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FEUP FACULDADE DE ENGENHARIA UNIVERSIDADE DO PORTO

Building Materials: The exposure to radon and gamma radiation

DEM FEUP - 2015
130/100 ANOS DE ENGENHARIA DE MINAS

Academia Politécnica do Porto (1885/1915)
Escola de Engenharia do Porto (1915-....)

Maria de Lurdes Dinis

Department of Mining Engineering (DEM FEUP) (*http://paginas.fe.up.pt/~deminas/*)

Faculty of Engineering, University of Porto (FEUP) (*www.fe.up.pt/*)

Research Centre for Natural Resources and the Environment (CERENA/FEUP) (*cerena.ist.utl.pt/*)

http://www.cerena.pt/
Twitter:@cerena_research

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IAEA GENERAL Safety Requirements - GSR Part 3

Directive 2013/59/EURATOM

DL Nº 108/2018, 03 December 2018

National Radon Action Plan

- *Strategy, including methods and tools, to prevent the penetration of radon in new buildings, including the identification of building materials with significant radon release.*

Indoor external exposure gamma radiation emitted by building materials

- *The reference level applying to indoor external exposure to gamma radiation emitted by building materials, in addition to outdoor external exposure, shall be 1 mSv per year.*

……

DL Nº 108/2018 , 03 December 2018

Natural Materials

- Alum-shale.

Building materials or additives of natural igneous origin

- Granitoides (such as granites, syenite and orthogneiss);
- Porphyries;
- Tuff;
- Pozzolana (pozzolanic ash);
- Lava.

Materials incorporating residues from industries processing NORM

- Fly ash;
- Phosphogypsum;
- Phosphorous slag;
- Tin slag;
- Copper slag;
- Red mud (residues from aluminium production);
- Residues from steel production.

Other identified by the competent authority

DL Nº 108/2018 , 03 December 2018

1 – ...activity concentration index (AI) are determined ... **before such materials** are placed in the market.

2 - ...before being placed in the market for the 1st time ... **and whenever** there is a modification of the factors that may affect the measured parameters.

3 - ...measurement results and the corresponding assessment of the AI, ... **provided** to the competent authority if requested.

4 - ...whenever **AI exceeds value of 1** - **must be communicated** to the competent authority which proceeds to dose estimative.

5 - ... building materials which are liable to give **doses exceeding RL**, the competent authority shall decide on appropriate measures, which may include specific requirements in building codes or restrictions on the use of such materials.

Natural stone and **other building materials** (concrete, brick, gypsum) - natural radionuclides ^{238}U, ^{232}Th + decay products, and ^{40}K.

- U238 ⟶ Ra226 ⟶ Rn222
- Proportion of Rn222 from building materials and natural stone in homes is small compared to the amount from soil.

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NORMs

In some branches of industry, natural radionuclides can accumulate in parts of the material flux.

Compared to the natural background content of soils, NORM show an enhanced content of naturally occurring radionuclides.

Some of them are used as secondary raw materials in the building and construction industry.

Pozzolana (pozzolanic ash)

Red mud (residue from aluminium production)

Construction/Building Materials:

- When using rocks and soils for building purposes, radionuclides contained therein or released from them may lead to a radiological exposure (U238, Th232 and K40).
- Radionuclides and radiation exposure – emission of gamma radiation and, in very particular cases, inhalation of radon released.

Construction Materials

Growing tendency to use new recycled materials

- Range 100-400 Bq/kg for U-238 and Th-232;
- Large-scale use of by-products, with enhanced levels of radioactivity, as a raw material in building products considered to increase considerably the exposure (Real Potential Risk?)

Research and studies - attention had been drawn to measurement of activity levels of construction materials

Several authors studied activity concentrations

CERAMIC TILES

GRANITE | SEDIMENT | CONCRETE
SOIL | MARBLE | SAND

EU national surveys - established radioactivity concentrations

Raw materials | Industrial by-products

Gamma radiation + Rn-222 exhalation rates

Construction materials

Elevated levels of natural radionuclides (annual doses of several mSv) worldwide - Brazil, France, India, Nigeria.

Different types of building materials - concentration of over 2 - 3 orders of magnitude between the minimum and maximum values.

Bricks: ^{226}Ra, ^{232}Th and ^{40}K activity concentration of 1916 samples from 25 EU MS and 55 samples from 3 non EU countries.

Bricks	n. samples	^{226}Ra (Bq kg^{-1})			^{232}Th (Bq kg^{-1})			^{40}K (Bq kg^{-1})		
		mean	max	min	mean	max	min	mean	max	min
EU countries										
Austria	32	38	71	20	45	112	16	635	880	520
Belgium	78	39	47	33	37	47	32	692	815	569
Bulgaria	1	42			43			600		
Cyprus	10	7	28	0.1	4	8	2	178	321	59
Czech Republic	516	69	113	46	61	86	48	498	616	312
Denmark	84	31	42	8	20	34	6	455	630	280
Estonia	6	20	27	13	19	30	13	275	440	145
Finland	42	51	80	23	41	62	21	804	986	622
France	12	69	133	24	67	106	18	59	118	21
Germany	188	84	281	8	72	233	6	463	719	115
Greece	86	52	93	35	41	52	24	685	860	551
Hungary	176	57	200	30	48	67	33	666	925	444
Ireland	14	42	139	7	31	50	8	482	1064	255
Italy	196	40	110	4	40	99	5	566	879	160
Lithuania	2	36	40	31	26	32	20	638	754	522
Luxembourg	3	68	83	53	102	147	58	805	1013	597
Netherlands	70	43	76	8	43	82	8	545	1030	150
Poland	6	16	20	11	19	33	6	514	825	204
Portugal	10	64	90	37	52	72	31	786	1098	473
Romania	76	53	139	8	47	66	12	549	1038	196
Slovakia	163	83	129	49	85	132	44	637	695	590
Slovenia	2	81	93	69	87	101	72	676	898	454
Spain	12	54	73	34	68	99	44	569	747	292
Sweden	71	75	98	10	94	127	7	734	960	162
United Kingdom	60	49	94	2	34	81	3	560	1000	12
Non EU countries										
Norway	19	83	104	63	69	74	63	1086	1136	1037
Switzerland	18	47	62	32						
Turkey	18	37	70	9	39	70	10	636	923	208
EU MS overall average and range		51 (7-84)			49 (4-102)			555 (59-805)		
CV		41%			52%			32%		
Non EU MS average		56			54			861		

Concrete: ^{226}Ra, ^{232}Th and ^{40}K activity concentration of 3458 samples from 24 EU MS and 171 samples from 3 non EU countries.

Concrete	n. samples	^{226}Ra (Bq kg^{-1})			^{232}Th (Bq kg^{-1})			^{40}K (Bq kg^{-1})		
EU countries		mean	max	min	mean	max	min	mean	max	min
Austria	1	15	21	7	14	57	3	164	382	16
Belgium	31	21	42	6	21	42	5	243	490	85
Bulgaria	1	19			17			200		
Czech Republic	1756	272	936	30	49	72	24	419	495	268
Denmark	22	72	670	10	30	53	10	685	1190	280
Estonia	1	35			11			207		
Finland	35	55	75	33	42	59	34	599	838	359
France	16	14	26	8	12	24	4	17	20	14
Germany	96	53	100	18	50	100	12	642	1100	193
Greece	71	35	140	7	5	17	3	79	383	23
Hungary	95	15	22	7	13	24	7	234	407	148
Ireland	8	29	68	18	12	43	3	217	1100	16
Italy	33	17	23	7	24	38	16	306	457	200
Lithuania	2	35	37	32	21	25	17	453	480	426
Luxembourg	5	87	163	4	66	99	6	323	707	110
Netherlands	66	66	710	10	32	132	6	239	870	111
Poland	678	115	200	65	72	127	36	666	1005	492
Portugal	11	53	98	8	47	86	7	404	529	278
Romania	135	78	118	17	138	556	16	459	918	163
Slovakia	150	32	107	10	21	41	5	345	664	214
Slovenia	2	21	22	20	11	11	10	124	143	105
Spain	33	30	38	21	24	32	18	244	283	204
Sweden	188	200	1300	3	51	100	3	432	770	20
United Kingdom	22	60	89	18	32	43	13	492	650	370
Non EU countries										
Norway	151	38	52	26	44	56	36	710	811	638
Switzerland	9	60	149	12						
Turkey	11	16	17	16	17	25	9	339	527	151
EU MS overall average and range		59 (14-272)			34 (5-138)			340 (17-685)		
CV		103%			85%			55%		
Non EU MS average		38			30			524		

Cement: ^{226}Ra, ^{232}Th and ^{40}K activity concentration of 2647 samples from 24 EU countries and 67 samples from 3 non EU countries.

Cement	n. samples	^{226}Ra (Bq kg^{-1})			^{232}Th (Bq kg^{-1})			^{40}K (Bq kg^{-1})		
EU countries		mean	max	min	mean	max	min	mean	max	min
Austria	18	27	49	11	14	26	10	210	286	89
Belgium	26	52	64	37	46	76	22	255	470	110
Bulgaria	1	29			19			160		
Cyprus	8	25	37	4	10	12	5	152	209	4
Czech Republic	506	39	46	31	20	20	19	215	237	193
Denmark	8	38	65	20	29	52	12	157	240	90
Estonia	1	47			21			587		
Finland	11	40	84	15	20	55	9	251	336	169
France	2	72	111	32	65	109	21	38	52	24
Germany	36	65	200	20	56	200	15	235	700	40
Greece	191	87	218	15	19	41	10	226	457	32
Hungary	525	25	61	8	22	53	11	207	402	95
Ireland	3	60	107	27	11	15	3	131	252	66
Italy	198	46	98	10	70	240	10	387	846	125
Latvia	1	28	51	5	48	93	3	175	320	29
Lithuania	1	70	465	4	30	211	3	268	1510	2
Netherlands	16	56	82	27	61	120	19	260	290	230
Poland	429	65	154	19	53	138	14	312	608	190
Portugal	8	31	40	22	19	23	15	256	276	235
Romania	57	69	178	4	48	206	12	225	421	50
Slovakia	383	43	61	23	28	67	15	281	613	190
Spain	180	66	422	7	49	266	2	270	599	12
Sweden	30	53	56	44	54	72	41	224	235	196
United Kingdom	8	65	109	22	23	28	18	160	160	160
Non EU countries										
Norway	8	61	96	30	39	59	19	410	815	259
Switzerland	10	20	29	11						
Turkey	49	31	40	22	19	26	14	197	317	99
EU MS overall average and range		50 (25-87)			35 (10-70)			235 (38-587)		
CV		36%			54%			43%		
Non EU MS average		37			29			304		

Fonte: Trevisi et al., 2012

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f + c = concrete floor + ceiling
ph.w. = phosphogypsum wall

Min. and max. of abosrved dose rate in indoor air

γ dose rate in 4 standard rooms, for a reasonable use of brick, concrete, cement and phosphogypsum.

433 nGy/h = 433 x 10^{-6} x 0.7 Sv/Gy x 7000 h/year = mSv/year

Dose to gamma radiation = 2.12 mSv/year

Country	Absorbed dose rate in air indoors (nGy h^{-1})							
	4 concrete walls + (f+ c)		4 brick walls + (f+ c)		3 concrete walls + (ph.w.) + (f+ c)		3 brick walls + (ph.w.) + (f+ c)	
	min	max	min	max	min	max	min	max
Austria	11	113	35	157				
Belgium	17	123	51	124	54	146	77	147
Bulgaria	52	115			47	137		
Cyprus[a]	4	15	6	33				
Czech Rep.								
Denmark	46	750	38	438				
Finland	96	166	85	169	84	231	77	234
France	16	242	39	176				
Germany	84	287	59	245	79	298	62	268
Greece	24	125	38	145	70	167	80	182
Hungary	40	87	58	146				
Ireland	21	160	24	184				
Italy	43	83	29	157				
Lithuania								
Luxembourg	186	204	184	191				
Netherlands	26	319	27	257	26	321	26	277
Poland	137	400	84	254	124	381	86	279
Portugal	3	433	41	320				
Romania	45	303	36	275	54	338	48	318
Slovakia	37	137	52	147				
Slovenia	37	353	88	286				
Spain								
Sweden	94	1338	60	780				
UK	59	177	33	187	110	287	92	293
Arithmetical average	51	282	53	234	72	256	69	250

[a] As data for Cyprus were lacking in the literature, 30% of cement activity concentrations were used as values for concrete.

Fonte: Nuccetell et al., 2012

Specific activities of natural radionuclides in natural stones, building materials and residues (Bq/kg)

(BfS – Federal Office for Radiation Protection)

Material	Radium-226 in Bq/kg Mean value (range)	Thorium-232 in Bq/kg Mean value (range)	Potassium-40 in Bq/kg Mean value (range)
Granite	**100 (30 - 500)**	**120 (17 - 311)**	**1000 (600 - 4000)**
Gneiss	75 (50 - 157)	43 (22 - 50)	900 (830 - 1500)
Diabase	16 (10 - 25)	8 (4 - 12)	170 (100 - 210)
Basalt	26 (6 - 36)	29 (9 - 37)	270 (190 - 380)
Gravel, sand, gravel sand	15 (1 - 39)	16 (1 - 64)	380 (3 - 1200)
Natural gypsum, anhydrite	10 (2 - 70) < 5	(2 - 100)	60 (7 - 200)
Tuff, pumice stone	**100 (< 20 - 200)**	**100 (30 - 300)**	**1000 (500 - 2000)**
Clay	< 40 (< 20 - 90)	60 (18 - 200)	1000 (300 - 2000)
Brick, clinker brick	**50 (10 - 200)**	**52 (12 - 200)**	**700 (100 - 2000)**
Concrete	30 (7 - 92)	23 (4 - 71)	450 (50 - 1300)
Sand-lime brick, porous concrete	15 (6 - 80)	10 (1 - 60)	200 (40 - 800)
Slag from Mansfelder copper-slate	**1500 (860 - 2100)**	**48 (18 - 78)**	**520 (300 - 730)**
Gypsum from flue gas desulfurisation	20 (< 20 - 70)	< 20	< 20
Brown coal filter ash	82 (4 - 200)	51 (6 - 150)	147 (12 - 610)

- Potential for radon release from building products - determined by specific activity of Ra226 and other material characteristics determining the radon transport (such as porosity).

- Results have shown that the traditional use of building materials such as:
    - Concrete,
    - Brick,
    - Porous concrete and,
    - Sand-lime brick.

- Generally **not the cause for the annual mean value of the indoor radon concentration!**

Higher radon concentrations in some building materials:

- Release rates that may result in higher indoor concentrations were occasionally measured in residues from the incineration of coals with enhanced U/Ra concentrations (formerly used locally to fill ceilings, referred as “coal slag”).

- Occurrences of above-average radon concentrations in the traditional mining regions, where residues from ore processing with enhanced radium concentration were used as building material: concrete or mortar additive or as foundation in house building.

How is the radionuclide content assessed?

- New Radiation Protection Law - requires for certain construction materials that the manufacturer or importer measure the concentrations of the natural radionuclides and must prove that the legal reference value for the exposure of the users of buildings is not exceeded by the building material.

- European Standards Institute CEN - developing standards measurement and evaluation of building materials within the framework of the Construction Products Regulation.
- Ultimately, the CE mark on a building material will also prove that the European radiation protection requirements have been met.

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- Reference level for exposure to radionuclides in construction materials is not to exceed 1 mSv/year.

- Activity Index (AI) = 1, ensures compliance with 1 mSv/year.

$$AI = \frac{C_{K40}(Bq/kg)}{3000} + \frac{C_{Ra226}\,(Bq/kg)}{300} + \frac{C_{Th232}\,(Bq/kg)}{200}$$

$$AI = \frac{313 \text{ x } C_{K40}(\%)}{3000} + \frac{12.5 \text{ x } C_{Ra226}\,(ppm)}{300} + \frac{4.06 \text{ x } C_{Th232}(ppm)}{200}$$

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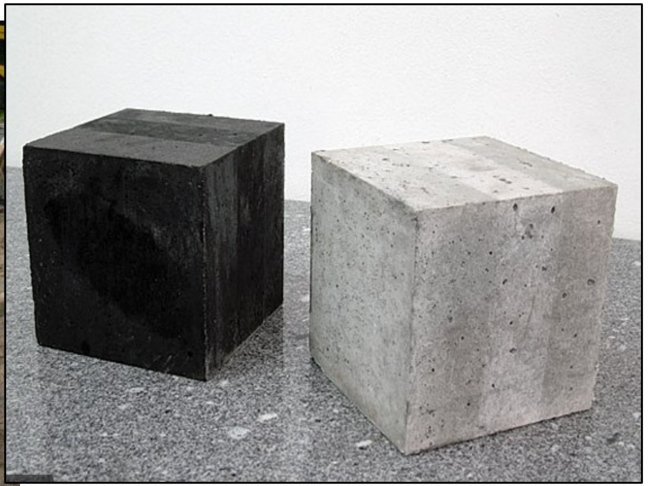
The European [EU28+EFTA, 2016] aggregates demand is 2.7 billion tonnes per year - annual turnover of ~€15-€20 billion.

What is needed:

- < 1 mSv/year gamma dose
- Testing NORM beginning cycle of the material
- Accredited test methods
- Certified factory control system – production and in situ tests
- Laboratory capacity
- High test capacity + quick and reliable results + low investment + low price per test.

Methodology:

- In situ testing – rock pit (considering eventually differences on the rock - heterogeneous).
- Production control – factories.
- Dose rate testing inside constructed buildings.
- Exact part the construction in the room/building is the main source of high dose rate (floor, wall and/or roof construction).



Method MMK 610 - Testing of building material raw rock/ballast/aggregate

- Accredited in accordance with the Construction Product Regulation (EU) No. 305/2011 - accreditation applies internationally throughout the EU.

- Activity Index, Radium Index and Gamma Radiation.

- Carried out in situ in the quarry with a non destructive method directly in the bore hole.

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Method MMK 605 – Determination of AI, RI, gamma radiation in construction materials

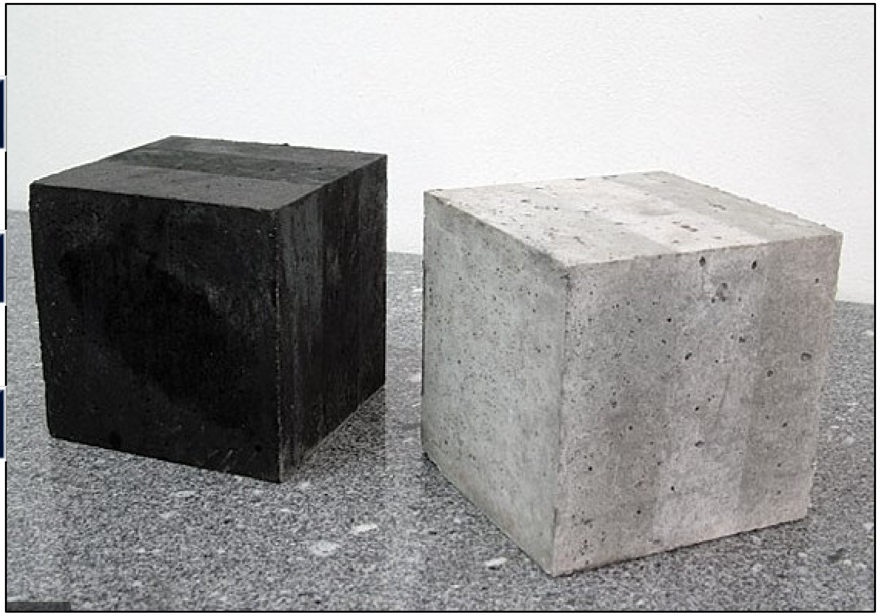
Scope

- Intended for laboratory and field laboratory for measure ^{40}K, ^{226}Ra and ^{232}Th (**suitable for daily factory control purposes**).
- Results from these radionuclides - activity index (AI), radium index (RI) and gamma radiation rate for the construction material.

- AI = tool for determining the suitability for a material regarding effective dose.

- RI = tool for determining the potential of radon production from the material.

- Gamma radiation = dose-rate from the material expressed in µSv/h.



Method MMK 605 – Determination of AI, RI, gamma radiation in construction materials

- Activity concentration of gamma-emitting radionuclides in construction material - determined by gamma spectrometry.
- Sample of 150 mm x 150 mm x 150 mm ± 5 mm.
- The building material in loose state consist of a volume of ≥ 10 litres.
- Minimum of two samples from the same production batch.

- Measurements carried out at least twice on each sample.
- Minimum measurement time = 300 seconds.

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Method MMK 605 – Determination of AI, RI, gamma radiation in construction materials

- Determination of activity concentration in a sample reflects the building material under the intended form of use.
- Instantaneous and can be performed at all times during the year.
- Natural nuclides: % for ^{40}K, ppm for ^{226}Ra and ppm for ^{232}Th.

- Average of activity concentration in one sample.
- Average of the activity concentration of all samples from the same batch.
- Calculation of AI, RI and gamma radiation dose rate from the building material.

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Activity Index (AI)

Element	Content	Activity concentration, Bq/kg
K - Potassium	1 %	313
U-Uranium 238/235	1 ppm	12,35
Th-Thorium	1 ppm	4,06

$$AI = \frac{C_{K40} \text{ x } 313}{3000} + \frac{C_{Ra} \text{ x } 12{,}35}{300} + \frac{C_{Th} \text{ x } 4{,}06}{200}$$

where:

C_{K40}= content of potassium, %.

C_{Ra}= content of radium (U238/235), ppm.

C_{Th}= content of thorium, ppm.

Radium Index (RI)

Element	Content	Activity concentration, Bq/kg
U-Uranium 238/235	1 ppm	12,35

$$RI = (C_{Ra} \text{ x } 12{,}35)/200$$

RI <= 1, 200 Bq/kg ^{226}Ra

where:

C_{Ra} = content of radium (U238/235), ppm.

Radiação Gama (µSv/h)

Element	Content	Dose rate, µSv/h
K - Potassium	1 %	0,0151
U-Uranium 238/235	1 ppm	0,0065
Th-Thorium	1 ppm	0,0029

$$\mu Sv/h = (C_{K40} \text{ x } 0{,}0151) + (C_{Ra} \text{ x } 0{,}0065) + (C_{Th} \text{ x } 0{,}0029)$$

where:

C_{K40} = content of potassium, %.

C_{Ra}= content of radium (U238/235), ppm.

C_{Th}= content of thorium, ppm

Principle for the method MMK 608 – Measurement of gamma exposure indoor inside buildings

Scope

In-situ method for determination of gamma exposure indoors inside buildings.

- Caused by NORM nuclides K40, Ra226 (U238 235) and Th232.

Principle for the method:

- Dose-rate is measured in the air in a room.
- Calculate effective dose.
- Instantaneous and can be performed at all times during the year.

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Principle for the method MMK 608 – Measurement of gamma exposure indoor inside buildings

Principle for measurement

- Air in the middle of a room with an instrument reporting dose-rate.
- Measurement is performed on c/c dimensions of the area (walls/floor/ceiling = 1 measurement points) in a room.

Measurement place

- Rooms where inhabitants stay more than temporarily ex. living rooms, bedrooms, and eating/cooking areas.
- Rooms/facilities where staff stays more than temporarily ex., facilities/rooms where daily operations are carried out, as well as other staff-spaces and dining rooms.

Measurement time

- Minimum = 300 seconds per measurement point.
- Measurement - at least twice on each point.

Principle for the method MMK 608 – Measurement of gamma exposure indoor inside buildings

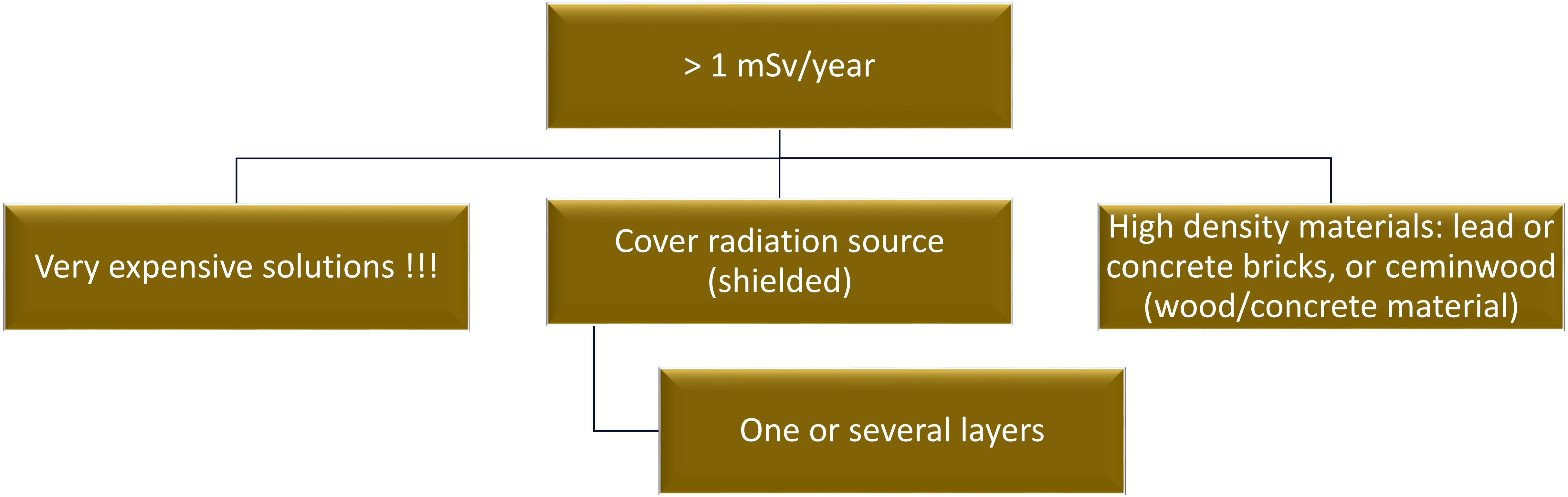
Calculation of effective dose
- Mean value for all measurement points in a room.

Step 1
- Measurement of individual measurement point = mean value for the individual point.
- Mean value for all measurement points in a room.

Step 2
- The effective dose is calculated.

Eeff = Dose-rate x Exposure time
- Exposure time residence/housing = 7000 h/y
- Exposure time workplaces = 2000 h/y

- If the result is >1 mSv/year in the room - a much more advanced and time consuming method has to be used. It is necessary to determine exactly which construction material is the problem: wall, floor and/or roof construction.



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Principle for the method MMK 606 – Activity Index of construction materials indoors of existing buildings

Scope

Measuring construction materials - ^{40}K, ^{226}Ra and ^{23}2Th.

→ determination of activity index (AI) for the construction material.

Activity concentration of gamma emitting radionuclides in construction materials - gamma spectrometry:

- Measured in a room directly on a surface of the building material.
- Measurement instantaneous, can be performed at all times during th
- Activity index is calculated.

Principle for the method MMK 606 – Activity Index of construction materials indoors of existing buildings

Measurement instrument

- Gamma spectrometer.

Measurement time

- Minimum = 300 sec/measurement point, at least twice on each point.

Principle for the measurement

- Directly on the surface of the construction material.
- Measurement performed on c/c dimensions of the area (walls/floor/ceiling = 6 measurement points) in a room.

Measurement place

- Rooms where inhabitants stay more than temporarily (living rooms, bedrooms, eating/cooking areas).
- Rooms/facilities where staff stays more than temporarily (facilities/rooms where daily operations are carried out, other staff-spaces and dining rooms).

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Principle for the method MMK 606 – Activity Index of construction materials indoors of existing buildings

Step 1
Measurement of individual measurement points = Mean activity concentration value of individual measurement point.

Step 2
Measuring mean value measuring point no (1 + 2 + 3 + 4 + 5 + 6) ÷ 6 = measured average value.

Step 3
The activity index is calculated:

$$AI = \frac{C_K \text{ x } 313}{3000} + \frac{C_{Ra} \text{ x } 12{,}35}{300} + \frac{C_{Th} \text{ x } 4{,}06}{200}$$

where:
C_K= content of potassium, %.
C_{Ra}= content of radium (U238/235), ppm.
C_{Th}= content of thorium, ppm.

$$AI = \frac{C_K}{3000} + \frac{C_{Ra}}{300} + \frac{C_{Th}}{200}$$

where:
C_K= content of potassium, Bq/kg.
C_{Ra}= content of radium (U238/235), Bq/kg.
C_{Th}= content of thorium, Bq/kg.

Final Remarks

Accredited/Validated methods:

- Measurement of NORM radionuclides (K-40, Ra-226 (U238/235) and Th-232)
- Estimation of activity index, radium index and gamma dose rate

Objective to fulfil a number of requirements set out in international directives, regulations, standards and DL 108/2018:

- IAEA Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards, No. GSR Part 3.
- EU Directive 2013/59/EURATOM.
- Regulation (EU) No 305/2011 on harmonised conditions for the marketing of construction products.
- Regulation (EC) No 765/2008 setting out requirements for accreditation and market surveillance relating to marketing of products.
- Radiological Protection Principles concerning the Natural Radioactivity of Building Materials, EC RP 112 (1999).

Thank you!

감사해요	谢谢	ありがとう