Ultrasound-Assisted Soil Washing Process for the Removal of Heavy Metals from Clays References

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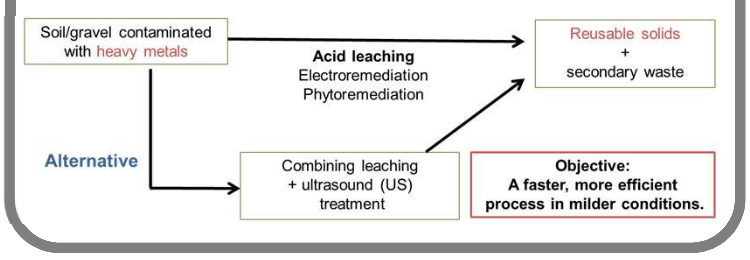
Context

Past human activities:

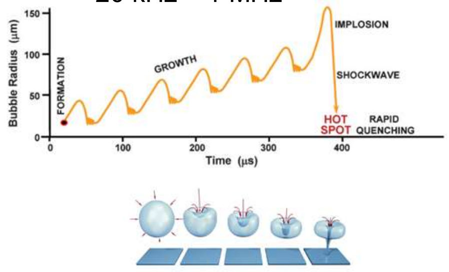
- Agriculture (pesticides, fertilizers…)
- Industrial activity (mining, paint production, battery production...)

are source of trace metals harmful to humans and the environment



Acoustic Cavitation

20 kHz – 1 MHz



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

A few recent studies combined leaching with ultrasound

Reference	Target metal(s)	Solvent
Choi et al., 2021	Cu, Pb, Zn	EDTA/HCl
Son et al., 2019	Cu, Pb, Zn	HCl
Park et al., 2017*	Cu, Pb, Zn	HCl
Kim et al., 2016	Cu, Zn	HCl
Hwang et al., 2007	Cu, Pb, Cd, Zn	EDTA/citric acid

US/mixing > conventional mixing in terms of

✓ removal efficiency

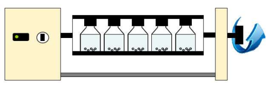
✓ consumption of chemicals

Attributed to better agitation (macroscale) & sonophysical effects (microscale)

However:
- Kinetic aspects neglected
- Poor soil characterisation

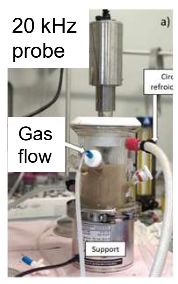
Methods

- **Vermiculite clay** was grinded, sieved (<100 µm) and contaminated with Zn or Ni by contacting it with Zn or Ni nitrates + $NaNO_3$ 10^{-2} M solutions at pH 4
- Initial metal content: 30-38 mg/g
- ICP-AES was used to quantify metal in solution
- Leaching solution HCl 0.1 M
- Temperature was kept around 20° C
- Tessier sequential extraction protocol was used to monitor metal repartition in the solid
- Laser granulometry for size distribution



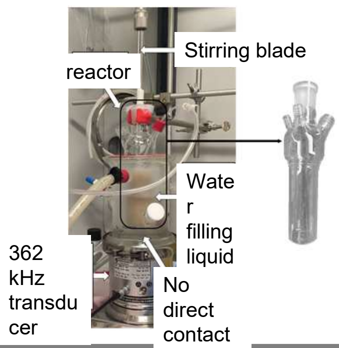
Silent H

Low frequency US: 20 kHz



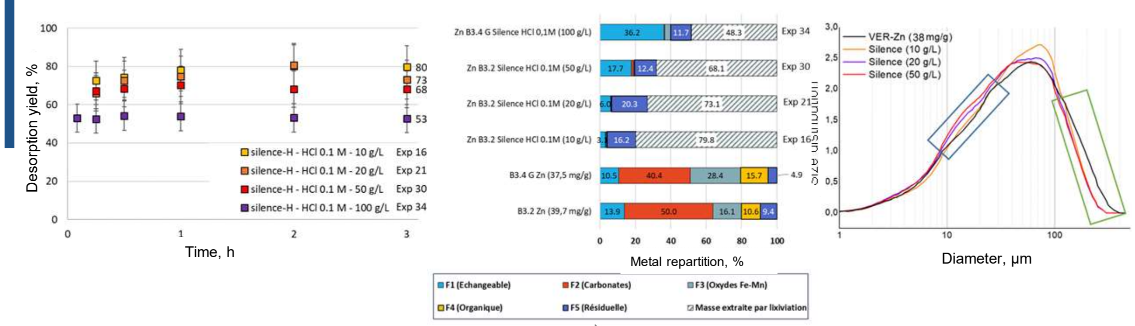
Sonics & Materials, Vibracell VCX 750
300 mL
P_{ac} = 34 W

High-frequency US: 362 kHz



ELAC NAUTIK
P_{ac} 25-30 W
120 mL
Stirring 300-400 rpm
Stirring alone = silent V

Results - Zn

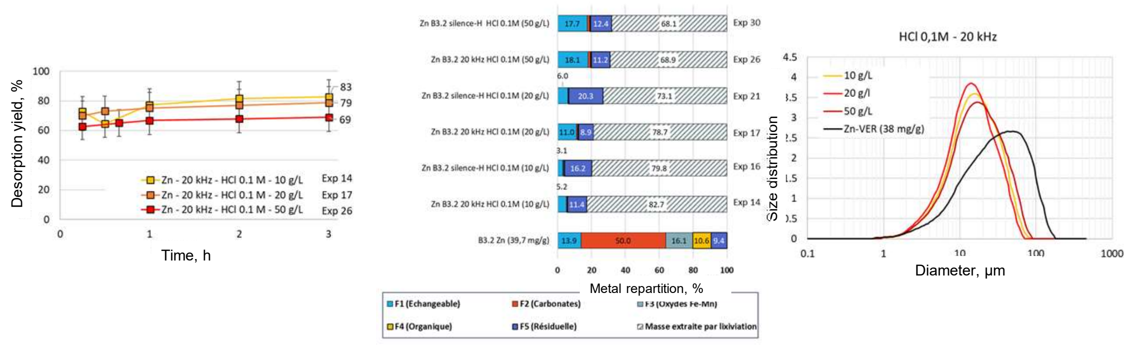


Initially, Zn mostly in F2 (+ F3)

Silent, HCl 0.1M: removal of F2-F3-F4

F1 ↓ at low m/V, ↑ at high ones due to fragmentation creating new adsorption sites

F5 ↑ indicating readsorption



20 kHz, HCl 0.1 M: removal of F2-F3-F4, similarly to silent conditions

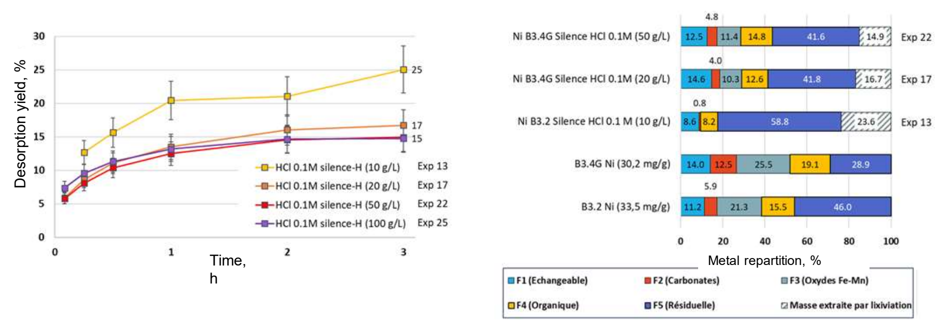
50 g/L similar to silent conditions

10-20 g/L: higher desorption from F5, but F1 ↑ due to fragmentation (new sorption sites)

At high-frequency US, fragmentation is limited, negative effects of US disappear.

362 kHz similar to silent conditions, in terms of depollution yields, Zn repartition and size distribution.

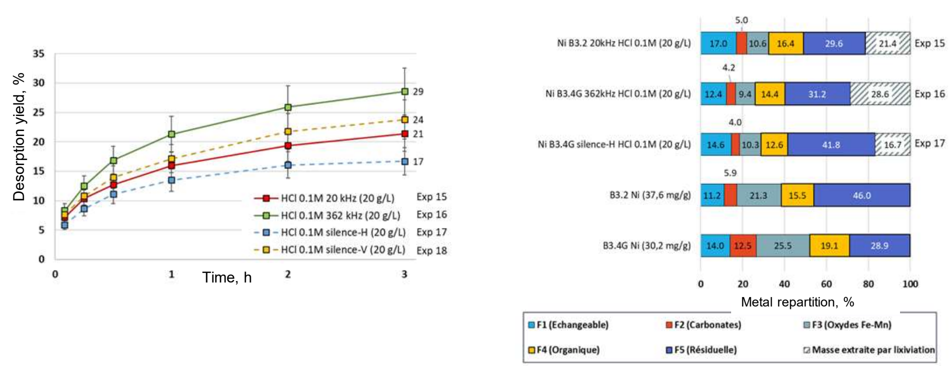
Results - Ni



Very different behaviour compared to Zn: Ni mostly in F5 (+ F4)

⇒ Much lower desorption yield

⇒ HCl 0.1 M silent conditions: removal of only part of F2-F3-F4



In the presence of US, higher desorption from F5

20 kHz: F1 ↑ due to fragmentation

362 kHz: after 1 hour +30% compared to silent conditions

Outlook and conclusion

- **Zn and Ni show very different repartitions in the clay** (Tessier sequential extraction protocol) and consequently **very different leaching behaviours** with HCl 0.1 M: fast and high yield for Zn, slow and less efficient for Ni.
- **Zn:** negative impact of 20 kHz, due to creation of new adsorption sites; non-significant one from 362 kHz.
- **Ni:** similar negative impact of 20 kHz, though higher desorption from F5; positive effect (+30% after 1 hour) of 362 kHz US.

References

Choi, J., Lee, D., & Son, Y. (2021). *Ultrason Sonochem, 74*, 105574.
*Park, B., & Son, Y. (2017). Ultrason Sonochem, 35(Pt B), 640-645.
Son, Y., Lee, D., Lee, W., Park, J., Hyoung Lee, W., & Ashokkumar, M. (2019). *Ultrason Sonochem*, 58, 104599.
Kim, S., Lee, W., & Son, Y. (2016). *Jpn. J. Appl. Phys.*, 55(7S1), 07KE04.
Hwang, S.-S., Park, J.-S., & Namkoong, W. (2007). *J Ind Eng Chem*, 13(4), 650-656

Acknowledgements

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