SunShot
U.S. Department of Energy

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Predictive physico-chemical modeling of intrinsic degradation mechanisms for advanced reflector materials

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Outline

- Project objectives
- Approach
  - Kinetics and transport
  - Mechanical properties
  - Aging of reflector stacks
- Summary and outlook

Project Team and Acknowledgments

NREL:

- Ross Larsen
- Ray Grout
- Chao Zhang
- Andrew Ferguson
- Robert Tirawat

- Matthew Gray (now with SkyFuel)
- Vince Wheeler (now at Australian National University)

Cost share partners: DLR

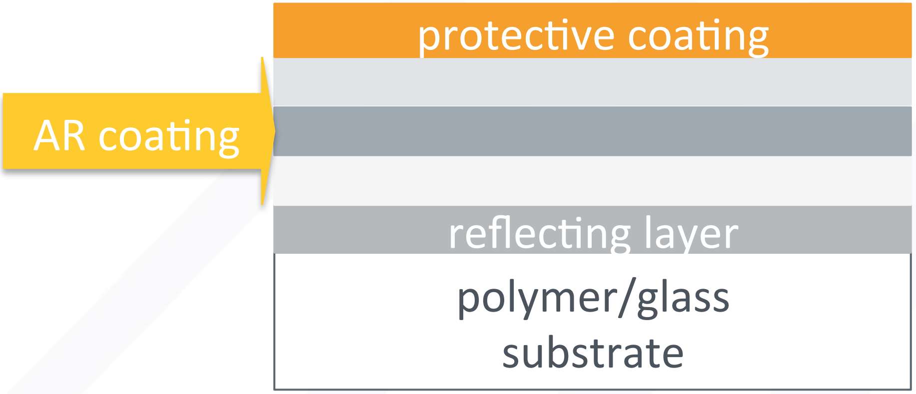
- Peter Heller
- Florian Sutter

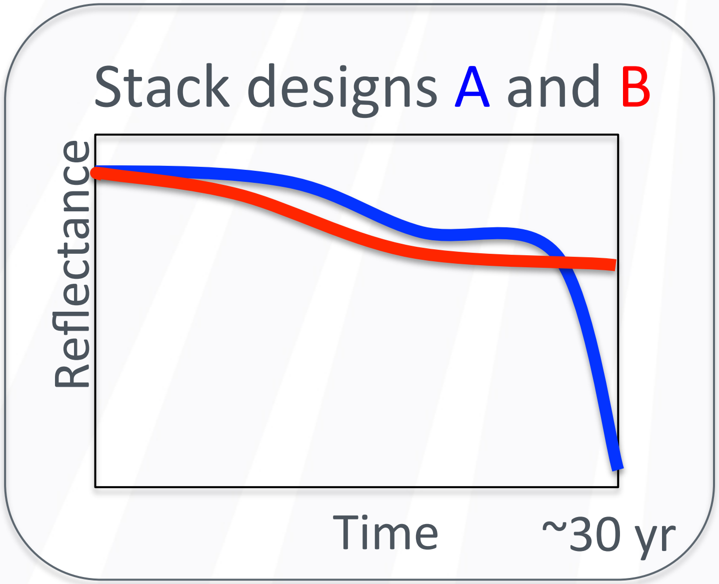
DOE CSP

- DE-FOA-0000861

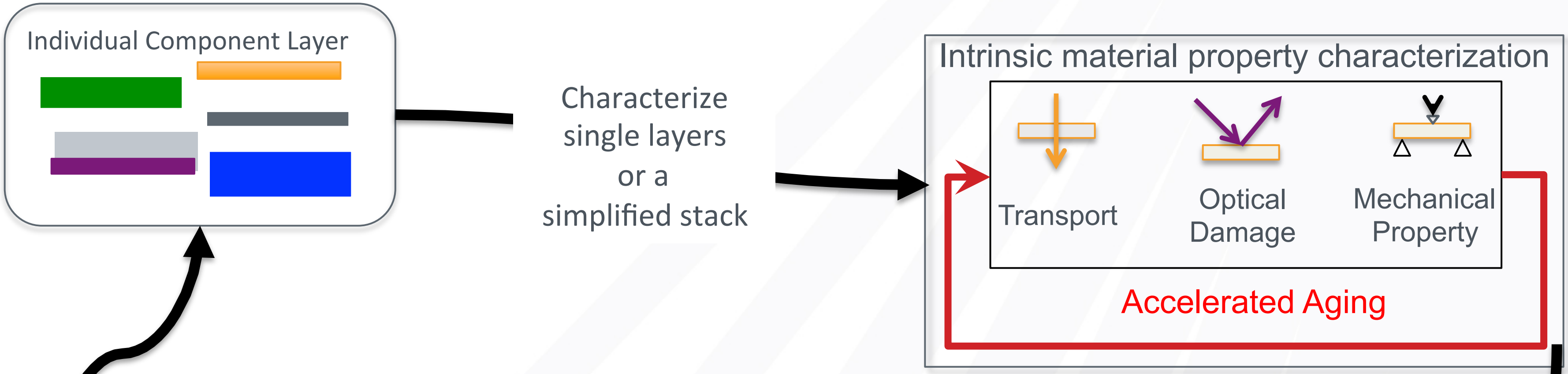
Project Objectives

- Develop novel modeling capability for understanding reflector degradation that combines a reaction-diffusion-optics formalism with experiments to identify underlying rates and mechanisms for degradation
- Create and disseminate a tool, validated by testing against multiple experiments, that can be used by the CSP community to evaluate degradation mechanisms





Project Approach



Individual Component Layer

Characterize single layers or a simplified stack

Intrinsic material property characterization

Transport

Optical Damage

Mechanical Property

Accelerated Aging

Simplified example photo-kinetic scheme

Micro-kinetic Description

$$\frac{\partial n_C(\boldsymbol{r},t)}{\partial t} = D_C \nabla^2 n_C(\boldsymbol{r},t) - \int \alpha(\lambda) I_\lambda(\boldsymbol{r},t)\, d\lambda \left[n_C(\boldsymbol{r},t) - n^*(\boldsymbol{r},t) - n_{bl}(\boldsymbol{r},t) \right]$$

$$\frac{\partial n^*(\boldsymbol{r},t)}{\partial t} = D_* \nabla^2 n^*(\boldsymbol{r},t) + \int \alpha(\lambda) I_\lambda(\boldsymbol{r},t)\, d\lambda \left[n_C(\boldsymbol{r},t) - n^*(\boldsymbol{r},t) \right] - k_{Ox} n_{Ox}(\boldsymbol{r},t) \cdot n^*(\boldsymbol{r},t)$$

$$\frac{\partial n_{bl}(\boldsymbol{r},t)}{\partial t} = +k_{Ox} n_{Ox}(\boldsymbol{r},t) \cdot n^*(\boldsymbol{r},t)$$

$$\frac{\partial n_{Ox}(\boldsymbol{r},t)}{\partial t} = D_{Ox} \nabla^2 n_{Ox}(\boldsymbol{r},t) - k_{Ox} n_{Ox}(\boldsymbol{r},t) \cdot n^*(\boldsymbol{r},t) + S(\boldsymbol{r},t)$$

$$I_\lambda(\boldsymbol{r},t) = I_\lambda(z=0,t) \exp[^{-\alpha(\lambda)\int_0^L \left[n_c(z,t) - n^*(z,t) \right] dz}]$$

$$n_{Ox}(0,t) = kP_{Ox}$$

$$k_{Ox} = k_{Ox}(n_C, n_{bl}, T)$$

$$k = k(T,P)$$

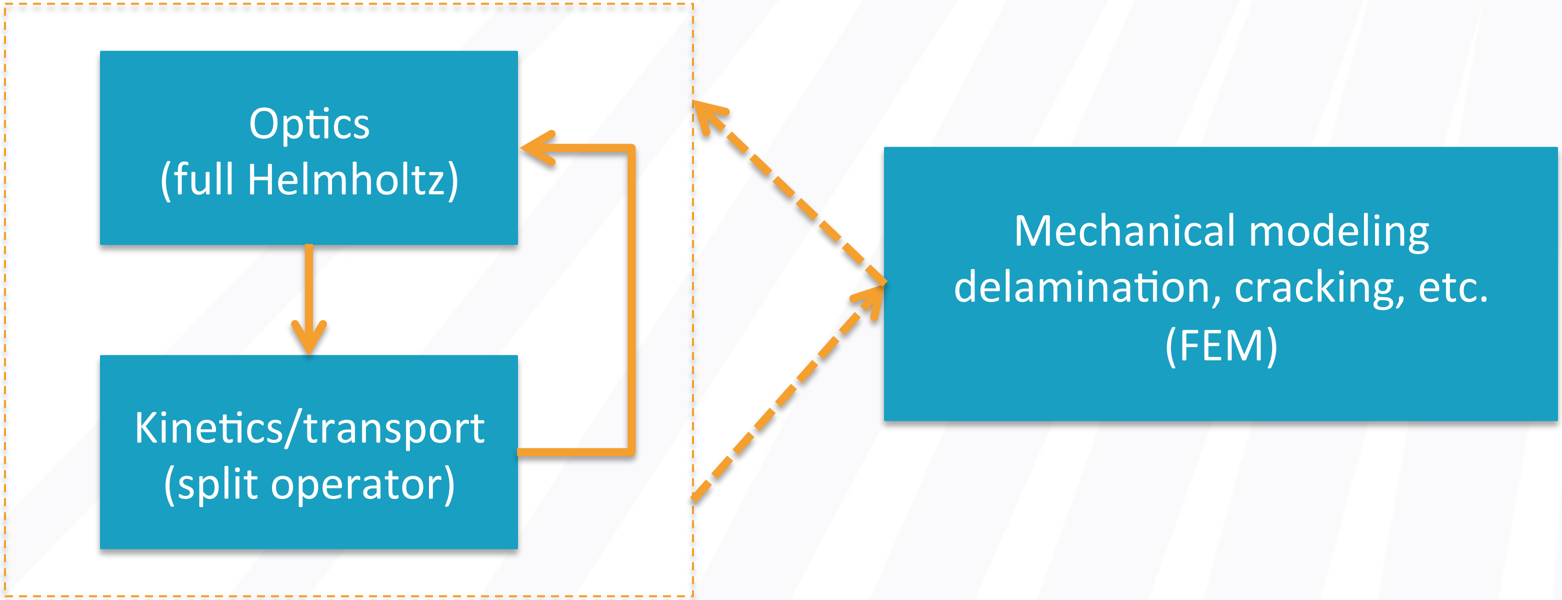
$$D = D(n_C, n_{bl}, n_{Ox}, n_{H_2O}, T)$$

$$\sigma_c = \sigma_c(n_C, n_{bl}, n_{Ox}, n_{H_2O}, T)$$

$$\alpha_\lambda = \alpha_\lambda(n_C, n_{bl}, n_{Ox}, n_{H_2O}, T)$$

Project Approach

- Create and validate models and software to predict and understand degradation of advanced reflectors spanning decades
  - Optics, reaction-diffusion kinetics solver (feedback)
  - Finite element mechanical modeling



Optics
(full Helmholtz)

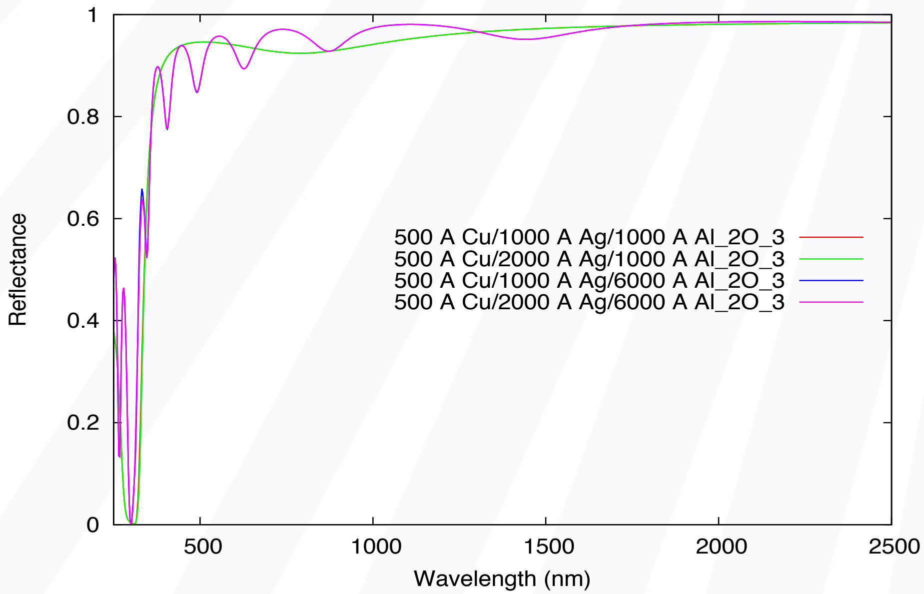
Kinetics/transport
(split operator)

Mechanical modeling
delamination, cracking, etc.
(FEM)

Optics solver

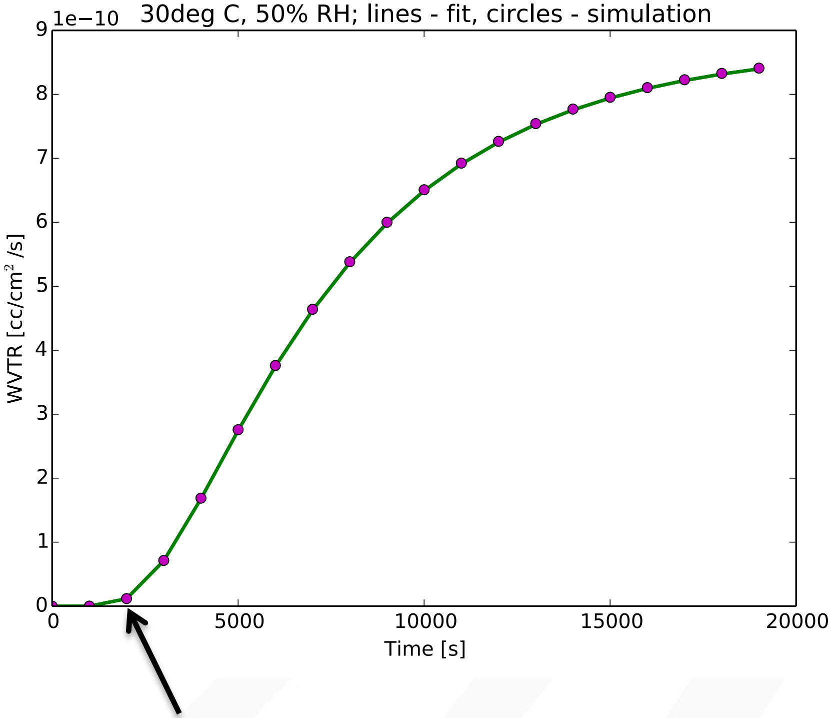
- Transfer matrix method for 1D reflector stacks
  - Fully vectorial (s and p polarization)
  - Numerically exact for piecewise constant stacks
  - Exact reflectance without need for upwinding, boundary elements, etc. (3D version: boundary elements)

Unpolarized reflectance from alumina-coated silver reflector



Kinetics-transport solver

Transient measurement of permeability of PET film to water



Delayed onset consistent with non-Fickian diffusion

Dual sorption:

$$\frac{\partial C}{\partial t} = D_D \frac{\partial}{\partial x}\left[\frac{1}{1+\frac{\alpha C_H'}{(1+\alpha C_D)^2}}\frac{\partial C}{\partial x}\right]$$

Non-linearity in transport and sorption requires novel solver for boundaries

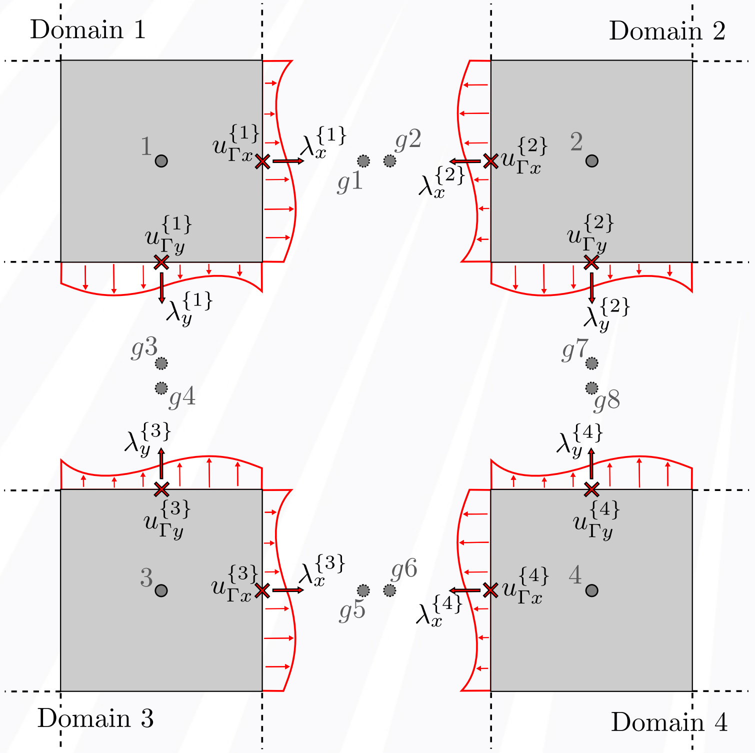
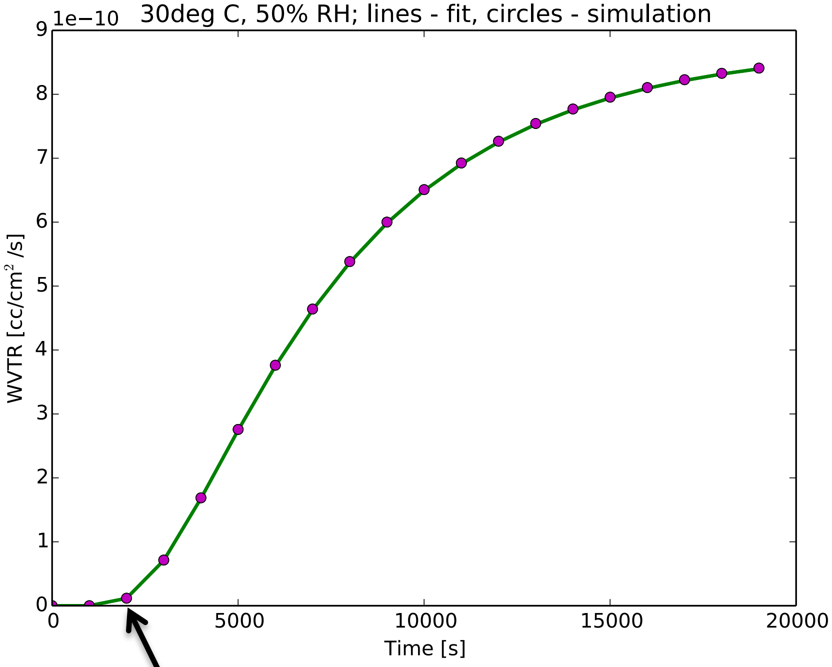


Figure 3: Four domains sharing a singe interface in the structured finite volume formulation. Many domains meeting at a single interface adds little or no difficulty to the use of the proposed interface residual method.

Kinetics-transport solver

Transient measurement of permeability of PET film to water



Delayed onset consistent with non-Fickian diffusion

Dual sorption:

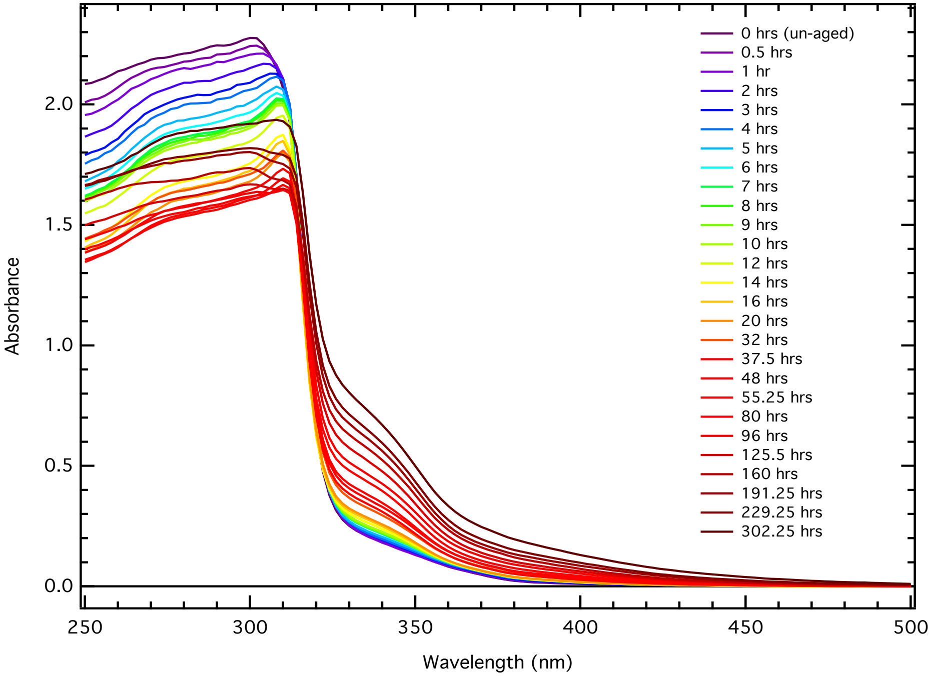
$$\frac{\partial C}{\partial t} = D_D \frac{\partial}{\partial x}\left[\frac{1}{1 + \frac{\alpha C_H'}{(1+\ \alpha C_D)^2}} \frac{\partial C}{\partial x}\right]$$

We have extracted permeability as a function of RH, T and P_{ox},T for PET, PMMA, with and without oxide coatings

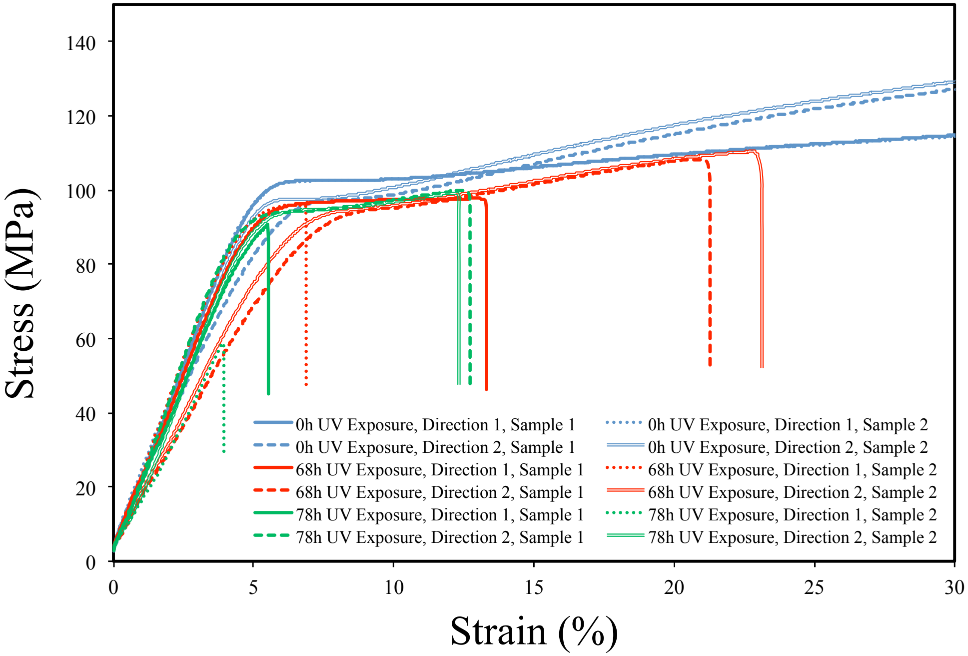
$$\mathrm{P} = (a_1 + a_2 p)e^{a_3/T}$$

Mechanical properties: UV degradation of poly(ethylene terephthalate) films

10X UVA/UVB under ambient conditions

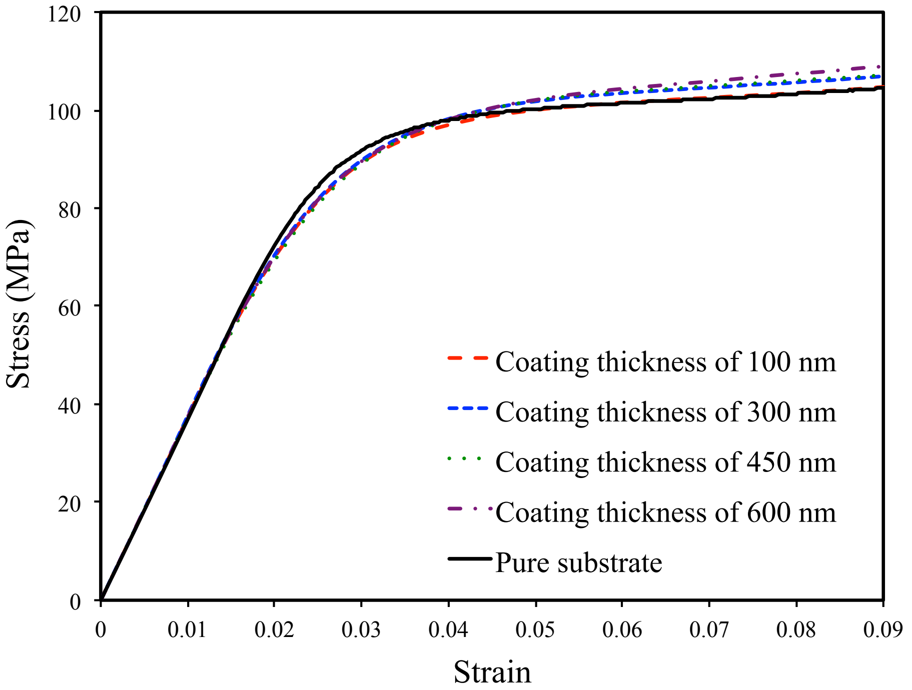


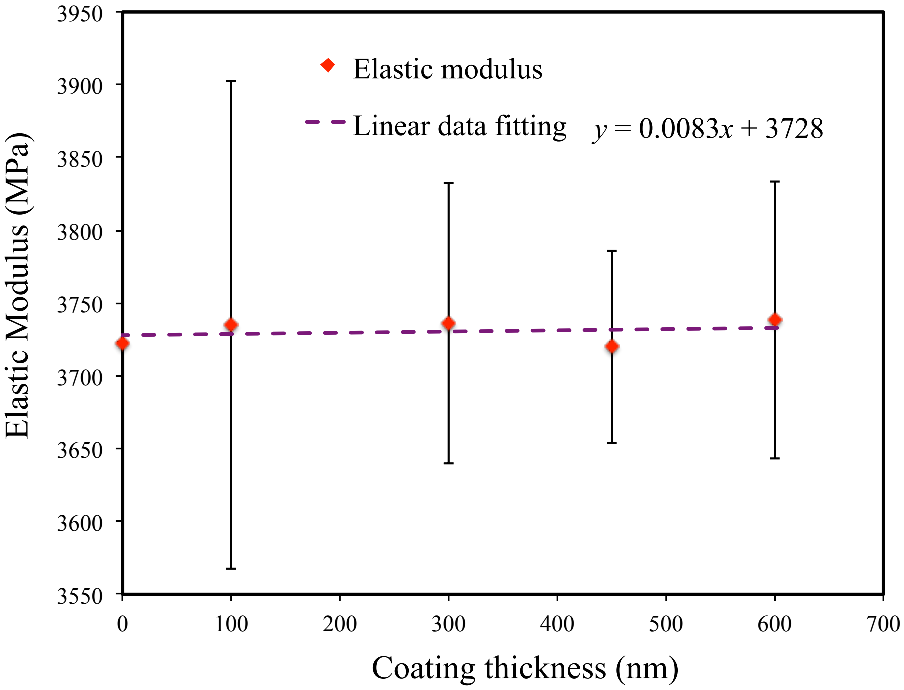
Aged films show minimal change in elastic constants, dramatic reduction in ultimate failure strain



Mechanical properties: Fracture of brittle coatings on strained PET films

- Tensile properties of TiO_2/PET thin film system

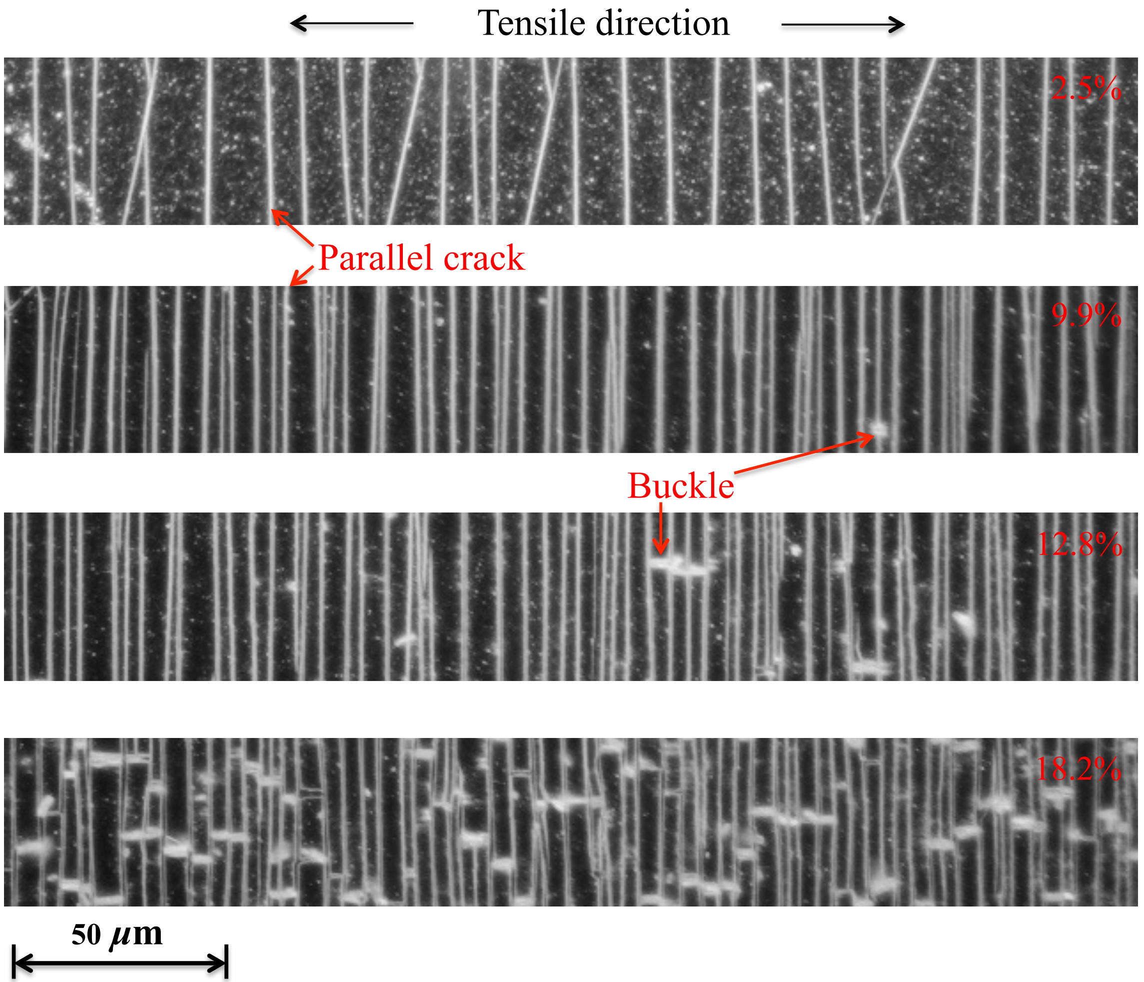




- ➢ Tensile load is transferred through the polymer substrate.
- ➢ Coated sample shows an earlier yielding.

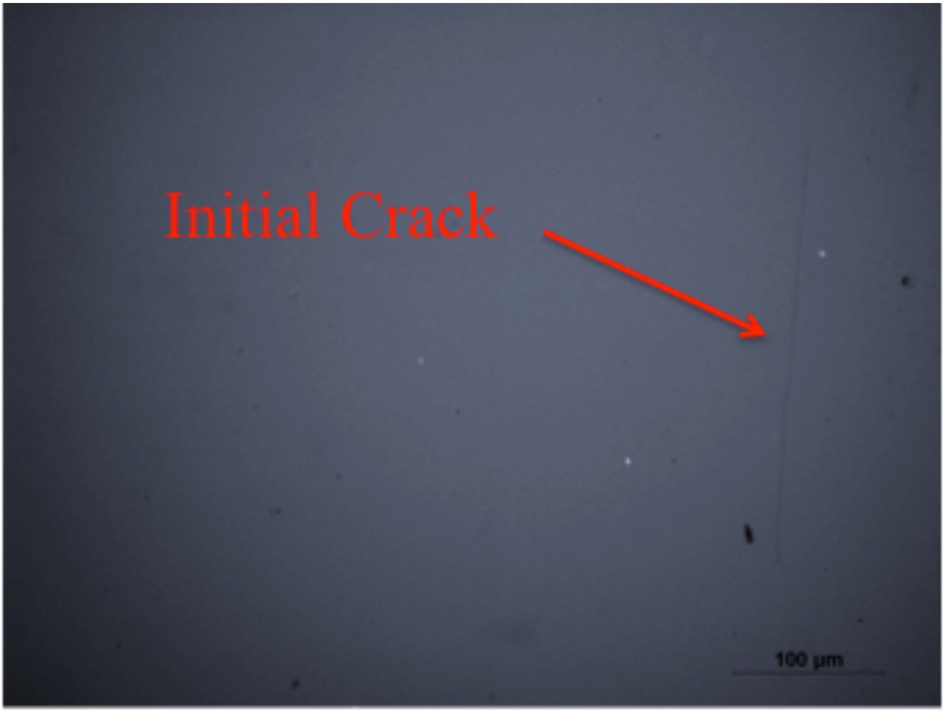
Mechanical properties: Fracture of brittle coatings on strained PET films

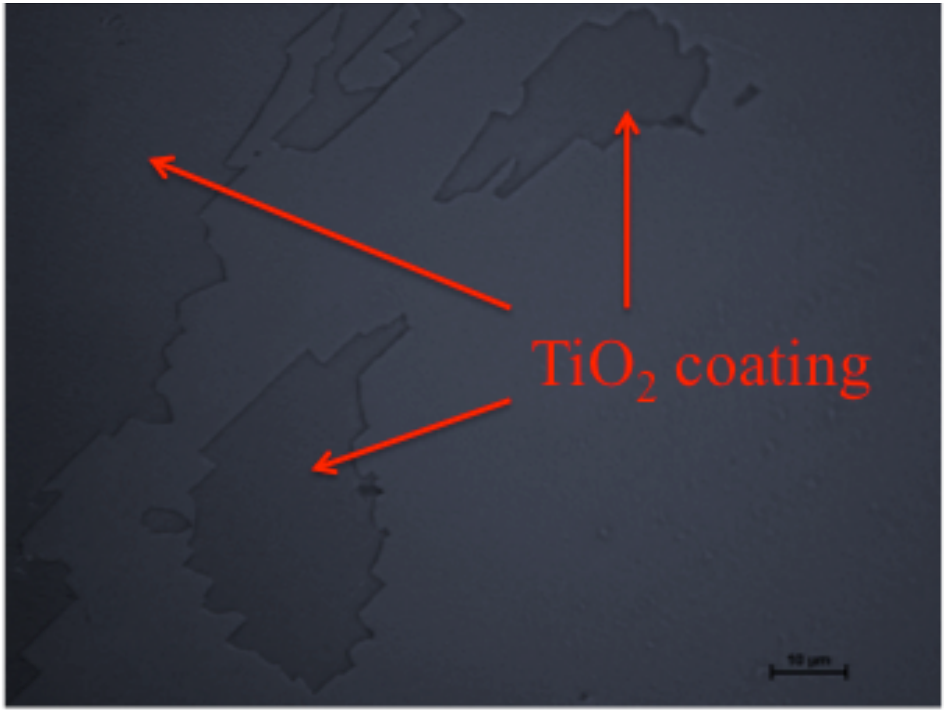
- In situ tensile test shows progressive cracking behavior



Mechanical properties: Fracture of brittle coatings on strained PET films

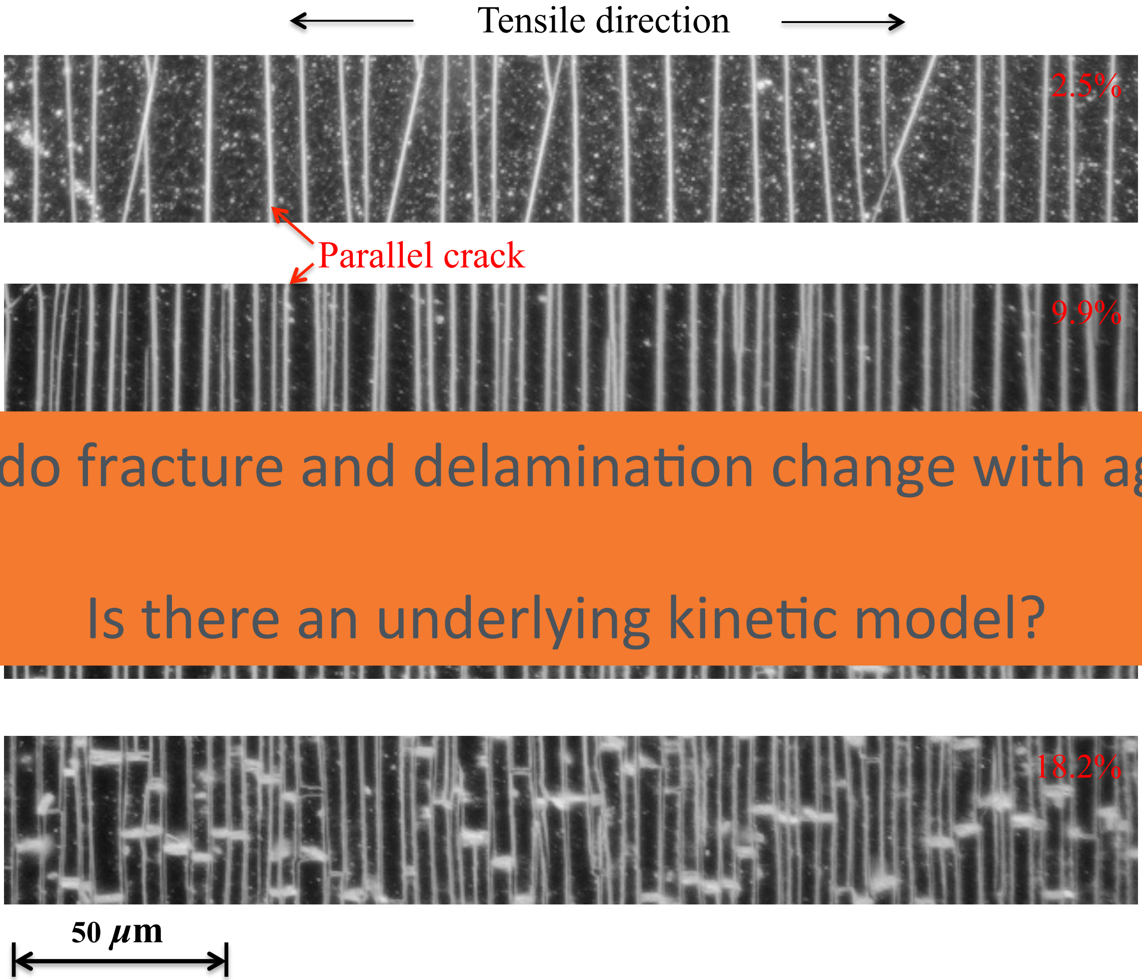
- Tape test shows delamination of TiO_2 for UV aged sample





Mechanical properties: Fracture of brittle coatings on strained PET films

- In situ tensile test shows progressive cracking behavior



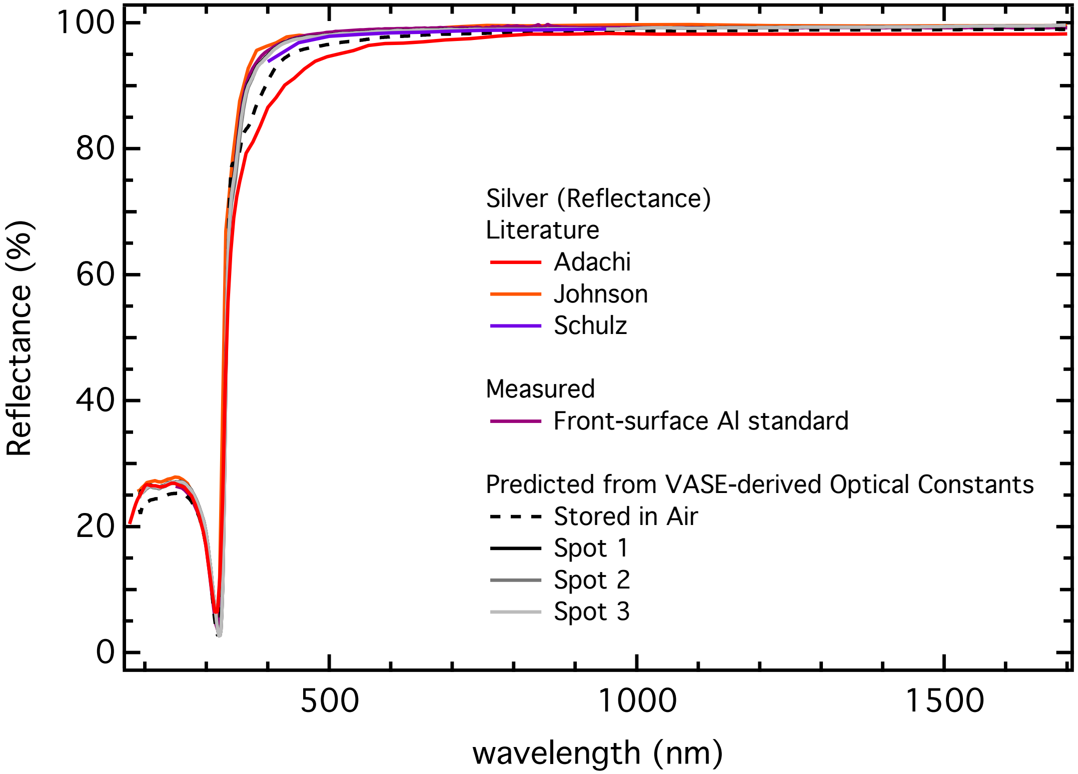
How do fracture and delamination change with aging?

Is there an underlying kinetic model?

Aging of simplified reflector stacks

Aging of simplified reflector stacks: Un-aged

Caution: there is not necessarily “canonical” silver

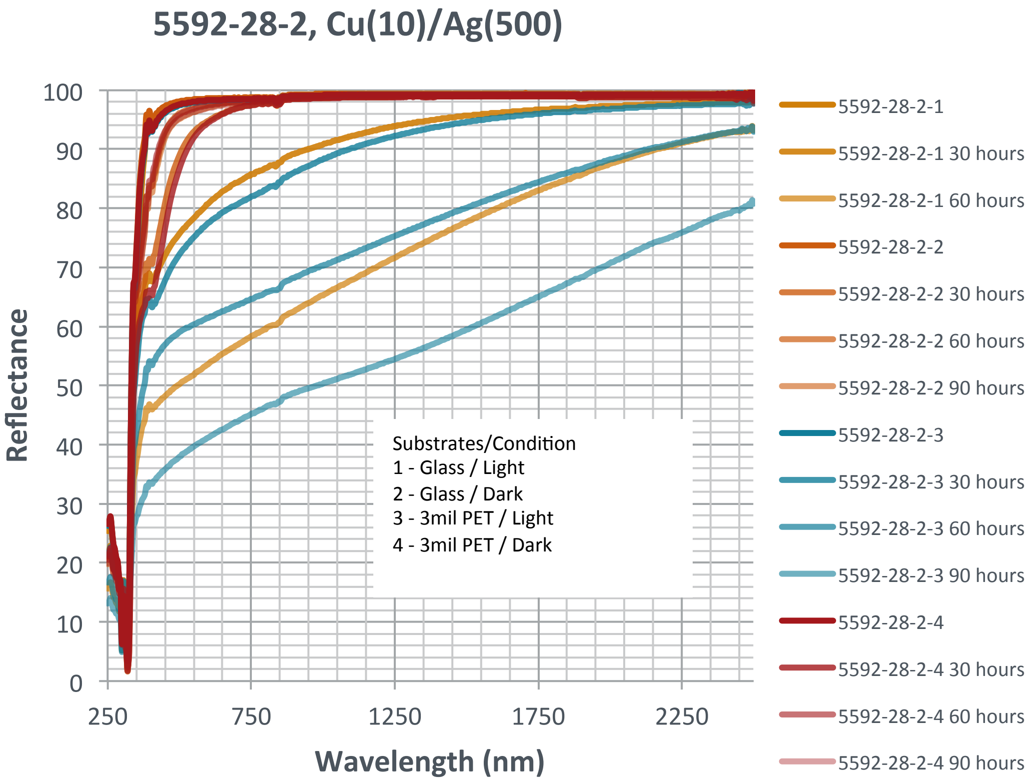


Our models now have “thin” and “thick” NREL silver optical constants

Aging of simplified reflector stacks: Un-coated

Degrade samples under conditions of 45 C, 40% RH 2X sunlight (or dark)

Hemispherical R

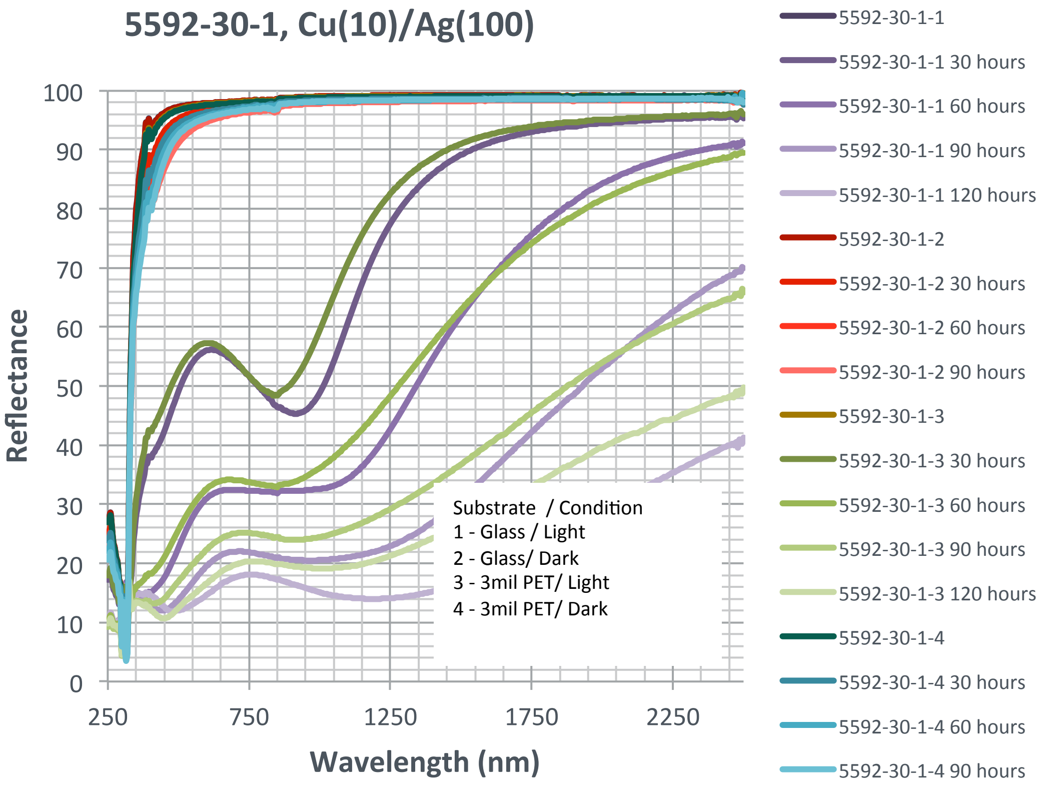


Silver degrades differently in the dark than under illumination

Aging of simplified reflector stacks: Un-coated

Degrade samples under conditions of 45 C, 40% RH 2X sunlight (or dark)

Hemispherical R



Substrate / Condition
1 - Glass / Light
2 - Glass/ Dark
3 - 3mil PET/ Light
4 - 3mil PET/ Dark

Silver degrades differently in the dark than under illumination
Reflectance changes differently with Ag thickness: thin film interference

Aging of simplified reflector stacks

Identify what is being formed with a combination of:

- Variable angle spectroscopic ellipsometry
- Rutherford backscattering
- X-ray photoelectron spectroscopy
- Specularity and roughness measurements

Identify relevant time scales and plausible kinetic models

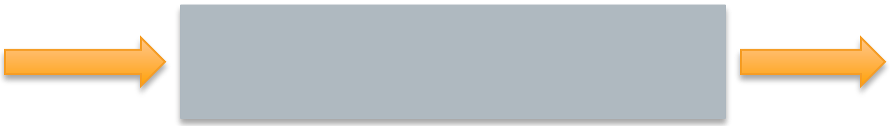
- Forward predict chemical changes and compare to exp’t

Ongoing work to correlate chemistry/material identity with optical/mechanical properties to build predictive model

Silver corrosion kinetics: Insights from the literature

- Quantitative extraction of silver sulfidation kinetics with *reaction/advection model* of degradation of silver in denuder tube

$$2\mathrm{Ag} + \mathrm{H_2S} + \tfrac{1}{2}\,\mathrm{O_2} \rightarrow \mathrm{Ag_2S} + \mathrm{H_2O}$$



Input gas mixture
Fixed flow rate

Exiting gas mixture

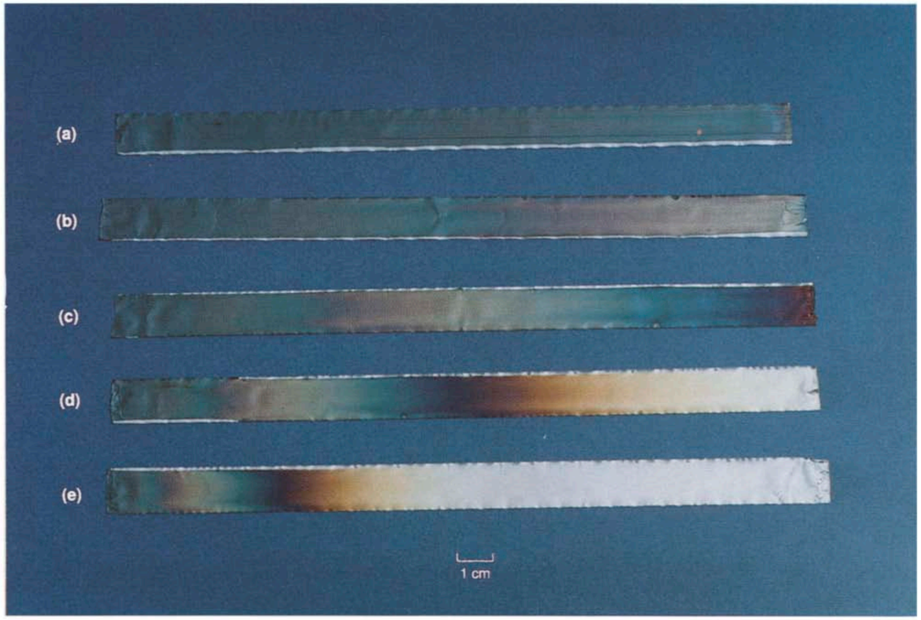


Fig. 6. Photographs of Ag foils after 48 h exposure to corrosion-chamber air. Foil leading edge on the left. Flow rates through tube (cm^3(STP) s^{-1}): (a) 51.8, (b) 27.3, (c) 14.3, (d) 7.67, (e) 3.83.

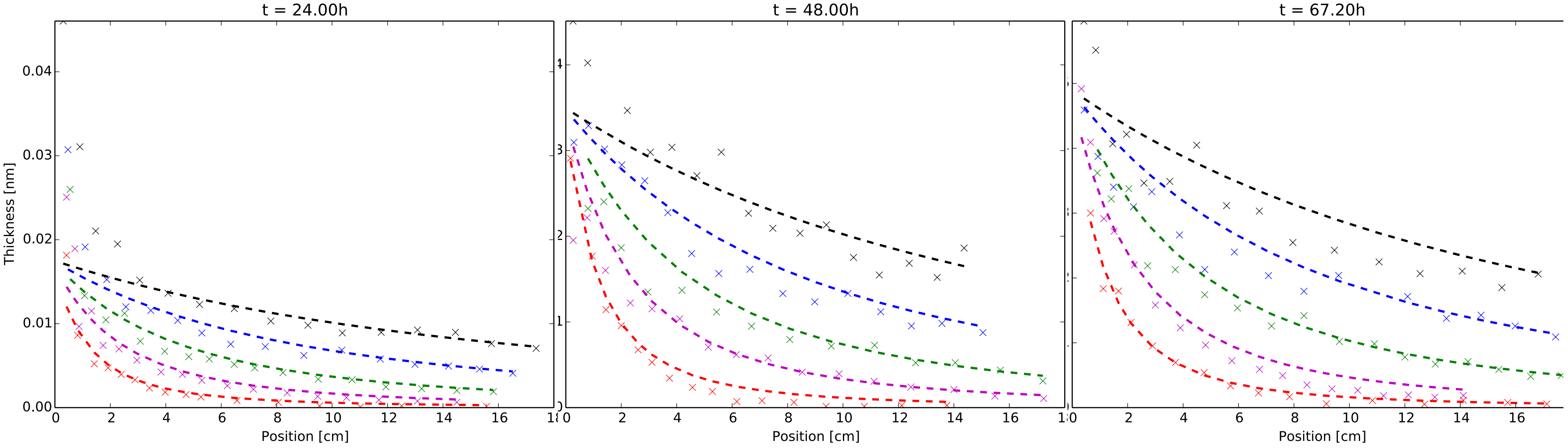
Volpe et al., *Corrosion Science* **10**, 1179-1196 (1989)

Silver corrosion kinetics: Insights from the literature

- Quantitative extraction of silver sulfidation kinetics with *reaction/advection model* of degradation of silver in denuder tube

$2Ag + H_2S + ½ O_2 \rightarrow Ag_2S + H_2O$

Flow rates: 3.83, 7.67, 14.3, 27.3, 51.8 cm^3(STP)/s



Volpe et al., *Corrosion Science* **10**, 1179-1196 (1989)

Silver corrosion kinetics: Insights from the literature

- Light dependence: are we photolyzing O_3?
  - Are we making Ag_2O, AgO
- Evolution of Ag_xO to Ag_2O (aging) as more oxide forms?
- Are we forming a heterogeneous mixed layer of oxides and sulfides? Chlorides?

Summary and Outlook

- Central hypotheses
  - Fundamental chemical events can be measured and mechanisms inferred
  - Predictive models can be constructed based on these measurements
- Measurements show complex degradation kinetics even for simplified geometries and materials
  - Q: Is full chemistry needed? Can one get away with a simplified pseudo-chemistry?
- Attention to roughening/loss of specularity
- Combined optics/kinetics/transport code to be made available to the community