Institute of Fundamental Technological Research
Polish Academy of Sciences

# LOW - ACTIVATION CONCRETE
# preliminary results

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# Outline

- Goal of the research
- Literature review
- Research plan: materials
- Preliminary results
- Expected results

# Goal of the research

**Estimation of the activation of concrete constituents: cement and aggregate**

↓

**Assessment of the ability to activate shielding concrete**

↓

**Reduction of the potential activity of shielding concrete due to the permissible level at the liquidation of Nuclear Power Plant (NPP)**

# Literature review – Japan 1

**About 300 samples of concrete materials in Japan were collected.**

**136 aggregates, 97 fine aggregates, 66 cements, and 7 concrete pieces were collected.**

| | |
|---|---|
| Aggregates | granite 6, gneiss 2, crystalline schist 5, sandstone 23, hard sandstone 5, sand 27, shale 6, breccia 1, quartzite 2, slate 3, quartz porphyry 3, porphylite 2, andesite 20, porphyry 1, trachyte 1, diabase 2, basalt 7, amphiborite 4, gabbro 4, peridotite 4, serpentnite 1, limestone 7 |
| Cement | normal portland 32, moderate heat 10, fly ash 13, aluminous 1, desulfurization from exhaust gas 2, Portland blast-furnace slag 8 |

**Suzuki, A., Iida, T., Moriizumi, J., Kameyama, T., Sakuma, Y., Takada, J., Yamasaki, K., & Yoshimoto, T. (2000). Quantitative measurements of trace elements with large activation cross section for concrete materials in Japan. IRPA-10 Proceedings of the 10th international congress of the International Radiation Protection Association on harmonization of radiation, human life and the ecosystem, (p. 1v). Japan: Japan Health Physics Society.**

# Literature review – Japan 1

**Nineteen nuclides were detected with activation analysis of the samples.**

**Cobalt-60, Europe-152 and Cesium-134 were investigated.**

| nuclide | Half life (y) |
|---|---|
| Na-24 | 0.002 |
| Ca-47 | 0.012 |
| Sc-46 | 0.229 |
| Cr-51 | 0.076 |
| Mn-54 | 0.856 |
| Fe-59 | 0.122 |
| Co-60 | 5.270 |
| Se-75 | 0.328 |
| Rb-86 | 0.051 |
| Sb-122 | 0.007 |
| Cs-134 | 2.062 |
| Ba-131 | 0.036 |
| La-140 | 0.005 |
| Sm-153 | 0.005 |
| Eu-152 | 13.300 |
| Tb-160 | 0.197 |
| Yb-175 | 0.066 |
| Lu-177 | 0.018 |
| Hf-181 | 0.116 |

Table 4 The concentration of $^{60}Co$, $^{152}Eu$ and $^{134}Cs$ forming nuclides in each concrete materials.

| Material | No.of Samples | **Co-60** [ppm] (range) | S.D. | R | **Eu-152** [ppm] (range) | S.D. | R | **Cs-134** [ppm] (range) | S.D. | R |
|---|---|---|---|---|---|---|---|---|---|---|
| Granite | 6 | 7.6 (4.2-11.7) | 3.1 | 0.96 | 0.91 (0.43-1.27) | 0.30 | 0.98 | 3.9 (2.1-6.9) | 1.9 | 0.94 |
| Gneiss | 2 | 9.7 (9.6-9.8) | 0.1 | 1.0 | 0.86 (0.74-0.97) | 0.17 | 1.0 | 5.3 (4.5-6.1) | 1.2 | 1.0 |
| Crystalline schist | 5 | 12.4 (7.1-19.4) | 4.9 | 0.98 | 1.2 (0.68-1.8) | 0.51 | 0.93 | 3.3 (1.9-5.0) | 1.8 | 0.93 |
| Hard sandstone | 5 | 14.2 (6.9-9.4) | 10.2 | 0.84 | 0.75 (0-1.1) | 0.46 | 0.87 | 4.4 (0-7.5) | 3.3 | 0.97 |
| Sandstone | 23 | 10.2 (2.5-47.7) | 7.1 | 0.79 | 0.90 (0-2.3) | 0.39 | 0.92 | 3.9 (0-11.6) | 2.2 | 0.93 |
| Sand | 27 | 6.7 (0.72-19.3) | 4.6 | 0.95 | 0.73 (0-1.9) | 0.49 | 0.97 | 2.5 (0-8.7) | 1.5 | 0.90 |
| Shale | 6 | 9.7 (5.2-12.6) | 2.8 | 0.95 | 1.1 (0.9-1.6) | 0.43 | 0.99 | 6.7 (2.8-13.5) | 4.5 | 0.92 |
| Breccia | 1 | 19.4 | | | 1.2 | | | 1.7 | | |
| Doromite | 1 | 0.45 | | | 0.13 | | | 0 | | |
| Quartzite | 1 | 4.2 | | | 0 | | | 0.90 | | |
| Slate | 3 | 11.1 (10.5-11.6) | 0.8 | 1.0 | 1.5 (1.1-1.7) | 0.30 | 0.94 | 6.5 (6.2-7.5) | 0.88 | 0.96 |
| Quartz porphyry | 2 | 6.5 (6.3-6.7) | 0.27 | 1.0 | 1.0 (0.9-1.2) | 0.16 | 1.0 | 7.9 (7.5-8.3) | 0.60 | 1.0 |
| Porphyrite | 1 | 31.5 | | | 3.1 | | | 0 | | |
| Andesite | 20 | 15.4 (0.45-43.2) | 11.7 | 0.92 | 1.2 (0.13-3.2) | 0.52 | 0.88 | 4.5 (0-18.3) | 4.0 | 0.90 |
| Porphyry | 1 | 2.5 | | | 0.58 | | | 1.9 | | |
| Trachyte | 1(2) | 0.66 | | | 0 | | | 1.6 | | |
| Diabase | 1 | 44.6 | | | 1.6 | | | 0 | | |
| Basalt | 7 | 26.6 (0-67.3) | 23.1 | 0.96 | 1.8 (0.77-3.1) | 0.75 | 0.99 | 1.6 (0-6.9) | 2.2 | 0.84 |
| Amphibolite | 4 | 15.4 (0-43.1) | 15.9 | 0.93 | 0.97 (0-1.92) | 0.57 | 0.93 | 2.0 (0-8.7) | 1.2 | 0.96 |
| Gabbro | 4 | 88.1 (42.8-127) | 39.8 | 0.93 | 0.28 (0-0.41) | 0.13 | 0.91 | 0.53 (0-2.1) | 0.85 | 0.79 |
| Peridotite | 4 | 107 (106-113) | 4.3 | 0.94 | 0.11 (0-0.43) | 0.17 | 0.79 | 0 | 0 | 0 |
| Serpentnite | 1 | 41.7 | | | 1.4 | | | 2.7 | | |
| Limestone | 7 | 1.8 (0-7.2) | 2.5 | 0.81 | 0.17 (0-0.75) | 0.27 | 0.85 | 1.4 (0-6.7) | 2.3 | 0.74 |
| Average in aggregate | | 17.3 | 19.1 | 0.78 | 0.93 | 0.61 | 0.86 | 3.2 | 2.7 | 0.93 |
| Portland cement | 32 | 9.4 (5.4-14.2) | 3.0 | 0.92 | 0.68 (0.39-0.94) | 0.21 | 0.89 | 5.9 (1.6-15.1) | 2.9 | 0.95 |
| Moderate heat | 10 | 23.9 (6.1-51.5) | 16.3 | 0.95 | 1.17 (0.49-6.4) | 1.2 | 0.63 | 9.8 (2.9-13.8) | 8.7 | 0.82 |
| Fly ash | 13 | 16.5 (6.6-31.6) | 7.2 | 0.95 | 2.4 (0.94-4.6) | 0.83 | 0.94 | 7.4 (4.1-15.7) | 3.6 | 0.90 |
| Aluminous | 1 | 3.4 | | | 1.2 | | | 1.4 | | |
| Portland blast-furnace slag | 8 | 5.3 (2.7-9.0) | 1.9 | 0.97 | 1.6 (1.3-2.3) | 0.30 | 0.89 | 3.9 (1.6-10.8) | 2.8 | 0.87 |
| Desulfurrization from exhaust gas | 2 | 0.4 (0.15-0.60) | 0.33 | 1.0 | 0.06 (0.05-0.07) | 0.01 | 1.0 | 0.05 (0-1.0) | 0.08 | 1.0 |
| Average in cement | | 12.1 | 8.6 | 0.89 | 1.2 | 0.92 | 0.85 | 6.3 | 4.39 | 0.87 |

S.D.: Standard deviation, Relative coefficient: deviation of their provability distribution from normal distribution

# Literature review – Japan-2

- The concentration of Co-60 formed in basic rock (gabbro, basalt) was the highest.
- The concentration **of Co-60 formed in carbonate rock was the lowest**.
- Weathered aggregates had lower concentration of 60-Co, because cobalt is easy to dissolve in water.

- The concentration of Eu-152 formed in intermediate rock (diorite, andesite) was the highest.
- The concentration of **Eu-152 formed in carbonate rock was the lowest**.
- Europium in the aggregate was a little decreased by weathering.

- The combination of the lowest Co-60, Eu-152 and Cs-134 concentrations in the concrete was achieved with limestone as aggregate and white Portland cement produced in particular places.

  These concentrations were about 1/25–1/100 of the mean value in ordinary concrete or less.

**A.Suzuki, T.Iida, J.Moriizumi, T.Kameyama, Y.Sakuma, J.Takada, K.Yamasaki and T.Yoshimoto, Trace Elements with Large Activation Cross Section in Concrete Materials in Japan, Journal of Nuclear Science and Technology, 2001, 38:7, 542-550**

# Literature review – Japan-2

- Though the serpentinite concrete was the most excellent with respect to neutron shielding effect, it had very high concentrations of Co-60.

  Therefore, in the viewpoint of reducing the radioactive waste, serpentinite concrete was not suitable for biological shield.

- Co-60, Eu-152 and Cs-134 formed in aggregates were decreased by weathering, however, these aggregates were not suitable for concrete because of their lower strength.

- The fly ash cement was not suitable for the biological shielding concrete, because fly ash is easy to activate, and the natural radionuclides concentration in fly ash are also very high.

**A.Suzuki, T.Iida, J.Moriizumi, T.Kameyama, Y.Sakuma, J.Takada, K.Yamasaki and T.Yoshimoto, Trace Elements with Large Activation Cross Section in Concrete Materials in Japan, Journal of Nuclear Science and Technology, 2001, 38:7, 542-550**

# Literature review – Japan-2

- According to the recommendations of the International Atomic Energy Agency, the material is classified as radioactive waste due to its **clearance level (*CL*)** of each radioactive nuclide ($C_i$ [Bq/g]).

  CL Co-60 = 0.4 Bq/g

  CL Eu-152 = 0.4 Bq/g

  CL Cs-134 = 0.5 Bq/g

- To classify **radioactive/non-radioactive materials, $\Sigma(D_i/C_i)$** is calculated, where “Di” indicates concentration of each residual radioisotope and ($_i$) indicates each radioisotope.
- When the **$\Sigma(D_i/C_i)$ of waste is less than 1**, the waste can be treated as non-radioactive waste.


- **IAEA, *Clearance Levels for Radionuclides in Solid Materials*, IAEA-TECDOC-855, IAEA, Vienna, (1996)**
- **Kimura, K., Hasegawa, A., Hayashi, K., Uematsu, M., Ogata, T., Tanosaki, T., Yoshino, R., Sato, M., Saito, M., Kinno, M. "Development of Low-Activation Design Method for Reduction of Radioactive Waste Below Clearance Level." *Proceedings of the 16th International Conference on Nuclear Engineering*. *Volume 1,* Orlando, Florida, USA. May 11–15, 2008. pp. 617-626. ASME**

# Literature review – Japan-3

When the Σ(Di/Ci) of waste is less than 1, the waste can be treated as non-radioactive waste.

$\Sigma^3 Di/Ci = D_{Eu\text{-}152}/C_{Eu\text{-}152} + D_{Eu\text{-}154}/C_{Eu\text{-}154} + D_{Co\text{-}60}/C_{Co\ \text{-}60}$

Di: Concentration of radionuclide of Eu-152, Eu-154 and Co-60 induced under $2.0\times10^{5}$ n $cm^{-2}sec^{-1}$ thermal neutrons, 40 years of operation, 6 years of cooling.

Ci: Clearance level referring CL for Eu-152, Eu-154 and Co-60.

International Atomic Energy Agency, Application of the Concepts of Exclusion, Exemption and Clearance Safety Guide No.RS-G-1.7, 2004

**Ken-Ichi Kimura, Akira Hasegawa, Katsumi Hayashi, Mikio Uematsu, Tomohiro Ogata, Takao Tanosaki, Ryoetsu Yoshino, Mituru Sato, Minoru Saito and Masaharu Kinno, Development of Low-Activation Design Method for Reduction of Radioactive Waste below Clearance Level, 2008**

# Literature review – Japan-3

D- Concentration of radionuclide
C - Clearance level

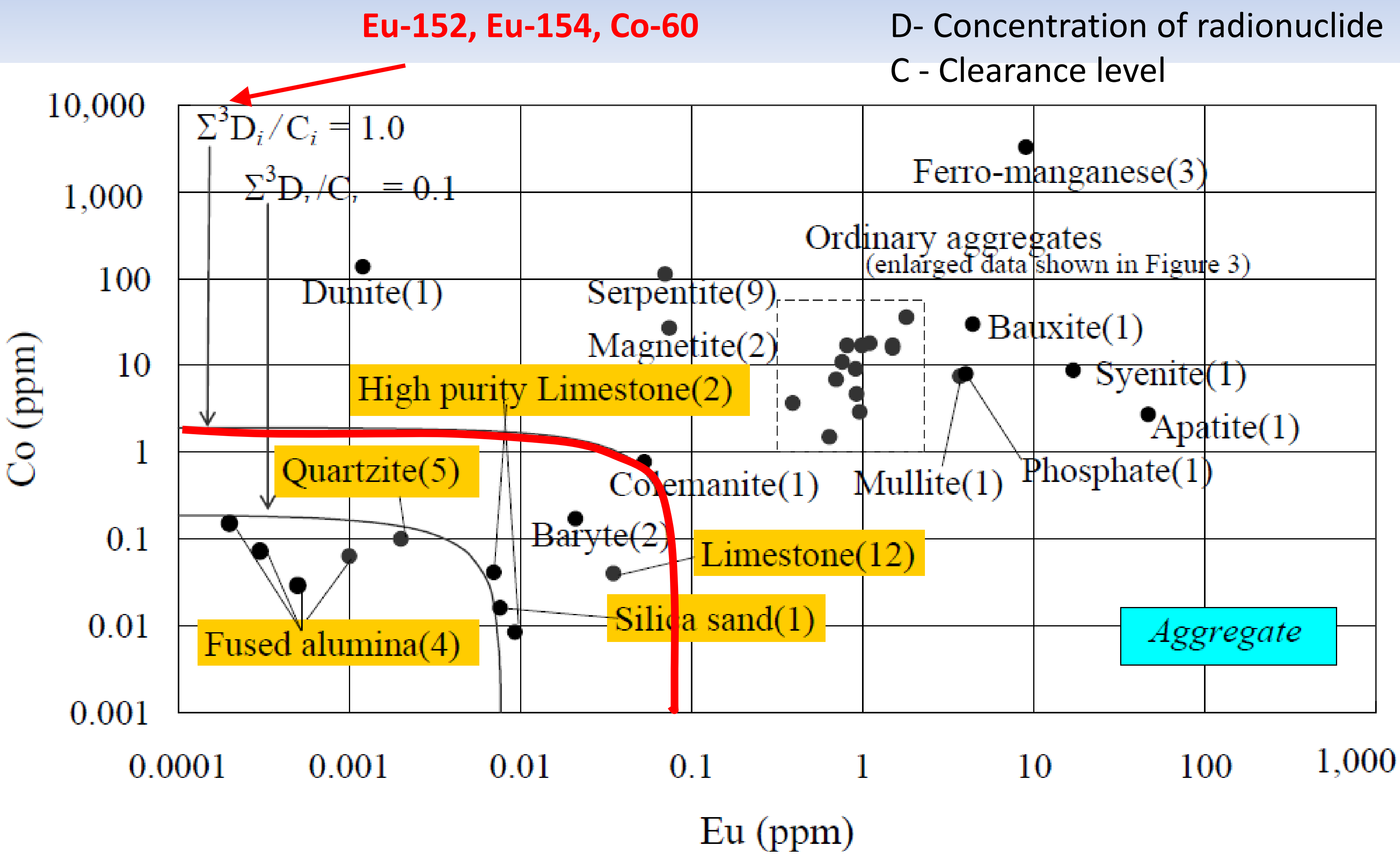



**Figure 3 Distribution of quantities for Eu and Co in aggregates with enlargement of ordinary aggregates** (Numbers in parentheses indicate the number of specimens )

# Literature review – Japan-3

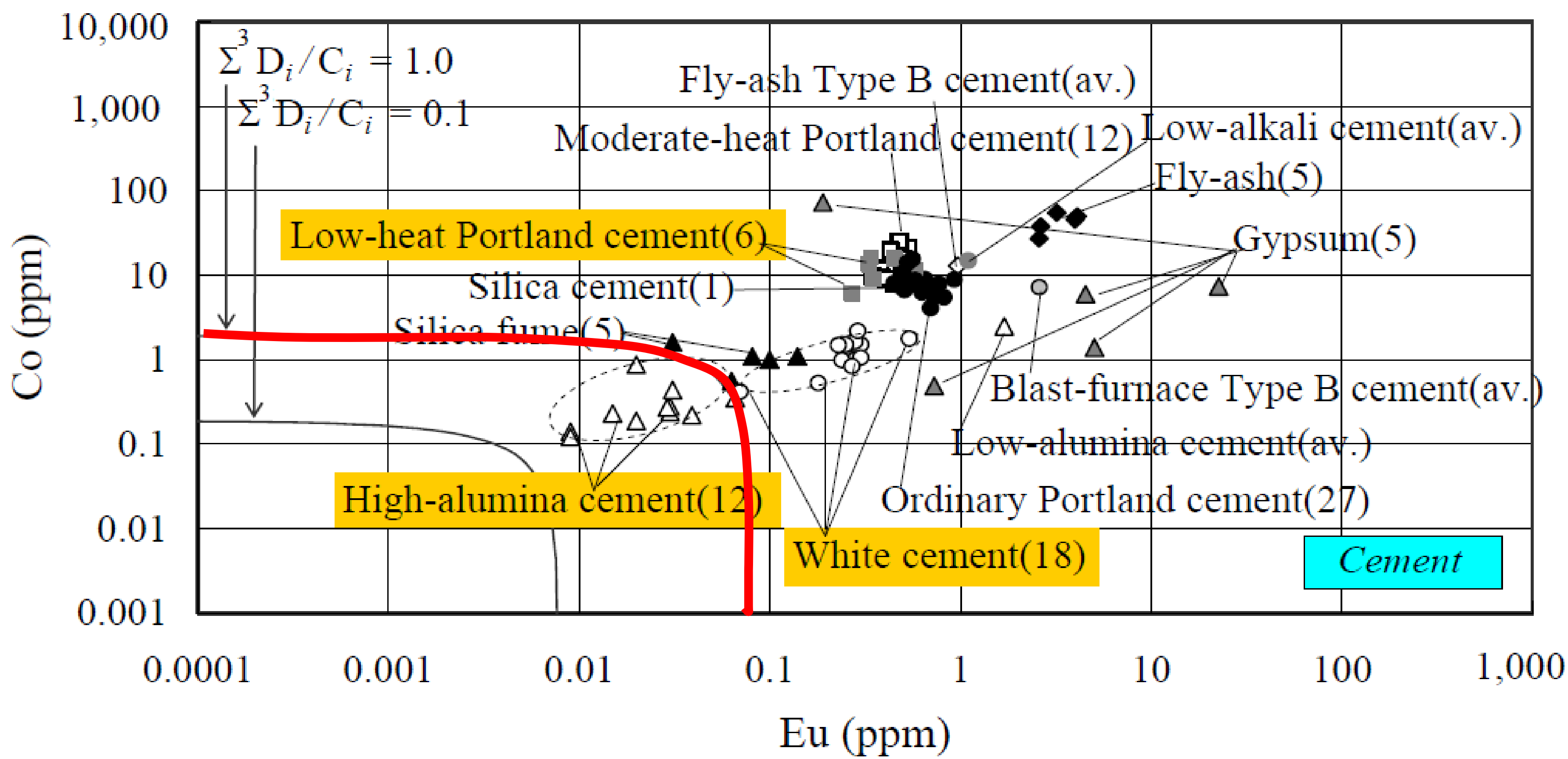
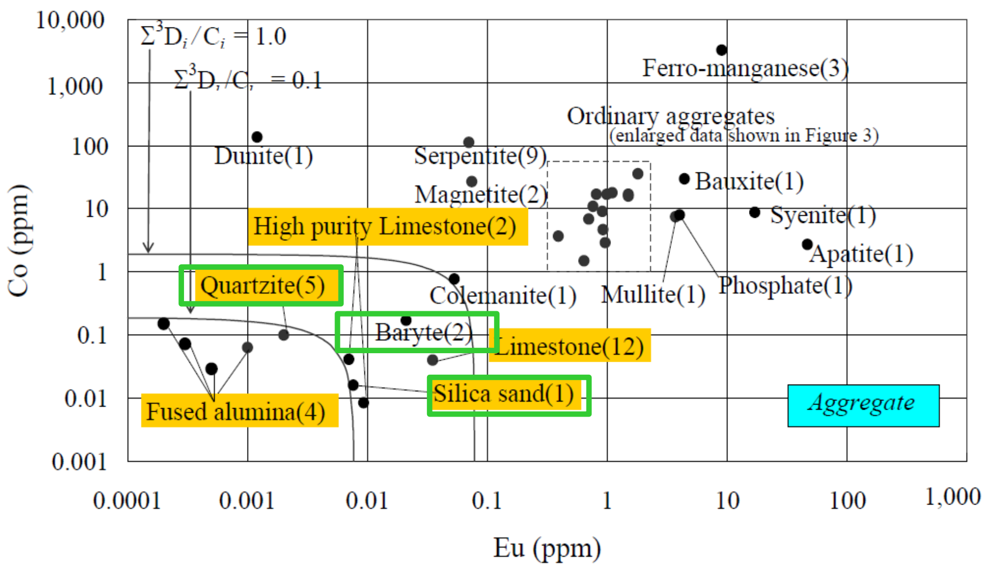



**Figure 5 Distribution of quantities for Eu and Co in cement**
(Numbers in parentheses indicate the number of specimens)

# Research plan: materials

## Estimation of the activation of concrete constituents: cement and aggregate



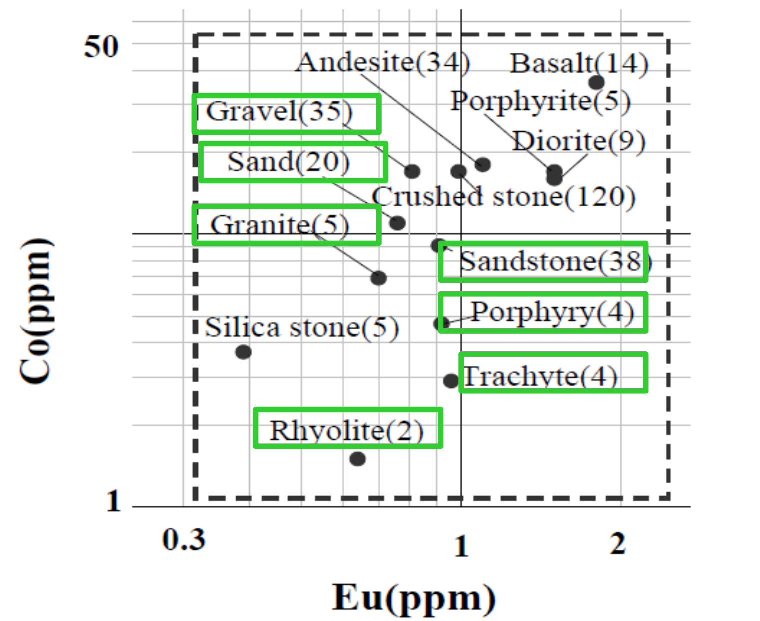
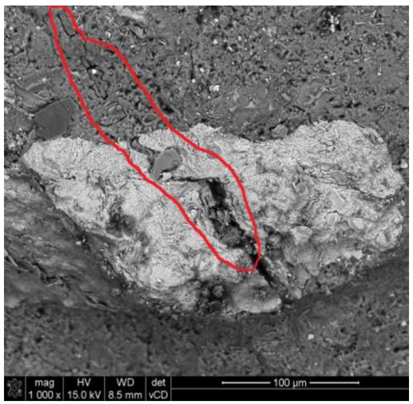
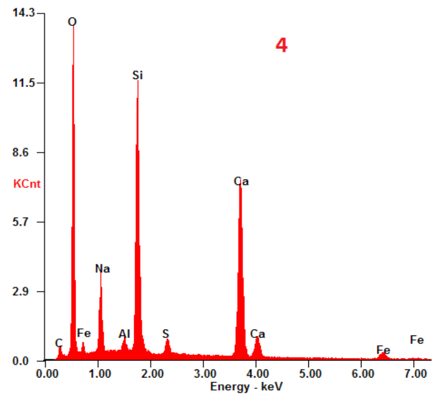



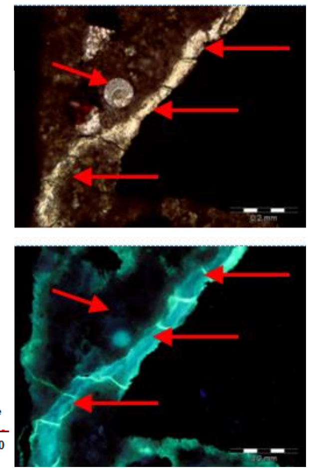

**Figure 4 Enlargement of the distribution for Ordinary aggregates from Figure 2**
(Numbers in parentheses indicate the number of specimens)

# Research plan: materials

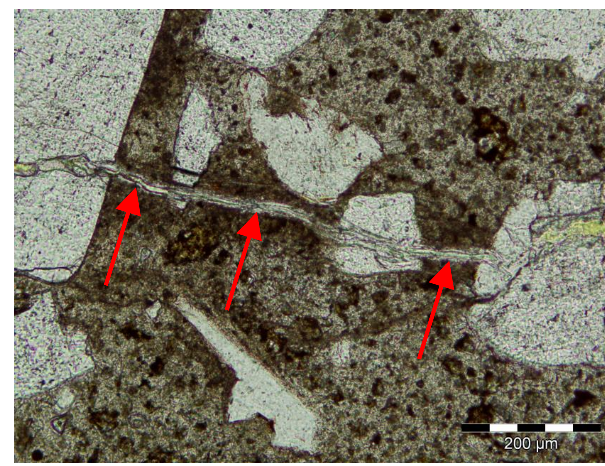
Evaluation of the threat of premature degradation due to Alkali-Aggregate Reaction in shielding concrete



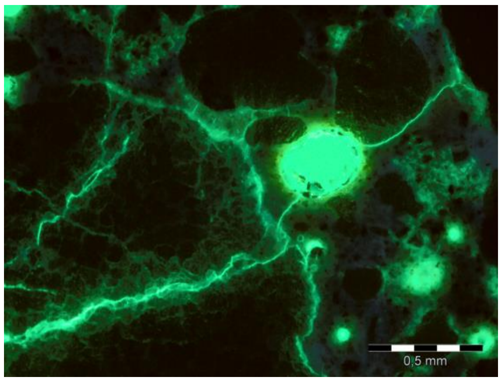
Hematite

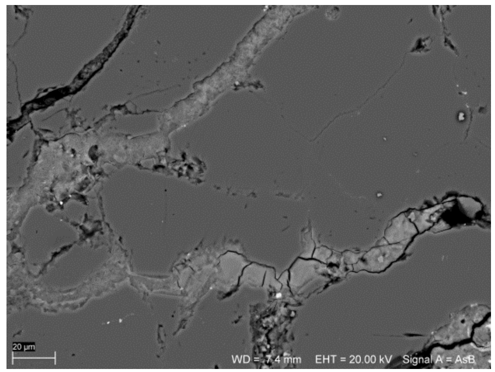


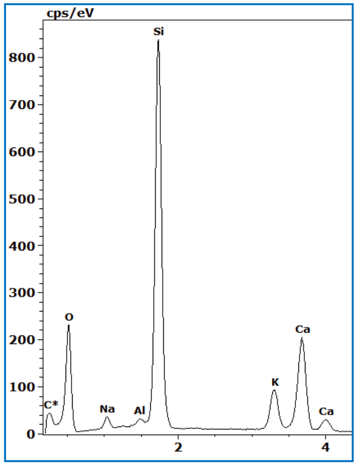

Barite

# Research plan: materials

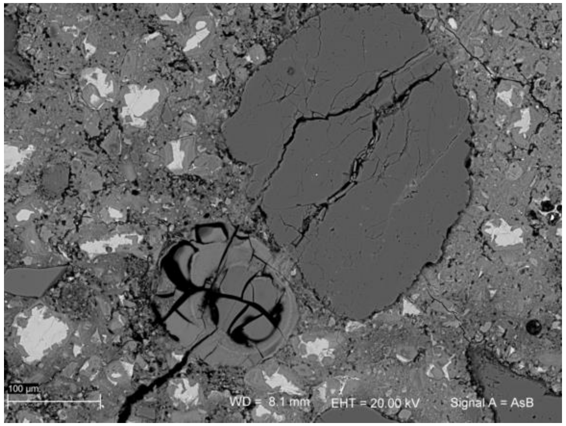
ASR - RID The alkali reactivity of domestic aggregates

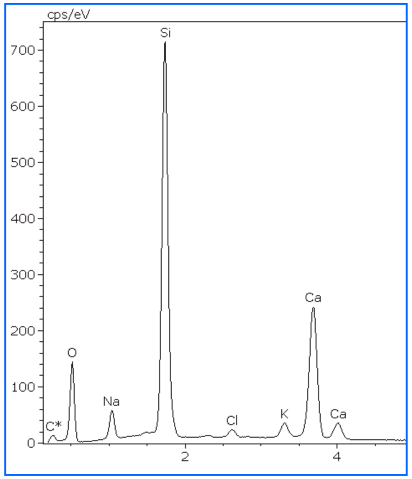



**Quartzite**




**Siliceous sand**

# Research plan: materials

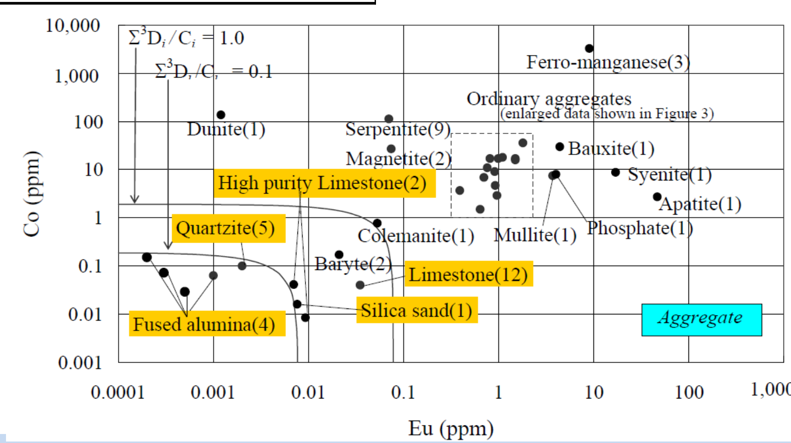
**LOW - ACTIVATION SHIELDING CONCRETE**

**ALKALI-SILICA RESISTANT SHIELDING CONCRETE**

# Estimation of the activation of concrete constituents

## Fine aggregate

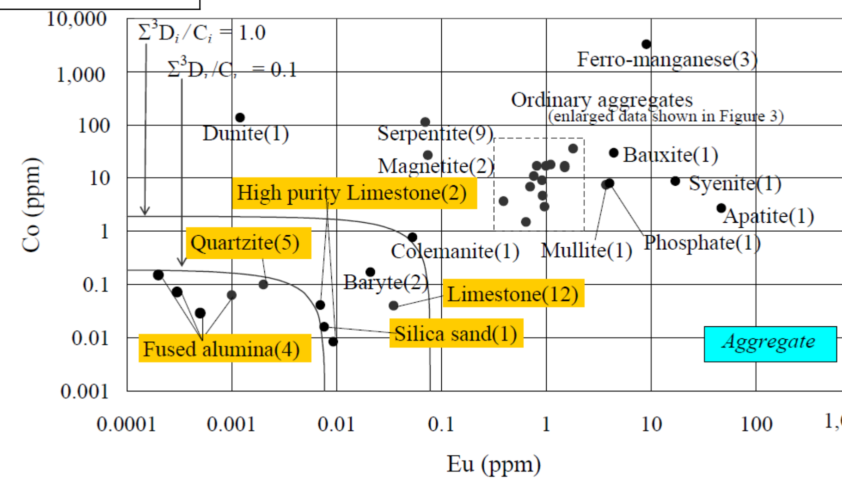
| | | | aggregate reactivity class |
|---|---|---|---|
| Siliceous sand | river | *Góra Kalwaria* | R2, highly reactive |
| | | *HB-1* | R1, moderately reactive |
| | fossil | *Borowce* | R0, non reactive |
| Limestone sand | fossil | *Miedzianka* | R0, non reactive |



# Estimation of the activation of concrete constituents

## Coarse aggregate

| | | aggregate reactivity class |
|---|---|---|
| Quartzite | *Wiśniówka* | R2, highly reactive |
| Barite | *Wolfach* | R0, non reactive |
| Granite | *Rogoźnica* | R0, non reactive |
| Limestone | *Jaźwica* | R0, non reactive |



# Estimation of the activation of concrete constituents

## Cement CEM I

**Content of $Na_2O_{eq}$**

| | | | |
|---|---|---|---|
| Portland Cement | CEM I 42,5R | Górażdże Cement (Poland) | 0.56 % |
| | CEM I 52,5R | Lafarge Małogoszcz (Poland) | 0.89 % |
| | CEM I 42,5R | Norcem - Heidelberger Zement (Norway) | 1.12 % |
| White Cement | CEM I 52,5R | Aalborg White (Denmark) | 0.23 % |

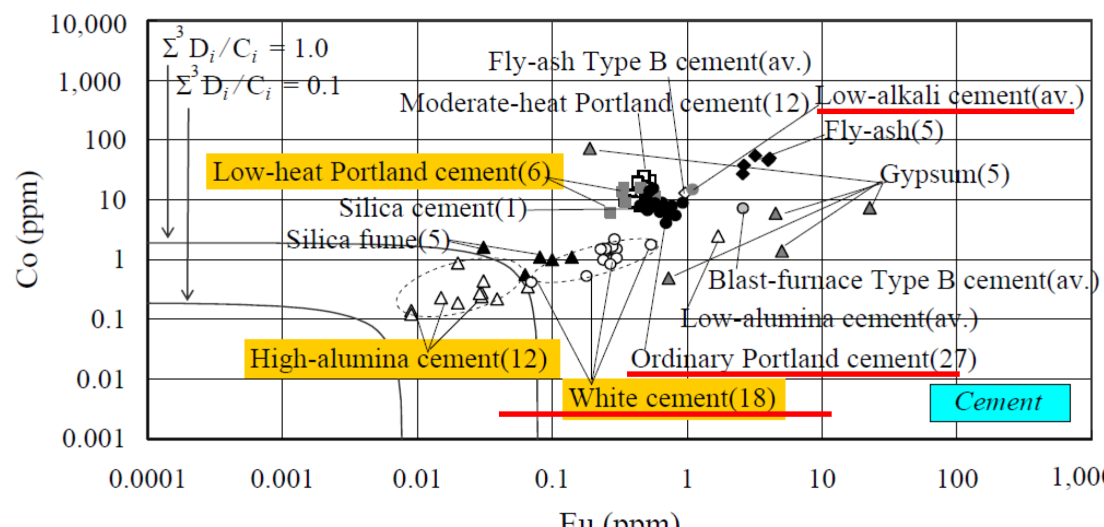
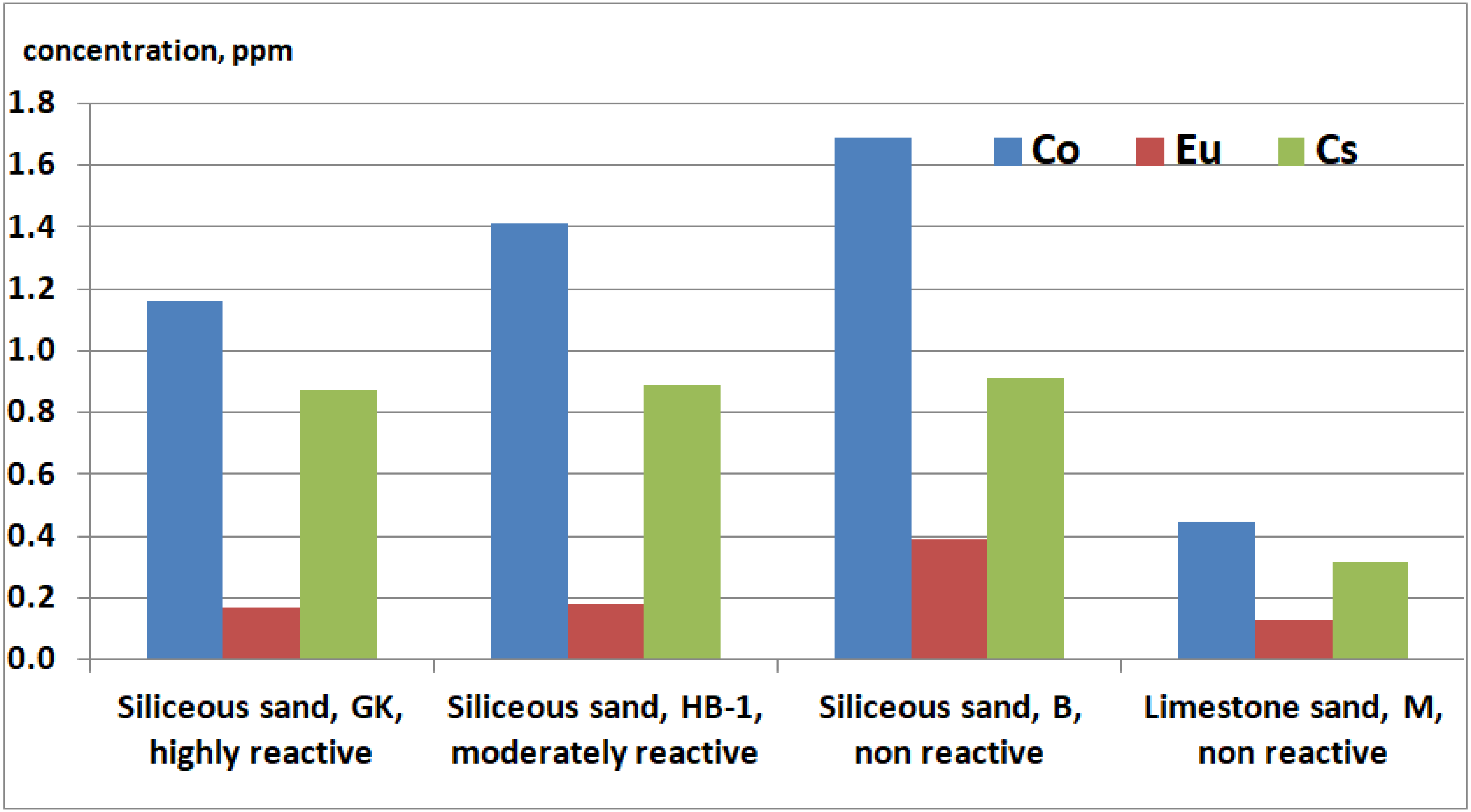


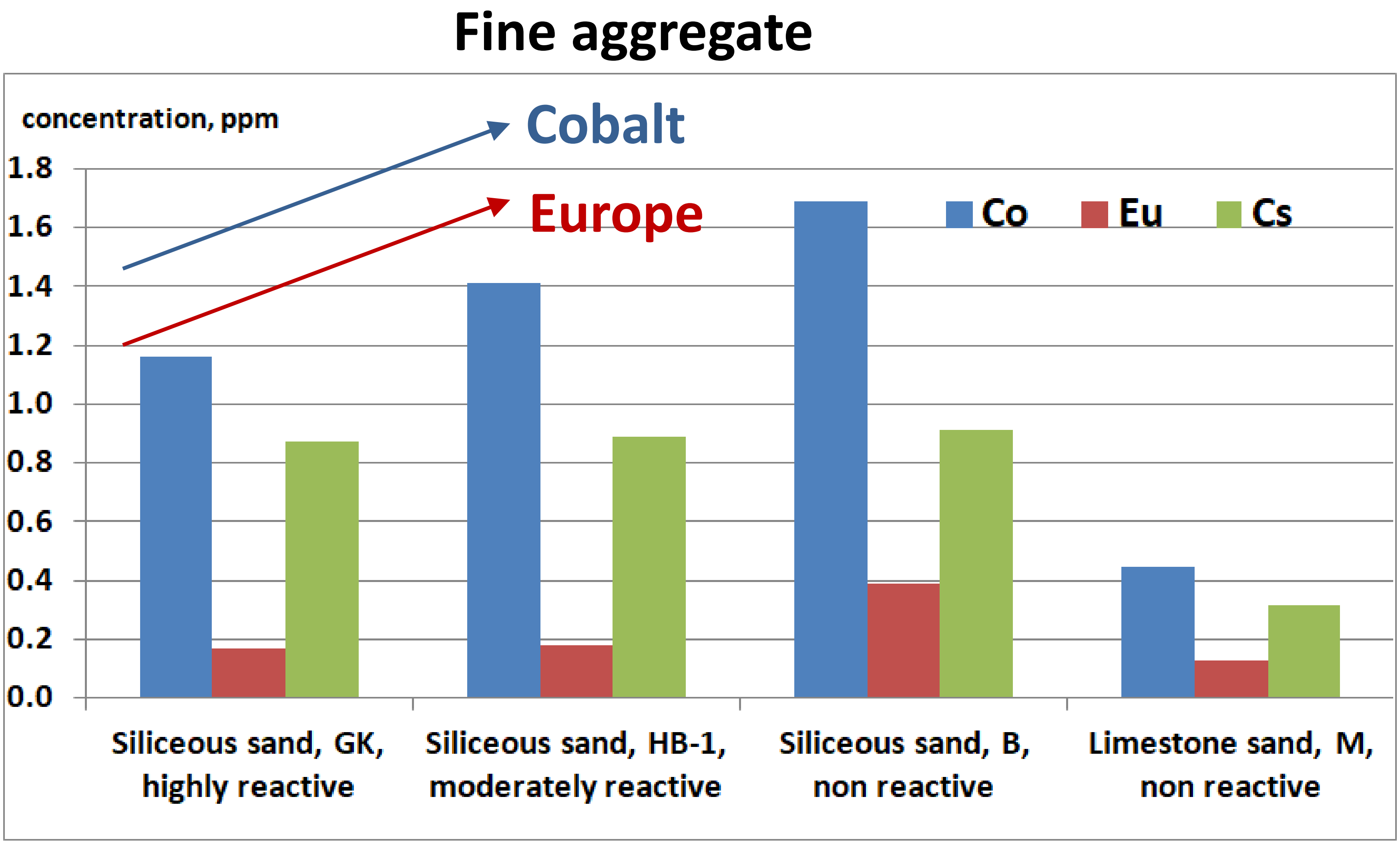

**Figure 5 Distribution of quantities for Eu and Co in cement**
(Numbers in parentheses indicate the number of specimens)

# Preliminary results: Neutron activation analysis

## Fine aggregate

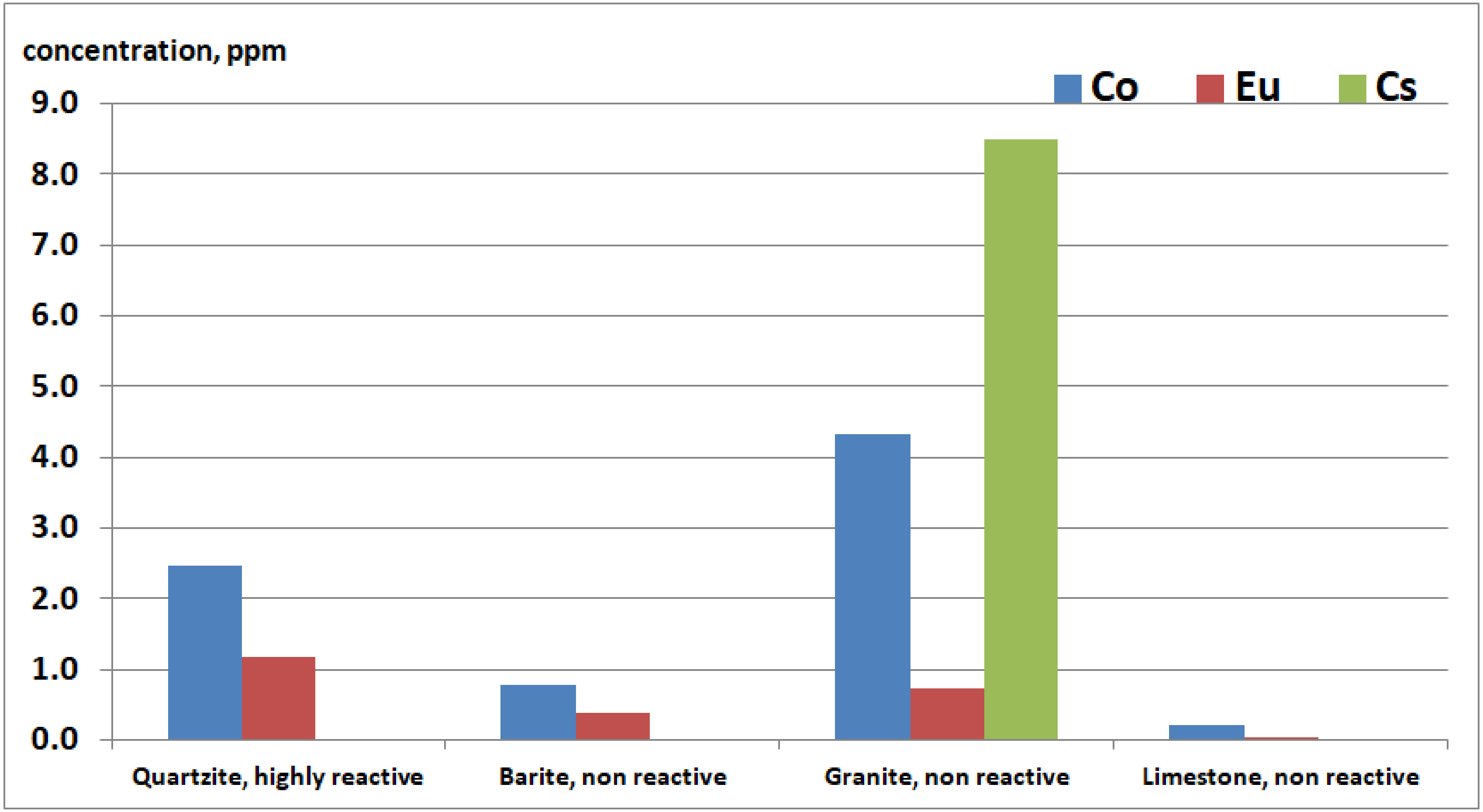


# Preliminary results: Neutron activation analysis

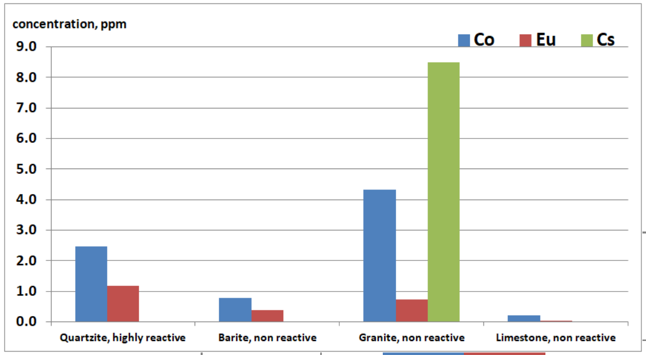


# Preliminary results: Neutron activation analysis

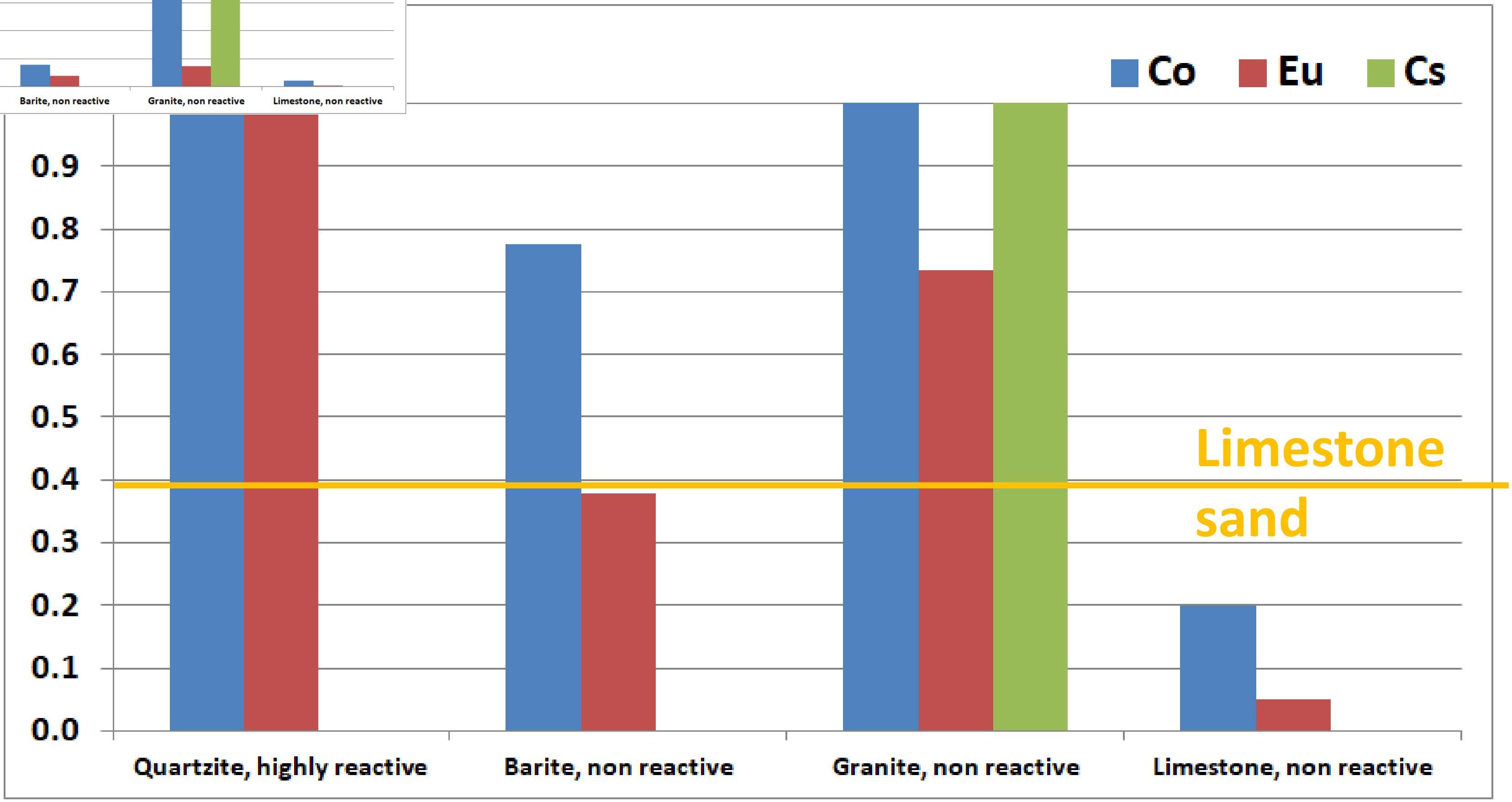
## Coarse aggregate



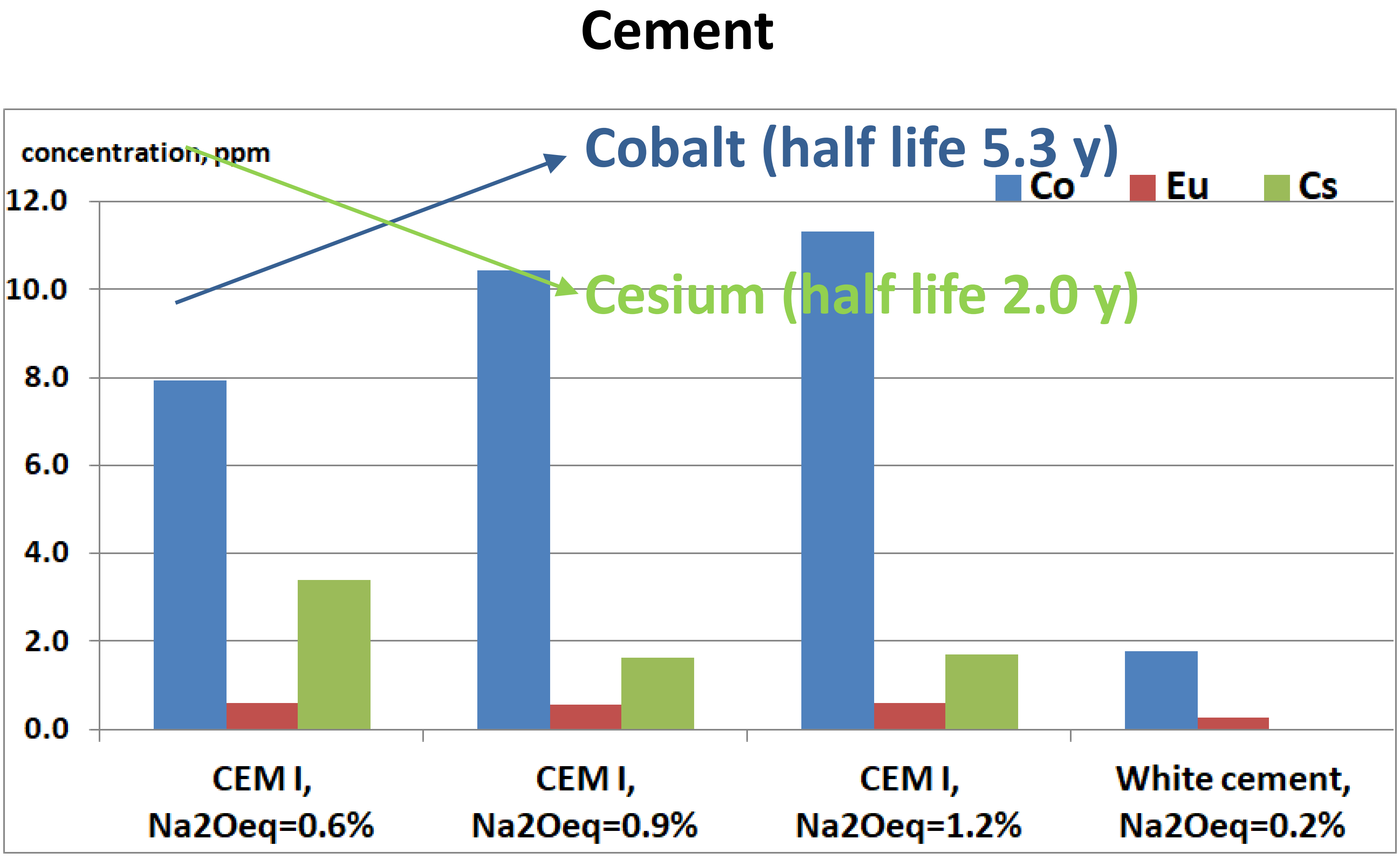
# Preliminary results: Neutron activation analysis




## Coarse aggregate



# Preliminary results: Neutron activation analysis



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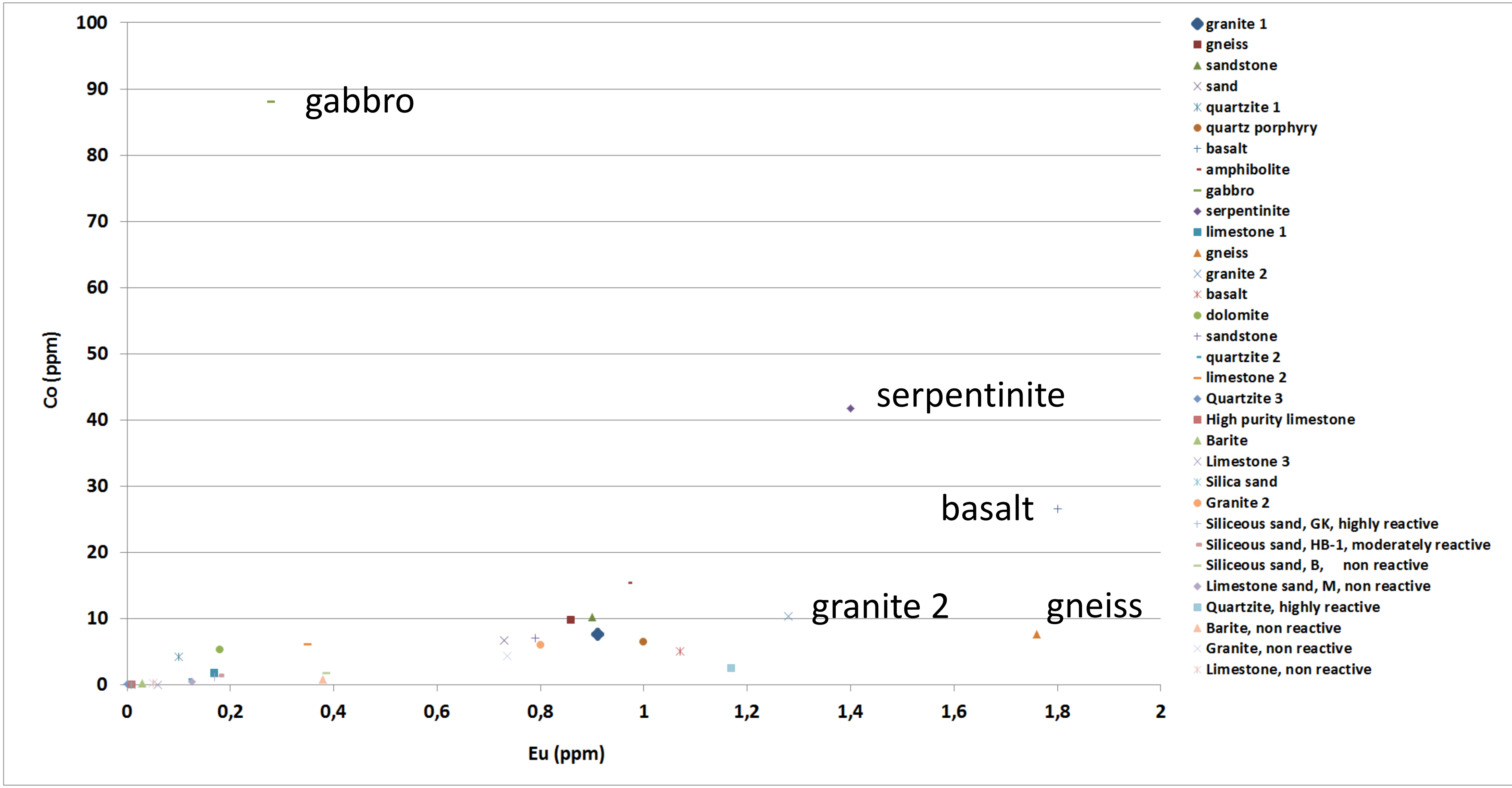
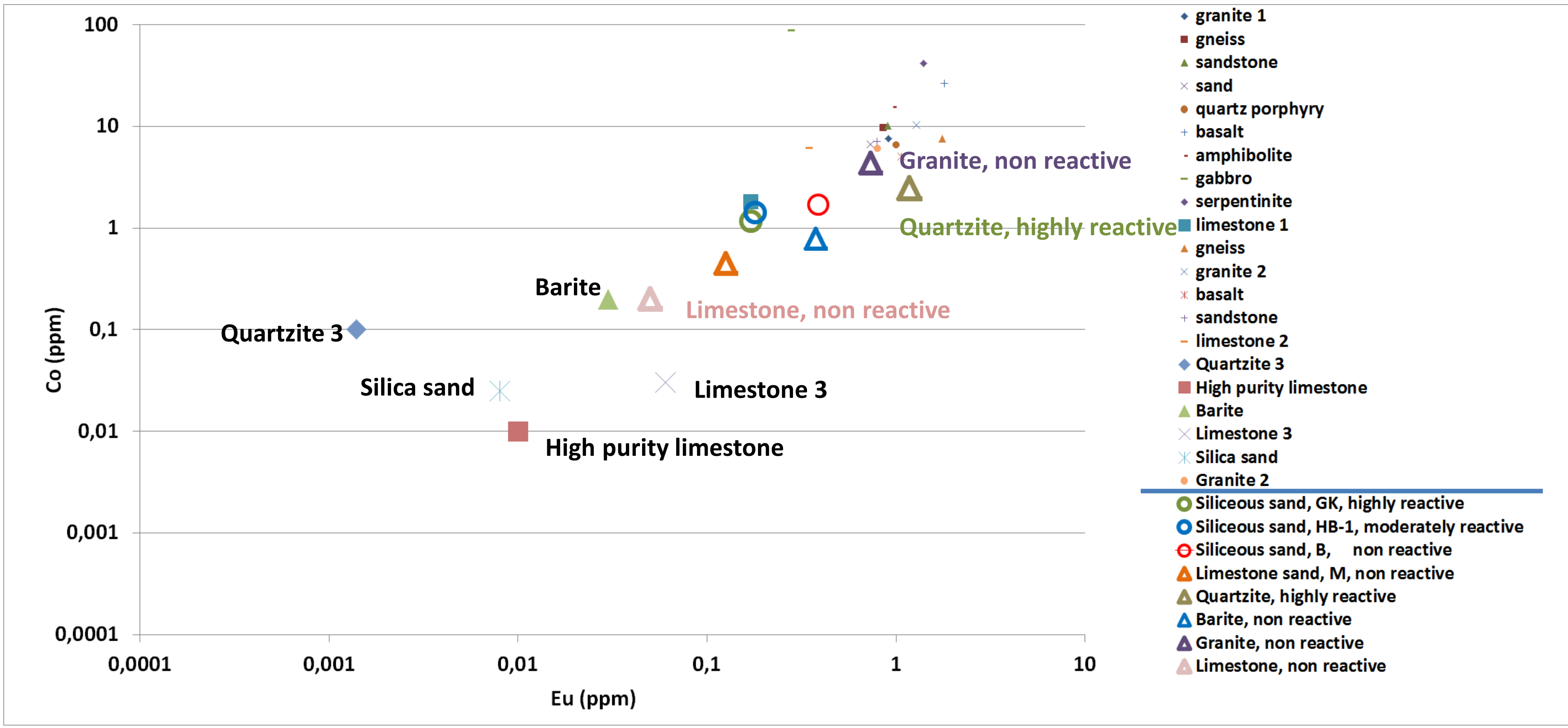
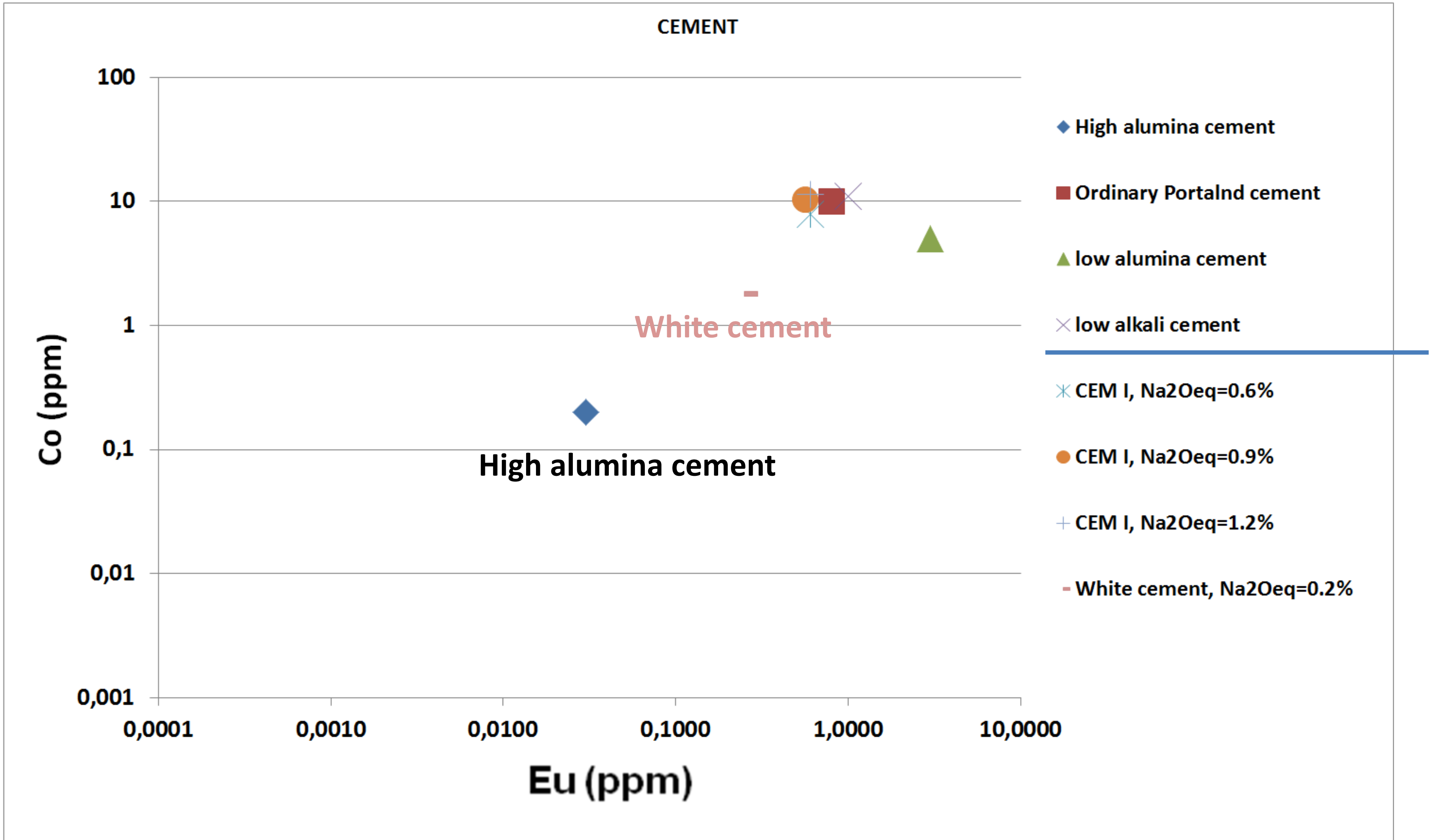


# Preliminary results: Neutron activation analysis



# Preliminary results: Neutron activation analysis



# Preliminary results: Neutron activation analysis

**Low – activation materials:**

- **Cement**
  - **White cement CEM I 52,5 R**
- **Aggregate**
  - **Quartzite**
  - **Limestone**
  - **Granite**

**Low-activation material**

**vs**

**low-activation cement matrix composite**

# Influence of irradiation on the ASR potential

White cement 52,5 R ($Na_2O_{eq}$=0.2%)

- ✓ Quartzite
- ✓ Limestone
- ✓ Granite

} Low-activation materials

- ✓ Opal
- ✓ Trachybasalt
- ✓ Flint

} ASR potential !!! vs Low-activation materials ???

- NaOH added (4 kg/$m^3$)
- w/c=0.47

**ALKALI-SILICA RESISTANT SHIELDING CONCRETE ???**

# Influence of irradiation on the ASR potential

- ✓ Opal
- ✓ Trachybasalt
- ✓ Flint

} siliceous minerals
but
different forms of $SiO_2$

- Amorphous
- Chalcedony, opal
- Cryptorcystaline (chalcedony, trydimite)

**Minerals without Fe, without heavy elements, ...**

**Influence of mineral composition on the … disolution rate?**

# Influence of irradiation on the ASR potential

## Mortar bar specimens: different time of irradiation

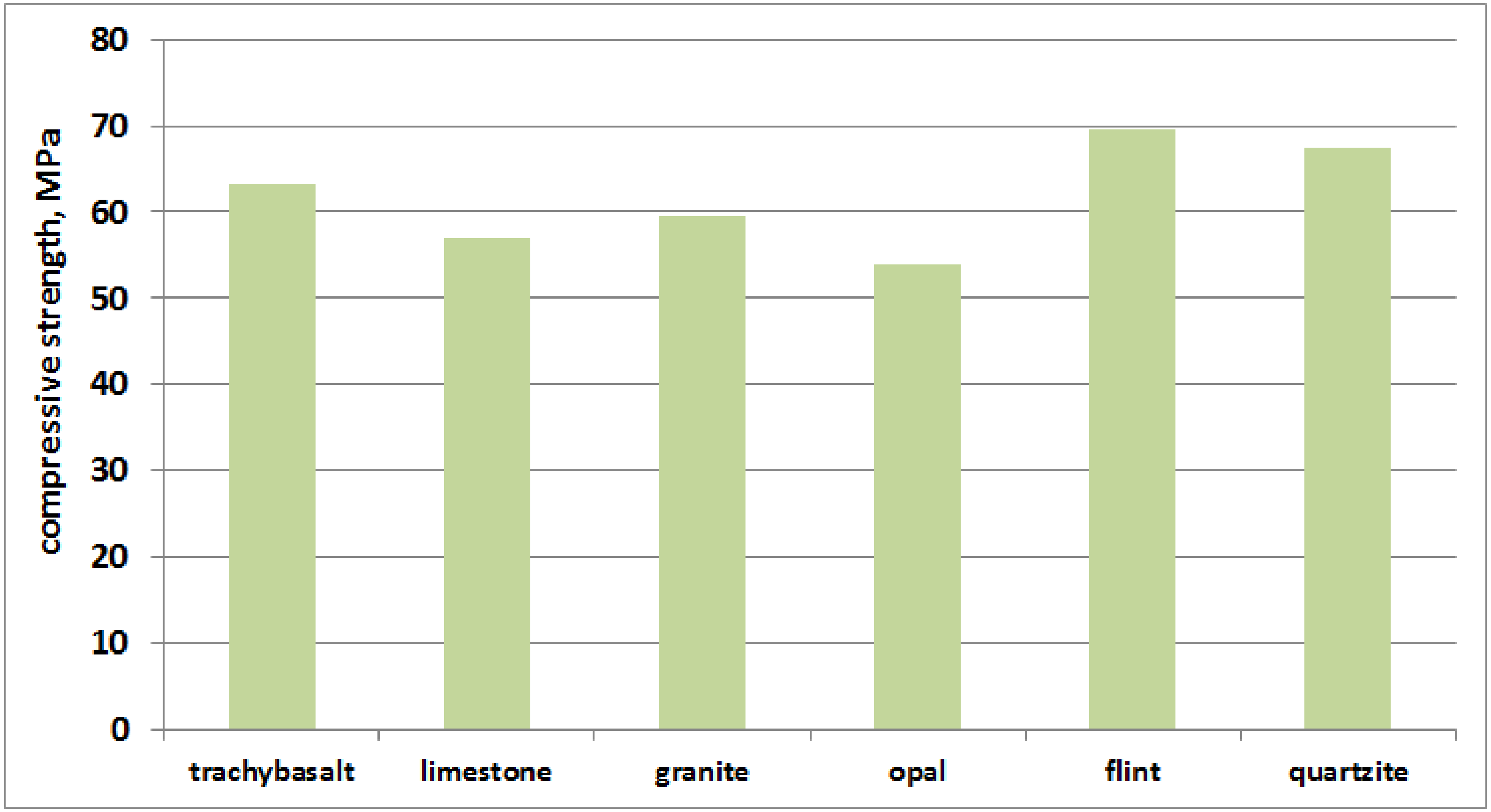
## 1 day, 1 week, 1 month

18x18x80 mm


Humid conditions

# Influence of irradiation on the ASR potential

## Mortar bar specimens, 28 days
## w/c=0.47



# Influence of irradiation on the ASR potential

**Expected analysis on mortar bar specimens after irradiation:**

- **Mechanical propreties**
- **Microstructure: aggregate, cement matrix**

- **Compressive strength**
- **Modulus of elasticity**
- **Microindentation**
- **SEM**
- **Thin sections**
- **… ?**

# Instead of summary …

**Materials characterisation using radiation**

↓

**low-activation concrete**
**(decommissioning)**

# Instead of summary …

**Materials characterisation using radiation**

⬇

**low-activation concrete (decommissioning)**

**Effect of neutron radiation**

⬇

**ASR potential**

# Thank you for your attention

**Acknowledgements**
The financial support by Polish National Centre for Research and Development (Project V4-506 Korea/2/2018) is gratefully acknowledged.