Automated hydrogen permeation analysis using machine learning-enchanced optical microscop

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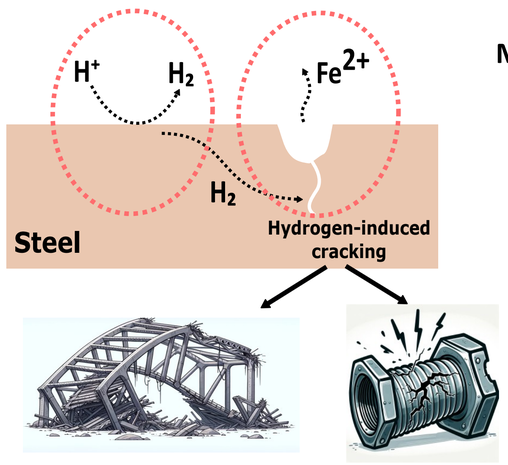
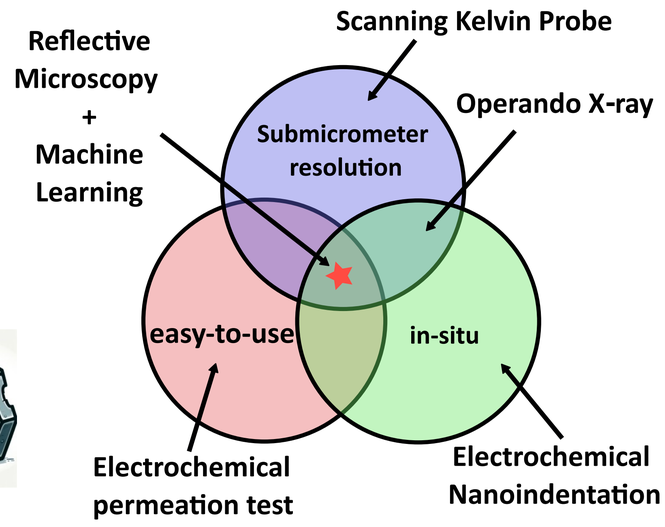
Source codes, cell designs and more

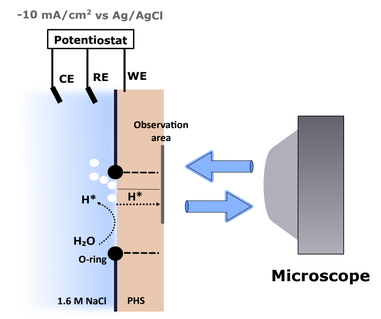
Hydrogen permeation detection



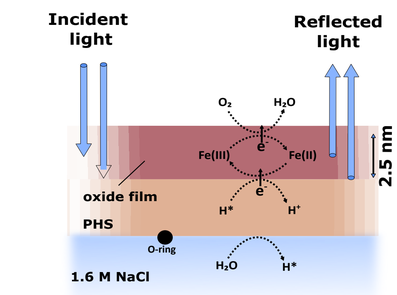


Hydrogen embrittlement is a bulk effect, but hydrogen alters surface films—detectable via surface analysis

Reflective microscopy for hydrogen permeation

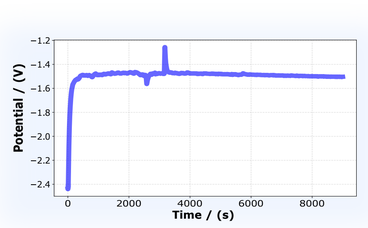


Schematic representation of the RM setup

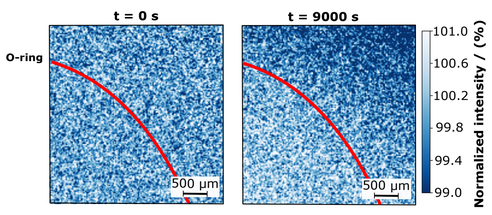


Schematic representation of interfacial electron transfer reactions

- Reflectivity depends on film thickness and composition
- Hydrogen reduces Fe(III) → Fe(II) in film oxides
- This change affects the film's optical properties
- Initial film is composed of Fe_2O_3 (confirmed with XPS) and has thickness of 2.5 nm (confirmed with ellipsometry)

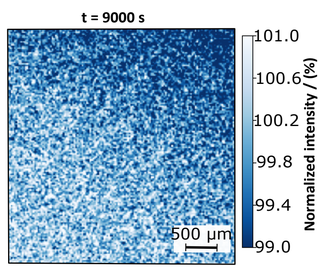


Potential-time curve

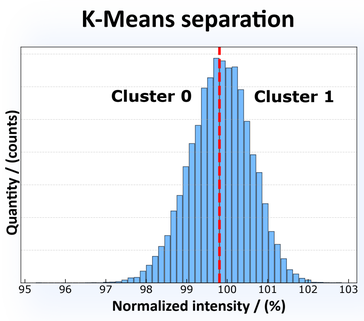


Normalized images before and after polarization

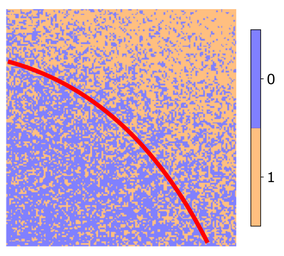
From static image analysis to time-resolved pixel data clustering



Final normalized image
Surface shows faint spatial variations

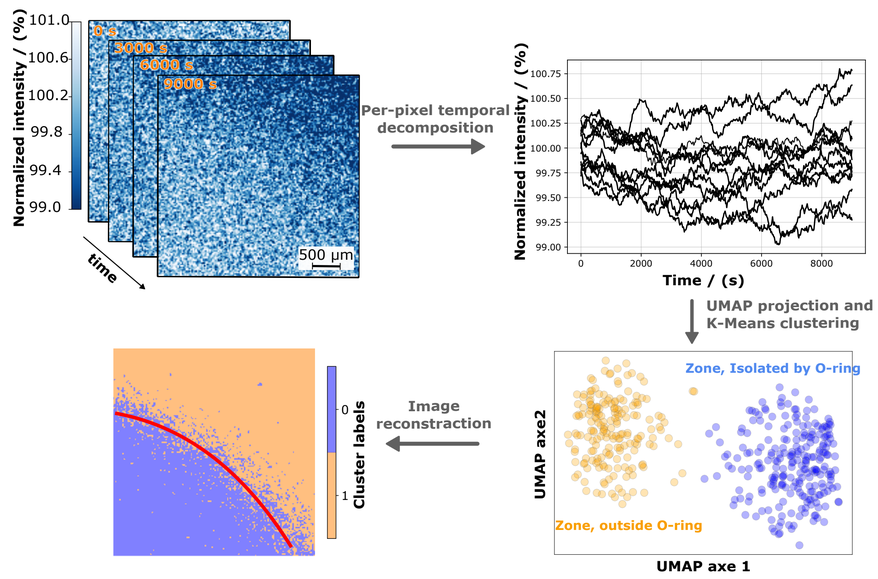


Histogram of pixel intensities is gaussian-like



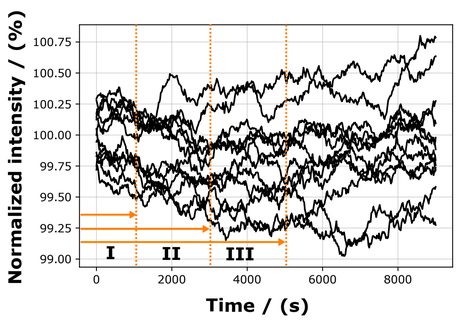
Binarized image
Histogram-based binarization shows two zones

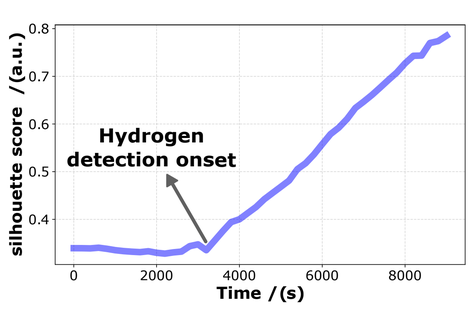
While slight differences are visible, they are too subtle and ambiguous for reliable detection using static image analysis

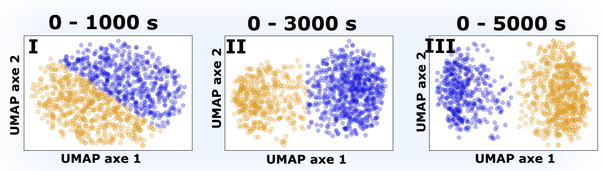


Analysis of pixel-wise time series clearly highlights area, affected by hydrogen

Diffusion coefficient estimation







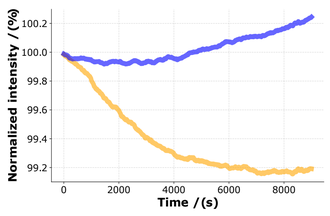
Diffusion estimated using

$$D = \frac{L^2}{6(t_1 - t_0)}$$

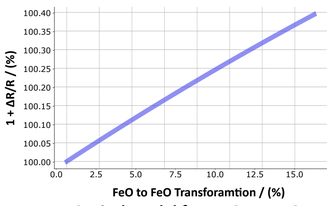
Where L - membrane thickness,
t1 - time, at which hydrogen is detectable
t0 - start time of experiment

Estimated diffusion coefficient (28 $\mu m^2/s$)
ia close to known from literature (32$\mu m^2/s$)

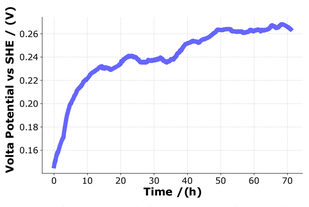
Mechanistic interpretation



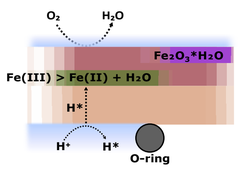
Reflectivity changes inside the O-ring (blue) and outside (orange)



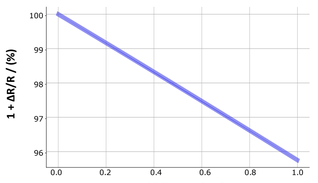
Optical model for Fe_2O_3 to FeO transformation in 3 nm film



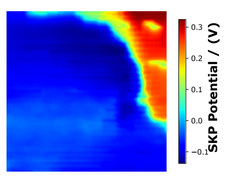
Volta potential vs SHE of the surface during Fe_2O_3 growth



Proposed mechanism: Fe_2O_3 transformation inside O-ring and Fe_2O_3 growth outside



Optical model for Fe_2O_3 growth



SKP map recorded immidiately after hydrogen charging

Optical modeling and SKP analysis confirms the proposed mechanism

Conclusions

- Hydrogen permeation can be quantitavely assessed using reflective microscopy
- UMAP + K-Means pipeline improves sensitivity of microscopy and allows automated hydrogen detection; RM with statistical analysis allows quantification of changes in oxidation state in nanometric layers
- RM is easy to couple with other techniques

Acknowledgements & references

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ANR JCJC, program (OCTAWA project, ANR-34 15 22-CE29-0010-01).

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