Assessment of long-lived residual radioisotopes in cement induced by neutron radiation

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- Goal of the research
- Materials
- Testing methods
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Motivation

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- All stages of Nuclear Power Plant (NPP) operation and maintenance are directly related
- Decommissioning is about 1/3 of the entire life of the NPP structure and is heavily influenced by design
- During the decommissioning of NPP, a significant amount of cement based composites should be disposed as radioactive waste
- The potential activation of elements in concrete should be minimalize
- The concrete constituents should be carefully selected to contain as low as possible concentration of dominant long-lived residual radioisotopes, while maintaining all the required parameters related to strength and durability

Goal of the reserach

After decommissioning, the massive concrete should be treated as radioactive waste

The use of low-activation constituents could effectively reduce radioactivity of concrete

An assessment of long-lived residual radioisotopes in cement intended for shielding concrete in nuclear power plant

Europium-152, $T_{1/2}$ = 13.54 years

Europium-154, $T_{1/2}$ = 8.59 years

Cobalt-60, $T_{1/2}$ = 5.28 years

Caesium-134, $T_{1/2}$ = 2.07 years

Materials

Four Portland cements were taken into consideration.

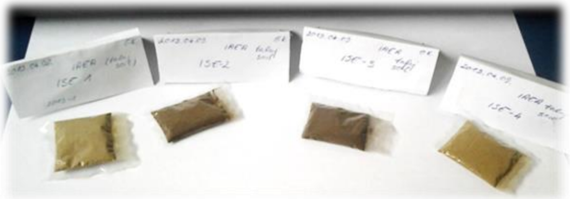
XRF method, wt %

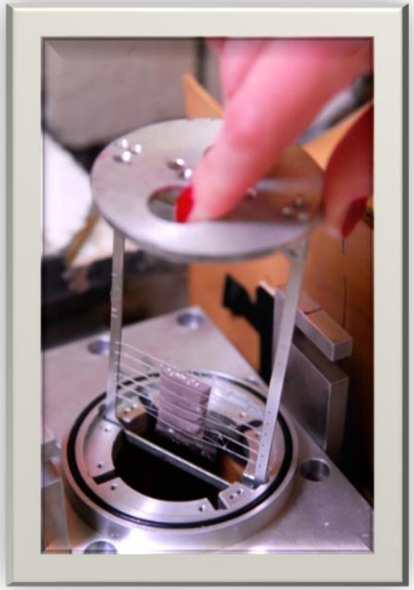
Constituent	C1	C2	C3	C4
	CEM I 42.5R	CEM I 52.5R	CEM I 42.5R	CEM I 52.5R
SiO_2	19.03	19.42	19.43	24.40
Al_2O_3	4.84	5.45	4.84	2.11
Fe_2O_3	3.22	2.94	3.18	0.30
CaO	63.64	64.10	61.81	68.40
MgO	1.15	1.75	2.56	0.66
SO_3	2.97	3.50	3.93	2.09
Na_2O_{eq}	0.56	0.88	1.12	0.23
LOI	3.34	3.50	2.67	1.22

Testing methods

Concentrations of major and trace elements in cements were determined by Prompt Gamma Activation Analysis (PGAA)

2-3 g for PGAA





- The thickness of the specimens was kept at the lowest possible (2-3 mm) to minimize the effect of self-shielding and self-absorption
- The irradiation and the collection of prompt-gamma spectra took place in one hour

PGAA

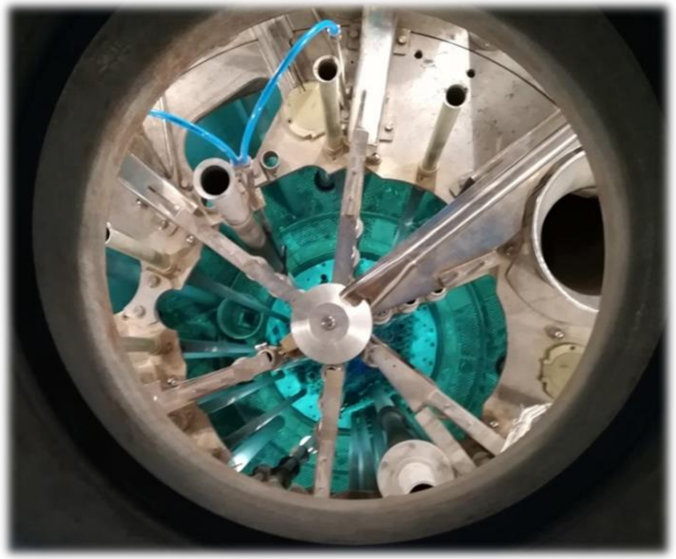
Major elements (11) and some trace (~5):

- Si, Ti, K, Fe and also H
- B, Cl, and Gd

Testing methods

The assessment of trace elements with large activation cross section using Neutron Activation Analysis (NAA) was performed.

150 mg for NAA



NAA
Trace elements (~20):
- 14 long-lived radionuclides

- Each cement was heat-sealed in high-purity quartz. The quartz ampules were wrapped in aluminum foil and encapsulated in an aluminum container
- The irradiation process took 3 hours and was performed in a channel of the Budapest Research Reactor
- The thermal equivalent neutron flux in the irradiation channel was 1.86×10^{13} cm^2 s^{-1}

Results

The major elemental composition of cement measured by PGAA, wt. %

Element	C1	C2	C3	C4
Ca	45.00	45.00	44.00	47.88
Si	9.70	9.40	9.30	11.99
Al	3.00	2.80	2.60	1.75
Fe	1.71	2.00	2.10	0.23
S	1.21	1.47	1.72	0.86
Mg	1.30	1.20	1.70	0.00
K	0.83	0.97	0.99	0.11
Cl	0.09	0.33	0.32	0.03
Mn	0.58	0.27	0.49	0.01
Na	0.17	0.25	0.38	0.00
H	0.17	0.19	0.17	0.14
Ti	0.17	0.19	0.17	0.04
B	0.06	0.07	0.05	0.01

Results

23 radionuclides were detected using the Neutron Activation Analysis

Caesium-134, $T_{1/2}$ = 2.07 years

Cobalt-60, $T_{1/2}$ = 5.28 years

Europium-152, $T_{1/2}$ = 13.54 years

The concentrations of radionuclides in cements induced by NAA, µg/g

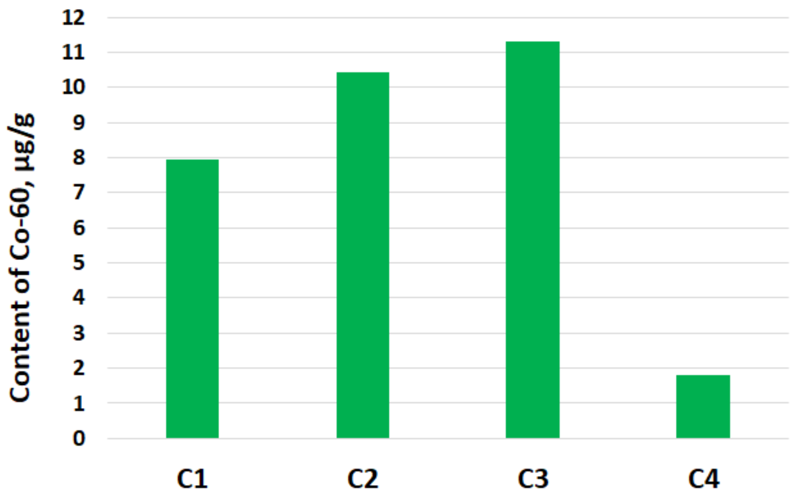
Isotope	Half-life, days	C1	C2	C3	C4
K-42	0.5	3 787.00	8 525.00	9 079.00	891.00
Zn-69m	0.6	558.50	845.30	401.80	16.56
As-76	1.1	5.27	4.45	12.84	0.80
Br-82	1.5	17.02	5.41	10.02	0.00
La-140	1.7	15.14	16.35	15.12	7.57
Sm-153	1.9	1.97	2.30	1.92	0.80
Yb-175	4.2	1.18	1.19	1.15	0.55
Ca-47	4.5	491 900.00	484 800.00	483 800.00	465 300.00
Nd-147	11.0	16.30	0.00	15.59	0.00
Ba-131	11.5	291.70	185.90	185.50	112.40
Rb-86	18.6	45.21	38.82	40.94	0.00
Cr-51	27.7	62.35	111.10	104.10	16.00
Ce-141	32.5	31.54	26.65	33.49	8.72
Hf-181	42.4	1.82	2.25	2.38	0.99
Fe-59	44.5	18 250.00	21 910.00	21 000.00	2 261.00
Sb-124	60.2	8.18	8.56	13.08	0.52
Sr-85	64.9	1 057.00	827.30	810.10	1 253.00
Tb-160	72.3	0.38	0.36	0.40	0.16
Sc-46	83.8	6.03	5.93	5.66	1.29
Ta-182	114.4	0.40	0.53	0.47	0.14
Cs-134	751.9	3.40	1.63	1.69	0.00
Co-60	114.4	7.93	10.44	11.30	1.79
Eu-152	4941.0	0.61	0.56	0.60	0.25

IPPT PAN

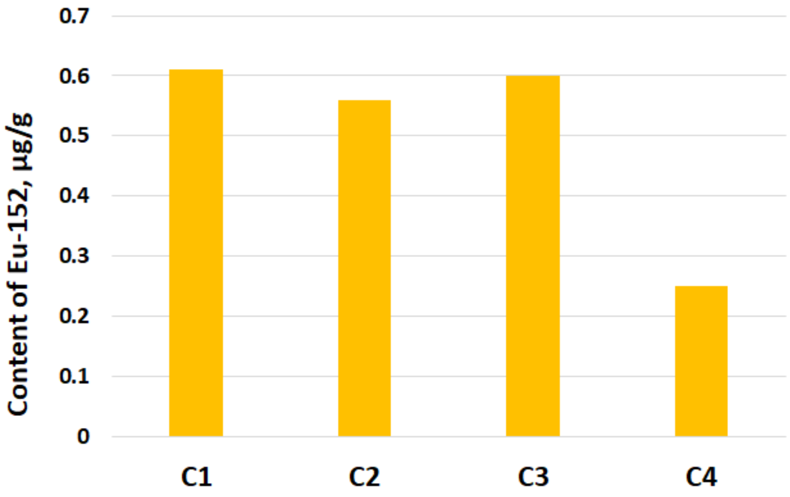
Results

Content of ^{60}Co, ^{152}Eu and ^{134}Cs in Portland cements

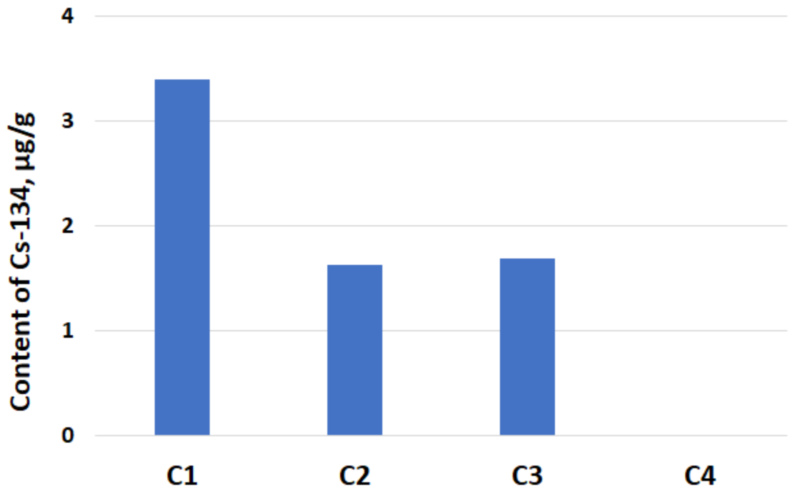
Cobalt 60



Europium 152

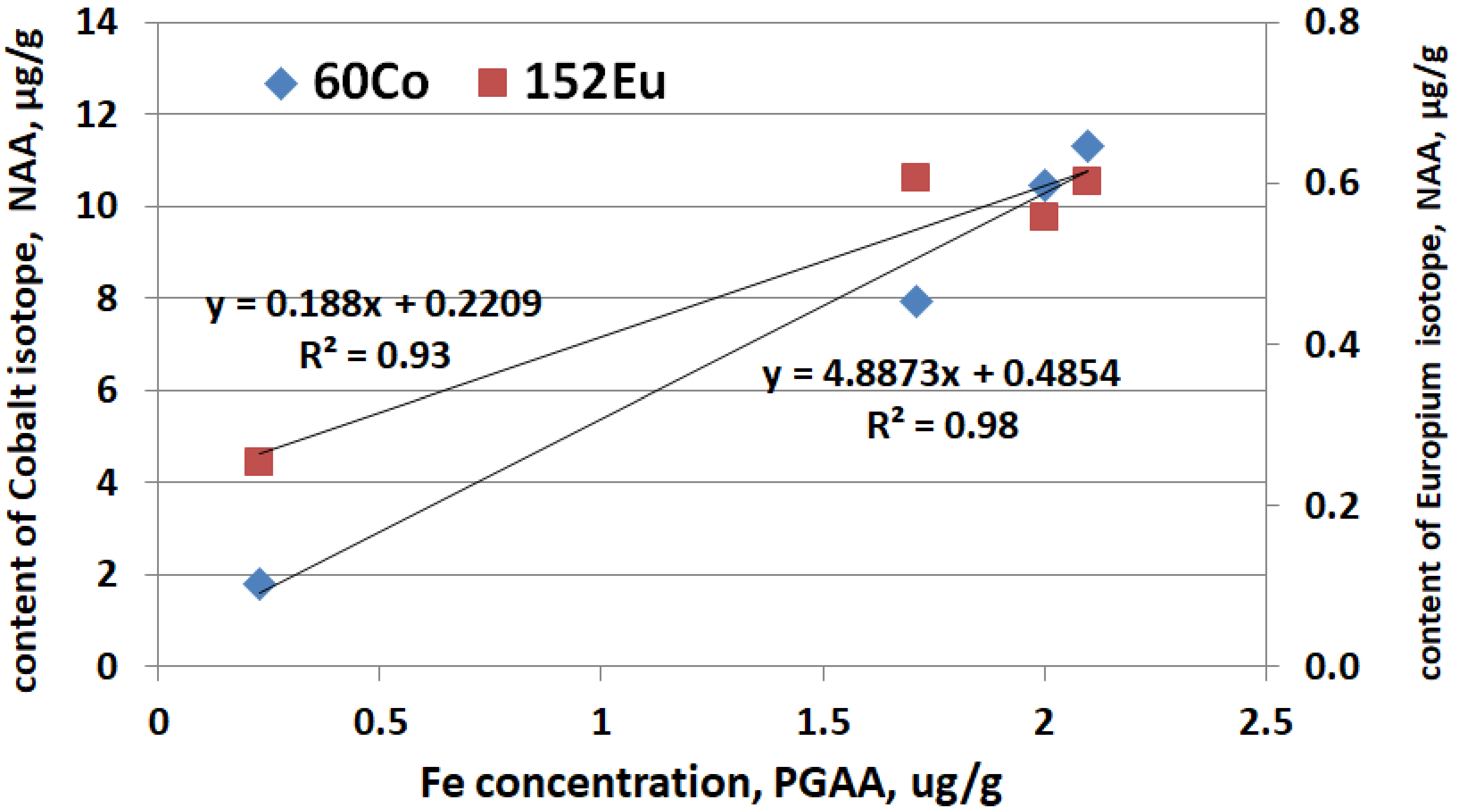


Caesium 134



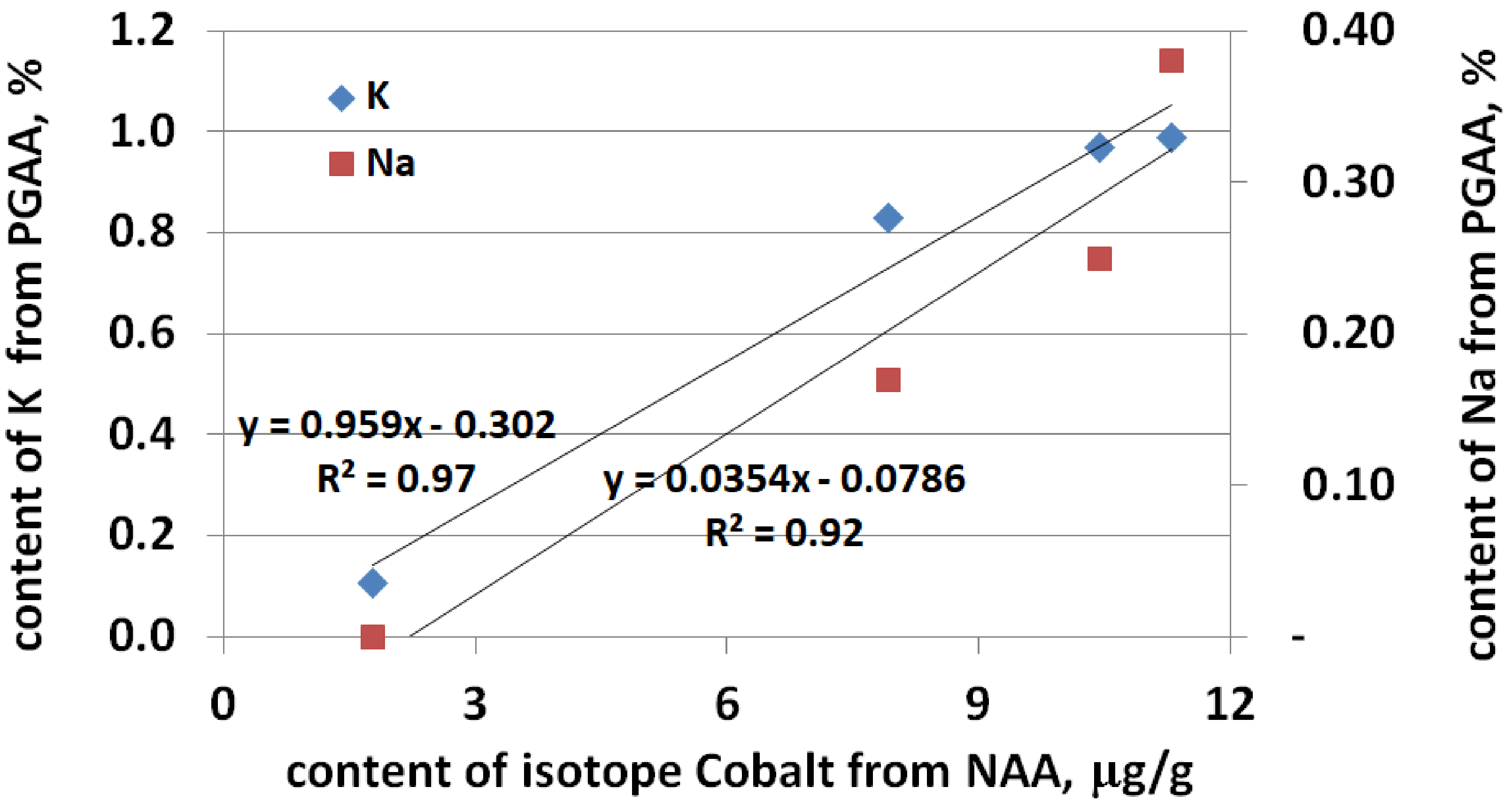
Results

Content of Europium and Cobalt isotopes vs Fe concentration in cement



Results

Concentration of the Cobalt isotope vs alkalis content in cement



Conclusions

- Application of prompt gamma activation analysis and neutron activation analysis allows for a pre-selection of low-activation cement based on its elemental composition.
- Three major long-lived residual radioisotopes were found in cements: ^{152}Eu, ^{60}Co and ^{134}Cs.
- For the considered range of Portland cements CEM I the content of Cobalt was proportional to the content of alkalis and the content of Europium as well as Cobalt concentartion was proportional to the total content of Ferrum.
- The cement with the low content of Fe and Al (white cement) was characterized by the lowest content of long-lived residual radioisotopes, making it suitable for low-activation concrete.

Future research

- Investigations on special aggregate and concrete activation are currently ongoing.
- New criterion for concrete mix design to reduce radioactive waste will be considered.
- Results of concrete constituent activation will be used to facilitate a multicriteria selection of constituents for shielding concrete.

Thank you for your attention !!!

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The research was supported by:

- Polish National Centre for Research and Development (Project V4-Korea/2/2018)
- National Research, Development and Innovation Fund of Hungary (NN_17 V4-Korea funding scheme, project no. 127102)

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