The application of quartz crystal microbalance to measure sludge rheology

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Introduction

The UK nuclear industry is currently transitioning into a stage of decommissioning and post-operational clean out.

The Sellafield site is the UK's largest nuclear site:

- **Over 10,000 employees;**
- **Over 1,000 nuclear facilities;**
- **Site measures 6 km^2 across.**

Clean-up and remediation costs are estimated to be <u>£53 bn</u> over the next 100 years.

New techniques and technologies need to be developed in order to address the decommissioning challenges.

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The sludge problem…

Ponds

First Generation Magnox Storage Pond:

1) 14,000 m^3 of contaminated water;
2) 1,200 – 1,500 m^3 of radioactive sludge;
3) Mostly composed of $Mg(OH)_2$.

<u>Rheology is a key physical parameter needed in the design of mobilization and transfer processes.</u>

HAST

Silos

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Objective

The current study assesses the applicability of Quartz Crystal Microbalance (QCM) as a technique to generate reliable and meaningful sludge rheology data.

Advantages of QCM:
1] Few mechanical parts;
2] Simple to use;
3] Portable.

SRS QCM 200

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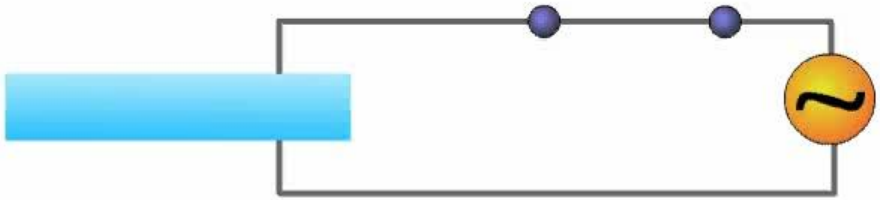
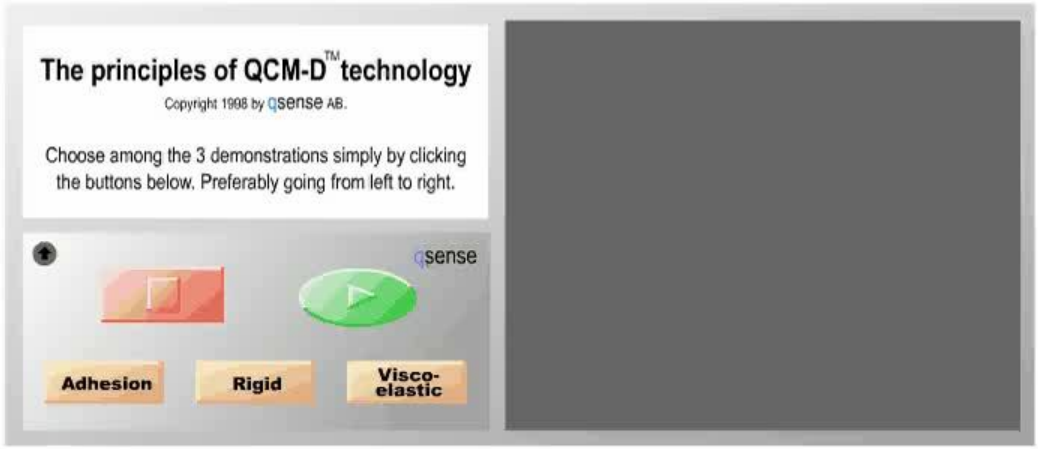
Quartz Crystal Microbalance





$$\Delta \boldsymbol{f} = -2\Delta \boldsymbol{m} f_0^2 / \left(A\sqrt{\mu_q \rho_q} \right)$$

Where Δf = change in frequency of oscillation, Δm = change in mass of the system, f_0 = fundamental mode oscillation frequency, A = contact unit area, μ_q = shear modulus of quartz and ρ_q = density of quartz.

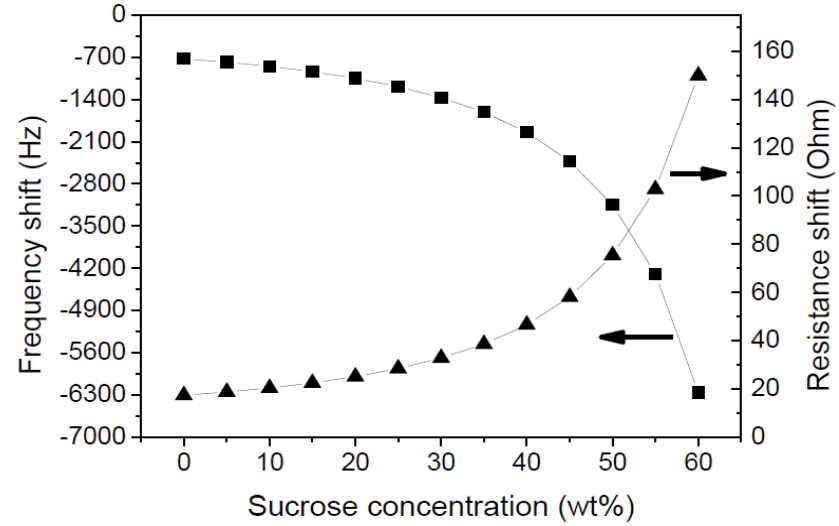
1. Sauerbrey, G., *Verwendung Von Schwingquarzen Zur Wagung Dunner Schichten Und Zur Mikrowagung.* Zeitschrift Fur Physik, 1959. 155(2): p. 206-222.

Quartz Crystal Microbalance

<u>Bulk effects (non-adsorbing)</u>: Frequency and resistance is dependent on the fluid density and viscosity.

$$\Delta f = f_0^{3/2}\left({\eta_s\rho_s}/{\pi\mu_q\rho_q}\right)^{1/2}$$

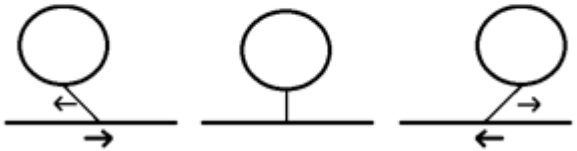
$$\Delta R = \left(\frac{\omega_s L_u}{\pi}\right)\left(\frac{2\omega_s\eta_s\rho_s}{\rho_q\mu_q}\right)^{1/2}$$



<u>Coupled resonance model</u>: Stationary particles weakly linked to the crystal surface increases its frequency of vibration.

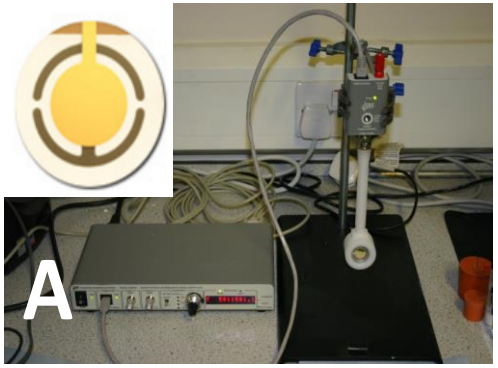
$$\Delta f = m\omega_s^2/\pi A\mu_q\rho_q$$

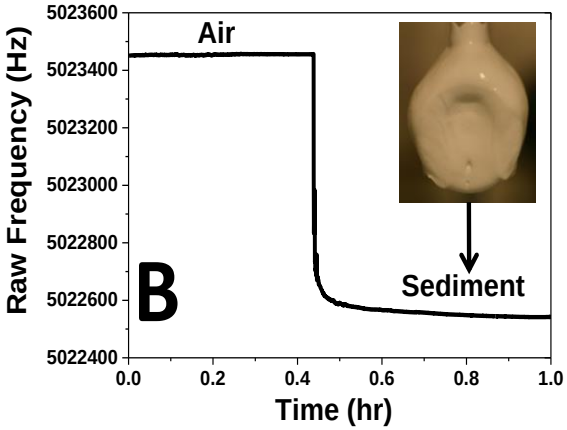
Weakly bound particle link pulls vibrating crystal back to its original position - this acts as a 'restoring force'



1. Kanazawa, K. K. and J. G. Gordon (1985). "Frequency of a Quartz Microbalance in Contact with Liquid." <u>Analytical Chemistry</u> 57(8): 1770-1771.
2. Martin, S. J., et al. (1991). "Characterization of a Quartz Crystal Microbalance with Simultaneous Mass and Liquid Loading." <u>Analytical Chemistry</u> 63(20): 2272-2281.

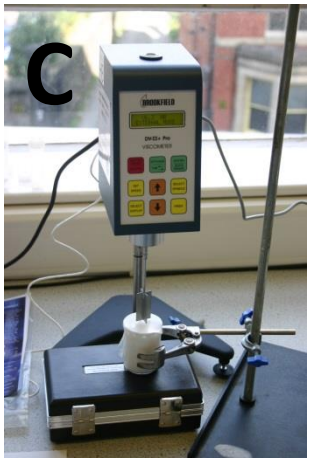
Materials and methods





A] Quartz crystal microbalance (SRS QCM200, 5 MHz)

B] Raw resonance frequency as the sensor is submerged into the sludge.



C] Brookfield DV II+ Viscometer

$$T(max) = \frac{\pi D^3}{2}\left(\frac{H}{D} + \frac{1}{6}\right)\tau_y$$

Where $T(max)$ = maximum torque, D = vane diameter, H = vane height and τ_y = shear yield stress of the test material.

A] Versamag A (Rohm and Haas)

B] Versamag B (Martin Marietta)

C] Kaolinite

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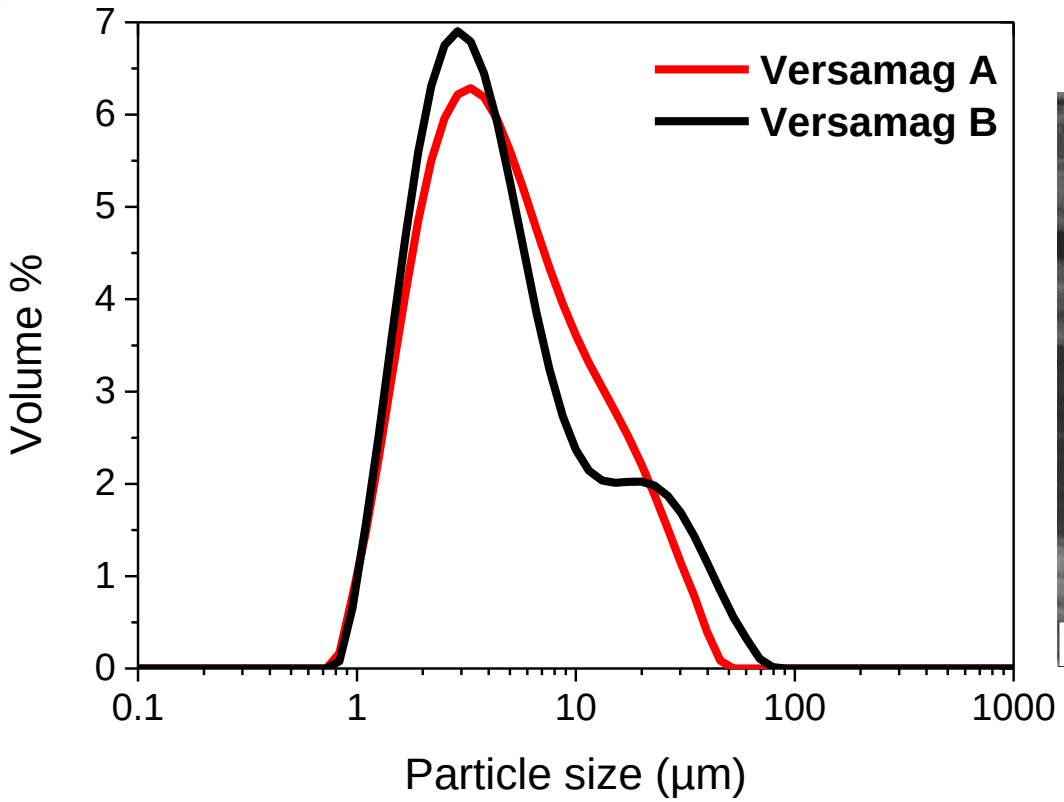
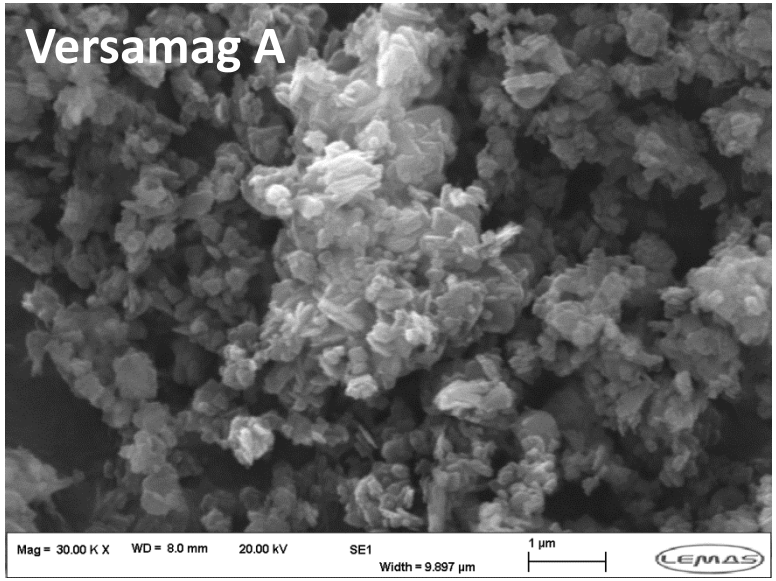
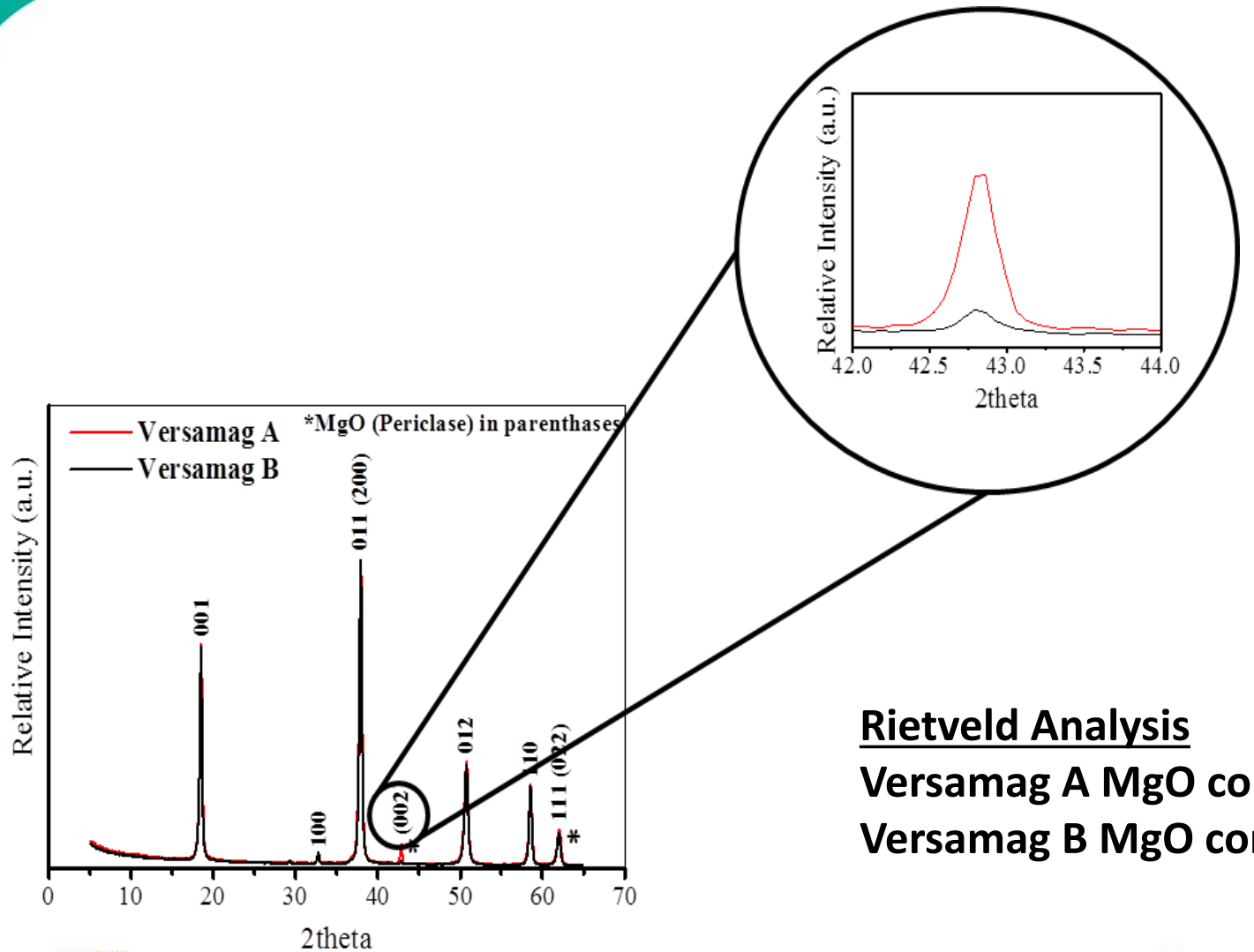
Versamag A and B characterisation





1) Zeta potentials for both samples are -7 mV ± 4 mV at a saturation pH = 10.2.

Versamag A and B characterisation



Rietveld Analysis

Versamag A MgO content = 2.6 wt%

Versamag B MgO content = 0.2 wt%

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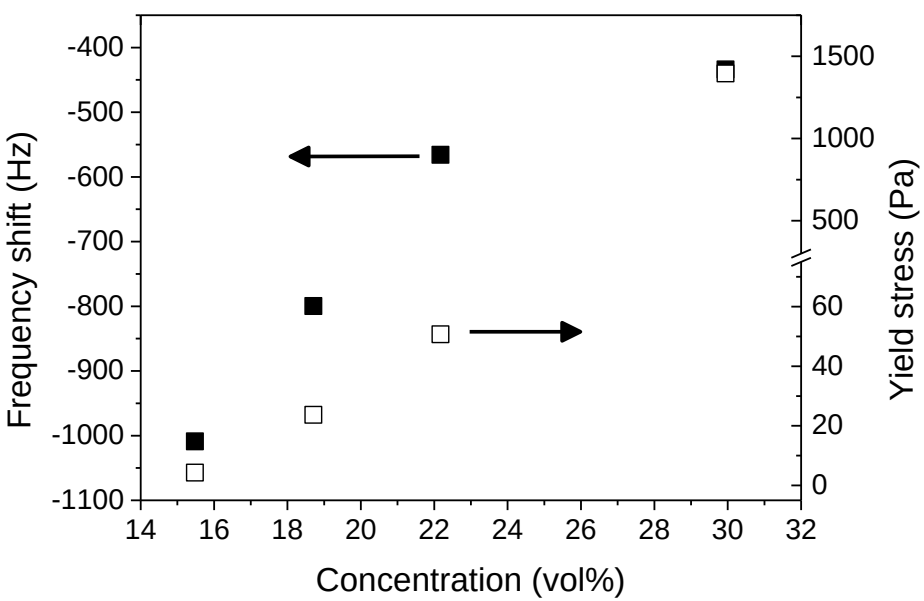
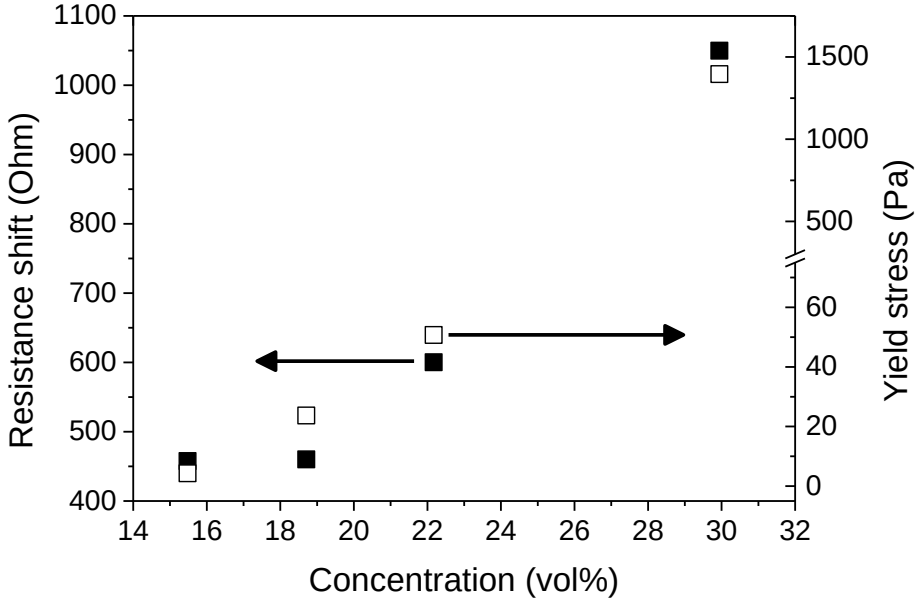
Comparison of vane yield stress and QCM

Versamag A





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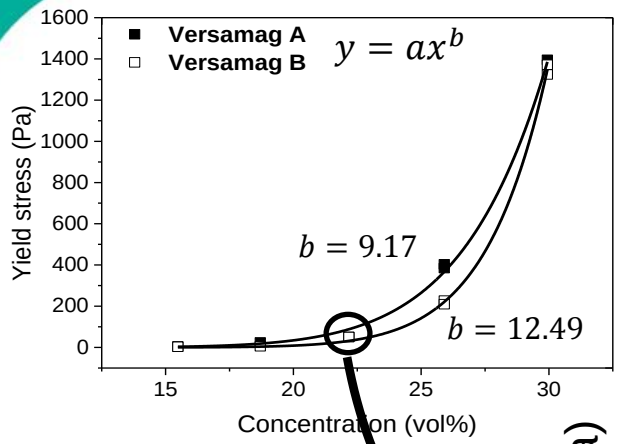
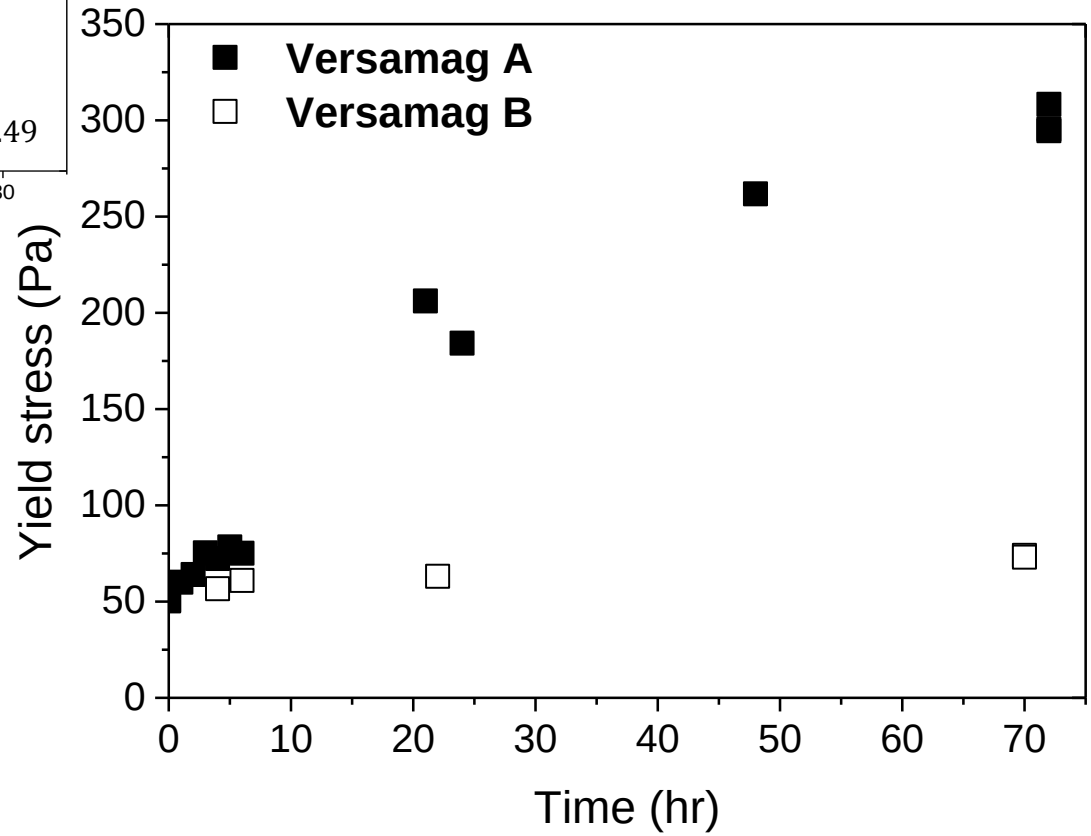
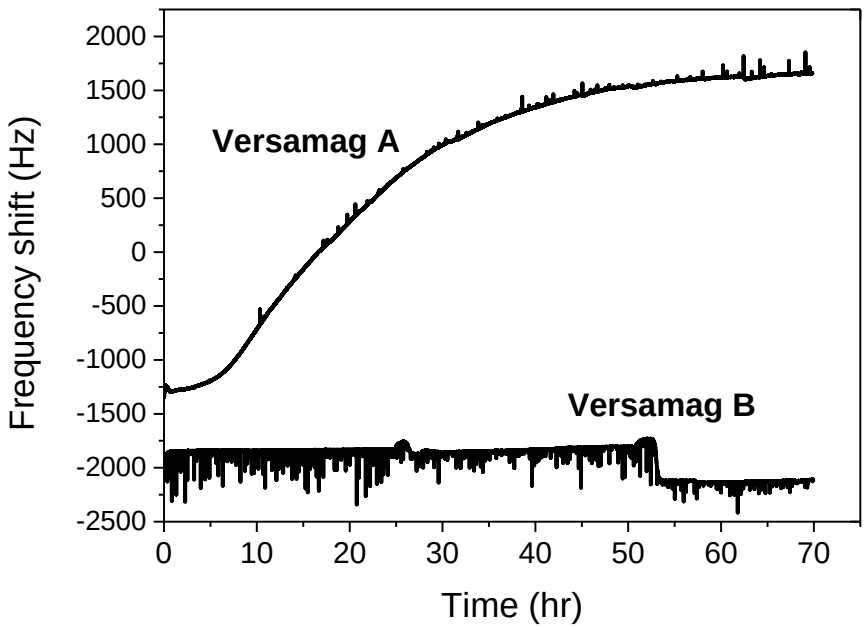
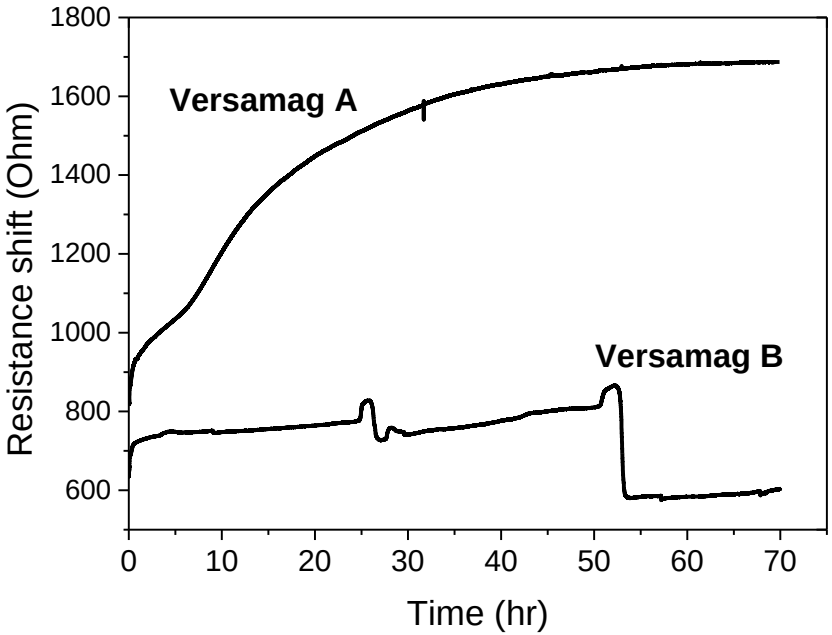
Yield stress: time-dependence





QCM: time-dependence (22 vol%)

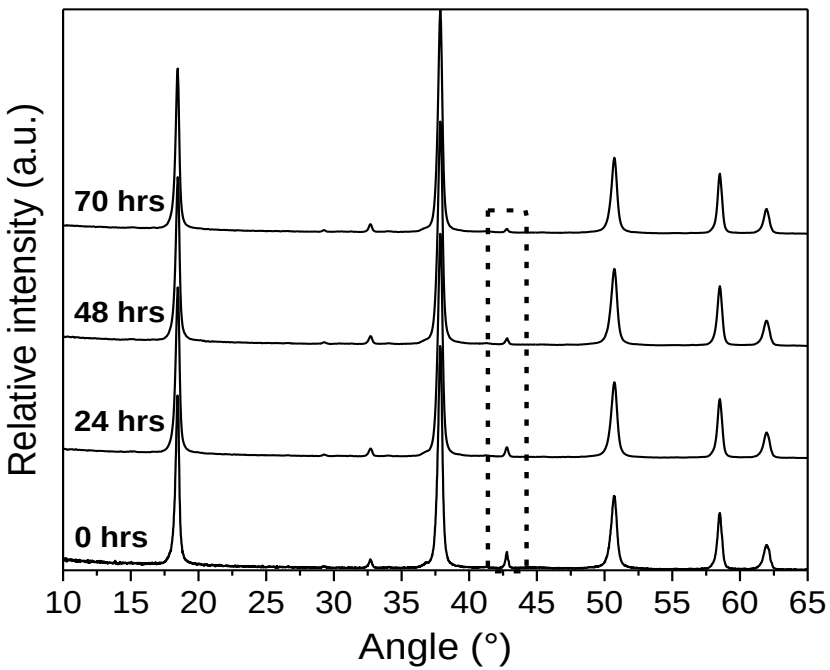


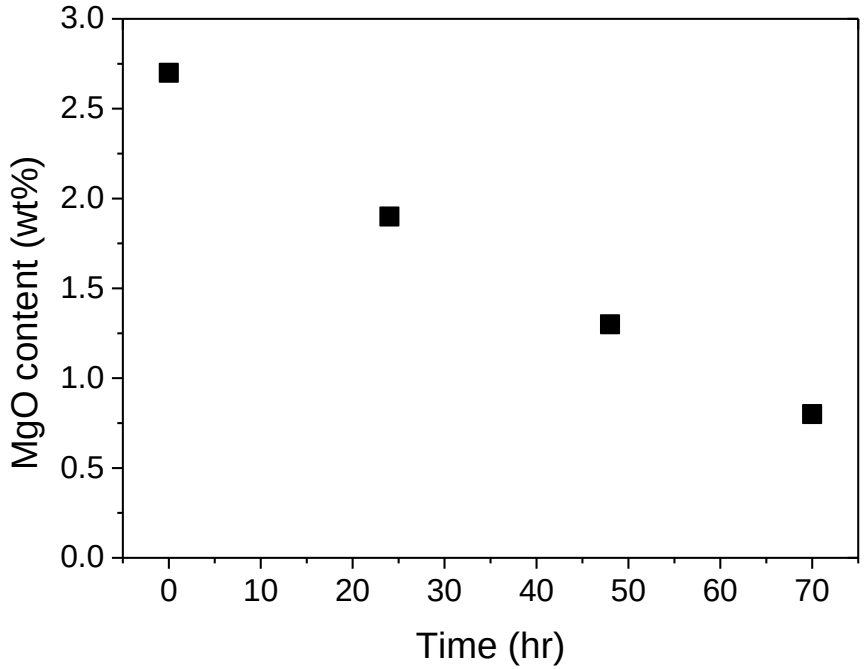


T = 30°C

X-Ray Diffraction: Versamag A

Rietveld analysis on Versamag A samples aged for 0, 24, 48 and 70 hrs showed a decrease in the MgO content as a function of aging time.





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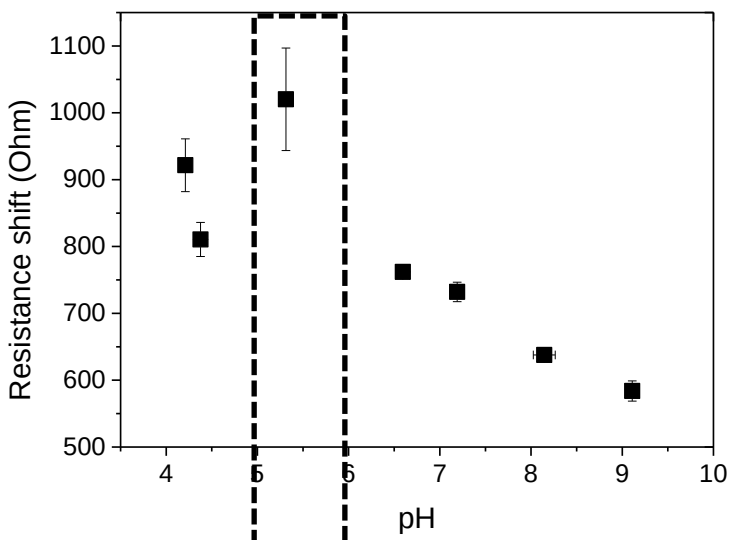
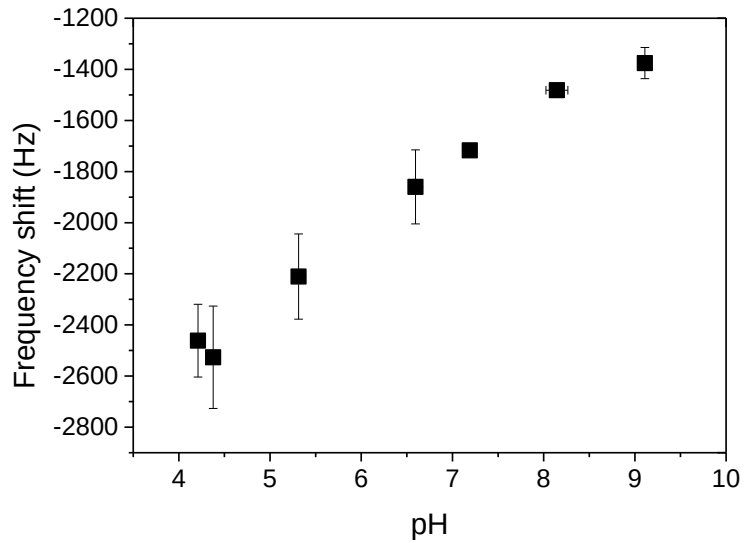
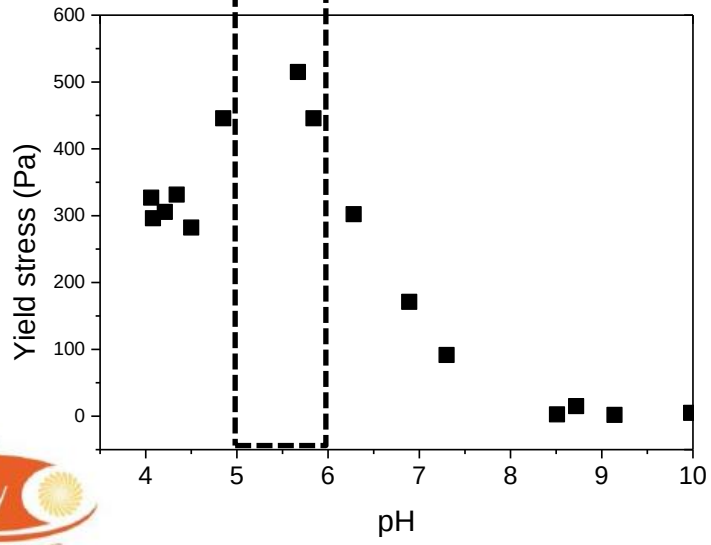
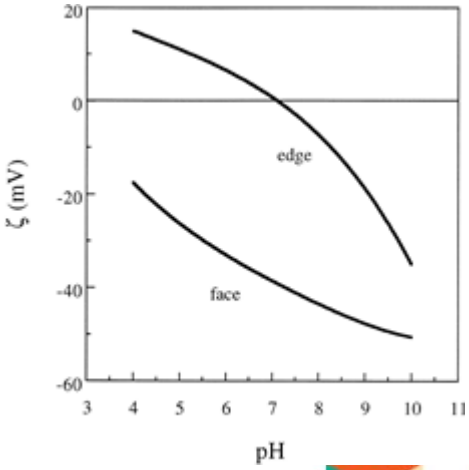
Comparison of vane yield stress and QCM

22.5 vol% Kaolinite









1. Johnson, S.B., et al., *Surface chemistry-rheology relationships in concentrated mineral suspensions.* International Journal of Mineral Processing, 2000. 58(1-4): p. 267-304.

Conclusions

1) As the shear yield stress of the sludge increases the frequency and resistance of the resonating sensor become more positive.

2) The relationship between frequency or resistance and the sludge shear yield stress is comparable at lower yield stresses. At high yield stress the relationship changes and this may result from a change in the sludge-sensor contact mechanics.

3) The QCM resistance response for kaolinite sludge shows good agreement to its yield stress. The frequency response appears sensitive to changes in shear yield stress, although the behavior is not fully consistent.

4) Versamag A has been shown to age to a greater extent than Versamag B. The aging effect is attributed to the higher MgO content.

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Acknowledgements

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