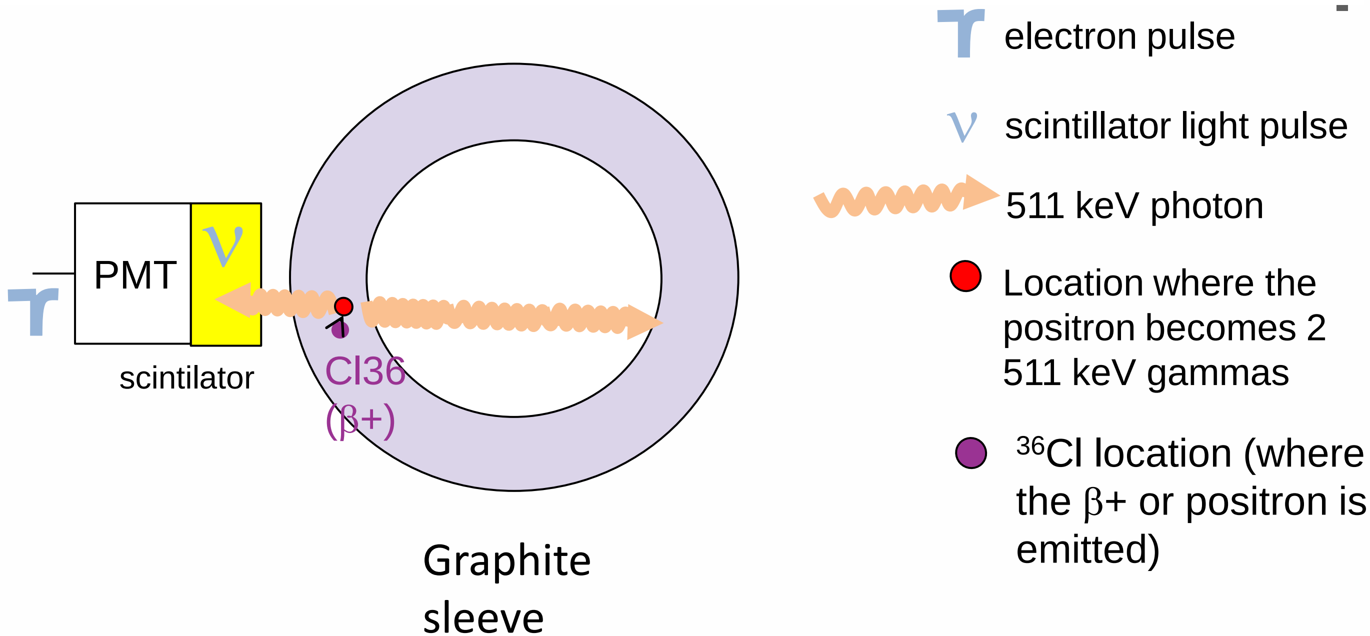
^{36}Cl

- Half life : 301 300 years
- Mostly created by neutron capture of stable isotope ^{35}Cl
- Beta emitter (710 keV, 98.1%)
- Positron emitter (50 keV, 0.014%)
- Level of activity in i-graphite : from 1 Bq/g to 1000 Bq/g

- For the measurement of the ^{36}Cl activity, we have decided to explore the ways of using the 511 keV resulting from the annihilation of the ^{36}Cl positron.
- 2 a priori drawbacks :
  - distinguishing the ^{36}Cl positrons from those of other isotopes
  - The signal will be very rare…

^{36}Cl Conventional positron measurement by a gamma spectro

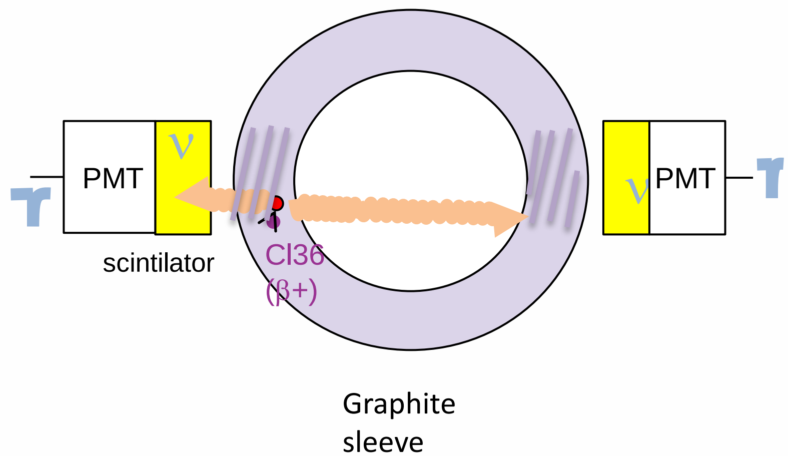
- Gamma detector in front of the object.
- Counting the 511 keV events to know the number of positons
- Exploiting the spectrum to know present gamma emitters
- Deducing from all that the ^{36}Cl activity



In short : - this would work well if ^{36}Cl is the only isotope in the graphite
- It would work less good if
  - ^{36}Cl is a marginal β+ producer (among others)
  - High energy gamma flux is high (specific enemy: e+e- pair production)

^{36}Cl Gamma-Gamma coincidence detection system

- Gamma detectors in front of the object and face to face
- Counting the 511 keV coincidence events to know the number of positons
- Exploiting the spectrum to know present gamma emitters
- Deducing from all that the ^{36}Cl activity

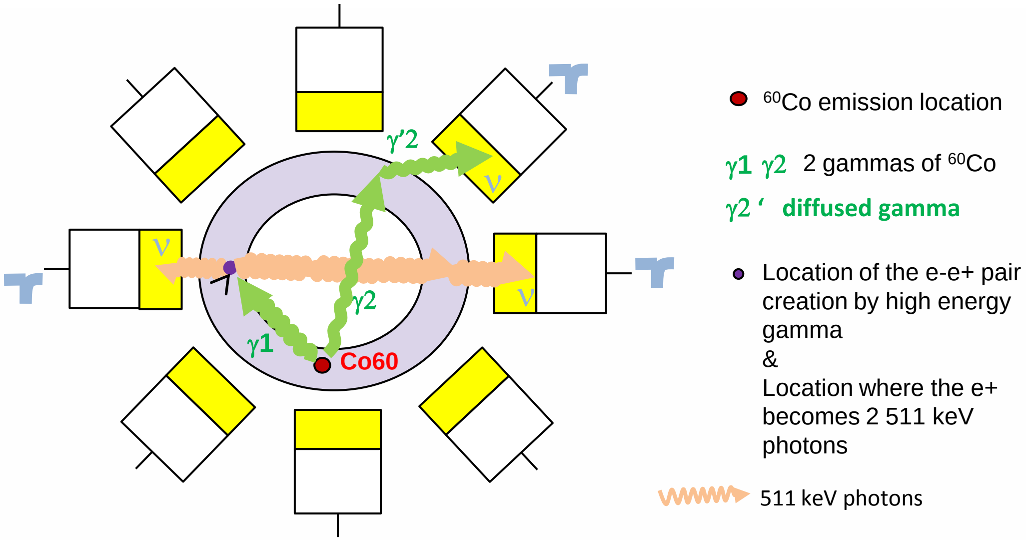


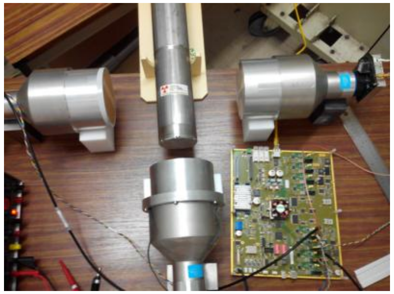
- electron pulse
- scintillator light pulse
- 511 keV photon
- Location where the positron becomes 2 511 keV gammas
- ^{36}Cl location (from where the β+ or positron is emitted)

Pros	**The noise is dramatically reduced** The counted events corresponds only to positrons annihilated in the hatched areas //// (positrons in other sleeve parts eliminated by geometry, positrons in detectors eliminated by time of flight)
Cons	**The signal is also reduced and the instrument is more complicated** The drawback of other direct or indirect β+ emitters remains

^{36}Cl Operational coincidence detection system

- At least 8 gamma detectors (LaBr3:Ce preferred – 5’’x2,5’’)
- Each detector provides a spectrum allowing measurement (with some spatial resolution) of gamma isotopes
- Detection of any type of coincidences
- high flux capabilities required (2 M-in-events per channel)
- High time resolution required : 1 ns typical





Basic lab equipment used for experimental validation

The figure shows how positron noise due to ^{60}Co gamma rays (creating positrons by pair production) can be well estimated by detection of « triplets ». Calculations (and experience) show that with a eight 5’’detectors system around a typical Bugey-1 sleeve detection of triplets occurs about 5 times for 100 60Co-due registered coincidences, which is a good result.

^{36}Cl

How the ^{36}Cl activity is calculated

- The total number of coincidences is counted
- The number of false coincidences (random coincidences) is estimated and subtracted
- The number of coincidences due to to β+ γ emitters is estimated and subtracted

  Thanks to activity measurement of these emitters by γ spectroscopy
- The number of coincidences due to pair production positrons is calculated and subtracted

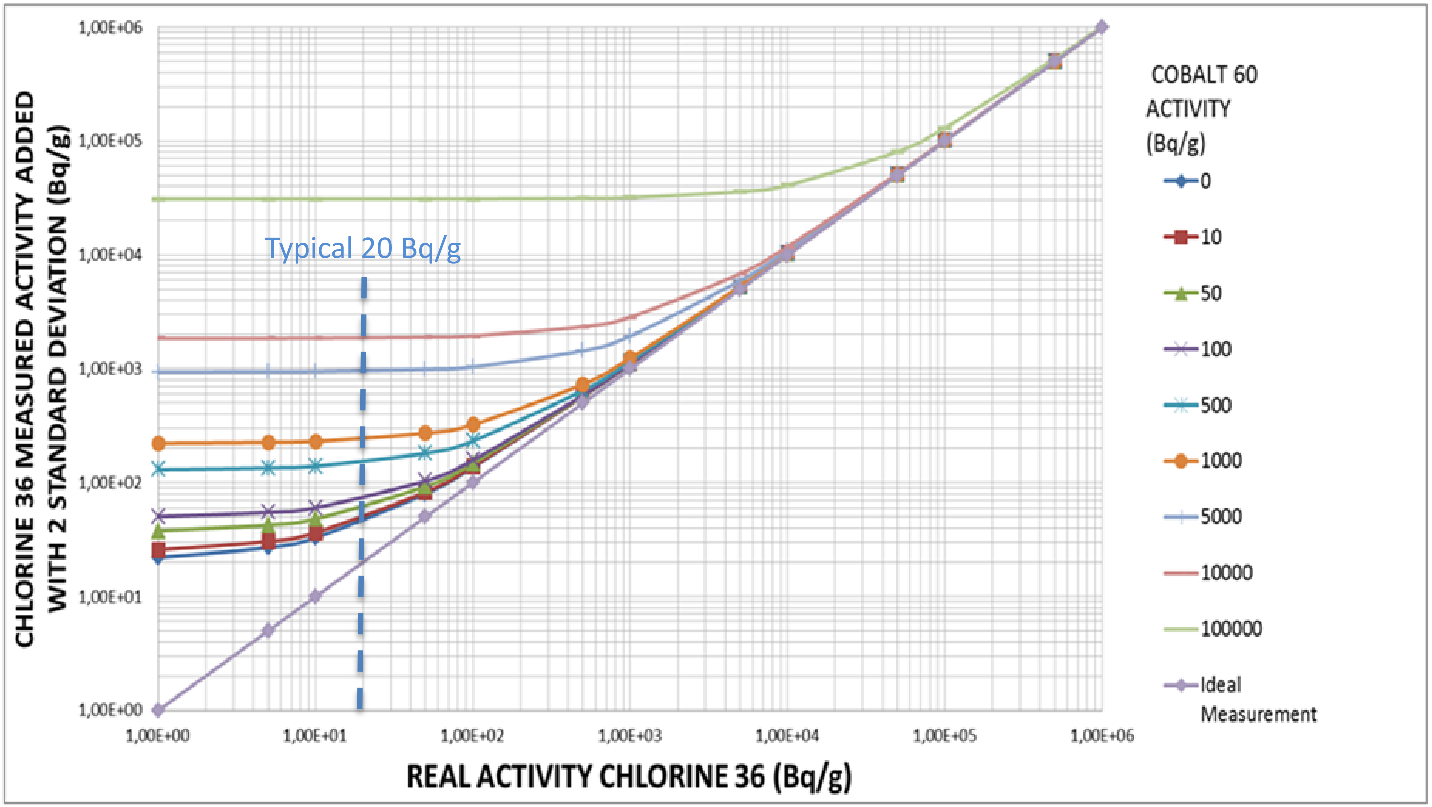
  The calculation is made on the basis of the spectroscopy result and on the basis of the « triplets » measurement

The net number of coincidences remaining from the 3 subtractions is then converted in Becquerel of ^{36}Cl thanks to a calibration function.

if there are others pure β+ emitters, the 36Cl activity is overestimated. It seems to be rare in typical i-graphite

^{36}Cl

Estimated performances



Hypothesis : measurement time 1200 s ; sleeve geometry 600mm high /220mm OD /158mm ID /20kgs; Eu152 : 50 Bq/g ; 60Co, 152 EU, fortuitous events standard deviations : 5% ; Configuration 8 detectors LaBr3 5’’

^{14}C

- Half life : 5730 years
- Mostly created by neutron capture of stable isotope ^{13}C or (n,p) reaction on ^{14}N
- Pure beta emitter (E_{mean} 50 keV, 100%)
- Level of activity in i-graphite : about 10^4 to 10^5 Bq/g typical

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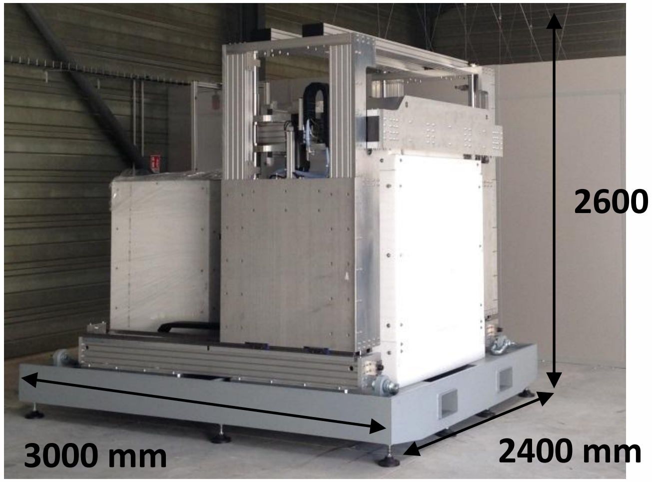
- We have decided to explore the conventional way of calorimetry but with a new instrument able to lower the usual detection limits
- 2 a priori drawbacks :
  - Being able to measure very low heating powers
  - Being able to subtract the other isotopes heating contribution

^{14}C

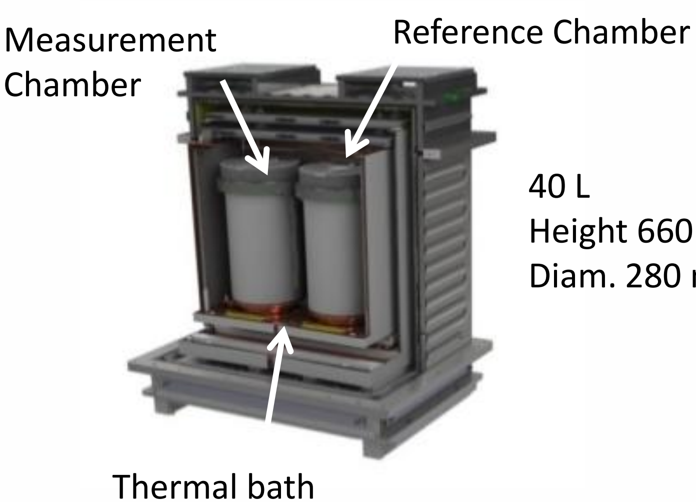
µLVC Large Volume Calorimeter

- KEP Nuclear developed a new Large Volume Calorimeter: **µLVC**
  - Differential isothermal heat flow calorimeter
  - Sample inside a vacuum chamber
  - Effective power range 50 µW – 100 mW
  - Measurement Mean Random Uncertainty on the first prototype:

Expected Uncertainties	Measured Uncertainties
±10% between 10 µW and 100 µW	±20% between 50 µW and 250 µW
±1% between 100 µW and 1 mW	±5% between 250 µW and 1 mW
±0.20% between 1 mW and 10 mW	±1% between 1 mW and 100 mW



Weight:
9 Tons



40 L
Height 660 mm – Diam. 280 mm

^{14}C

- Calorimetric assay of graphite is appropriate but:
  - Relative low energy quantity released by ^{14}C decay:

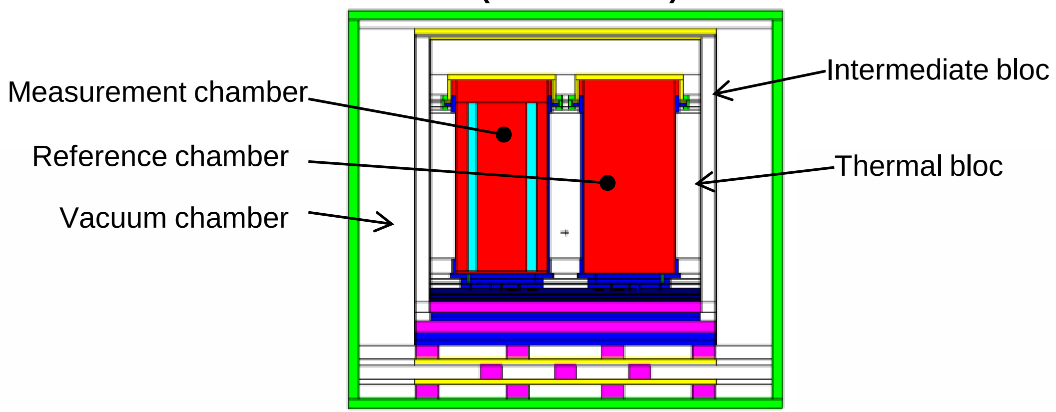
  50 keV in average ⇔ 8E-15 W/Bq ⇔ 8 µW per GBq.
  - Irradiated graphite does not contain only ^{14}C → obliged to calculate the contribution of other isotopes for correction

- The identification of beta emitters is done by gamma spectrometry:
  - Identifying charged particle emitters and quantifying gamma rays.
  - Estimation of the power brought to the object by the gammas thanks to modeling.

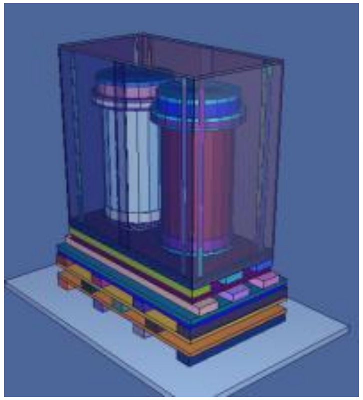
^{14}C The gamma-heating Issue

- The energy deposit in graphite and calorimeter due to gamma photons is estimated using MCNPX code.

Cross section of the µLVC (MCNP Model)



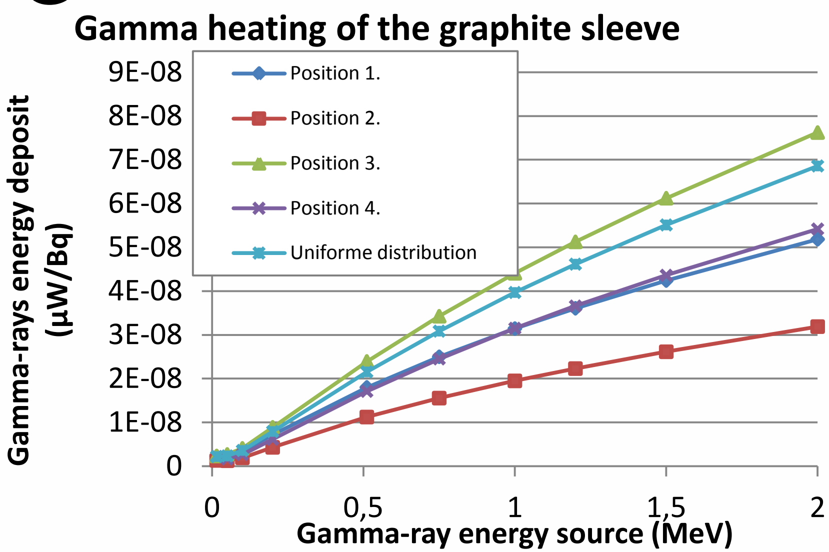
3D view of the µLVC

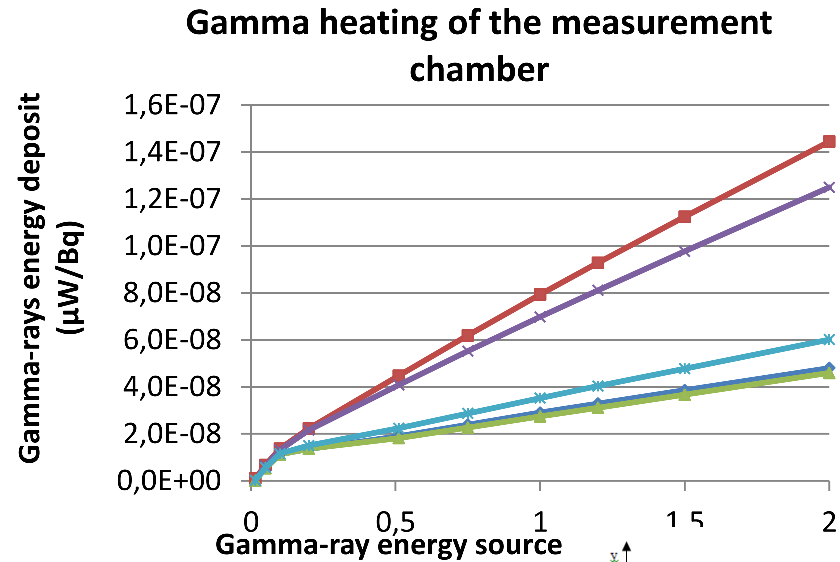


- 2 MeV gamma rays source at 1E9 Bq (the worst case):
  - The gamma-heating of the intermediate bloc = 20 µW
  - The gamma-heating of the thermal bloc = 50 µW.
  - Heat produced by the temperature sensors ~1 mW >> 50 µW

→ The heating of the regulation blocs is not measured during the assay, and does not disturb the calorimeter working.

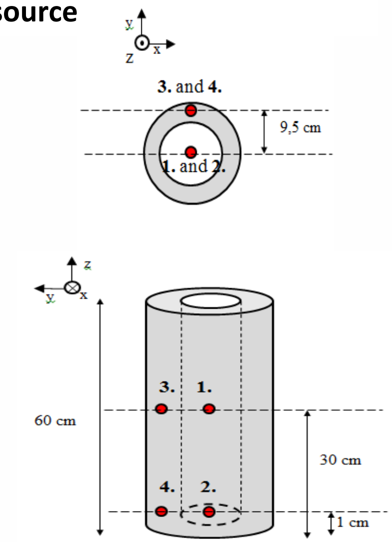
^{14}C Gamma-Heating calculation





The gamma-heating inside the graphite sleeve and the measurement chamber depends of the source location :

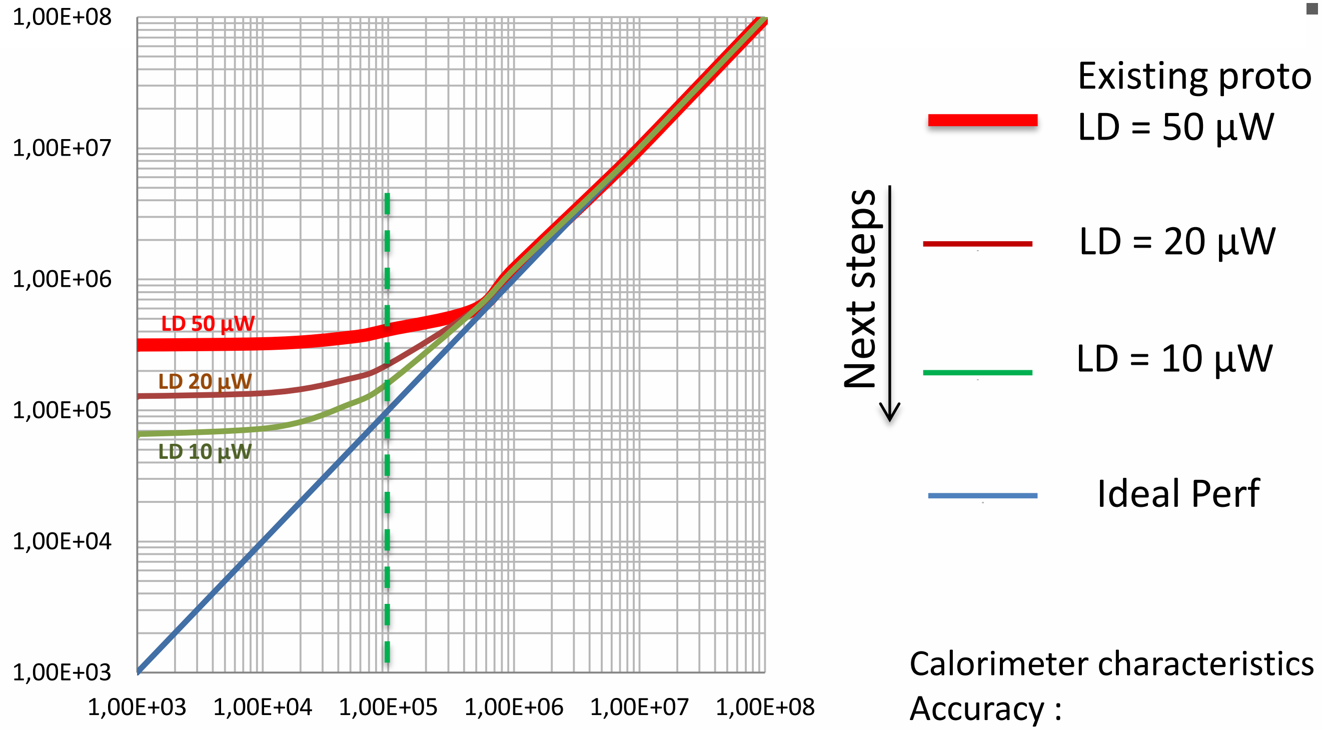
- In graphite sleeve: if the source is closed to the outside → gamma rays can easily leak → not induce heating inside the sleeve.
- The measurement chamber:
  - If the source in close to the lateral wall → gamma rays go through the thin copper wall → not induce heating inside the chamber.
  - If the source is close to the low part → gamma rays are absorbed by the thick back part of the chamber. → induce heating inside the chamber.



^{14}C

^{14}C measurement estimated performances

Measured activity increased of its error (Bq/g)



14C real activity (Bq/g)
Graphite sleeve 20 kgs
(typical value 10^5 Bq/g)

Calorimeter characteristics
Accuracy :
50µW-250 µW : 20%
250 µW- 1mW : 5%
1 mW – 100 mW: 1%

Gamma heating error not taken into account
(10^8 Bq/sleeve makes 100 µW, ie error about 10µW)

^{14}C & ^{36}Cl

Conclusion & perspectives

- Feasibility of instruments dedicated to field measurements of ^{14}C and ^{36}Cl in rough (not any preparation or sampling) i-graphite objects has been assessed. On site real measurement tests remain to do.
- Limits of detection seem to be near a practical use for ^{36}Cl but yet to be worked for ^{14}C through improvement of a new low heating power calorimeter.
- Processes and waste management scenarios which could use such equipments have to be thought for evaluation of interest.

^{14}C & ^{36}Cl

THANKS FOR YOUR ATTENTION