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Natural radioactivity of Portland cement mortars made with granite aggregates

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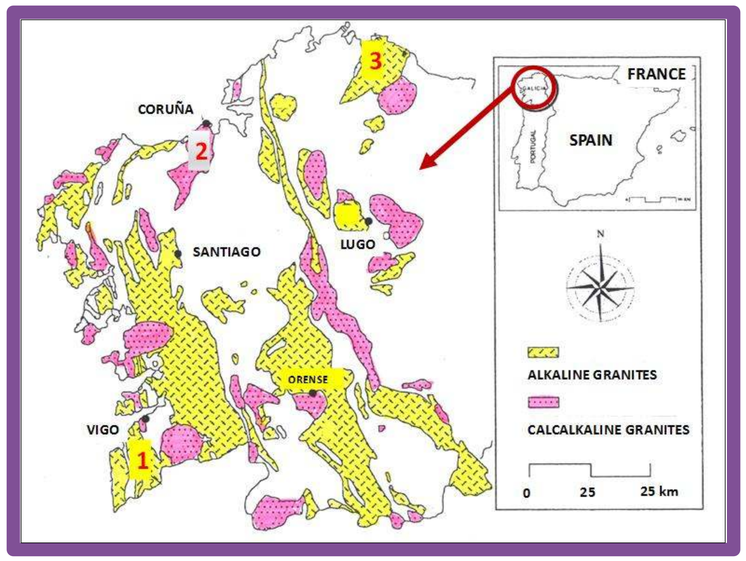
Objectives

The goals of this study are:

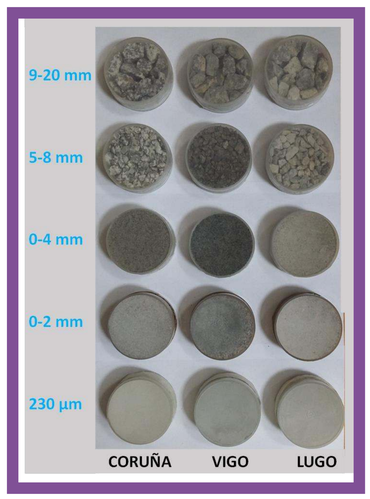
i) To determine the specific radioactivity concentrations of ^{238}U, ^{232}Th, and ^{40}K in local granites provided by three different quarries (Spain).
ii) To study the effect of the aggregate grain size in the gamma radiation.
iii) To determine the specific radioactivity concentrations of ^{238}U, ^{232}Th, and ^{40}K in mortars and to assess the effect of the granite aggregates on Portland cement mortars.

Location of granite quarries



Materials

Granite samples



Chemical composition (XRF)

	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	Na_2O	K_2O	TiO_2	P_2O_5	LOI
CEM I 52,5 R	19.36	4.42	2.62	62.32	1.86	3.23	0.09	0.94	0.23	0.16	2.75
Coruña	72.20	17.11	2.45	0.75	0.62	0.03	2.35	2.67	0.28	0.10	1.23
Vigo	75.57	13.89	3.25	0.89	0.51	0.03	2.54	2.34	0.32	0.09	0.46
Lugo	71.65	13.73	3.61	1.97	1.10	0.06	2.33	2.35	0.33	0.09	2.70

Methodology

Gamma spectrometry analysis

Each granite aggregate sample was packed in a cylindrical (Φ 76 mm x 30 mm) plastic beaker, sealed for three weeks to achieve secular equilibrium between ^{226}Ra and ^{232}Th with their decay products. Later, the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K for all the aggregate samples were measured by a gamma ray spectrometry by using High Purity Germanium (HPGe) detector.
The Council Directive 2013/59/EURATOM defines the Activity Concentration Index (ACI) for the gamma radiation emitted by building materials. This Directive stipulates that building materials ACI must lie below the reference level of 1 mSv year^{-1}.
ACI was calculated for each aliquot and the mean determined for each type of material from the following equation:

$$ACI = \frac{C_{226_{Ra}}}{300} + \frac{C_{232_{Th}}}{200} + \frac{C_{40_K}}{3000}$$

where C_{226Ra}, C_{232Th} and C_{40K} are the activity concentrations in Bq kg^{-1} for radium (equivalent to uranium under secular equilibrium conditions), thorium and potassium, respectively. The activity concentration of ^{40}K was determined directly from the 1460 keV photopeak.

Testing mortars were mixing according to the European standard EN 196-1:2016, and moulding was made directly into the testing plastic beakers. They were made with w/c of 0.5 and cement/sand ratio of 1/3. They were cured at 21 ± 2 ºC and 99 % RH for 28 days. Activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K and ACI for the cement mortars made with quartz and granite aggregates were determined.

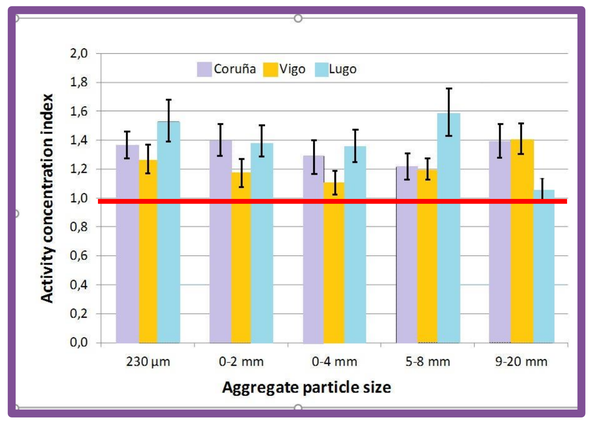
Results

Influence of aggregate particle size on ACI

Activity concentration of the radioisotopes ^{40}K, ^{214}Pb and ^{212}Pb for all the samples.

Sample	Size	Activity concentration (Bq kg^{-1})		
		^{40}K	^{214}Pb (^{226}Ra)	^{212}Pb (^{232}Th)
Coruña	230 µm	1015 ± 43	200 ± 30	70.5 ± 5.7
	0-2 mm	1030 ± 44	207 ± 31	72 ± 12
	0-4 mm	1081 ± 92	180 ± 27	65 ± 11
	5-8 mm	1206 ± 104	148 ± 22	63 ± 11
	9-20 mm	1021 ± 88	172 ± 26	95 ± 16
Vigo	230 µm	945 ± 81	135 ± 20	99 ± 16
	0-2 mm	875 ± 75	128 ± 19	90 ± 15
	0-4 mm	841 ± 72	115 ± 17	86 ± 14
	5-8 mm	1244 ± 106	99 ± 15	88 ± 14
	9-20 mm	1191 ± 103	120 ± 18	120 ± 20
Lugo	230 µm	1032 ± 88	114 ± 17	161 ± 26
	0-2 mm	933 ± 80	96 ± 15	148 ± 24
	0-4 mm	1050 ± 90	90 ± 14	139 ± 23
	5-8 mm	1347 ± 115	111 ± 17	152 ± 33
	9-20 mm	1073 ± 92	90 ± 14	79 ± 13
Siliceous standard aggregate	0 - 2 mm	147 ± 13	4.2 ± 0.71	7.2 ± 1.2
Cement		205 ± 18	32.0 ± 4.9	15.0 ± 2.4

Activity concentration index (ACI) for tested granite aggregate types in function of the particle size



ACI values of cement mortars with/without granite aggregates

Activity concentration of ^{40}K, ^{214}Pb and ^{212}Pb and ACI of cement mortars made with granite and siliceous aggregates.

Mortar	Activity concentration (Bq kg^{-1})			Gamma activity concentration index (ACI)
	^{40}K	^{214}Pb (^{226}Ra)	^{212}Pb (^{232}Th)	
CEM I 52.5R & (0-4 mm) Coruña	685 ± 59	116 ± 18	41.6 ± 7.1	0.822 ± 0.070
CEM I 52.5R & (0-4 mm) Vigo	608 ± 52	75 ± 11	59 ± 10	0.748 ± 0.091
CEM I 52.5R & (0-4 mm) Lugo	633 ± 55	60.1 ± 9.1	89 ± 15	0.8565 ± 0.0091
CEM I 52.5% & (0-2mm) siliceous aggregate	130 ± 11	7.3 ± 1.2	8.9 ± 1.4	0.1120 ± 0.0086

Conclusions

1. All granite aggregates gave ACI values greater than 1, and therefore would not meet the requirements established in European Directive 2013/59/EURATOM

2. The aggregate particle size affects directly to the activity concentration of ^{40}K, ^{232}Th and ^{226}Ra.

3. Portland cement acts a thinner constituent in mortars and concretes allowing to the granite aggregates to be used in mentioned building materials.

4. The additive effect of all the mortar constituents on the activity concentration of all the radioisotopes has been established.

5. The dilution of the aggregate in the mix leads to gamma activity concentration index values <1. Thus, the granitic aggregates can be used in mortars.

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