Desorption of Cs from vermiculite clay with ultrasound under ambient and hydrothermal conditions

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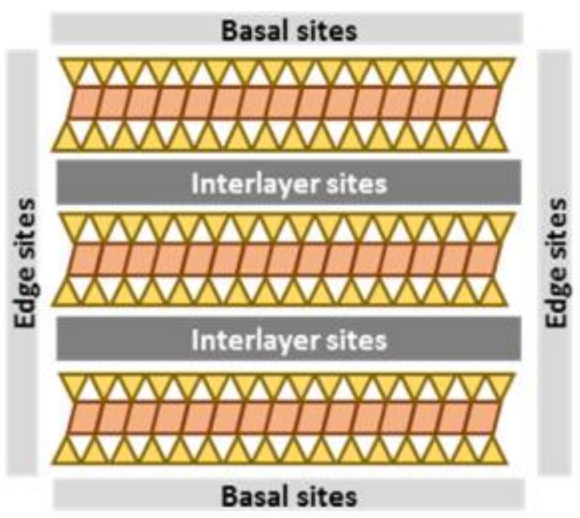
Scope

Following the power plant accident of Fukushima Dai-ichi (Japan, 2011), a significant amount of radioactive cesium (^{137}Cs) has been released into the environment, leading to the removal of important volumes of contaminated soil. Among remediation technologies, soil washing is the most used technology and several studies focused on Cs^+ desorption through ion exchange. Ultrasound, which is often used in surface cleaning or delamination, is here combined to the ion exchange process in order to enhance the desorption rates.

Vermiculite (verm.) clay and ion exchange

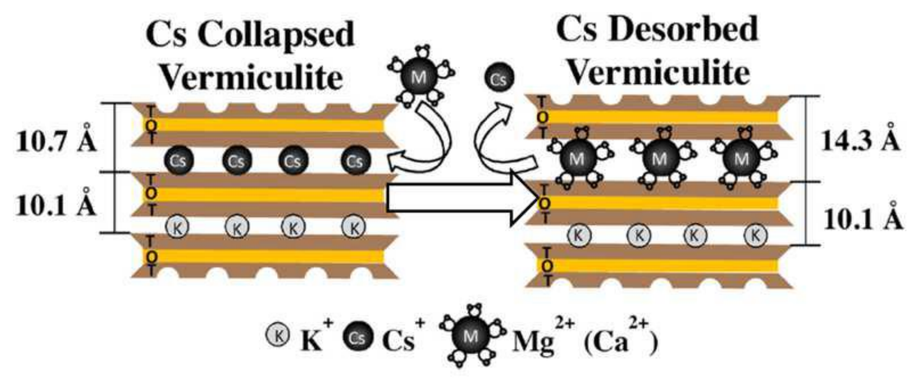
Vermiculite clay in which **sites (basal, edge and interlayer)*** are available for **ion exchange** or **complexation.**



Cs^+ extraction is **challenging** because of its **high affinity** (esp. in the **interlayer space**) and the **layers collapse** it involves**.

Cs^+ desorption can be **promoted** by:

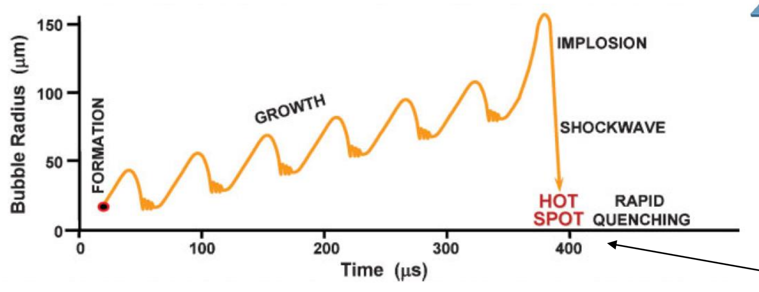
- Use of **divalent cation**
- **Diminution** in **particle size**
- **Elevated temperatures**

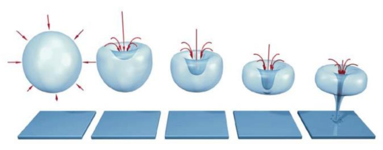


Acoustic Cavitation for the Decontamination of Solids

Physical effects – Low frequencies (20 – 100 kHz)

- **Erosion** and **Fragmentation**
- **Diminution** of the **diffusion layers**
- **Acceleration** of the **mass transfer**





Bubble implosion near a surface → **micro-jet**

Thousands of Kelvin and hundreds of atmosphere

Chemical effects – High frequencies (100 kHz – 1 MHz)

- **H_2O_2** and **radicals** formation
- **Sonochemical reactions**

$H_2O \rightarrow))) \rightarrow H + OH^\bullet$

Ultrasound

Under Ar atmosphere

$H + OH^\bullet \rightarrow H_2O$

$OH^\bullet + OH^\bullet \rightarrow H_2O_2$

$H + H \rightarrow H_2$

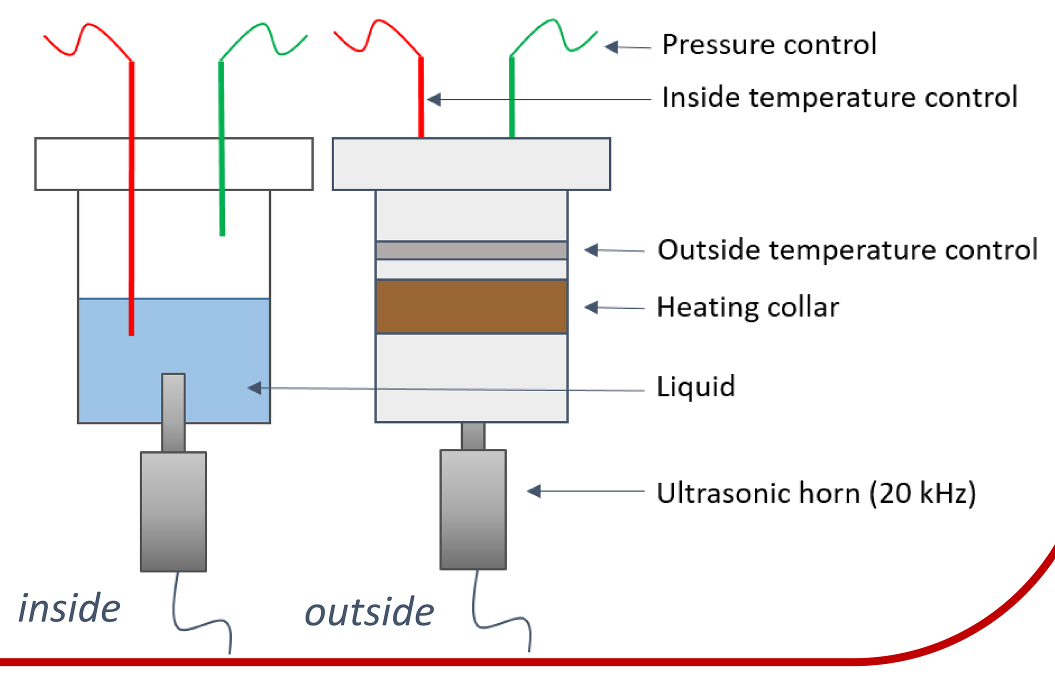
Experimentals

Ambient temperature set-up

← **Conditions:**
20/362 kHz,
20/60 °C,
Ar bubbling,
3/2 g clay powder in 300/200 mL $MgCl_2$ solution

↓ **Conditions:**
20 kHz, 100-200 °C,
autogenic pressure,
0.5 g clay powder in 50 mL $MgCl_2$ solution

Sonohydrothermal reactor

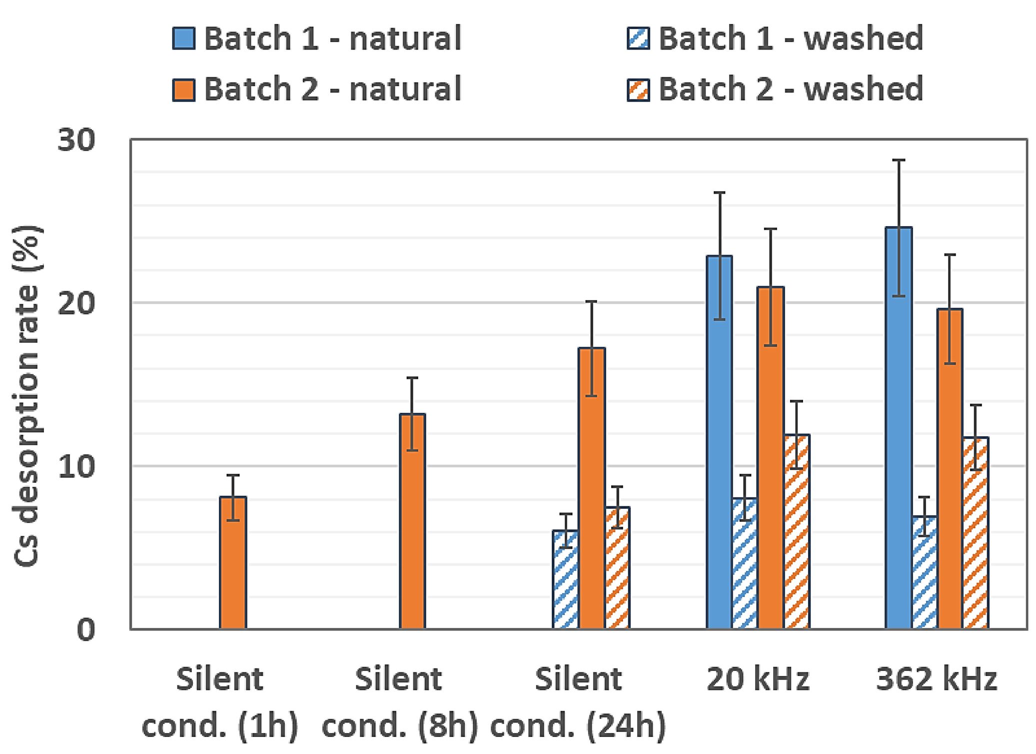


* Dzene, L., *J. Colloid Interface Sci.* **2015**   ** Yin et al., *J. Hazard. Mater* **2017**

Cs desorption with $MgCl_2$ under …

Ambient temperature treatment

10 g verm. /L – 1 M $MgCl_2$ – 1h – US (Ar, 20 °C)

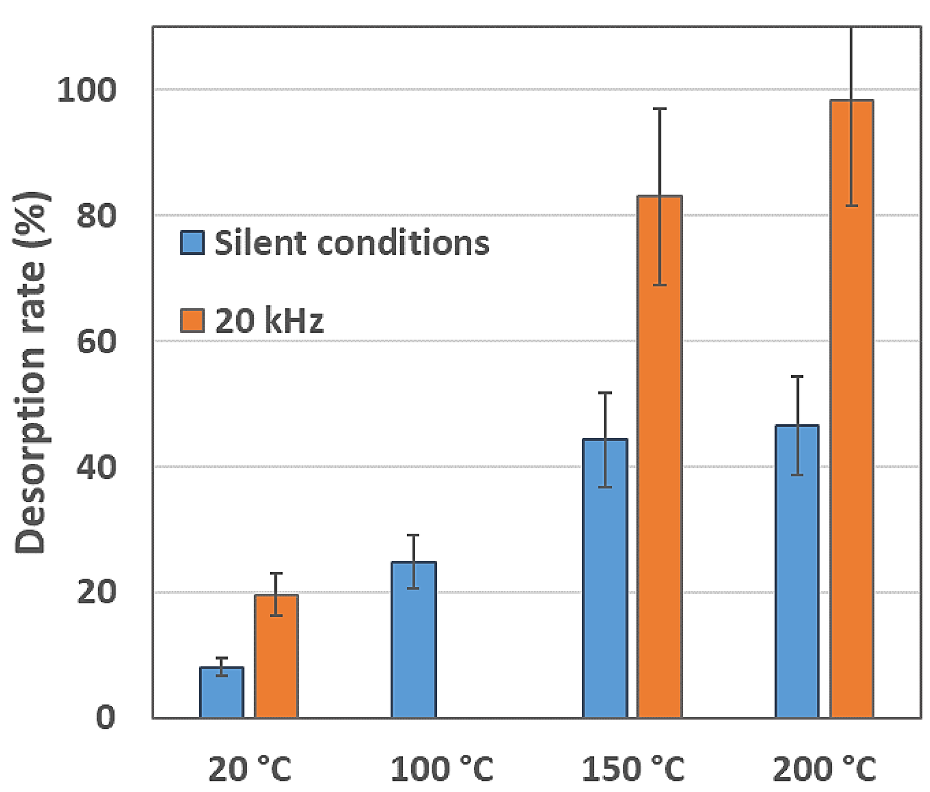


Batch 1 (7 mg/g), Batch 2 (25 mg/g),
washed = natural form washed with CH_3COONH_4 (1M)

At **room temperature**, desorption is **limited** but **kinetic rates** are **enhanced by ultrasound**

Hydrothermal (HT) and Sonohydrothermal (SHT) treatments

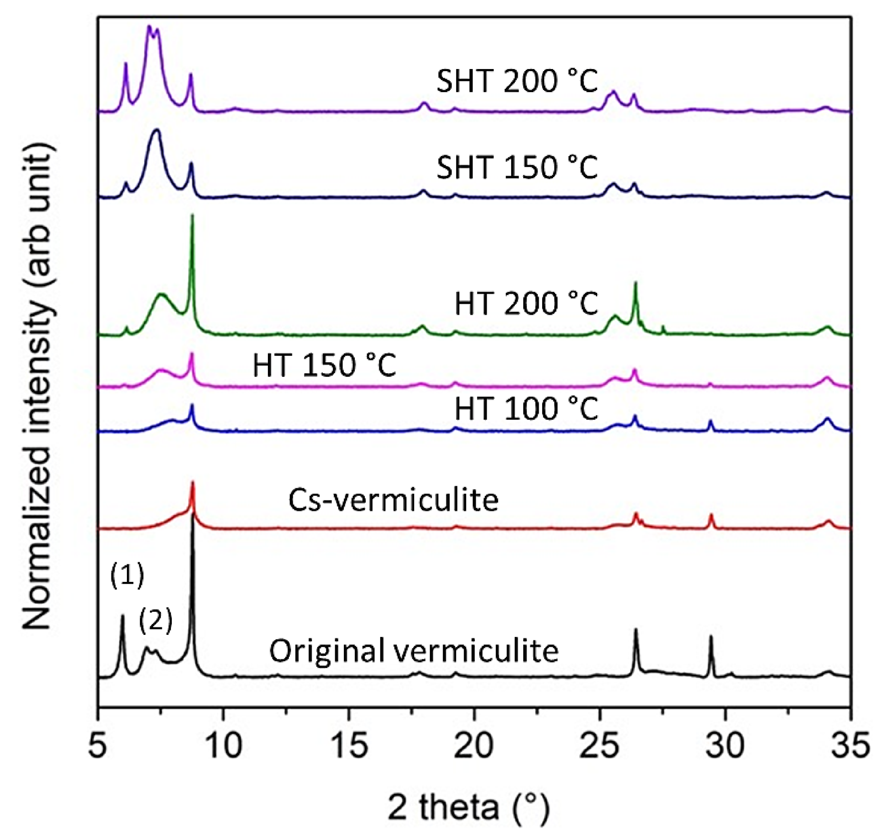
10 g verm. /L – 1 M $MgCl_2$ – 1h – US (Ar, 20 kHz)



With increasing temperature:

- desorption rate **increases**
- **sonohydrothermal** treatment shows **greater efficiency** than simple **hydrothermal** treatment

X-ray powder diffractograms



(1) : Mg^{2+} interlayer form, (2) : Mg^{2+}/K^+ interlayer form

(1) and (2) peaks, which **disappear** upon Cs^+ adsorption (preferably **replacing Mg^{2+}**), are **again visible** after treatment with Mg^{2+} at 200°C

Concluding remarks

Ultrasound allows to **accelerate** the desorption process at ambient temperature compared to silent conditions

Quantitative desorption of Cs^+ is observed under **hydrothermal conditions**

This process is currently **transposed** to the **depollution** of **heavy metals** (Ni^{2+} and Zn^{2+}) from the same matrix