LEACHING BEHAVIOR OF CEMENTITIOUS MATERIAL IMMOBILIZING Cs-CONTAINING B-10 ENRICHED BORIC ACID WASTE

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Introduction

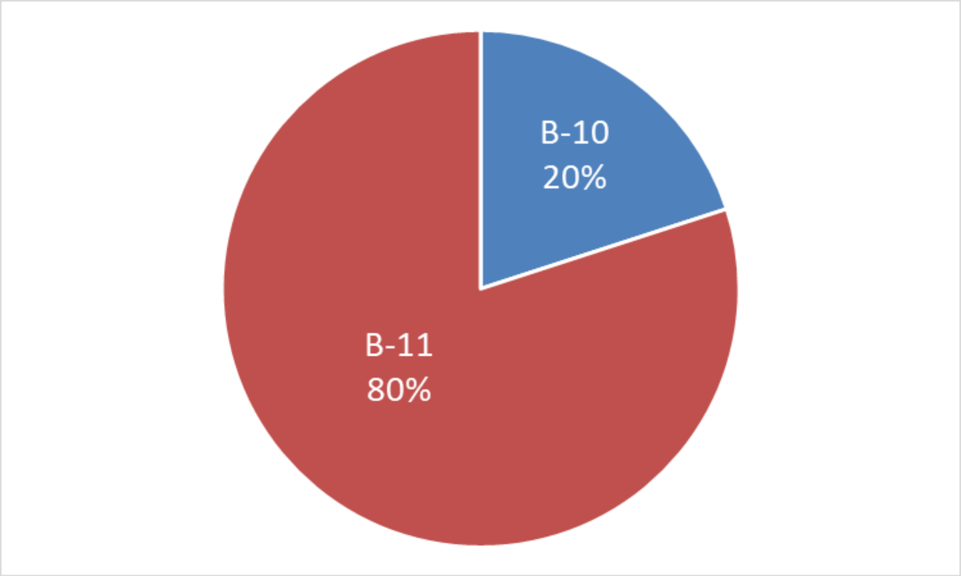


Fig.1. Natural isotopic abundance of B-10 and B-11

Boric acid enriched in B-10 isotope is a new type of neutron absorber which is already used in some nuclear power plants. Boric acid with natural isotopic abundance of B (**Fig. 1)** is the conventionally used solution. In both solutions, the B-10 isotope is responsible for neutron absorption due to its large cross section with thermal neutrons (3337 vs. 0.005 barns, Xu & Zhang, 2010)

Advantages connected to the use of B-10 enriched boric acid in nuclear power plants are the

(1) increase in the controllability of the nuclear reactor with a lower amount of boric acid solution, and therefore , the
(2) significant reduction in the volume of boric acid liquid radioactive waste, consequently, a decrease in the management cost and environmental risks.

Several radioisotopes of radiological and health concern, i.e. Cs-137 and Sr-90 are also present in significant amounts in boric acid liquid radioactive waste.

Prior to disposal of liquid radioactive waste, cementation is an acceptable and widely used immobilization technique (Hyatt & Ojovan, 2019)

Assurance of radioactive waste immobilization by cementitious materials calls for attention to the study of cement – radioactive waste system.

Status of studies about cement - boric acid radioactive waste

Natural boric acid

Cement - natural boric acid system has been studied extensively: fates of different radioisotopes, optimization and how natural boric acid affects waste form durability and performance, i.e. compressive strength, porosity and leachability.

Enriched boric acid

There is no published data about this new cementitious system and if B-10 enrichment has any effect on the fate of radioisotopes, waste form optimization and durability.

Aim of the study

In this ongoing experimental work, we study the behavior of Cs in boric acid containing cement paste specimens, aiming to shed light on the fate of Cs-137 radioisotope in the solidified cementitious waste forms of B-10 enriched boric acid.

Materials and methods

SAMPLE PREPARATION

- Portland cement and simulated liquid waste mixing (**Fig. 2**)
- Casting of cement paste in cylindrical molds (**Fig. 3**)
- Curing - incubator
- Demolding to obtain waste form

LEACHING TEST

- ASTM standard (2017)
- Immersing waste form specimen in leachante (demineralized water, **Fig. 4**)
- Leachante renewal and leachate extraction

CHARACTERISATION

- XRD and SEM for analysis of solid samples
- ICP-OES and ICP-MS for analysis of extracted leachates (**Fig. 5**)

Fig. 2

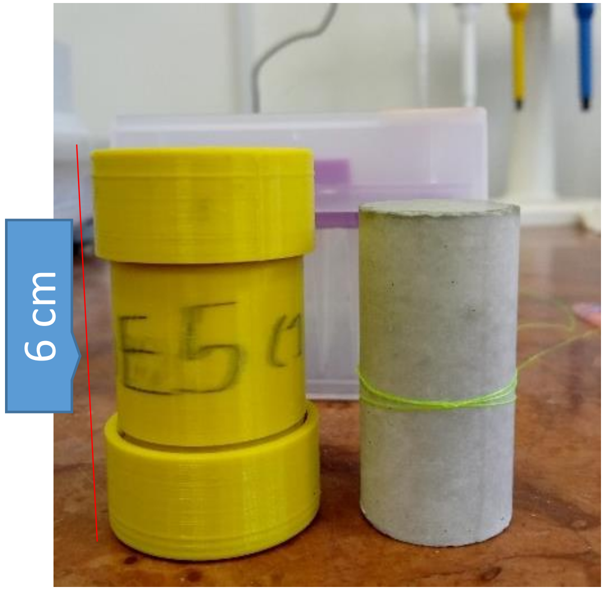


Fig. 3

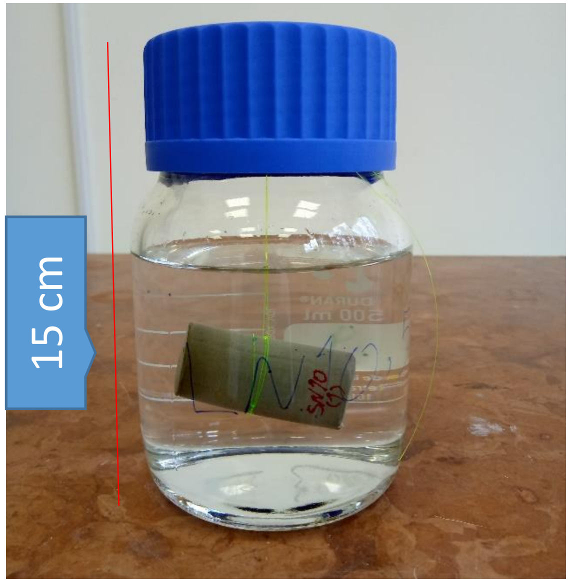


Fig. 4

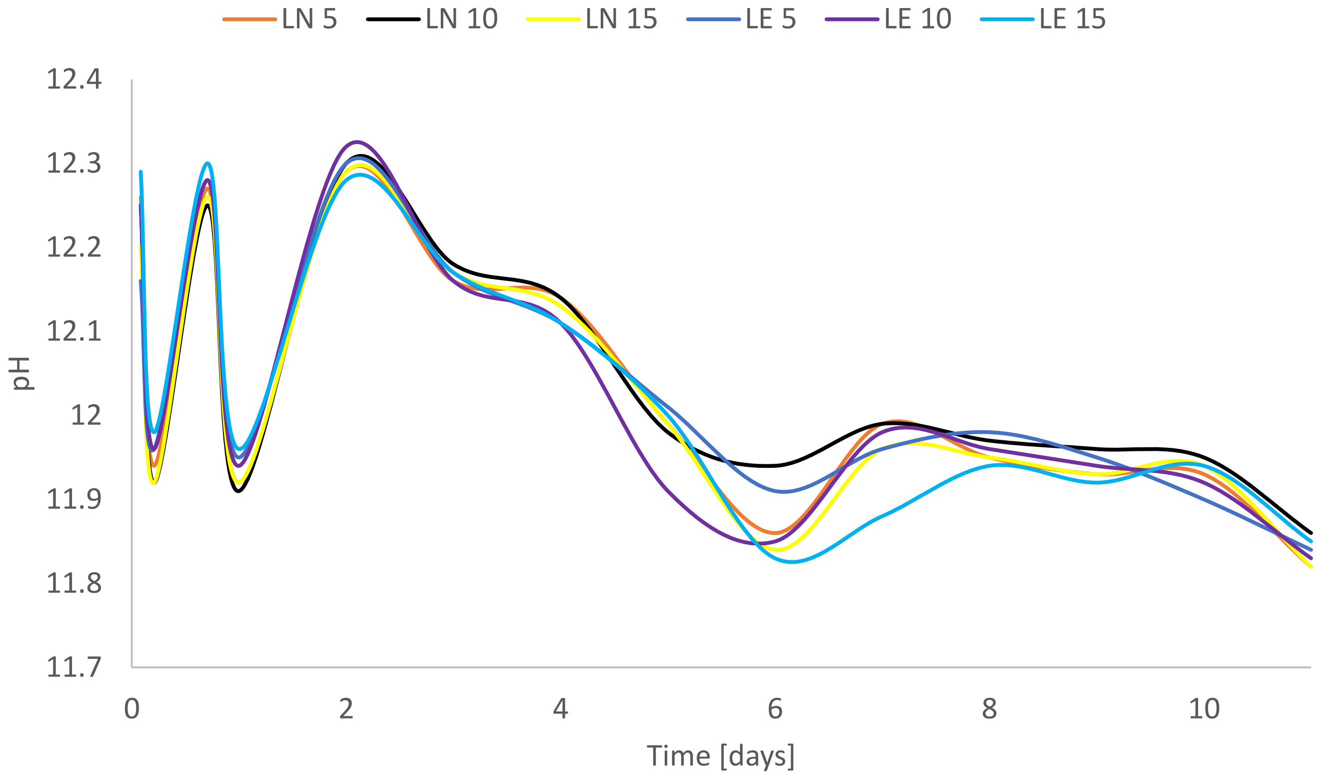
Fig. 5

Preliminary results (a) XRD analysis of 28 days cured samples

Table1. Mineral phases (names and chemical formula) of B and Cs containing cement paste specimens before leaching (SE and SN samples of this work) and reference sampes (FF samples from the work of Rostamiparsa et al., 2020). The reference samples were made from the same cement a few months earlier.

Sample name	Alite C_3S	Inyoite $(CaB_3O_3(OH)_5{\bullet}4H_2O$	Portlandite $Ca(OH)_2$	Akermanite $Ca_2Mg\ (Si_2O_7)$	Brownmillerite $Ca_2(Al,Fe^{3+})_2O_5$	Hydrocalumite $Ca_4Al_2(OH)_{12}(Cl,CO_3,OH)_2{\bullet}4H_2O$	Gowerite $CaB_6O_{10}\ {\bullet}4H_2O$	Amorphous C-S-H	Biringucci $Na_4[B_{10}O_{16}(OH)_2]{\bullet}2H_2O$	Kanemit $NaHSi\ _2O_5\ .3H_2O$	Ettringite $Ca_6Al_2(SO_4)_3(OH)_{12}{\bullet}26H_2O$	Sample details: B, Cs conc. in ppm
FF 1B	34		39	2	4	3		7			5	without Cs and B
FF 5D	52		19	3	2	7	1	9	5	2		40000 B, enriched, 0 Cs
SE 5	63	9	11	5	4	1	1	6				40000 B, enriched, 5000 Cs
SE 10	64	8	10	4	5	1	2	6				40000 B, enriched, 10000 Cs
SE 15	64	8	9	4	4	2	2	7				40000 B, enriched, 15000 Cs
FF 7D	46		18	4	3	8	2	7	6	6		40000 B, natural, 0 Cs
SN 5	62	8	11	4	5	1	2	7				40000 B, natural, 5000 Cs
SN 10	60	9	10	4	5	1	3	8				40000 B, natural, 10000 Cs
SN 15	69	7	9	2	4	1	3	5				40000 B, natural, 15000 Cs

(b) pH of the leachates (11 days leaching period)



High pH of the cementitious system is due to portlandite mineral phase release from the matrix:

$Ca(OH)_{2(s)} + H_2O_{(l)} \rightarrow 2Ca^{2+}{}_{(aq)} + OH^{-}{}_{(aq)}$

Different pH peaks are due to different portlandite release rate and different time elapsed in the steps of leaching.

Fig. 6. pH as function of time during leaching test. In sample names L refers to leachate, N to natural boric acid, E to boric acid enriched in B-10 isotope, and the numbers 5, 10 and 15 to the concentrations of Cs of 5 000, 10 000, 15 000 ppm in the simulated liquid waste.

(c) SEM analysis of 28 days cured samples

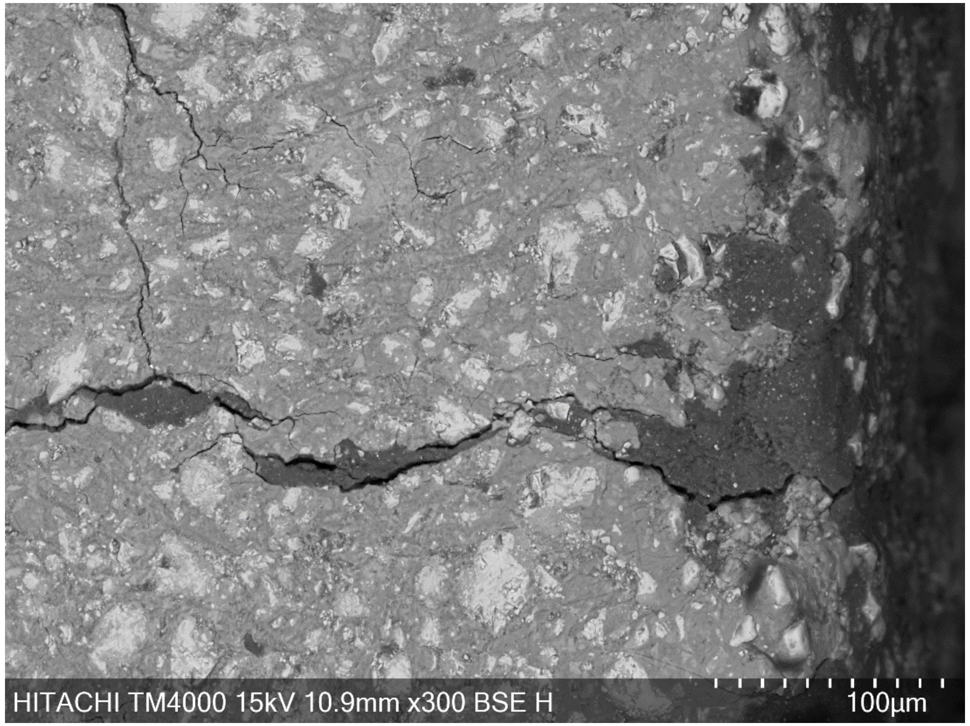


Fig. 7. SEM, BSE image of the edge of the sample SN 5 before leaching

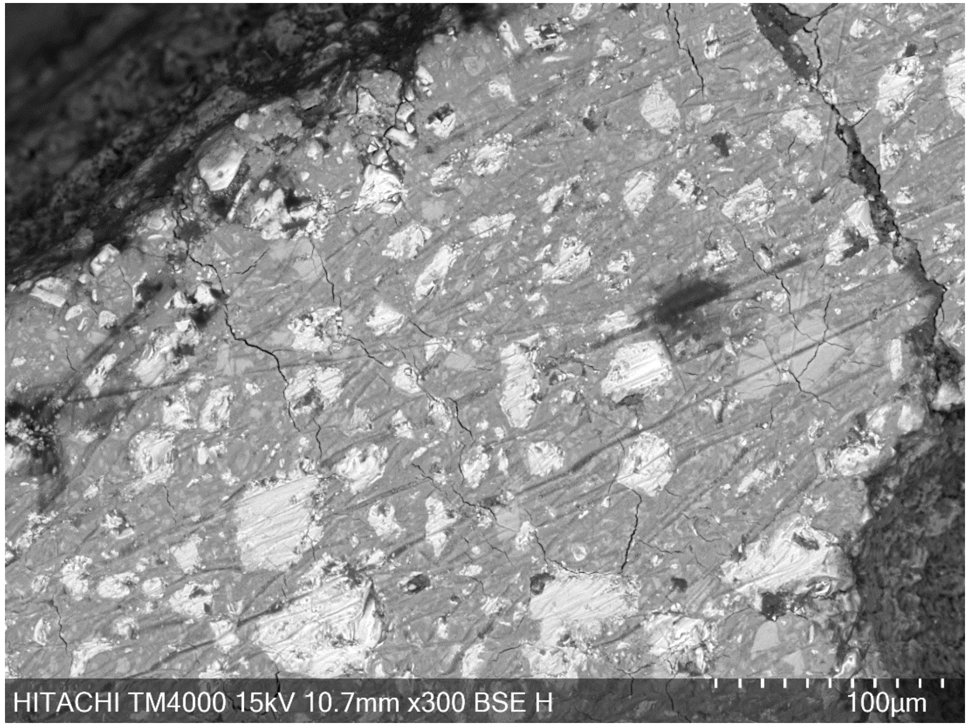


Fig.8. SEM, BSE image of the edge of the sample SE 5 before leaching

SN 5 and SE 5 sample names refer to the cement paste specimens made with 40000 ppm natural and enriched boric acid solutions, respectively, and 5000 ppm Cs.

Highlights

- Boric acid enriched in B-10 isotope is a new type of neutron absorber in nuclear power plants.
- Its immobilization calls for this study which investigates the fate of Cs in boric acid – cement systems.
- We apply standard leaching test, XRD, SEM, ICP-OES and ICP-MS techniques.
- We expect to contibute to better understanding of Cs-137 radioisotope immobilization in cementitious waste forms.
- Optimization of the new waste form performance can be achieved.

REFERENCES

- Hyatt, N. C., & Ojovan, M. I. (2019). Special issue: Materials for nuclear waste immobilization. Materials, 12(21), 12–15. https://doi.org/10.3390/ma12213611
- Rostamiparsa, M., Szabó-Krausz, Z., Fábián, M., Falus, G., Szabó, C., & Völgyesi, P. (2020, May). Experimental assessment of interaction between boric acid enriched in boron-10 and cementitious matrix. In EGU General Assembly Conference Abstracts (p. 19441), https://doi.org/10.5194/egusphere-egu2020-19441
- Xu, J., & Zhang, W. (2010). The application of 10B enriched boric acid in nuclear power industry. International Conference on Nuclear Engineering, Proceedings, ICONE, 3, 1–5. https://doi.org/10.1115/icone18-29042
- ASTM. (2017). ASTM C1308-08. Standard Test Method for Accelerated Leach Test for Diffusive Releases from Solidified Waste and a Computer Program to Model Diffusive, Fractional Leaching from Cylindrical Waste Forms. https://doi.org/10.1520/C1308-08.2

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