New perspectives and issues arising from the introduction of NORM residues in building materials

Cristina Nuccetelli, Rosabianca Trevisi^, Yiannis Pontikes+*

*National Institute of Health (ISS), Rome, Italy

^National Institute of Occupational Safety and Prevention (INAIL), M. Catone (Rome), Italy

+Katholieke Universiteit Leuven (KU Leuven), Heverlee, Belgium

Contents

- Database of byproducts used in building materials
  - activity concentrations in Member States
    - ✓ fly ash
    - ✓ blast furnace slag
    - ✓ phosphogypsum
    - ✓ red mud (from Australia, China, Greece, Hungaria, Jamaica, Turkey)
- Analysis of bulk / superficial products exceeding
  - RP112 criterion
  - RP112 criterion + Rn
  - Dose reference levels with *Room model*
- Conclusions

Database information

Database contains data of byproducts for 17 out of the 27 MS. This sub-database is part of the database of building materials in EU (but red mud).

For some MS information are not available for all types of byproducts, therefore number of MS involved are different for each material.

In order to estimate the radiological impact of the byproduct introduction in building materials, the database of cement and bricks in MS was used.

Different types of byproducts diluted in standard building materials or used as they are:

- ✓ fly ash (about 650)
- ✓ furnace slag (about 145)
- ✓ phosphogypsum (about 300)
- ✓ red mud (few data)

- ✓ others (about 270) not analysed

By products (I): fly ash (13 MS- 666 samples)

Country	N of samples	^{226}Ra (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40}K (Bq kg^{-1})
Bulgaria	1	120	60	500
Denmark	79	155	105	615
Estonia	1	27	14	301
Finland	58	125	62	495
Germany	2	141	76	424
Greece	172	768	58	350
Hungary	1	102	122	435
The Netherlands	4	133	111	537
Poland	108	235	118	798
Romania	157	210	123	592
Slovakia	1	83	68	496
Spain	81	69	67	799
United Kingdom	1	89	68	900
overall average		**174**	**81**	**557**

By products (II): furnace slags (8 MS – 146 samples)

Country	N of samples	^{226}Ra (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40}K (Bq kg^{-1})
Belgium	1	85	41	
Finland	5	117		176
Germany	15	378	61	321
Greece	1	15	1	20
Hungary	1	115	36	229
The Netherlands	1	1200	60	100
Poland	46	169	98	644
Romania	76	94	49	514
overall average		**272**	**50**	**286**

Fly ash and slag in building products

These byproducts are already used - for many years - as cementitious material:

- ✓ Fly ash - pozzolanic properties
- ✓ Blast furnace slag - reduction of permeability, increased density

In the last years research activities have been devoted to their use as aggregates (fine and/or coarse) in new materials maintaining at least characteristics and properties of standard concrete (Australia, India, Japan, US...)

Application of index I of RP 112 was carried out on the standard room (4x5x2.8 m^3) with the following hypotheses:

Concrete composition:
- 20% cement
- 35% fine aggregate
- 45% coarse aggregate

Byproduct use:

Fly ash -
- 25% in cement
- 25% in cement + 35% as fine aggregate

furnace slag -
- 50% in cement
- 50% in cement + 50% as coarse aggregate

Cement: ^{226}Ra, ^{232}Th and ^{40}K activity concentration (~2000 samples)

Country (21MS)	N of samples	^{226}Ra ($Bq\ kg^{-1}$)			^{232}Th ($Bq\ kg^{-1}$)			^{40}K ($Bq\ kg^{-1}$)		
		Average	Min	Max	Average	Min	Max	Average	Min	Max
Austria	19	27	11	49	14	10	26	210	89	286
Belgium	26	52	37	64	46	22	76	255	110	470
Bulgaria	1	29			19			160		
Cyprus	8	16	9	21	10	5	12	152	4	209
Czech Republic	496	46			19			237		
Denmark	6	20	9	30	12	4	21	90	20	140
Finland	11	40	15	84	20	9	55	251	169	336
France	1	35			21			24		
Germany	23	86	30	200	73	20	200	170	40	320
Greece	183	85	20	218	19	10	41	257	32	553
Hungary	400	30	8	61	22	13	53	218	95	402
Ireland	3	60	27	107	11	3	15	131	66	252
Italy	200	41	7	98	63	9	240	357	80	846
The Netherlands	17	62	27	82	64	19	120	271	230	305
Poland	344	73	17	154	66	20	138	353	204	608
Portugal	8	31	22	40	19	11	23	256	235	276
Romania	55	44	4	154	27	11	76	233	47	633
Slovakia	6	35	23	47	18	15	19	223	190	253
Spain	171	61	23	422	40	15	266	305	44	599
Sweden	30	53	44	56	54	41	72	224	196	235
United Kingdom	6	22			18			160		
Overall average		**45**			**31**			**216**		

An index accounting for Rn

The Index I from RP 112

$I = C_{Ra\text{-}226}/300 + C_{Th\text{-}232}/200 + C_{K\text{-}40}/300 \leq 1$

Gamma dose ≤ 1 mSv/y ($C_{Rn\text{-}222} \leq 200 Bq/m^3$ (ICRP 65/103))

With the new Rn nominal risk coefficient (ICRP statement 2009) the lower reference level in dwelling is 100 Bq/m^3, roughly corresponding to 3 mSv/y.

The Index I from RP 112 + Rn (a screening tool accounting for Rn)

Starting from the classical formula

$C_{Rn\text{-}222} = C_{Ra} * \lambda * \varepsilon * \rho * d/2 * S / V / \nu$

$$I_{+Rn} = C_{Ra\text{-}226}(1+\alpha)/300 + C_{Th\text{-}232}/200 + C_{K\text{-}40}/300$$

where α is a factor taking into account :

- outdoor ^{222}Rn background (10 Bq/m^3)
- emanation, density and wall thickness
- ^{222}Rn reference level = 100 Bqm^{-3}

Note: for fly ash in cement and concrete studies are underway in order to assess the emanation reduction in an experimental room made of massive concrete (Haquin-IRPA 13-P10.87).

Percentage of concrete samples with I and I_{+Rn} exceeding screening level

	Use in cement		Use in cement & aggregate	
	$I > 1$	$I_{+Rn} > 1$	$I > 1$	$I_{+Rn} > 1$
Fly ash concrete (651 samples in 13 MS)	0%	0%	40%	65%
Furnace slag concrete (146 samples in 8 MS)	0%	1%	32%	34%

By products (III): phospho-gypsum (12 MS - 310 samples)

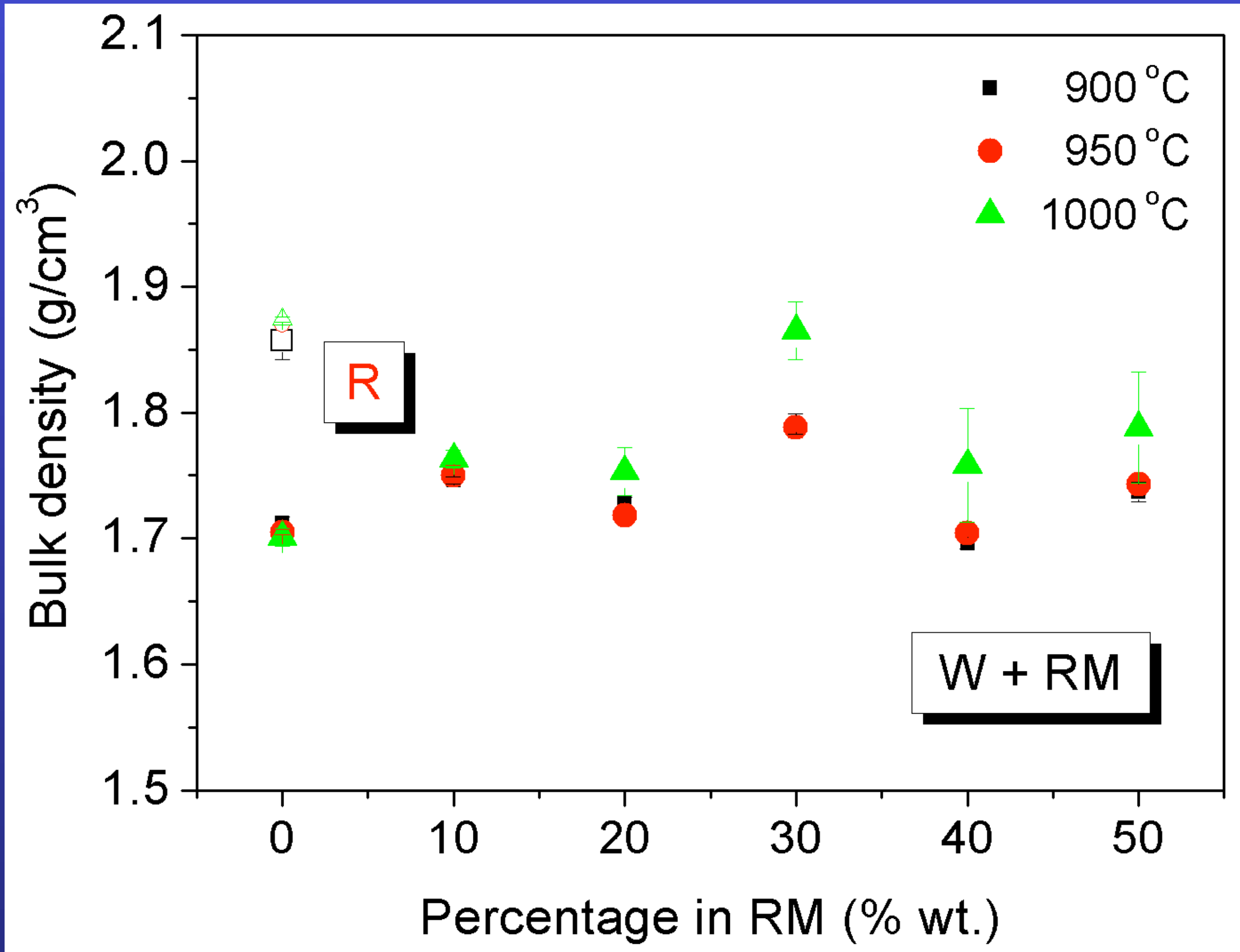
Country	N of samples	^{226}Ra (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40}K (Bq kg^{-1})
Belgium	32	272	21	
Bulgaria	2	209	17	3
Czech Republic	22	115	31	95
Finland	17	306	23	17
Germany	4	168	20	110
Greece	4	606	10	22
The Netherlands	20	223	24	50
Poland	28	267	17	72
Portugal	16	355	7	46
Romania	73	497	40	242
Slovenia	1	500	10	41
United Kingdom	91	1018	33	130
overall average		378	22	75

Gamma dose rate in standard room with 2 phosph. walls

Country	2 phosph. walls + 2 concrete walls + concrete f.c. (nGy/h)		
	average	max	min
Belgium	107	162	78
Bulgaria	98	153	44
Czech Republic	97		
Finland	155	274	77
Germany	173	306	76
Greece	132	196	101
The Netherlands	104	322	26
Poland	224	369	115
Portugal	209	361	60
Romania	188		
Slovenia	258	359	144
United Kingdom	107	162	78

In 6 countries out of 9 the max values exceed the reference level of 250 nGy/h corresponding to 1 mSv/y

Red mud - density of tiles vs RM percentage



R= clay ceramic giving red body

W=clay ceramic giving white body

RM= red mud

Dose from tiles (RM+W) vs RM %

Red mud activity concentration (Bq kg^{-1})			RM% = 10 ρ=1.75	RM% = 20 ρ=1.75	RM% = 30 ρ=1.85	RM% = 40 ρ=1.75	RM% = 50 ρ=1.75
Ra	Th	K	mSv/y				
379	472	21	0.05	0.10	0.16	0.21	0.26
310	1350	350	0.11	0.22	0.34	0.43	0.54*
326	1129	30	0.09	0.19	0.29	0.37	0.46
370	328	265	0.04	0.09	0.13	0.17	0.21
1047	350	335	0.08	0.16	0.25	0.32	0.40
477	705	153	0.07	0.15	0.23	0.29	0.36
300	260		0.03	0.07	0.11	0.13	0.17
250	400		0.04	0.08	0.13	0.16	0.20
210	539	112	0.05	0.10	0.15	0.19	0.24

average values

Ra	Th	K
408	615	181

* I = 4 < 6 reference level for superficial materials

Dose rate in standard room vs % of RM in bricks

(RM average activity concentrations)

Country (24 MS)	4 brick walls +concrete (f+c) in standard room (nGy/h)		
	10%	20%	30%
Austria	109	145	182
Belgium	114	150	187
Bulgaria	130	166	203
Cyprus	57	98	138
Czech Republic	140	176	212
Denmark	181	219	257
Finland	166	201	236
France	125	163	200
Germany	170	207	243
Greece	115	151	187
Hungary	126	162	198
Ireland	108	145	183
Italy	115	151	188
Lithuania	128	164	201
Luxembourg	242	272	303
The Netherlands	127	164	201
Poland	190	229	267
Portugal	190	224	259
Romania	171	208	244
Slovakia	140	176	211
Slovenia	193	226	259
Spain	140	175	210
Sweden	300	332	365
United Kingdom	149	186	222

Conclusions

Fly ash and furnace slag:

- radioactivity content is generally high, especially for ^{226}Ra
- when added to cement, on average they do not cause a significant increase in the radioactivity content of the concrete (I and $I_{+Rn} < 1$)
- different situation if byproducts are used as aggregate of concrete (coarse and/or fine): concern due to the increasing pressure of industry to use them as bulk cheap component of concrete.

Phosphogypsum:

- an extensive use of phosphogypsum walls (e.g. partition walls) can produce significant increase of dose exceeding the reference level of 1 mSv/y.

Red mud:

- the radioactivity content is generally high both for ^{226}Ra and ^{232}Th
- from the available data, the radiological impact of RM use in tiles does not seem to concern, but a problem exists for clearance of dismantled materials (radiological and toxic issue)
- the RM dilution in bricks (bulk use) presents a significant radiological problem.

A general comment: a responsible management of byproducts reuse for a sustainable resource exploitation might be possible, but some doubts arise due to:

- the non trivial radiation protection problems
- the risk of an unware and/or uncontrollable use (e.g. once by-products were placed on the market for proper applications/products – i.e. limited use and/or low density final materials – might be used without accountig for prescriptions)
- the conflict with the industry profit.

From the EU Directive proposal (Sept. 2011) - art. 29(4):

"... *The competent authority may authorise in specific situations the mixing of radioactive residues containing naturally occurring radioactive material with other materials to promote the reuse and recycling of these materials and* *to reduce public exposure*."

Thank you