eurofins

Determination of radioactivity concentrations in building materials with respect to NORM issue

eichrom
LABORATORIES

member of

eurofins | Nuclear

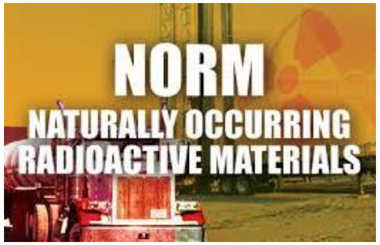
OUR MISSION

To provide reliable results
in an efficient and timely manner
to solve customer issue and constraints


Drinking Water


Foodstuff


NPPs surroundings



NORM

Summary

Determination of radioactivity concentrations

in building materials

with respect to NORM issue

- **Regulation of NORM in EC**
- **Two methods for efficiency calibration**
- **Results comparison**
- **Conclusion**

REGULATION OF NORM IN EC

Regulation of NORM in EC

- Potential risk of ionizing radiation from NORM (Naturally Occurring Radioactive Materials) identified by ICPR

- European Community directive 2013/59/EURATOM article 75 :

  External exposure of gamma radiation in building materials

  < 1mSv/year

- To prove this, European Committee of normalisation stated:

  technical standard CEN/TS 17216

  where

$$I = \frac{C_{^{226}Ra}}{300} + \frac{C_{^{232}Th}}{300} + \frac{C_{^{40}K}}{3000}$$

Materials

- The building industry needs to qualify all materials used to build flats and houses

- Building industry challenge:

  ✓ Reliable

  ✓ Quick

  ✓ Economically reasonable protocol

I formula

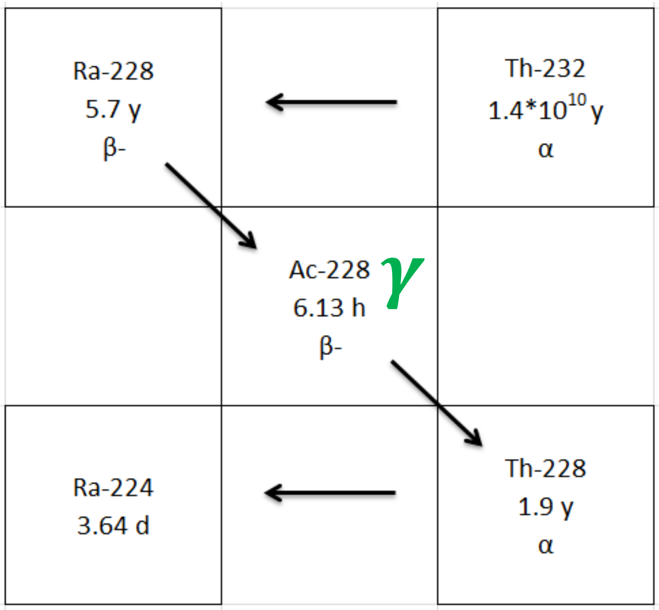
- $$I = \frac{C_{226_{Ra}}}{300} + \frac{C_{232_{Th}}}{300} + \frac{C_{40_{K}}}{3000}$$ Where C are mass activities in Bq/kg

- ^{40}K is β and γ emitter

- ^{226}Ra is α and γ emitter

- ^{232}Th is pure α emitter : long preparation #2weeks

I formula



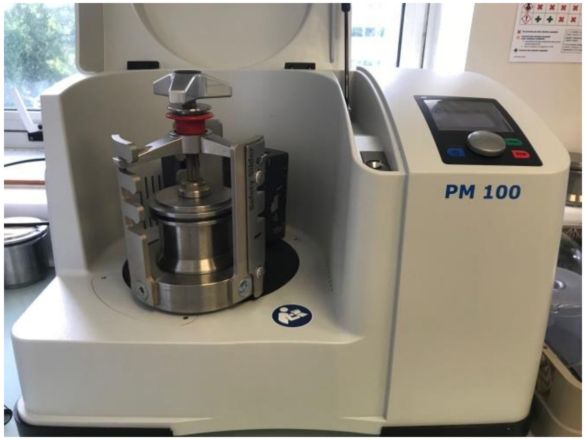
One of progenies of ^{232}Th is a gamma emitter. With an assumption of radioactive equilibrium we can deduce ^{232}Th from ^{228}Ac.

I formula

- $$I = \frac{C_{^{226}Ra}}{300} + \frac{C_{^{232}Th}}{300} + \frac{C_{^{40}K}}{3000}$$ Where C are mass activities in Bq/kg
- ^{40}K γ emitter
- ^{226}Ra γ emitter
- ^{232}Th with ^{228}Ac : γ emitter
- 1 measurement
- 1h Detection limit 0,2Bq/g max : quick and sufficient

Solids preparation

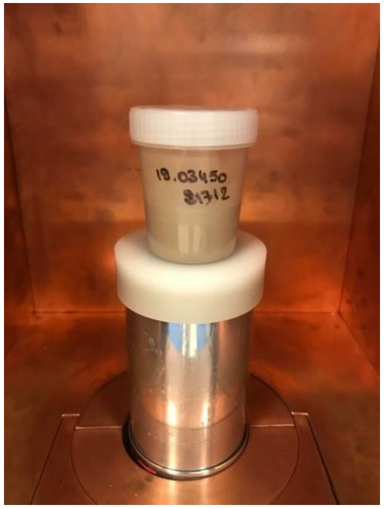
- Needs a preparation (crushing, sieving at 200µm) #2h if sample not in a powder form at reception



Crusher

Sieve

Gamma spectrometry

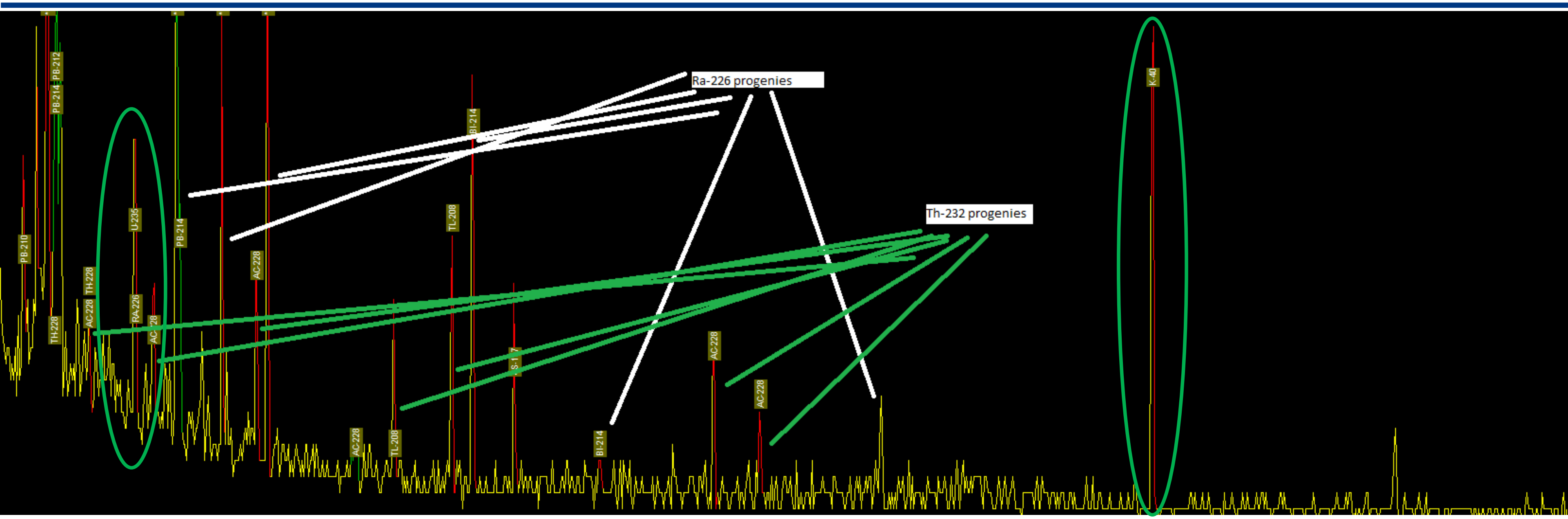


HPGe detector

Lead castle

Automatic sample loader

Gamma spectrometry



- Pay attention to peak interferences (for example ^{226}Ra/^{235}U close to 0,5keV), sum peak, self-absorption, …

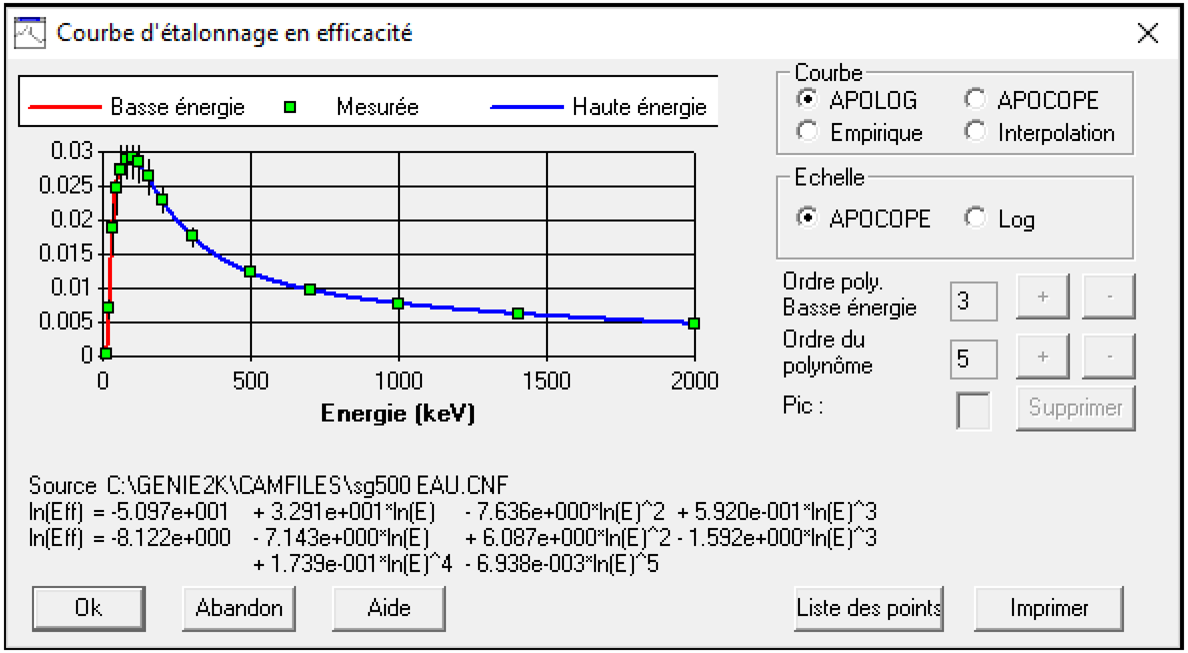
requires experience from qualified technicians

- To quantify a peak an efficiency calibration is needed. Trickiest part of this analyse.

TWO METHODS FOR EFFICIENCY CALIBRATION

Efficiency calibration for solids

- In most commonly used procedures, 2 methods can be used to determine efficiency curves for gamma spectrometry in solids:
  - ✓ Classical way with sealed sources to determine self-absorption factor C
  - ✓ Monte-Carlo simulation with software

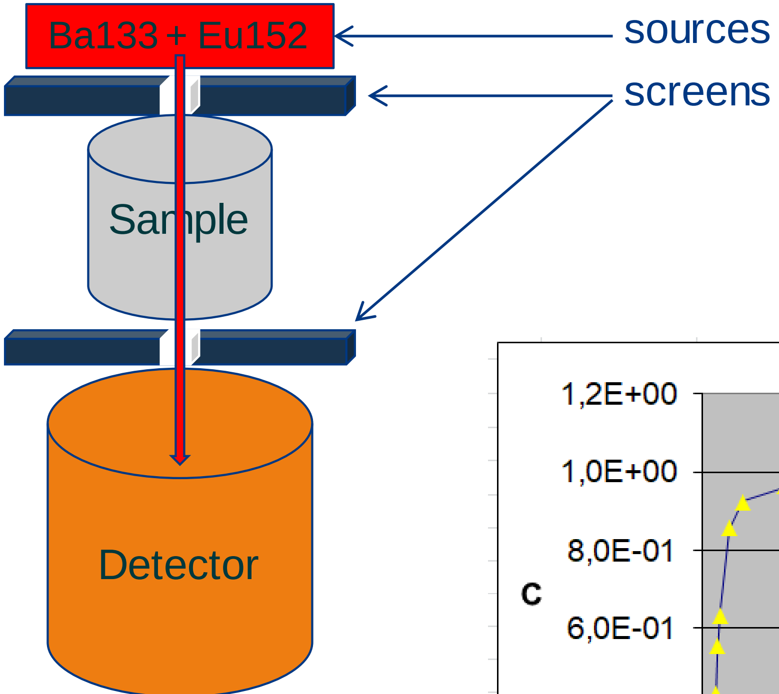


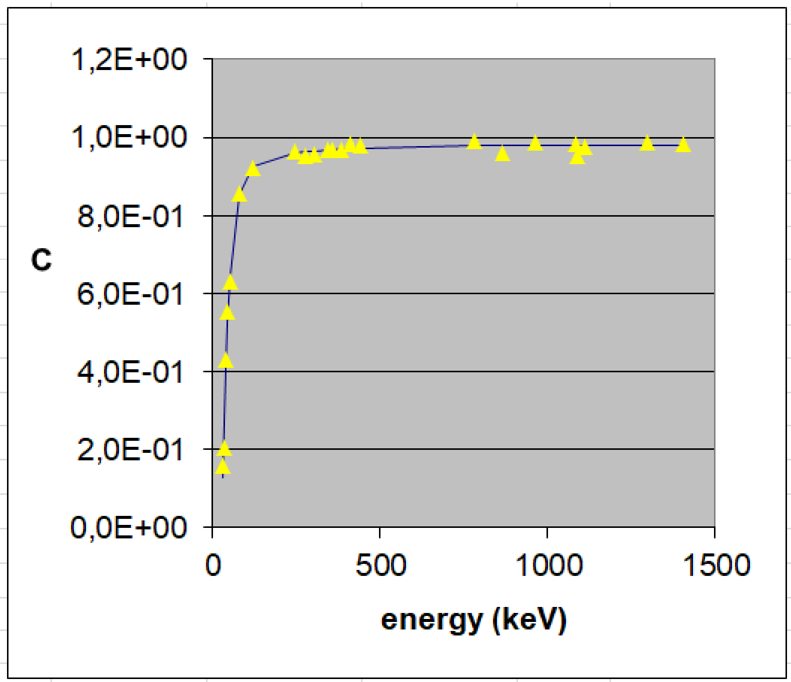
Self-absorption factor C

- Method used in many labs around the world

- Needs 4 measures:
    a) Empty beaker with sealed sources (4h)
    b) Beaker with same height of water with sealed sources (2h)
    c) Sample with sealed sources (2h)
    d) Sample alone (1h)

Self-absorption factor C





Origin regression	
y = y0+A1*(1-exp(-x/t1))+A2*(1-exp(-x/t2))	
y0 =	-2,45043
A1 =	0,09010
t1 =	163,61760
A2 =	3,33938
t2 =	21,02821

Geometry :	B50mL
Solid nature :	Concrete
Density :	1,3
Detector :	DET03

Self-absorption factor C

- Method used in many labs around the world
- Needs 4 measures:
    a) Empty beaker with sealed sources (4h)
    b) Beaker with same height of water with sealed sources (2h)
    c) Sample with sealed sources (2h)
    d) Sample alone (1h)

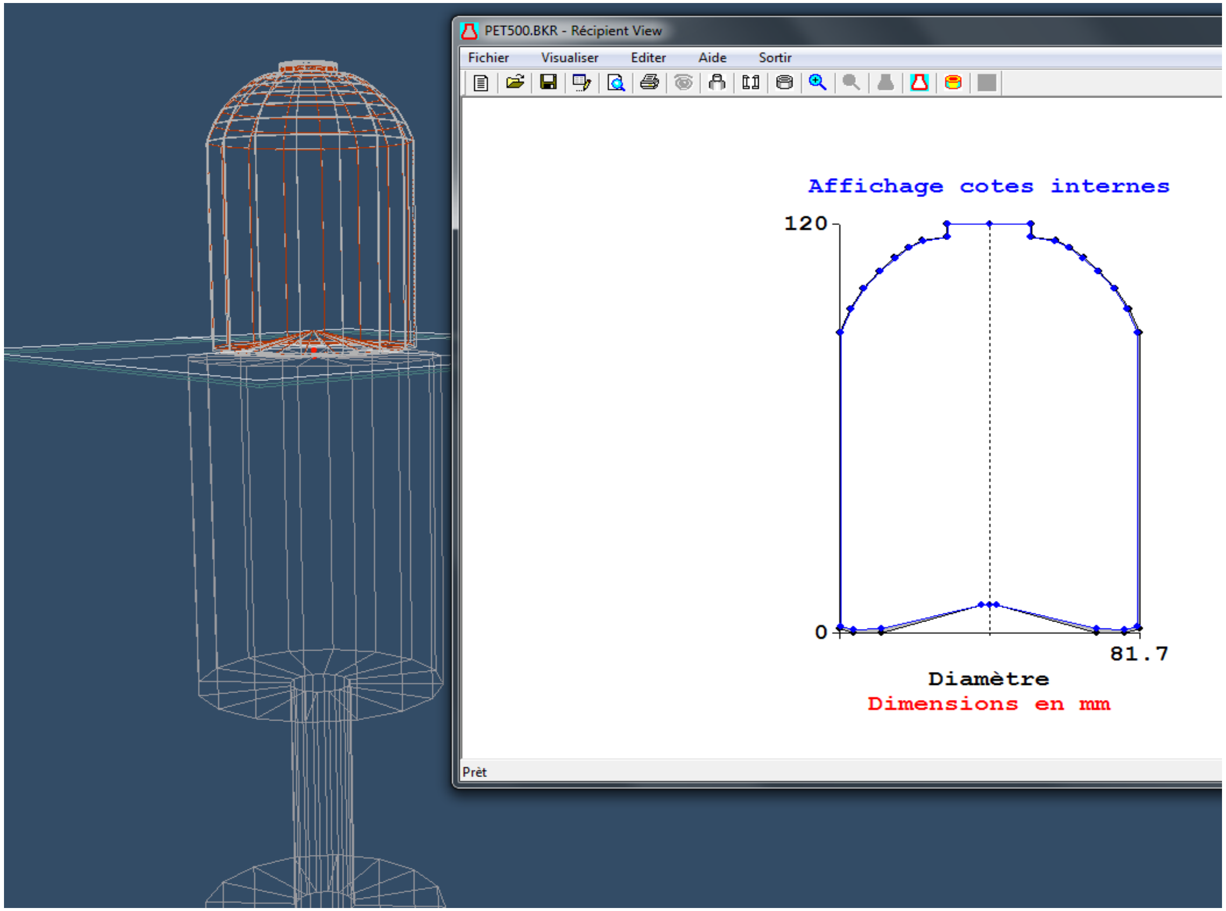
- Time of exploitation by a technician of the c spectrum : 45min

- Advantage : Faithfull to nature of sample

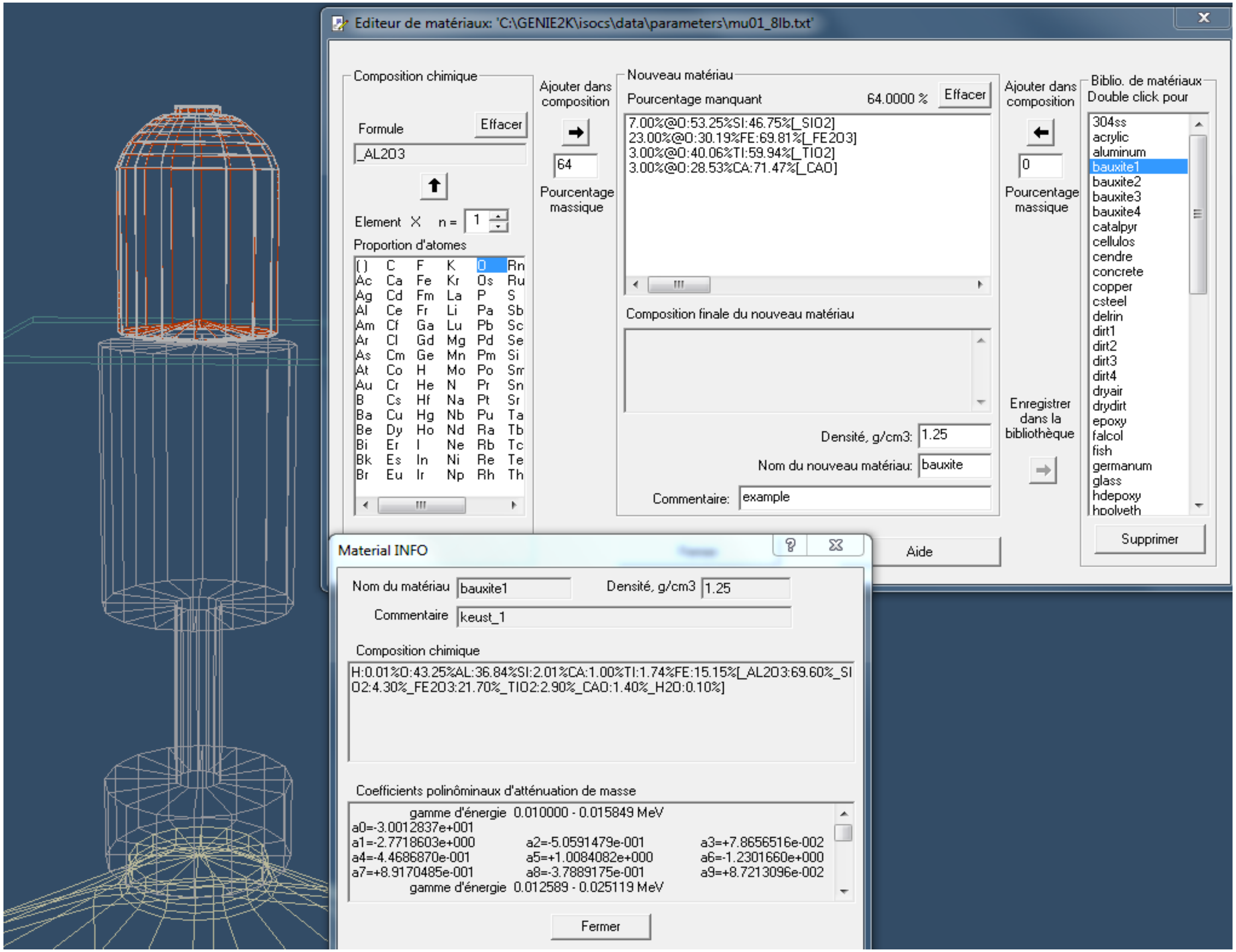
- Disadvantage : 3h45 per sample, lot of software work

Monte Carlo

- Use of Mirion' software LABSOCS
- Beaker need to be designed – reusable for every samples – usable for every detector



Monte Carlo



Editeur de matériaux: 'C:\GENIE2K\isocs\data\parameters\mu01_8lb.txt'
Composition chimique
Formule
Effacer
_AL2O3
Element X n = 1
Proportion d'atomes
Ajouter dans composition
64
Pourcentage massique
Nouveau matériau
Pourcentage manquant 64.0000 %
Effacer
7.00%@O:53.25%SI:46.75%[_SIO2]
23.00%@O:30.19%FE:69.81%[_FE2O3]
3.00%@O:40.06%TI:59.94%[_TIO2]
3.00%@O:28.53%CA:71.47%[_CAO]
Composition finale du nouveau matériau
Densité, g/cm3: 1.25
Nom du nouveau matériau: bauxite
Commentaire: example
Ajouter dans composition
0
Pourcentage massique
Enregistrer dans la bibliothèque
Biblio. de matériaux
Double click pour
304ss
acrylic
aluminum
bauxite1
bauxite2
bauxite3
bauxite4
catalpyr
cellulos
cendre
concrete
copper
csteel
delrin
dirt1
dirt2
dirt3
dirt4
dryair
drydirt
epoxy
falcol
fish
germanum
glass
hdepoxy
hpolyeth
Supprimer
Aide
Material INFO
Nom du matériau bauxite1
Densité, g/cm3 1.25
Commentaire keust_1
Composition chimique
H:0.01%O:43.25%AL:36.84%SI:2.01%CA:1.00%TI:1.74%FE:15.15%[_AL2O3:69.60%_SIO2:4.30%_FE2O3:21.70%_TIO2:2.90%_CAO:1.40%_H2O:0.10%]
Coefficients polinôminaux d'atténuation de masse
gamme d'énergie 0.010000 - 0.015849 MeV
a0=-3.0012837e+001
a1=-2.7718603e+000 a2=-5.0591479e-001 a3=+7.8656516e-002
a4=-4.4686870e-001 a5=+1.0084082e+000 a6=-1.2301660e+000
a7=+8.9170485e-001 a8=-3.7889175e-001 a9=+8.7213096e-002
gamme d'énergie 0.012589 - 0.025119 MeV
Fermer

Monte Carlo

- Use of Mirion' software LABSOCS
- Beaker need to be designed – reusable for every samples – usable for every detector
- Modelling matrix : chemical composition

Count = sample only

Matrix modelling = 5min

Advantage: very quick, 5min of technician work compared to 3h45

Disadvantage: if unknown matrix composition, need to use a similar matrix (e.g. sand, concrete, …)

RESULTS COMPARISON

Results comparison of unknown composition

from KRISS NORM MRC 2018 (3 samples)

No definitive values yet

Korea Research Institute of Standards and Science

➡ reference activities used for calculations = **means of labs**

Criteria retained = French criteria for ministry agreements

Normalized bias = $$\frac{value_{reported} - value_{target}}{\sqrt{(2*uncertainty_{reported})^2 + (2*uncertainty_{target})^2}}$$

Results **acceptable if Normalized bias < |1.0|**

Efficiency with LABSOCS, with chemical composition found in literature (BRGM)

Results comparison of unknown composition

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- Phosphogypsum:

	Labs mean (Bq/kg)		EICHROM (Bq/kg)			
Radionuclide	Activity	uncertainty (k=1)	Activity	uncertainty (k=1)	bias	**Normalized bias**
Th-234	50,5	4,3	47,8	4,2	-5,4%	**-0,23**
Th-230	411	26	370	67	-10,0%	**-0,29**
Ra-226	424	29	381	60	-10,0%	**-0,32**
Pb-214	424	29	384	42	-9,3%	**-0,39**
Bi-214	424	29	378	29	-10,7%	**-0,56**

- Zirconium silicate:

	Labs mean (Bq/kg)		EICHROM (Bq/kg)			
Radionuclide	Activity	uncertainty (k=1)	Activity	uncertainty (k=1)	bias	**Normalized bias**
U-235	166	17	188	12	0,0%	**0,53**
Pa-234m	3603	370	3838	170	6,5%	**0,29**
Th-234	3603	370	4003	311	11,1%	**0,41**
Th-230	3639	260	3948	643	8,5%	**0,22**
Ac-228	753	73	669	37	-11,1%	**-0,51**
Ra-226	3603	370	3747	596	4,0%	**0,10**
Pb-214	3603	370	3424	378	-5,0%	**-0,17**
Bi-214	3603	370	3524	270	-2,2%	**-0,09**
Pb-212	753	73	660	54	-12,3%	**-0,51**
Bi-212	753	73	740	54	-1,7%	**-0,07**
Tl-208	251	24	230	14	-0,9%	**-0,37**

Normalized bias < |1,0| ☺

Results comparison of unknown composition

- Bauxite:

4 different compositions found in literature (BRGM), with huge variations of silicon composition.

When unknown composition, impact on gamma and on I?

	Labs mean (Bq/kg)		EICHROM (Bq/kg)				
Radionuclide	Activity	uncertainty (k=1)	Activity	Uncertainty (k=1)	bias	Normalized bias	Standard deviation
K-40	< 50		< 50				
U-235	4,37	0,38	4,25	18,1%	-2,7%	-0,09	0,0%
Th-234	94,8	8,2	97,2	8,3%	2,6%	0,12	4,6%
Th-230	103,1	8,6	129	27,5%	25,2%	0,49	9,5%
Ac-228	153	11	146,0	5,7%	-4,3%	-0,27	0,0%
Ra-226	94,8	8,2	92	18,8%	-3,2%	-0,10	1,0%
Pb-214	94,8	8,2	89	11,3%	-5,8%	-0,25	0,1%
Bi-214	94,8	8,2	92,1	9,0%	-2,8%	-0,13	0,2%
Pb-212	153	11	152	7,9%	-0,5%	-0,03	0,3%
Bi-212	153	11	157	7,2%	2,8%	0,16	0,3%
Tl-208	50,8	3,5	54,3	6,3%	6,8%	0,41	0,0%
I	0,82		0,79		-3,9%		0,4%

Normalized bias < |1,0| ☺

Standard deviation < uncertainty ☺

Good agreement between expected values and experimental results

Results comparison of unknown composition

- Same Bauxite results with energy:

Radionuclide	energy (keV)	bias	Normalized bias	Standard deviation
Th-230	67,7	25,2%	0,49	9,5%
Th-234	92,6	2,6%	0,12	4,6%
U-235	185,7	-2,7%	-0,09	0,0%
Ra-226	186,2	-3,2%	-0,10	1,0%
Pb-212	238,6	-0,5%	-0,03	0,3%
Pb-214	351,9	-5,8%	-0,25	0,1%
Tl-208	583,2	6,8%	0,41	0,0%
Bi-214	609,3	-2,8%	-0,13	0,2%
Bi-212	727,3	2,8%	0,16	0,3%
Ac-228	911,2	-4,3%	-0,27	0,0%
K-40	1460,8	-	-	
I	-	-3,9%	-	0,4%

Energy increasing = Standard deviation decreasing

I is calculated with Ra-226 Ac-228 and K-40

Use of **average composition** from literature has **very small impact on dose I** with Labsocs

Monte Carlo

- Daily used and accredited method since 2011 (ISO 17025 ACCREDITED LABORATORY)
- Around 6 proficiency test per year to check validity of LABSOCS parameters, on many matrix (water, milk, air filter, vegetable, soil, sediment, concrete, …)
- All results are acceptable for PTE organizer criteria, especially for low energy emitters (^{241}Am, ^{210}Pb) and sum peaks emitters (^{60}Co, ^{228}Ac)
- Described method in environmental soils standard ISO 18589-3 but not yet in CEN/TS 17216 (building materials)

➡ **Very reliable**

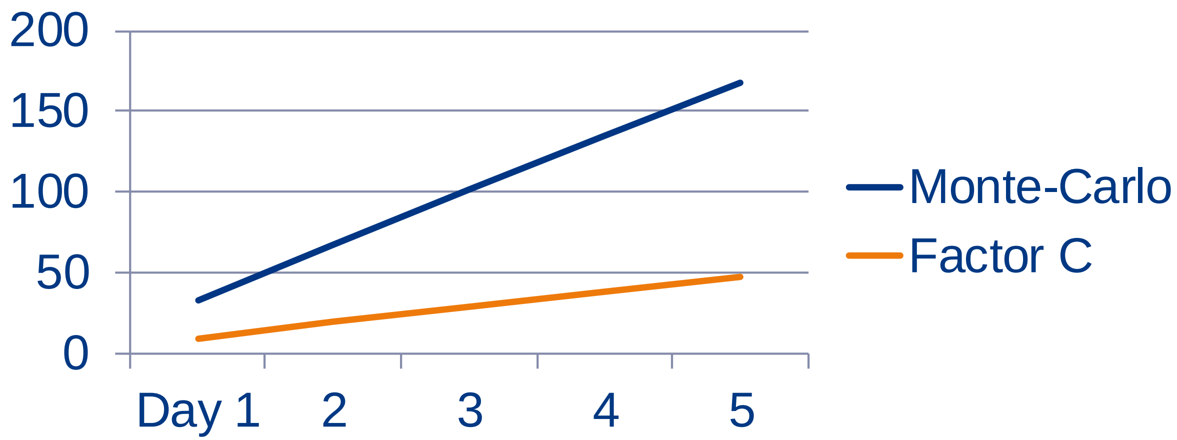
- Each detector we buy is LABSOCS characterized

CONCLUSION

Eurofins Eichrom answers different problematics

Gamma method used by EUROFINS EICHROM :

- ✓ measure ^{40}K, ^{226}Ra and ^{232}Th, and calculate dose assessment l of building materials
- ✓ in agreement with environmental standards
- ✓ Quicker than sources method (up to 5times!) :



- ✓ Our accredited method of calibration ensure reliable, affordable and quick results

CEN/TS 17216 “should” include Monte-Carlo method to answer industrial problematic

Merci

Dziękuję

Thank you

شكرا

Gracias

Obrigado

köszönöm

ありがとうございました

eichrom LABORATORIES

member of

eurofins | Nuclear

Patrice Letessier
Director of Nuclear Activities - President
+ 33 2 23 50 13 80

Patrice Letessier@eurofins.com

Benoît Daniel
Technical Manager
+ 33 2 23 50 15 82

BenoitDaniel@eurofins.com

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