





# Experimental assessment of interaction between boric acid enriched in boron-10 and cementitious matrix

#### Mojtaba Rostamiparsa\*

Lithosphere Fluid Research Lab, Department of Petrology and Geochemistry, Eötvös Loránd University, Budapest, Hungary (\*rostam@caesar.elte.hu)

#### Zsuzsanna Szabó-Krausz

MTA Premium Postdoctorate Research Program, Lithosphere Fluid Research Lab, Department of Petrology and Geochemistry, Eötvös Loránd University, Budapest, Hungary

#### Margit Fábián

Center for Energy Research, Budapest, Hungary

#### **György Falus**

Mining and Geological Survey of Hungary, Budapest, Hungary

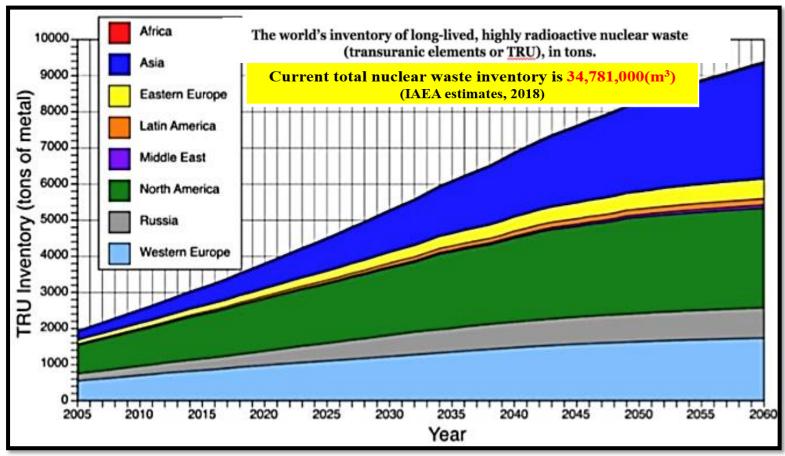
#### Péter Völgyesi

Center for Energy Research, Budapest, Hungary

EGU General Assembly 2020 (4<sup>th</sup> May)

## Introduction

- o Nuclear technology has many advantages and a significant disadvantage
- The main concern about nuclear technology is nuclear waste
- o The amount of waste is increasing in the world continuously



Trend of nuclear waste production (www.weforum.org)

# **Introduction -** boric acid in nuclear power plants

- o NPPs1 are the main applicants of nuclear technology
- o Boric acid is used because of its excellent neutron absorbing properties
- Main waste stream at a typical NPP is boric acid waste (volumetric)
- o Production around 200 m³/yr boric acid waste in a typical PWR² (Pacey et al. 2011)



Generation of huge amount of boric acid waste in NPPs (www.kaeri.re.kr/)

<sup>1 -</sup> Nuclear Power Plant

<sup>2 -</sup> Pressurized Water Reactor

## Introduction - waste management of boric acid

- o Boric acid is conditioned for easy, secure and safe handling, storaging and disposal
- o Common method for boric acid waste is stabilizing with cementitious matrix
- o **Boron has a high natural potential for releasing from the cementitious matrix** (Palomo and Palacios 2003)
- There are some approved methods for decreasing the leachability of boron



Packing the waste – Paks, Hungary



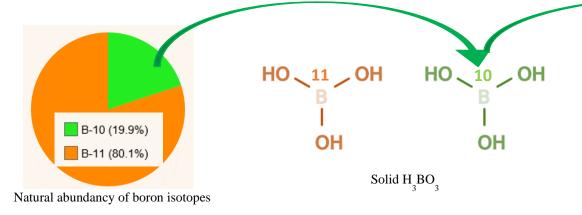
Cementation the packs – Bátaapáti, Hungary



Disposal at Bátaapáti, Hungary

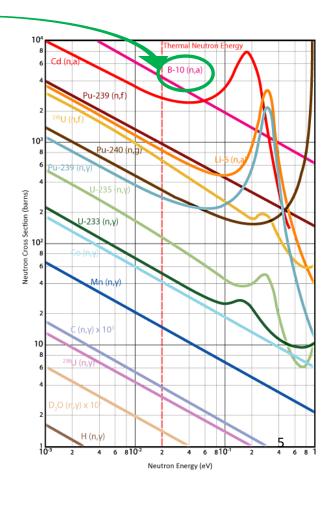
## Introduction - natural boric acid or enriched boric acid solution?

- Two ways for increasing the control ability of boric acid
  - $\times$  Increasing the boric acid concentration (corrosion and leachability  $\uparrow \uparrow$ )
  - ✓ Using enriched acid boric (recently used technology)
- What is the meaning of enriched boric acid?



B-10 cross section is 3840 barns B-11 cross section is 0.005 barns

- Natural boric acid has a fixed ratio of boron isotopes: [(B-10)/(B-11)] = 0.248
- $\triangleright$  Enriched boric acid in boron-10, i.e. changing the isotope ratio of [(B-10)/(B-11)] > 0.248



## **Questions - what is the effect of B-10 enrichment on cementitious matrix?**

- o Currently we have information for the durability of the waste-form with NBA<sup>1</sup>
- There is no published information about the effects of using EBA<sup>2</sup> in cementitious matrix
- What is the leachability and durability of waste-forms including EBA?

If the leachability of EBA/NBA is decreased in cementitious matrix, durability ↑↑



If the leachability of EBA/NBA is increased in cementitious matrix, durability  $\downarrow\downarrow$ 

 $<sup>\</sup>bigcirc$ 

<sup>1 -</sup> Natural Boric Acid (NBA)

<sup>2 -</sup> Enriched Boric Acid (EBA)

## Materials and methods

## Materials

- **Ordinary Portland Cement (European standard EN 197-1)**
- Natural boric acid (NBA) and enriched boric acid (EBA) (> 99% B-10)
- Demineralized water

## **Leachability test**

- o logical index for comparison of the durability of samples
- ASTM C1308-08 (2017) is an approved method for leachability test

# **Analytical methods**

- **Output** OF COMMON OF COMM
- **O XRD, ATR-FTIR, SEM and Raman-spectroscopy for phase analysis**
- Geochemical modeling with PHREEQC code

## **First experiments** — **sample preparation** (EK<sup>1</sup> & ELTE<sup>2</sup>)

- o Cement paste samples were prepared containing boric acid
- o Three w/c ratios of 0.35, 0.4 and 0.428 were used
- Samples were cured for 28 days
- **O Powdering for XRD and ATR-FTIR**
- **o Surfaces polished for SEM and Raman**



Mixing mortar with boric acid - EK



Removing samples from molds - EK



Preparing of samples for SEM - ELTE



Preparing of samples for XRD - ELTE

<sup>1 -</sup> Hungarian center for energy research

<sup>2 -</sup> Eötvös Loránd University

# First experiments — leaching test

- o Cured samples were soaked in leachantes (DM water) for specified times
- o pH detected for each batch and solution samples collected



Liquid – solid interaction for leachability test - EK



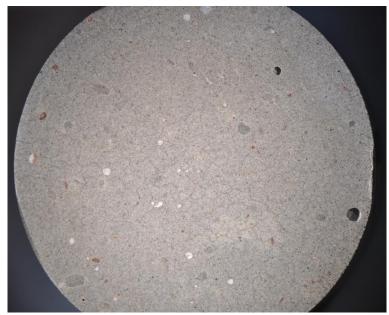
Leachate pH-metery - EK

# Preliminary results - phase analysis

- Alite, ettringite, larnite, brownmillerite, portlandite and inyoite are detected phases in the cementitious matrix by XRD
- **Output** The abundance of unhydrated phases are higher in lower w/c ratio samples



Cured sample -EK



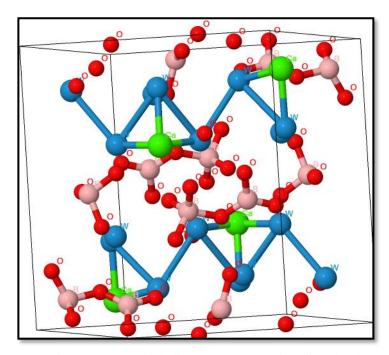
Microscopic view of samples surface - ELTE

# **Preliminary results - phase analysis**

- **Inyoite** (CaB<sub>3</sub>O<sub>3</sub>(OH)<sub>5</sub>·4H<sub>2</sub>O) is the only phase detected containing boron
- o Samples with higher w/c ratio, have lower concentration of inyoite
- Increasing of w/c can increase the boron release from the matrix



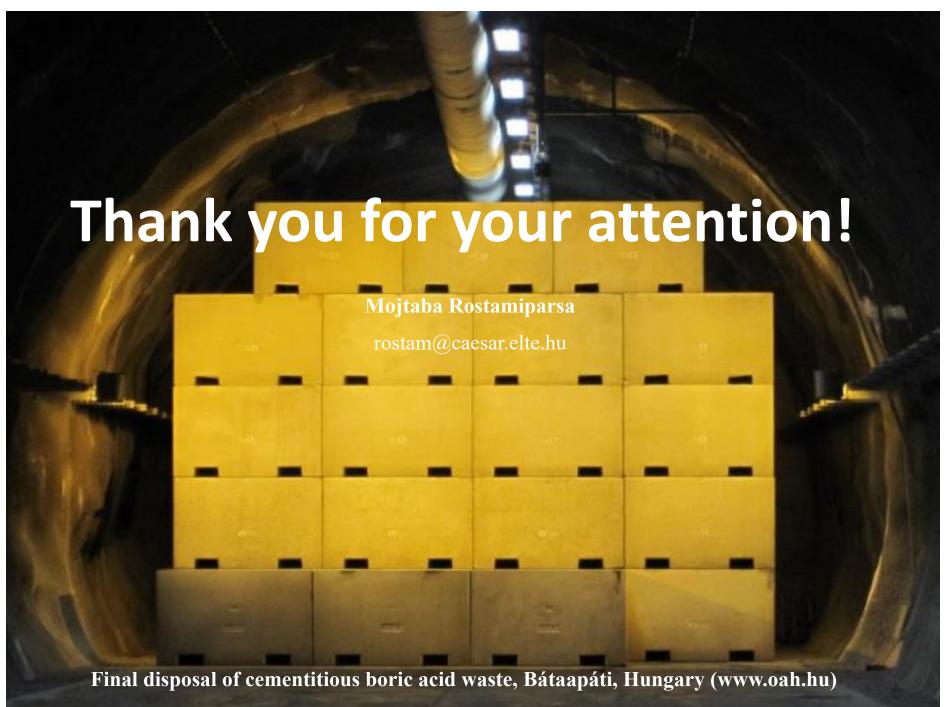
SEM analysis - ELTE



Inyoite 3D structure including boron (www.mineralienatlas.de)

## Summary

- o Boric acid is one of the most important waste streams in all kind of NPPs
- Its stabilization for long storage and disposal is done by cementation process
- Many tests for durability of the cementitious matrix have been done already with natural boric acid (NBA)
- Some modern NPP's have started to using enriched boric acid (in B-10) in recent years, while there is no published data about stabilizing it in cementitious matrix
- EBA and NBA might have different leachability and durability properties in the cementitious matrix (Marschall and Foster 2016)
- Experiments have already started in this doctorate project to better understand and predict geochemical processes in EBA vs. NBA cementitious systems



#### References

- Basis, Protein Dry, Lactose Monohydrate, Pesticides Other, Insolubility Index, Test Method, Phenol Sulphuric, and Mean Result. 2007. "Certificate of Analysis Certificate of Analysis." 86(06151):1–3.
- Marschall, H., and G. Foster. 2016. Advances in Isotope Geochemistry- Calcium Isotopes.
- Pacey, Nigel, Ian Beadle, Anna Heaton, and Laura Newsome. 2011. *Chemical Discharges from Nuclear Power Stations: Historic Releases and Implications for Best Available Techniques (BAT)*. Environment Agency.
- Palomo, A., and M. Palacios. 2003. "Alkali-Activated Cementitious Materials: Alternative Matrices for the Immobilisation of Hazardous Wastes Part II. Stabilisation of Chromium and Lead." *Cement and Concrete Research* 33(2):289–95.