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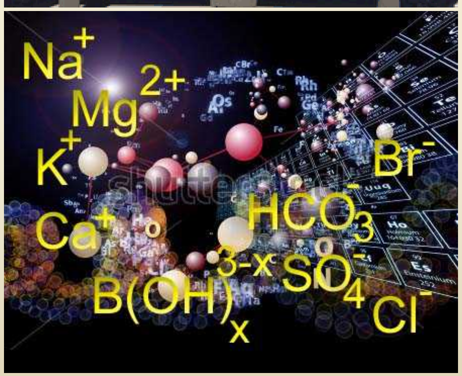

Dissolution of International Simple Glass (ISG) in Sodium Chloride Brine Solution

GOMD Annual Meeting

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U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

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Objectives

- Quantify the dissolution rate of the International Simple Glass (ISG) over a range of NaCl concentrations at 90^{o}C and pH(25^{o}C) = 9 using:
  - Chemical assay on powders
  - Chemical assay on monoliths
  - Interferometry on same monoliths.
- Powder dissolution rate vs. monolith rate; geometric or BET normalization?
- Compare the interferometry results with those of chemical assay.
- **Evaluate if dissolved NaCl enhances or suppresses rates.**

Motivations

- No decision yet on the geologic setting of the repository for high level waste.
  - Granite
  - Shales/mudrocks
  - Tuff
  - Salt (brine solutions)
- Repeated cycles of evaporation/condensation result in high ionic strength brines.
- Are there sufficient data to evaluate the effects of brines on glass dissolution rates?

Previous Work

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- Abdelouas, A., Crovisier, J. L., Lutze, W., Müller, R. and Bernotat, W. (1995) *European Journal of Mineralogy* **7**, 1101-1113.
- Grambow, B. and Strachan, D. (1984) In: *Scientific Basis for Nuclear Waste Management VII* (G. L . McVay, ed.) Elsevier Science Publication Co, New York, NY, pp. 623-634.
- Grambow, B. and Müller, R. (1990) In: *Scientific Basis for Nuclear Waste Management XIII* (V. M. Oversby and P. W. Brown, eds.) Materials Research Society, Pittsburgh, PA, pp. 229-240.
- Grambow, B., Loida, A., Kahl, L. and Lutze, W. (1995) In: *Scientific Basis for Nuclear Waste Management XVIII* (T. Murakami and R. C. Ewing, eds.). Materials Research Society, Pittsburgh, PA, pp. 39-46.
- Luckscheiter and Nesovic (1997) *Waste Management* **17**, 429-436.
- McGrail, B. P., Pederson, L. R. and Petersen, D. A. (1986) *Physics and Chemistry of Glasses* **27**, 59-64.
- Pederson, L. R., McGrail, B. P., McVay, G. L., Petersen-Villalobos, D. A. and Settles, N. S. (1993) *Physics and Chemistry of Glasses* **34**, 140-148.
- Strachan, D. M. (1983) *Nuclear and Chemical Waste Management* **4**, 177-188.
- Strachan, D. M., Krupka, K. M. and Grambow, B. (1984) *Nuclear and Chemical Waste Management* **5**, 87-99.
- Zimmer, P., Bohnert, E., Bosbach, D., Kim, J. I. and Althaus, E. (2002) *Radiochimica Acta* **90**, 529-535.

- Where rates were measured, massive dilution of solution was required, therefore large uncertainties in rates.
- No systematic evaluation of the effects of ionic strength.
- Both rate *enhancement* and rate *inhibition* have been proposed.
- Is there a better way to measure rates in brines?

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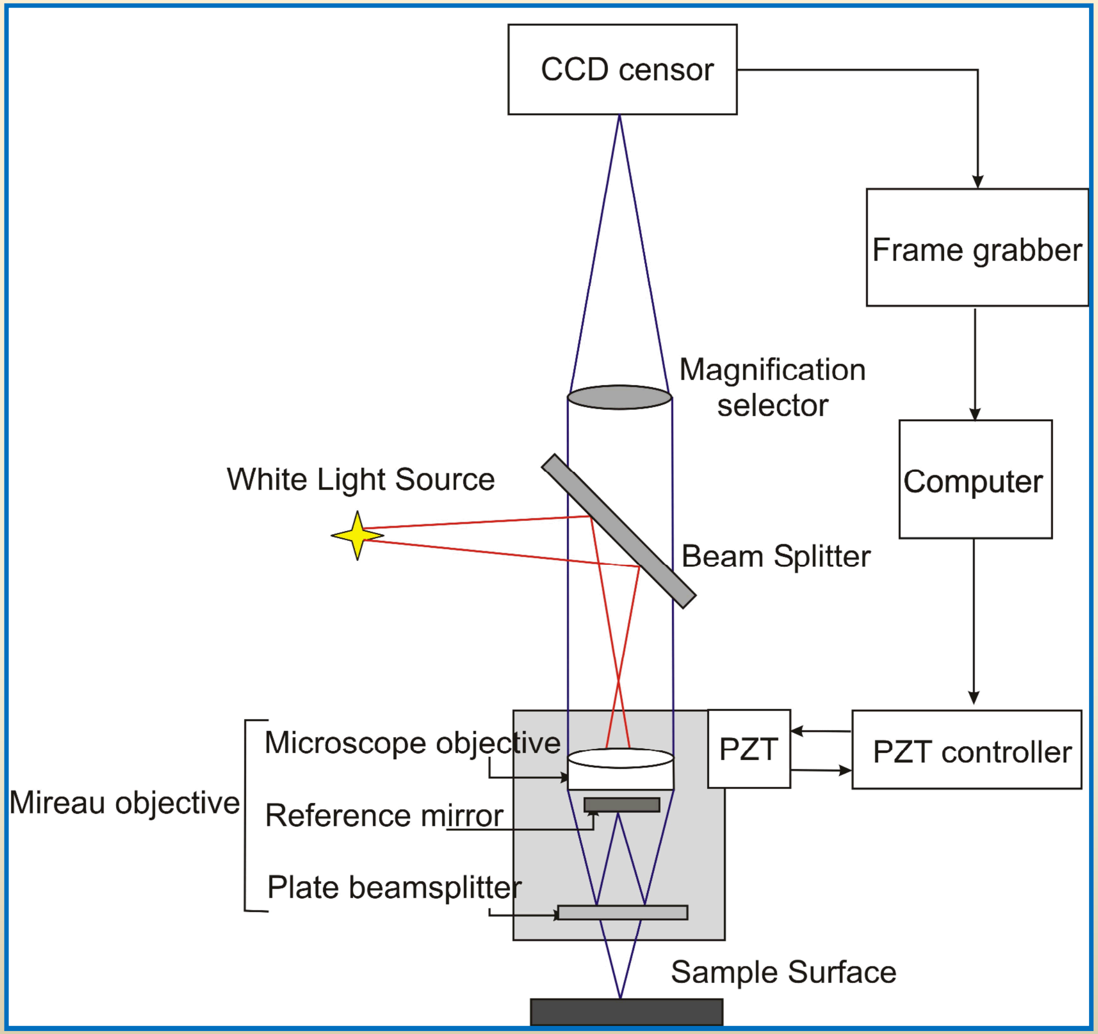
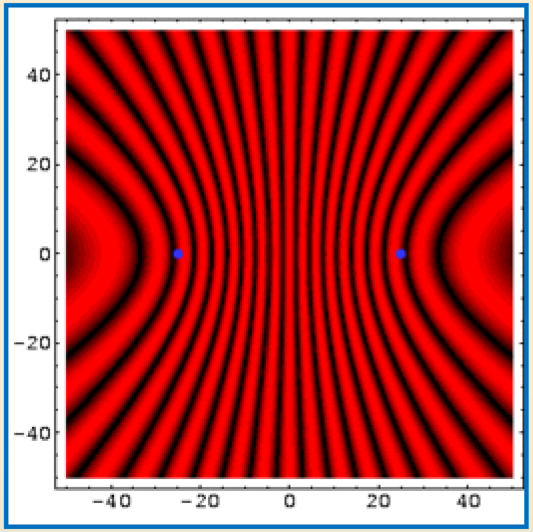
Interferometer

Interferometry measurements





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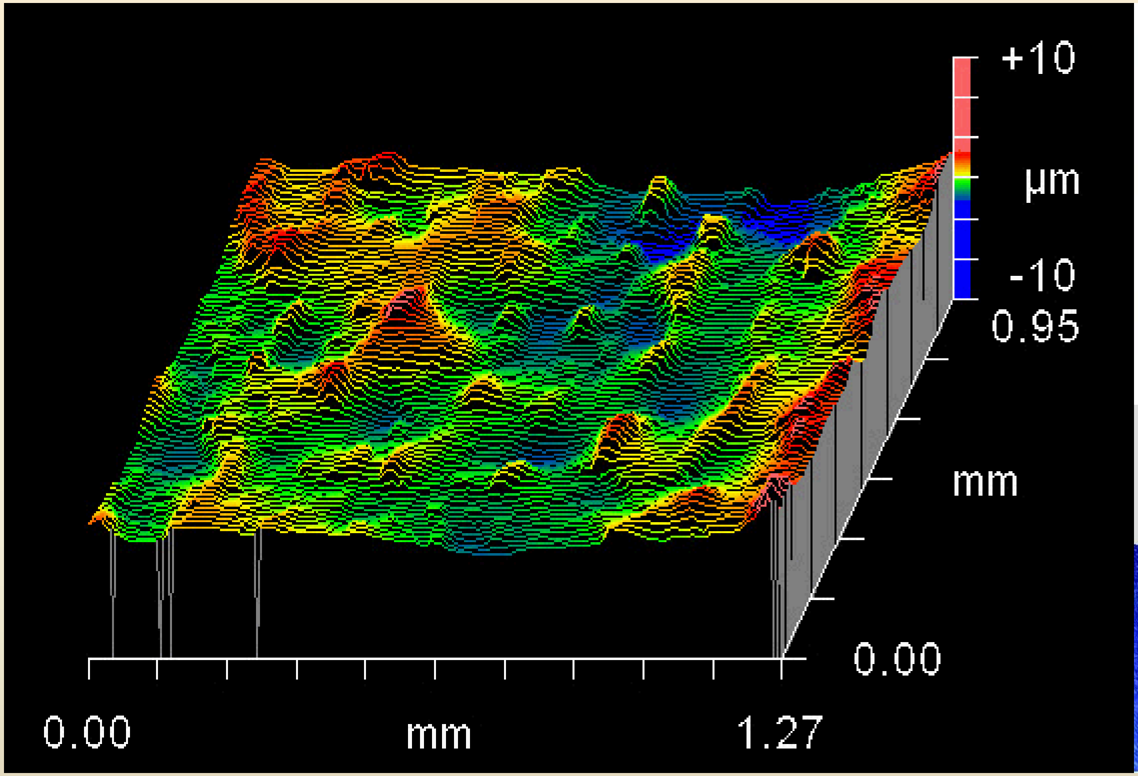
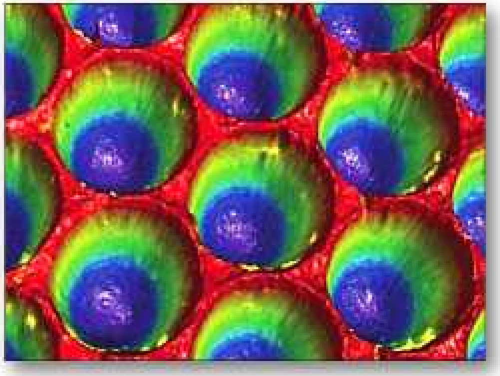
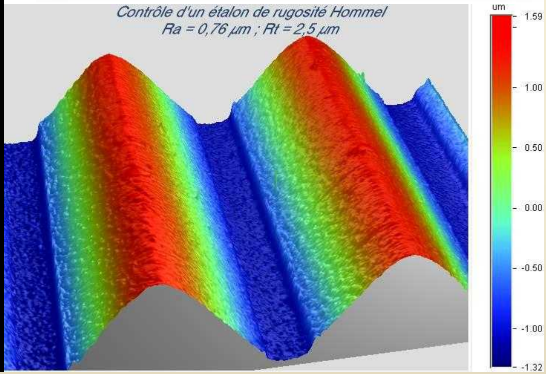
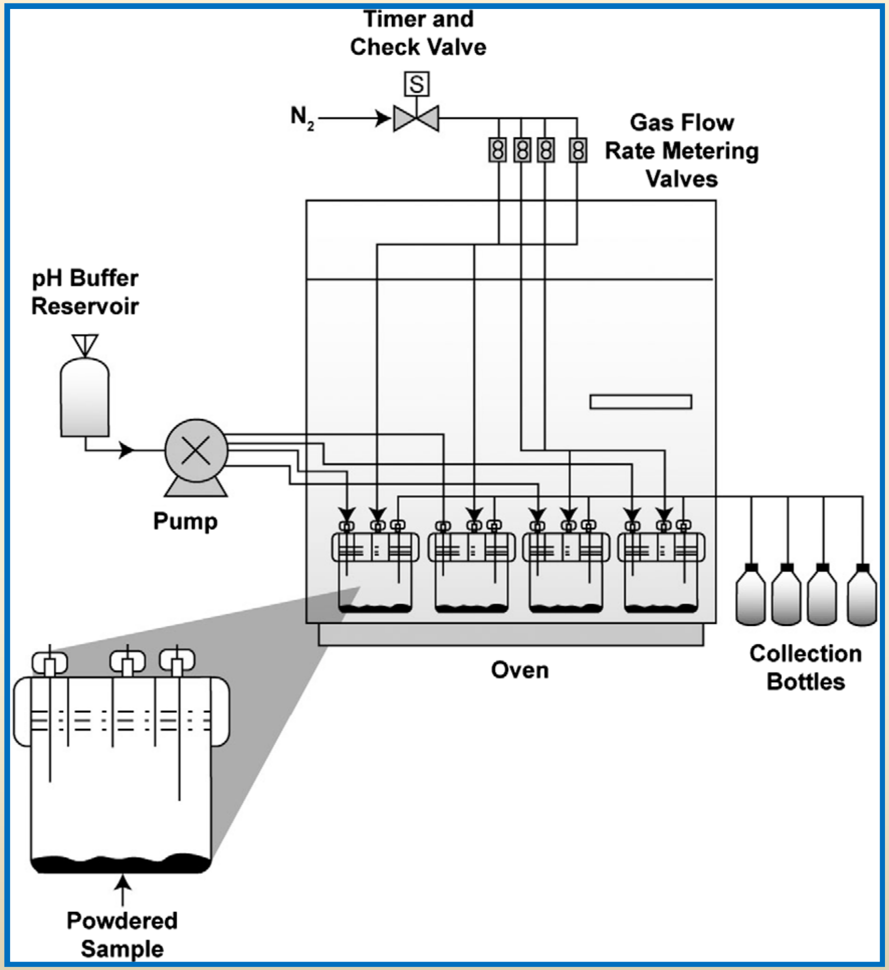
3-D mapping of a surface







Experimental Setup



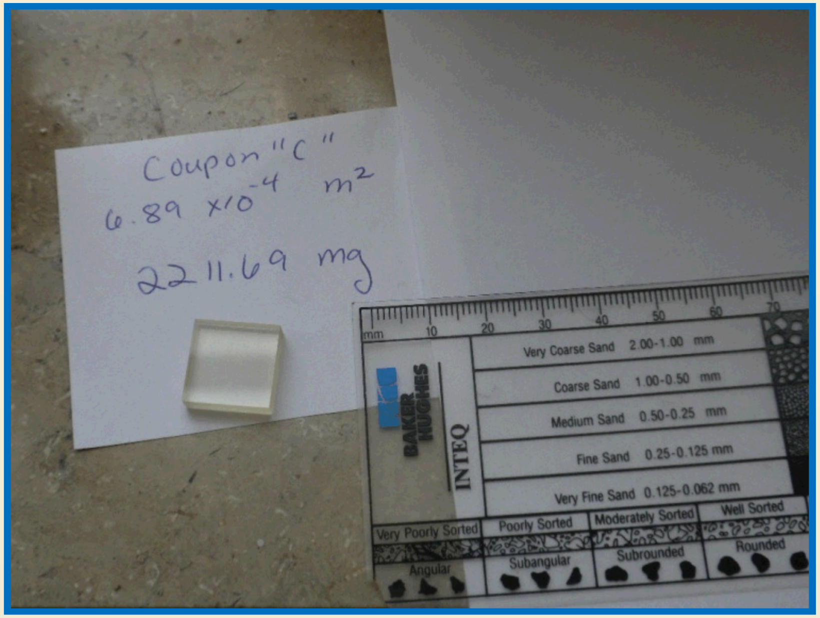
- Glass powder or monolith of known surface area.
- Solution flow rate constant.
- Reactors behave like a CSTR.
- Powder and monoliths in separate reactors, but at same *q*/S ratio.
- Effluent collected and analyzed for release of elements.

$$rate_i = \frac{(C_i^{out} - C_i^{b})q}{f_i S}$$

Solution Preparation

- Solutions buffered with 0.015 M TRIS solution.
- A range of NaCl concentrations (no NaCl and 0.5, 1.0, 2.0, 3.0 and 4.0 M NaCl)
- Initial solutions at pH(25^{o}C) = 9.0
  - Small correction factor required; determined analytically

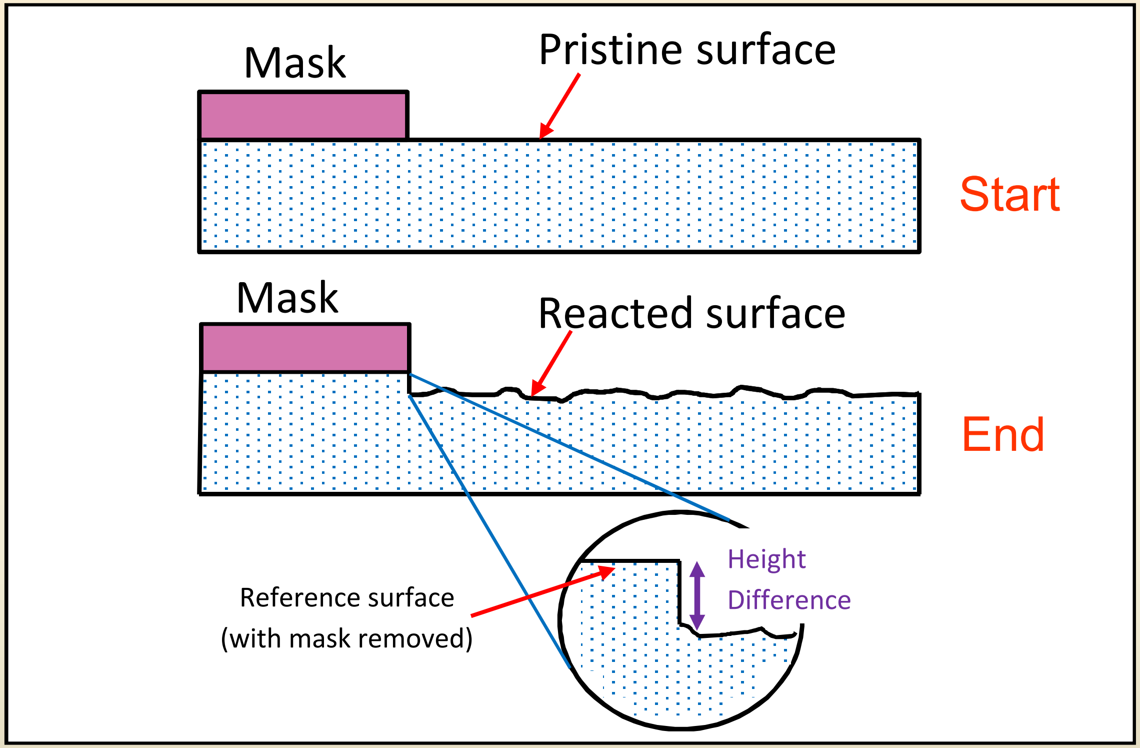
Interferometer rates



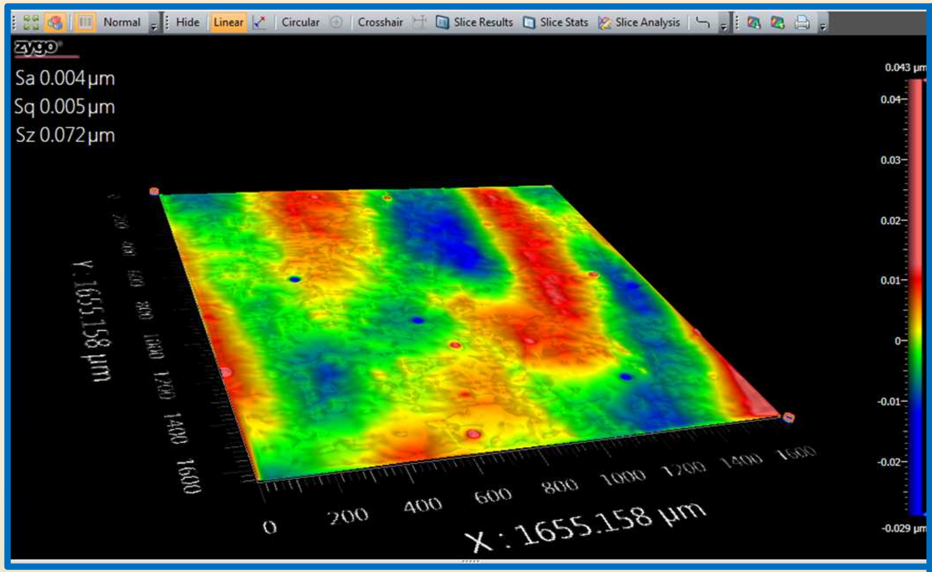
- Cut and polish glass monoliths.
- Measure monolith dimensions using calibrated electronic calipers.
- "Mask" a small portion of the monolith.
- Expose monolith to solution in flow-through reactor.
- Remove monolith from reactor, remove mask, measure the change in height between reference and reaction surface.

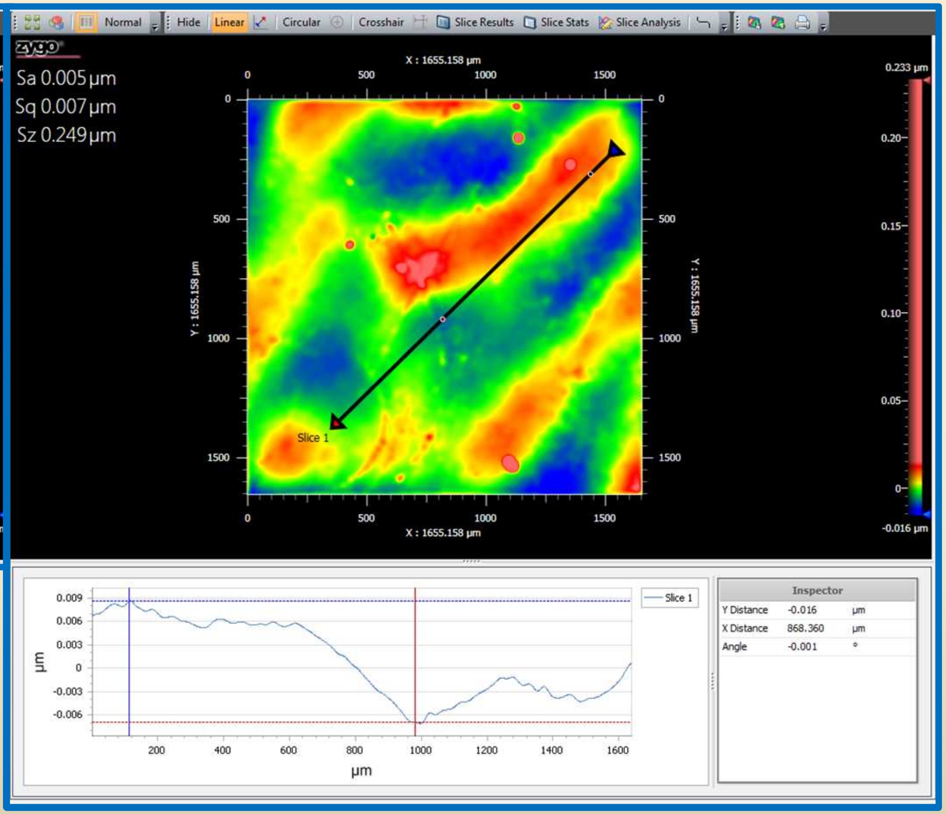
$$Rate = \rho \frac{\Delta h}{\Delta t}$$

Summary of interferometry strategy

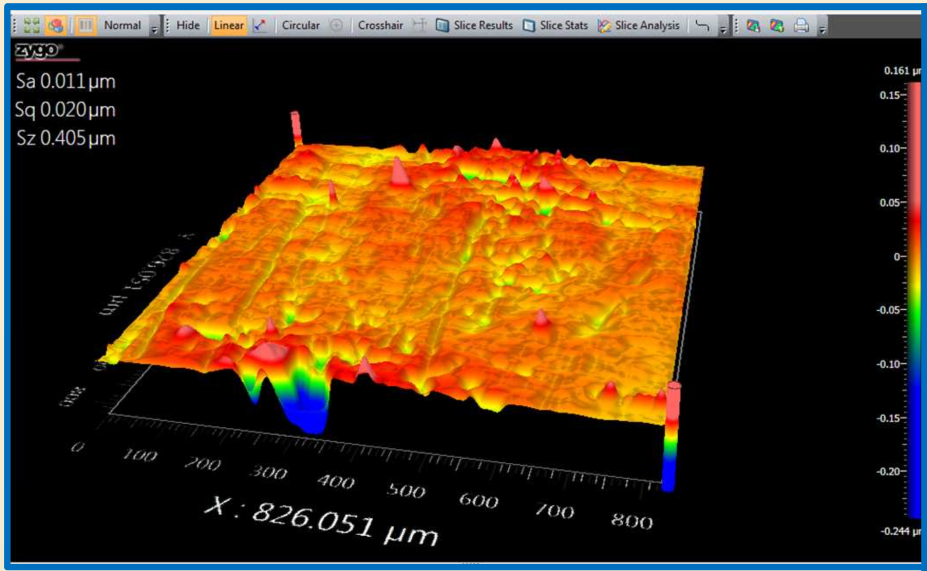


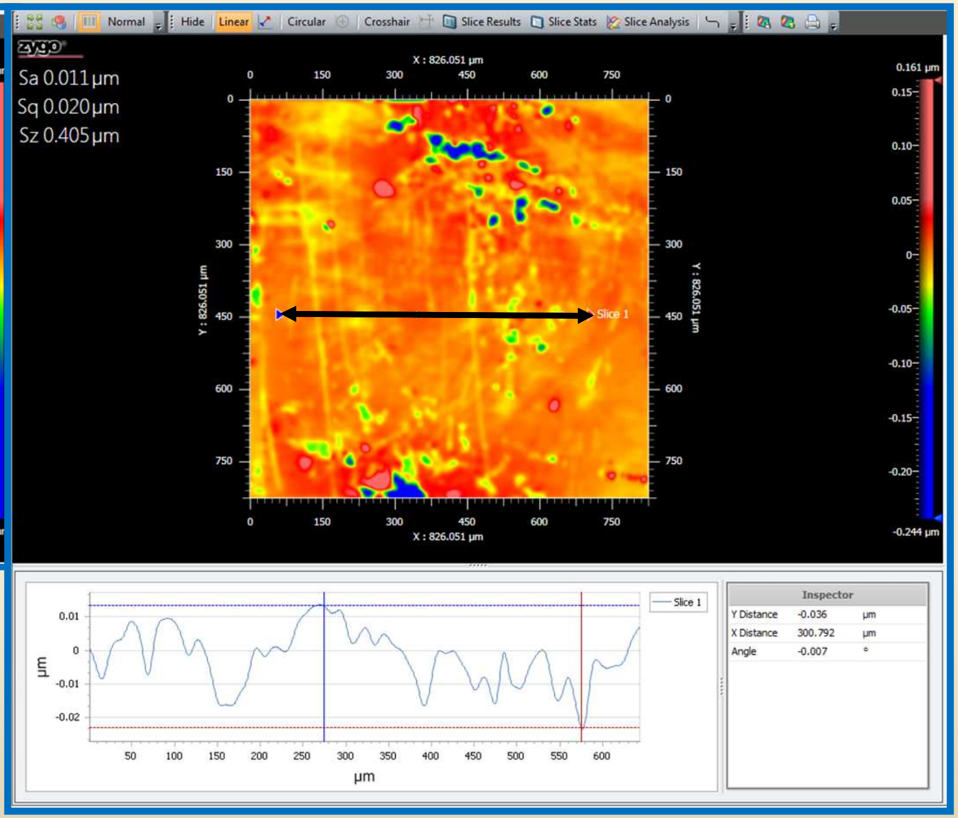
Unreacted surface



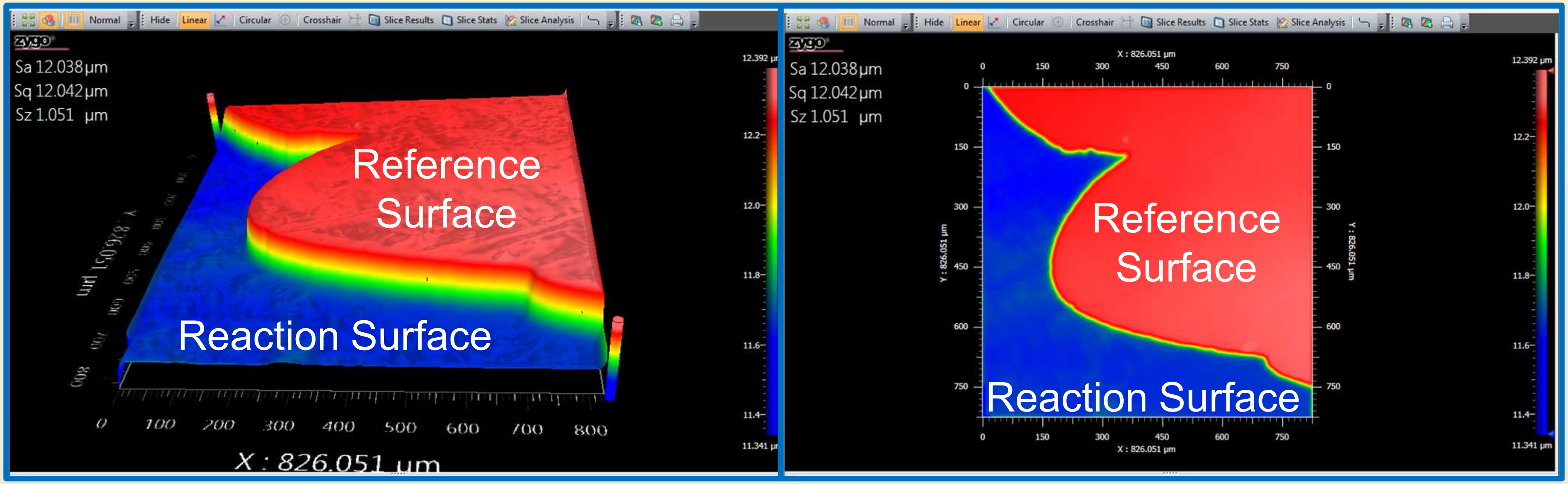


Reacted surface
(No NaCl, 14 days)

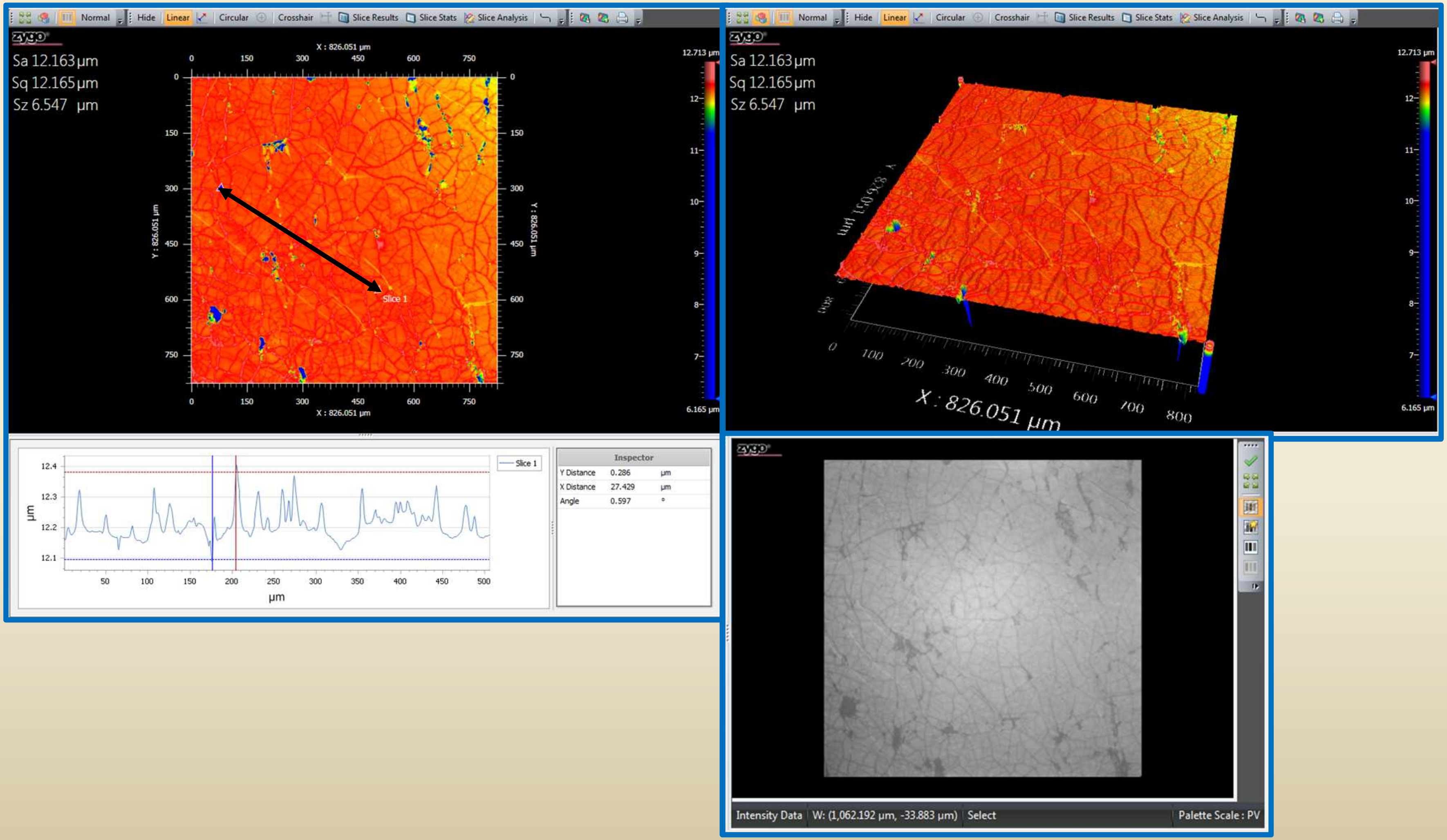




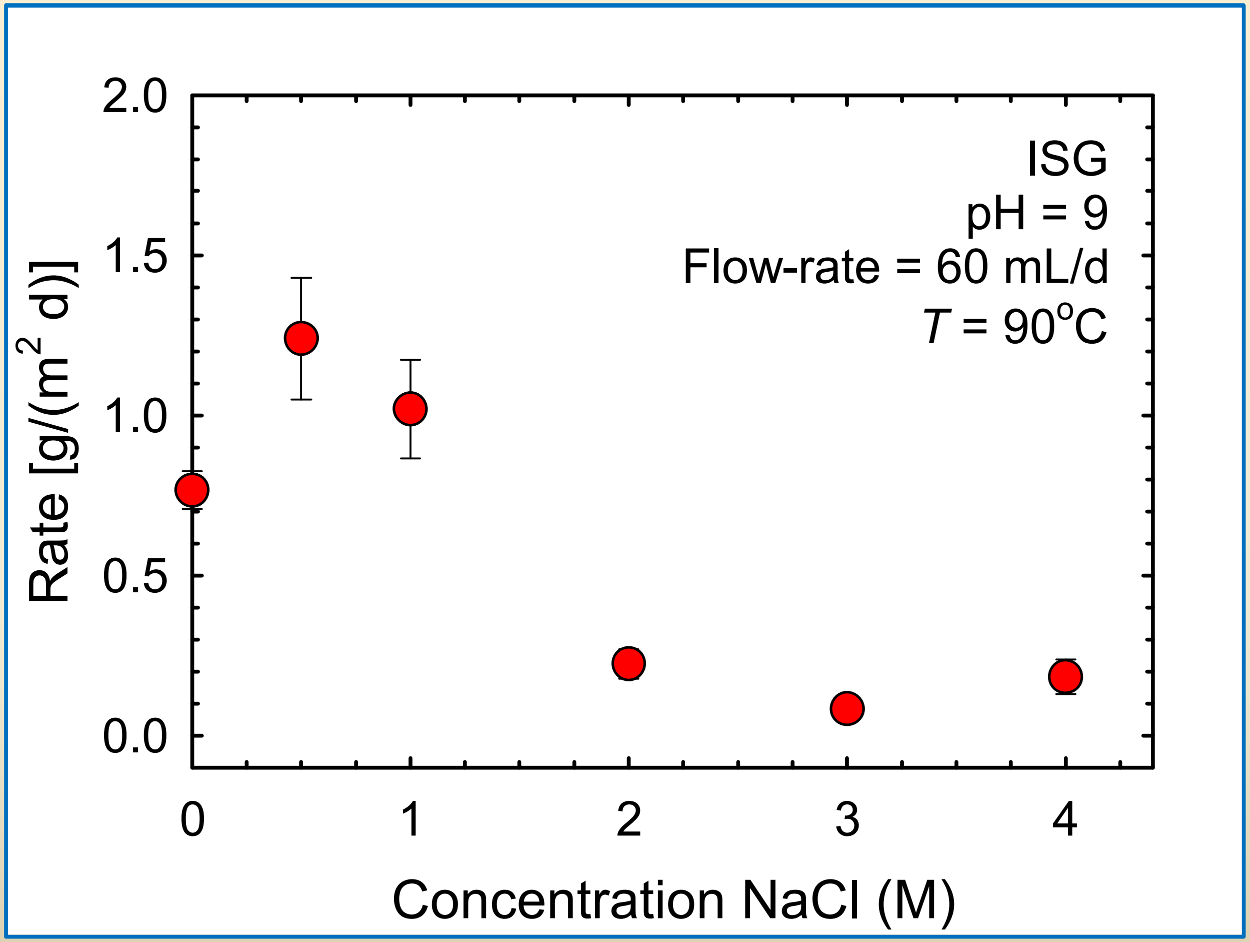
Determining surface retreat
(No NaCl, 6 days)



Reacted Surface in NaCl solutions
(0.5 M NaCl, 14 days)



Dissolution Rates in NaCl Solutions (14 days reaction time)



So, who's right about rates in NaCl solutions?

- The **preliminary** rate values indicate that rates both increase and decrease, depending on NaCl concentration.
  - Relatively low NaCl concentrations: Rate enhancement.
  - Relatively high NaCl concentrations: Rate inhibition.
- Conclusion: Everyone is right!

Acknowledgements

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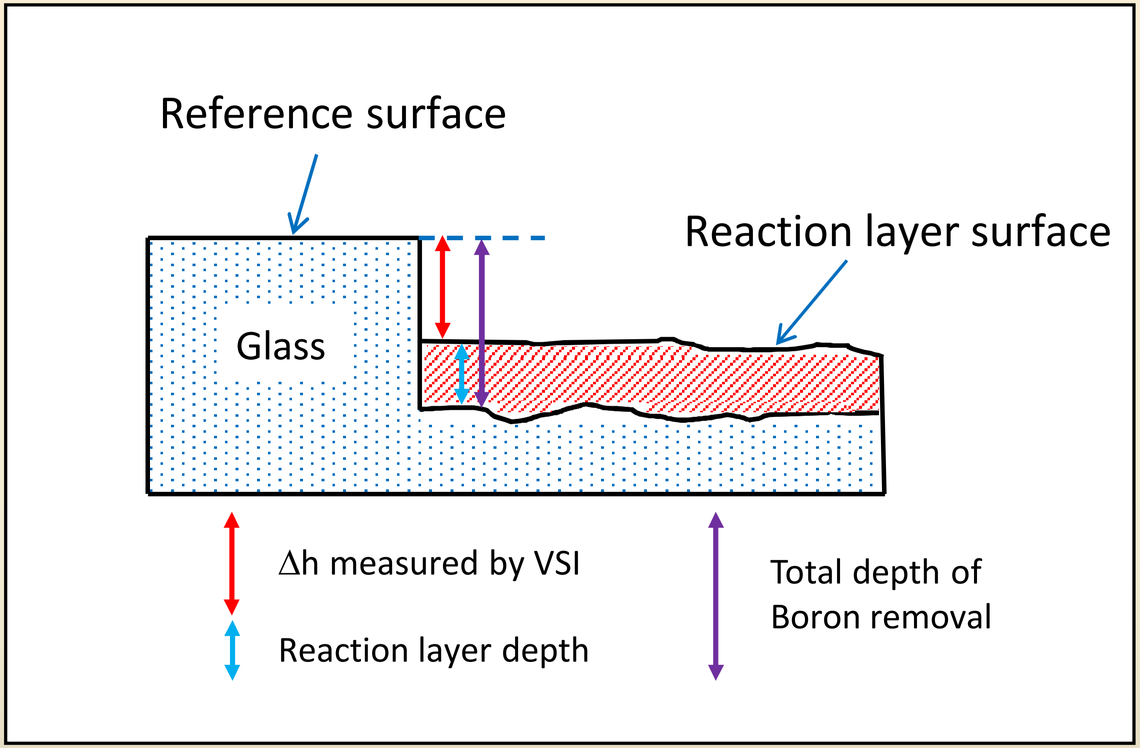
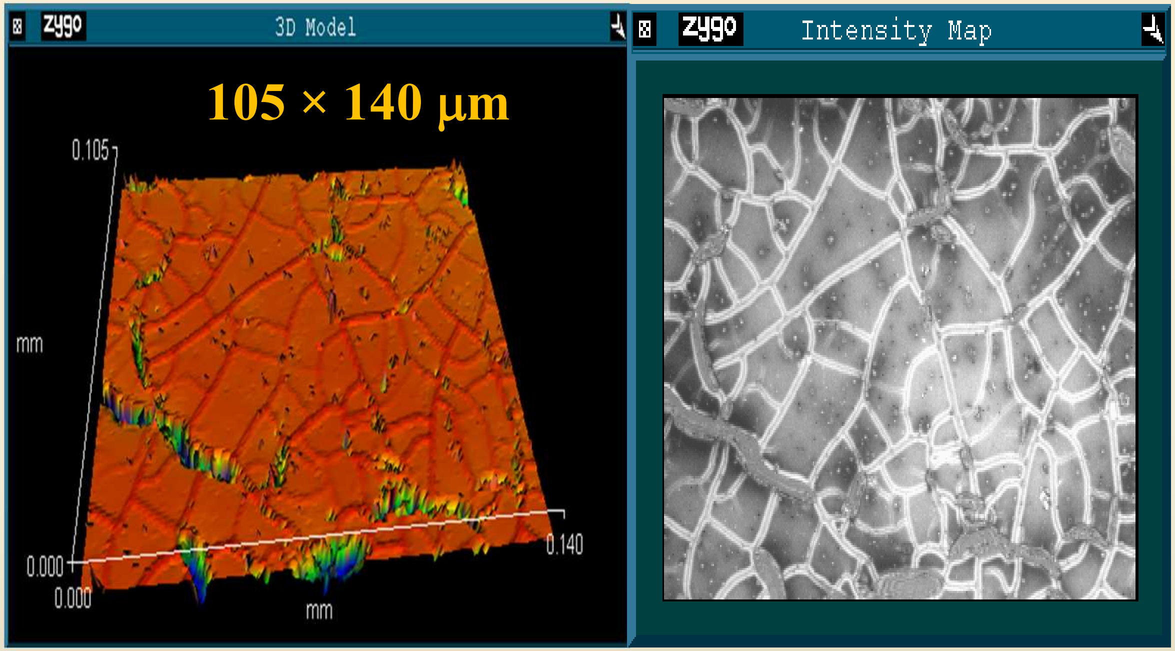
Effect of reaction layer on Interferometer-based rates



"Gel" dehydration effects



Interferometry measurements