SAND2016-2315C

An overview of DLO modeling and relevance for polymer aging predictions

"Reactive Diffusion Models for Spatially Resolved Polymer Aging"

Mathew Celina, Adam Quintana, and Nick Giron

3/23/2016

Service Life Prediction Conference, Santa Fe, NM

March 20 to 24, 2016

Sandia National Laboratories

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Polymer Aging and Materials Characterization

- Polymers will degrade over time
- How and where? How does it affect material performance attributes?
- When is engineering testing sufficient?
- How does material science contribute?

- History of Polymer Aging at SNL
- 1970s: Qualification of materials mostly for nuclear power applications
- 1980s: Good understanding of degradation processes and lifetime prediction approaches, mostly for elastomers
- 1990s: Heterogeneous degradation concepts and ox-rate data
- 2000s: Material requalification efforts, condition monitoring
- 2010 onwards : Fundamental understanding of degradation processes
  Cross-correlations between chemistry and physics

Applied Polymer Science supports Materials Engineering

Research Needs at SNL

- Focus: New Materials, Engineered Reliability and Performance, Aging/Degradation, Polymer Physics/Chemistry,

- In depth materials characterization, new techniques
- Understand convoluted processes for polymer degradation
- Provide a foundation for decisions regarding material reliability
- We engage in areas that are not being dealt with by industry/academia
- We need to understand science relevant to SNL mission needs
- Interdisciplinary phenomena: Couple chemistry/physics and mathematics for material lifetime prediction purposes

Material science enables engineering solutions for many application environments and demands

External Community Efforts

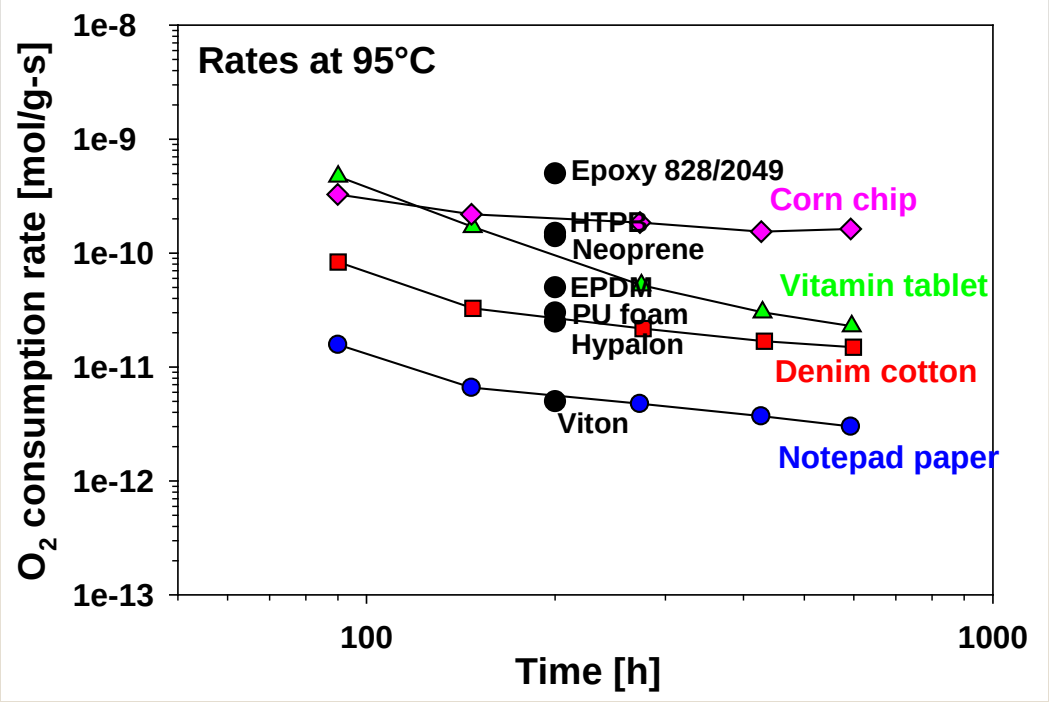
- Many basic polymer degradation studies in literature, 'cook and look'
- Lot's of high temperature TGA work. Do we care about 200C?
- Polymer degradation and numerical approaches for kinetics models are well established
- Composite R&D focuses on high temperature surface degradation, oxygen permeation and DLO is recognized as a complicating phenomenon
- Other researchers have similar interests, however many organic material applications are unique to SNL

- External work is complementary, but offers limited solutions for SNL needs
- We have become leaders in this field and often assist the external community [PhD reviews, aging R&D review for French nuclear plants, visiting academics and students, connections with US industry (WFO or CRADA)]

Some in depth polymer degradation needs are unique to SNL because of extended service life requirements

Polymers and Oxygen – Aging is Guaranteed

- SNL introduced sensitive methods to measure intrinsic oxidation rates
- Materials oxidize and change mechanical and other physical properties
- Oxidation is at the core of many reliability and degradation processes



Oxidation rates are a key parameter for any aging model

Celina MC, Dayile AR, Quintana A. A perspective on the inherent oxidation sensitivity of epoxy materials. Polymer 2013;54:3290.

Diffusion Limited Oxidation – What is It?

- Where do we see DLO in nature?
- DLO is the equivalent of surface corrosion for polymers
- What is the real color of rocks in the side canyons of the Colorado?

Freshly fallen fractured rock shows only surface color

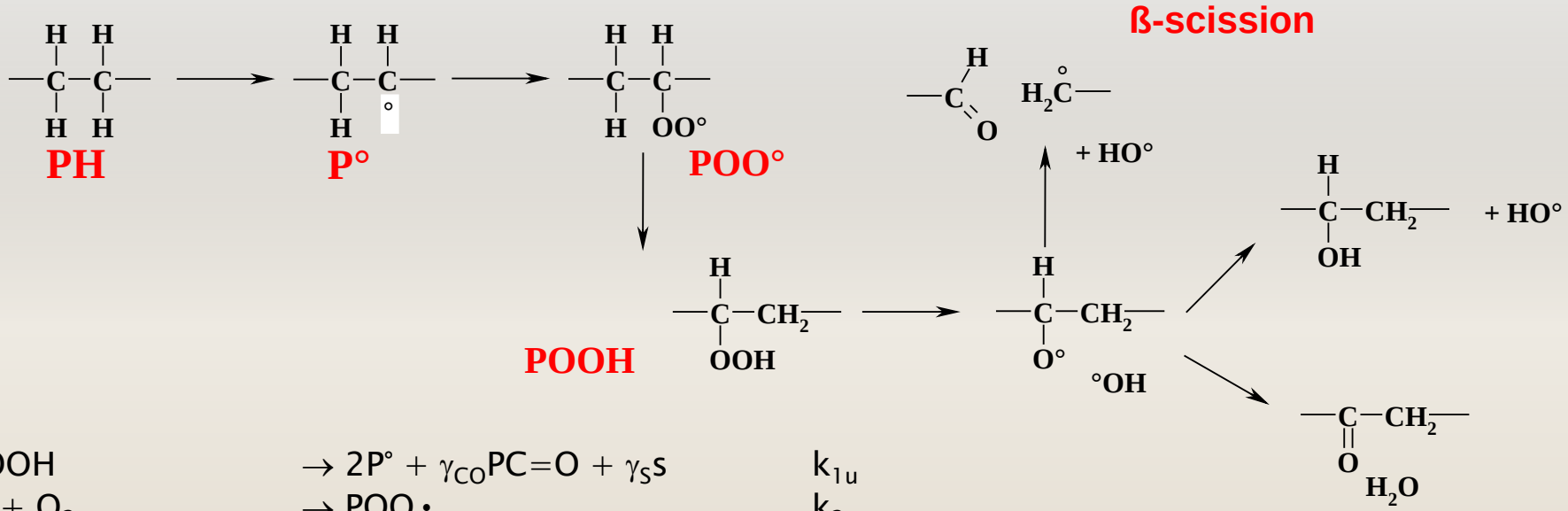
Thick polymers also age from the surface

- **Chemistry coupled with diffusion phenomena is real**
- **Beauty is at the surface, complexity is on the inside!**
- **Heterogeneous catalysis at interfaces has similar attributes**

Celina MC. Review of polymer oxidation and its relationship with materials performance and lifetime prediction. Polym Degrad Stab 2013;98:2419.

Free Radical Oxidation – Slow Combustion

- Polymer oxidation – Basic Auto-oxidation Scheme (BAS); hydrocarbon oxidation in the 1950s, Bolland, Bateman and Gee
- Degradation chemistry often leads to hardening (polymer crosslinking)



POOH	$\rightarrow 2P^{\circ} + \gamma_{CO}PC{=}O + \gamma_{S}S$	k_{1u}
$P^{\circ} + O_2$	$\rightarrow POO\cdot$	k_2
$POO^{\circ} + PH$	$\rightarrow POOH + P^{\circ}$	k_3
$POO^{\circ} + POO^{\circ}$	$\rightarrow$ Inactive products $+ O_2$	k_6

Polymer chain cleavage and oxidation

Polymer, peroxides, free radicals, scission, crosslinking, H-abstraction/transfer, carbonyls, aldehyds, alcohols, CO_2, H_2O

Gillen KT, Wise J, Clough RL. General solution for the basic autoxidation scheme. Polym Degrad Stab 1995;47:149.

Polymer Oxidation as Kinetic Model

- Free radical auto-oxidation; classic initiation, propagation, termination
- Reaction kinetics have been described
- Combinations of differential equations

POOH	$\rightarrow 2P\bullet$	(k_{1u})	$P\bullet + P\bullet$	$\rightarrow$	(k_4)
2POOH	$\rightarrow P\bullet + PO_2\bullet$	(k_{1b})	$P\bullet + PO_2\bullet$	$\rightarrow$ …	(k_5)
$P\bullet + O_2$	$\rightarrow PO_2\bullet$	(k_2)	$PO_2\bullet + AH$	$\rightarrow$	(k_{s1})
$PO_2\bullet + PH$	$\rightarrow POOH + P\bullet$	(k_3)	$PO_2\bullet + A\bullet$	$\rightarrow$ …	(k_{s2})
$PO_2\bullet + PO_2\bullet$	$\rightarrow$ …	(k_6)			

$$\frac{d[P\bullet]}{dt} = 2k_{1u}[POOH] + k_{1b}[POOH]^2 - k_2[P\bullet][O_2] + k_3[POO\bullet][PH] - 2k_4[P\bullet]^2 - k_5[P\bullet][POO\bullet]$$

$$\frac{d[POOH]}{dt} = -k_{1u}[POOH] - 2k_{1b}[POOH]^2 + k_3[POO\bullet][PH]$$

$$\frac{d[POO\bullet]}{dt} = k_{1b}[POOH]^2 + k_2[P\bullet][O_2] - k_3[POO\bullet][PH] - k_5[P\bullet][POO\bullet] - 2k_6[POO\bullet]^2$$

$$\frac{d[PH]}{dt} = -k_{1u}[POOH] - k_{1b}[POOH]^2 - k_3[POO\bullet][PH]$$

Oxidation rate

$$\frac{\partial[O_2]}{\partial t} = D_{O_2}\frac{\partial^2[O_2]}{\partial x^2} - k_2[O_2][P\bullet] + k_6[PO_2\bullet]^2$$

Extensive work by multiple research groups, chemistry is understood

Colin X, Fayolle B, Audouin L, Verdu J. The classical kinetic model for radical….. Int J Chem Kinet 2006;38:666.

Oxygen Activity Depends on Location

- Oxygen availability = surface partial pressure
- O2 concentration in polymer = solubility

Biopolymer oxidation depends on elevation

Burning glucose on Mt. Everest

In the deep too much O_2 is toxic

Depends on partial pressure

Same issues for polymers:

- **P_{O2} and S_{O2} (O_2 partial pressure and solubility)**
- **Material oxidation depends on depth, surface is often more oxidized**

Oxygen will degrade polymers – a matter of TIME and LOCATION

Gillen KT, Clough RL. Rigorous experimental confirmation of a theoretical model for DLO. Polymer 1992;33:4358.

Local O2 Partial Pressure Affects Rate

- Kinetic model; correlation between [O2] and Ø
- ß$_0$ material property, but depends on T, relates to auto-oxidation scheme

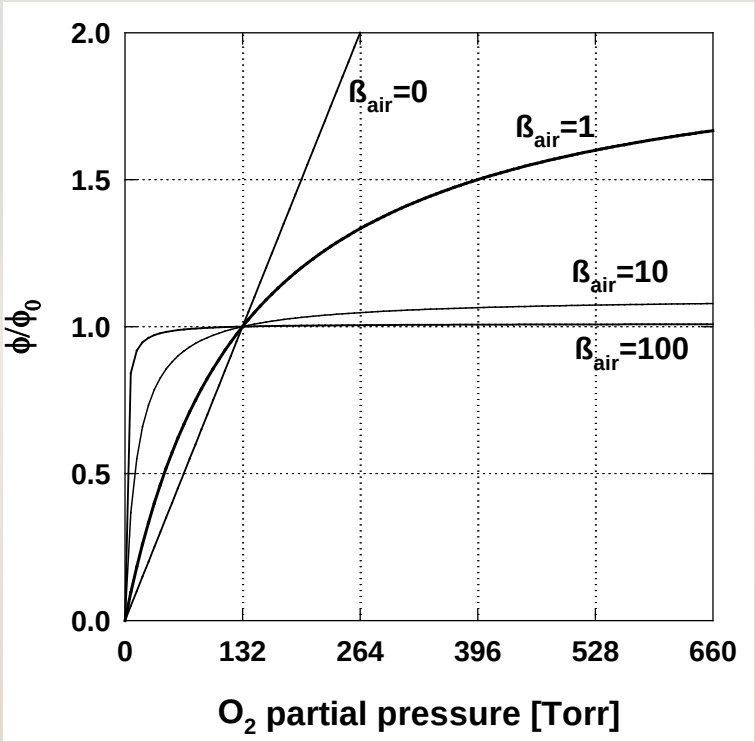
$$\beta_0 = f(C_2, [O_2]) = C_2 * S * (p_{O_2})_0$$

$$\beta = \beta_0 * \frac{P_{O_2}}{(P_{O_2})_0}$$

$$C_{1b} = k_2\left(\frac{R_i}{2k_4}\right)^{1/2} \quad C_{2b} = \frac{k_2}{k_3}\left(\frac{k_6}{k_4}\right)^{1/2}$$

$$C_{1u} = \frac{k_2 R_i}{k_8} \quad C_{2u} = \frac{k_2 k_7}{k_8(k_3 + k_7)}$$

$$\frac{\phi}{\phi_0} = \frac{1+\beta_0}{\frac{P_0}{P} + \beta_0}$$

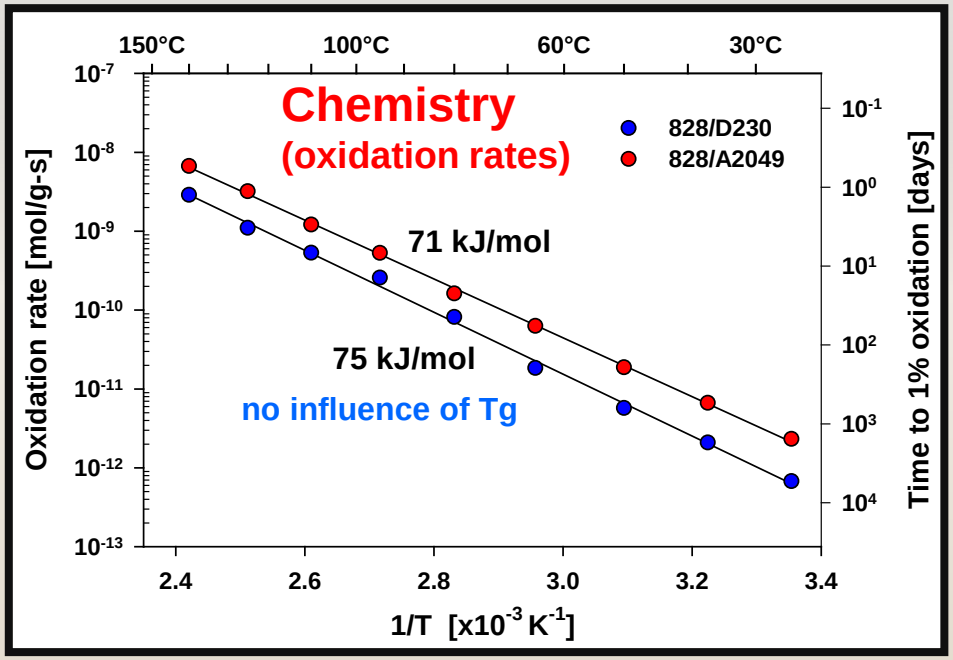


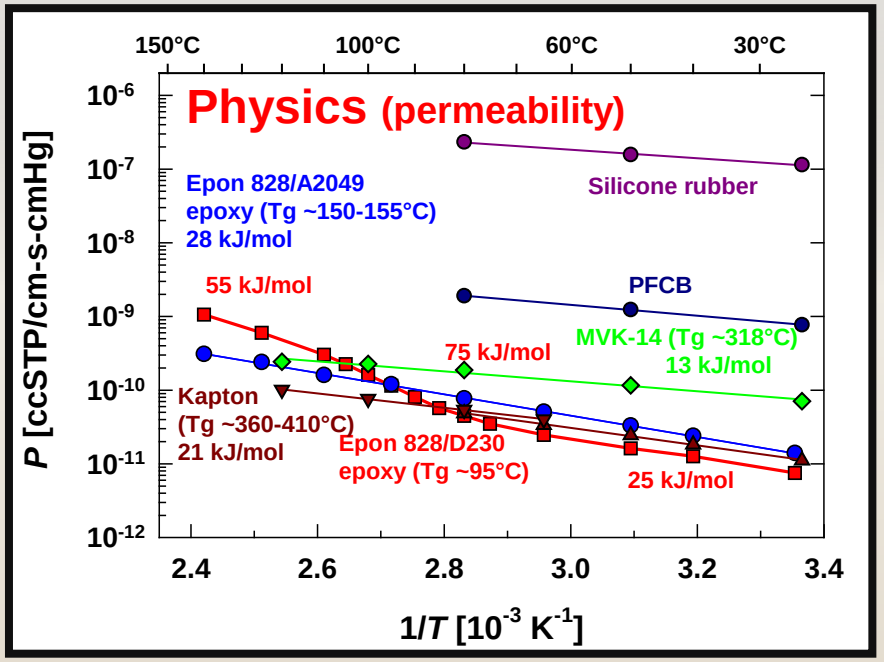
The description of polymer oxidation involves physical chemistry

Wise J, Gillen KT, Clough RL. Quantitative model for the time development of DLO profiles. Polymer 1997;38:1929.

Key Input: Oxidation Rates and Permeability

- Oxidation rates are experimentally determined for many polymers
- Consumed O2 under non-DLO conditions. SNL leads research activity.
- Oxygen permeability is obtained from flux measurements through thin films. O2 diffusion and solubility are available via Fickian diffusion fits.





- **Detailed material characterization is required**
- **Established new capability: Permeability with temperature**

Celina M, Gillen KT. O2 Permeability Measurements on Elastomers at Temperatures up to 225°C. Macromolecules 2005;38:2754.

DLO in Polymers – Mathematical Description

MODEL CONCEPT - OXIDATION & DIFFUSION

Oxidation rate derived from the Basic Auto-Oxidation Scheme (BAS):

$$-r_{O_2} = \phi = \frac{k_a[O_2]}{k_b[O_2] + 1}$$

Incorporation into Fick's second law:

$$\frac{\partial\theta}{\partial\tau} = \nabla^2\theta - \frac{\alpha\theta}{\beta\theta + 1}$$

Pressure dependence of oxidation rate:

$$\frac{\phi}{\phi_0} = \frac{\beta + 1}{\beta + {}^{1}/_{\theta}}$$

DLO model parameters:

$$\frac{\alpha}{\beta + 1} = \frac{\phi_0 L^2}{P_{ox} p_0}$$

$$\alpha = \frac{k_1 L^2}{D}$$
$$\beta = k_2 S p_0$$
$$\theta = {}^{p}/_{p_0}$$
$$\tau = \frac{tD}{L^2}$$
$$\chi = {}^{x}/_{L}$$
$$P_{ox} = DS$$
$$[O_2] = Sp_0 \text{ (Henry's Law)}$$

MODEL PARAMETERS

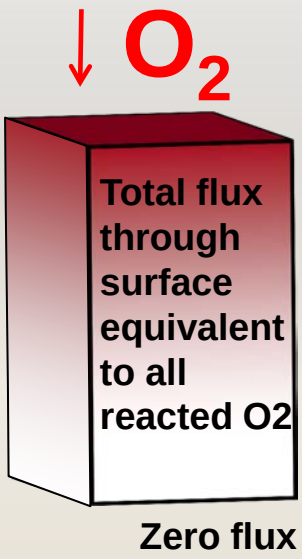
k_a, k_b . .	BAS overall rate constants
α, β. . . .	DLO model parameters
ϕ	Oxidation rate [mol/g/s]
θ	Relative oxygen partial pressure
P_{ox}	Oxygen Permeability [ccSTP/cm/s/cmHg]
D	Diffusivity [cm^2/s]
S	Solubility [mol/cc/cmHg]
p_0	Oxygen partial pressure (reference) [cmHg]
χ	Relative position
L	Thickness (reference) [cm]
τ	Normalized time
t	Time [s]
Ω	Spatial integrator depending on dimension
φ	Simplex interpolation function

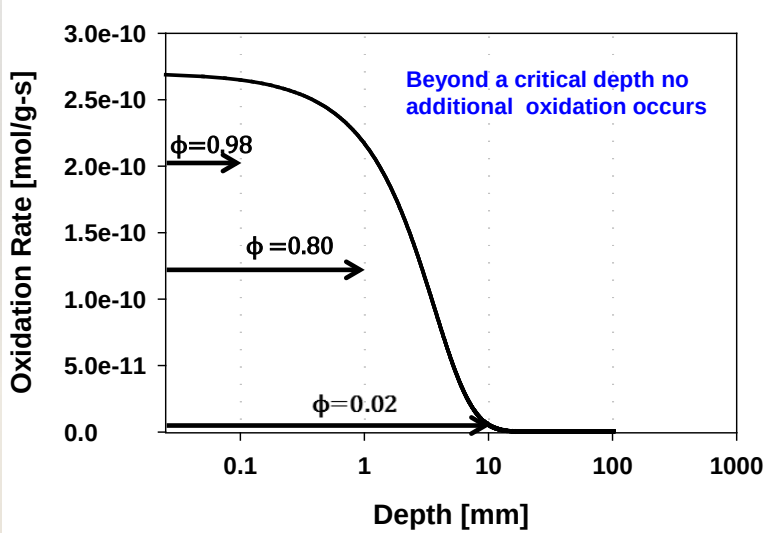
Many years of developmental work – SNL is the global leader

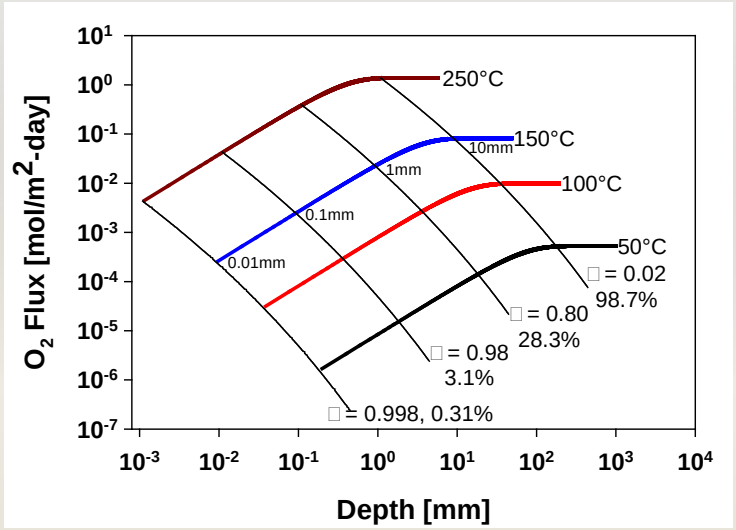
Gillen KT, Clough RL. Rigorous experimental confirmation of a theoretical model for DLO. Polymer 1992;33:4358.

1D Cross-section Models

- Local rates are used to estimate total O2 flux into specific materials
- Prediction of oxygen flux and oxidation rate with respect to depth into an EPDM material for total material oxidation reactivity







$$J_{ox}{}^2 = \phi P_{ox} p_0 \gamma, \qquad \gamma \equiv 2\frac{(\beta+1)(\beta-\ln(\beta+1))}{\beta^2}$$

$$\frac{\partial[O_2]}{\partial t} = D\nabla^2[O_2] - \frac{k_1[O_2]}{k_2[O_2]+1}$$

Generalized DLO equation

Polymer degradation is position-dependent as a function of local oxygen concentration

New SNL Capability: FEM Solutions

- Mathematical Approach uses Galerkin FEM solutions

Application of Galerkin Weighted Residual Finite Element Method (FEM)
Solution for the weak formulation of the partial differential equation
FEM uses triangle simplex subspaces; Approximate solution: $\theta_{i,j} \approx u_{i,n} = \sum_{j=1}^{n} u_{i,j}\varphi_j$
DLO weak formulation after divergence theorem:

$$\int_{\Omega_j} \boldsymbol{\varphi}^T \frac{\partial \boldsymbol{u}}{\partial \tau} d\,\Omega = \int_{n_j} (\boldsymbol{\varphi}^T \nabla \boldsymbol{\varphi} \cdot \boldsymbol{u}) \cdot d\boldsymbol{n} - \int_{\Omega_j} \nabla \boldsymbol{\varphi}^T \nabla \boldsymbol{\varphi} \cdot \boldsymbol{u}\, d\,\Omega - \int_{\Omega_j} \boldsymbol{\varphi}^T \frac{\alpha \boldsymbol{\varphi} \cdot \boldsymbol{u}}{\beta(\boldsymbol{\varphi} \cdot \boldsymbol{u}) + 1} d\,\Omega$$

Summation over all simplex interpolation functions: $\mathbb{M}\frac{\partial \boldsymbol{u}}{\partial \tau} = (\mathbb{f} + \mathbb{K})\boldsymbol{u} - \boldsymbol{r}$

- $\mathbb{M}$ **is the mass matrix (**$\mathbb{M} = \underline{0}$ **for steady state)**
- $\mathbb{f}$ **defines the surface flux (**$f_n = \frac{P_{ox\,i}}{P_{ox\,l}}$ **is necessary for the laminar boundary condition, which is 0 for non-boundary)**
- $\mathbb{K}$ **is the stiffness matrix**
- $\boldsymbol{r}$ **is summation of averaged element rate expressions using mean value theorem**

Complex numerical analysis enables 2D DLO models for any geometry and boundary conditions

$$\mathbb{M} = \sum_{n=1}^{N} \int_{\Omega_n} \boldsymbol{\varphi}^T \boldsymbol{\varphi}\, d\,\Omega \qquad \mathbb{f} = \sum_{n=1}^{N} f_n \overline{\boldsymbol{\varphi}^T} \nabla \boldsymbol{\varphi}\big]_{\varphi_n}$$

$$\mathbb{K} = -\sum_{n=1}^{N} \Omega_n \nabla \boldsymbol{\varphi}^T{}_n \nabla \boldsymbol{\varphi}_n \qquad \boldsymbol{r} = \sum_{n=1}^{N} \overline{\boldsymbol{\varphi}^T}_n \overline{\frac{\alpha \boldsymbol{u}_n}{\beta \boldsymbol{u}_n + 1}}$$

Science Foundation Enabling This Work

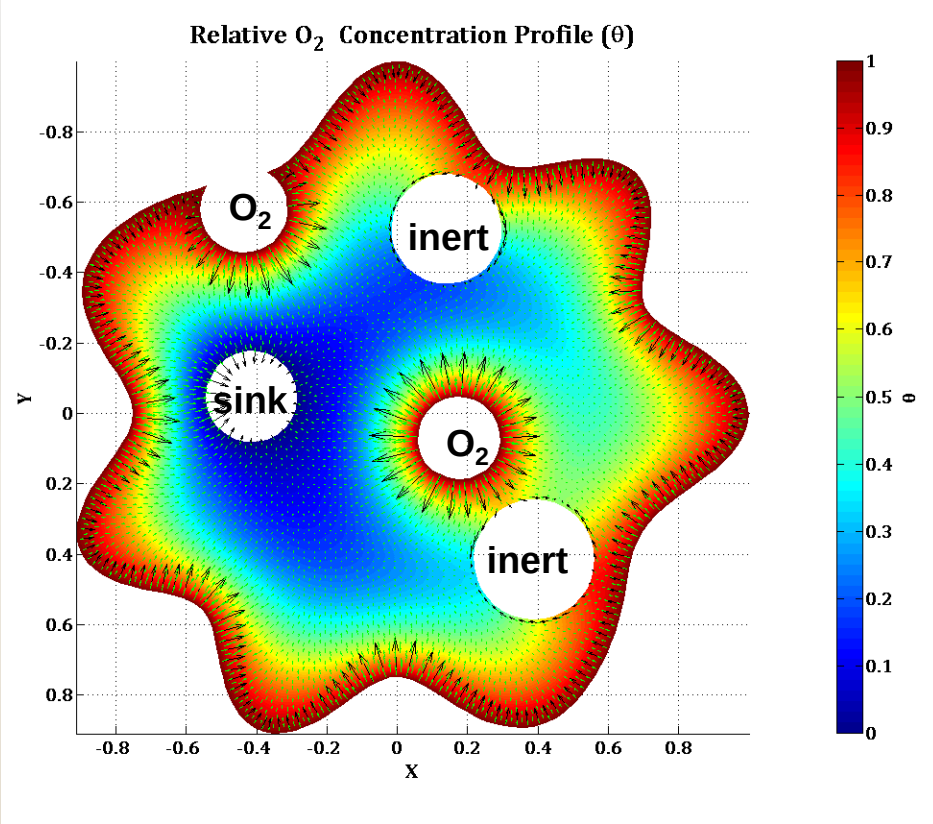
- Experimental challenges
  - Oxidation rates, measurement approaches, first ever paper to quantitatively describe oxidation of epoxies (resulted in outside customer contact)
  - Permeability at high T, elimination of oxidation effects during measurement, no literature data available, we customized instrumentation
  - If oxidation applies, O2 transmission data do not yield permeability constant
  - Partial pressure dependence, experimentally very difficult to address
- Coupling of experiments with models based analysis
  - Beta factor determination through multi-parameter analysis of permeation data, need to understand the combined effect of multiple behaviors
- Model development
  - Analytical solutions, semi-numerical as well as FEM approaches
  - Boundary conditions for FEM codes, flux, local concentration and rate
  - Multi-material assemblies and 3D are mathematically challenging

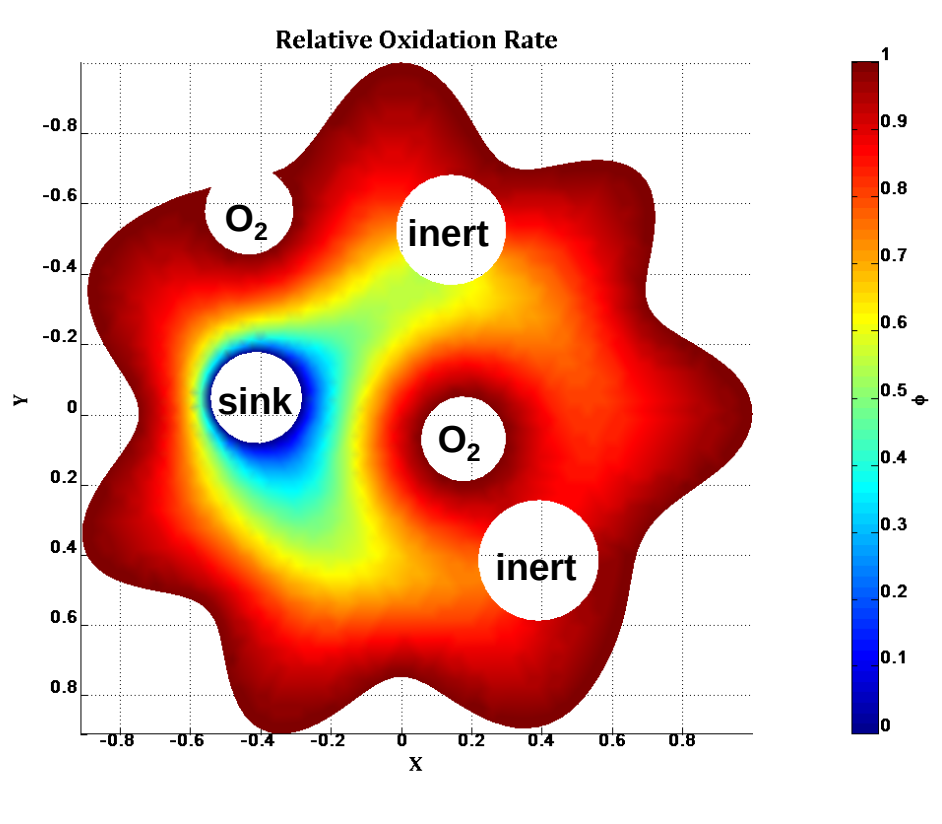
No other group has developed a comprehensive science basis

Celina MC, Dayile AR, Quintana A. A perspective on the inherent oxidation sensitivity of epoxy materials. Polymer 2013;54:3290.

FEM Codes – Evaluate Boundary Conditions

- Model evaluation with multiple boundary conditions. Example of irregular domain, surface O2 exposure, with internal inert voids, additional O2 source and sink.

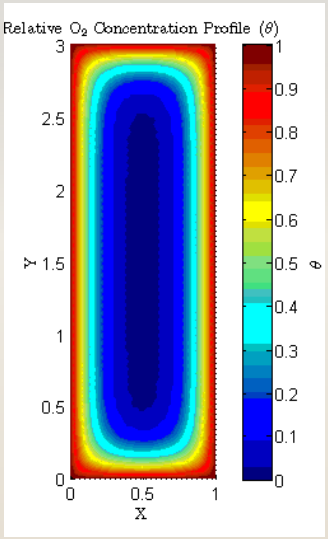


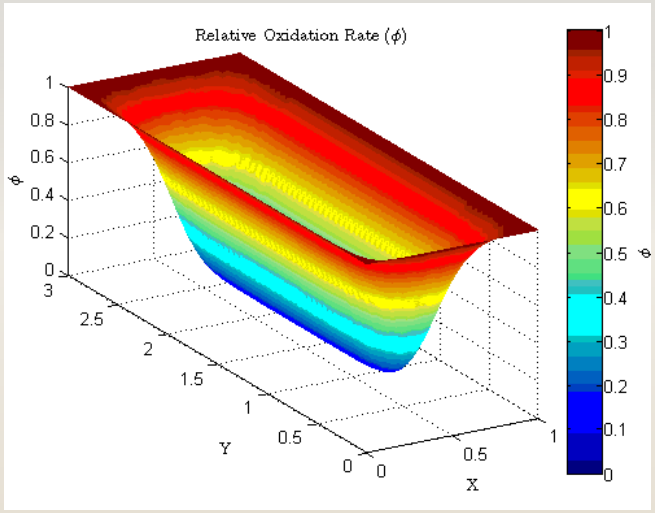


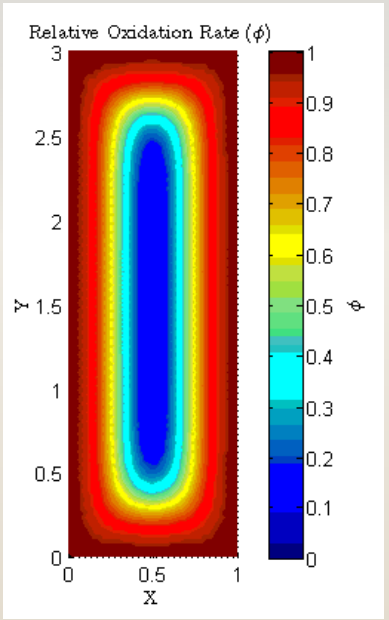
Discontinuities due to changes in material composition and geometry

DLO – 2D FE model

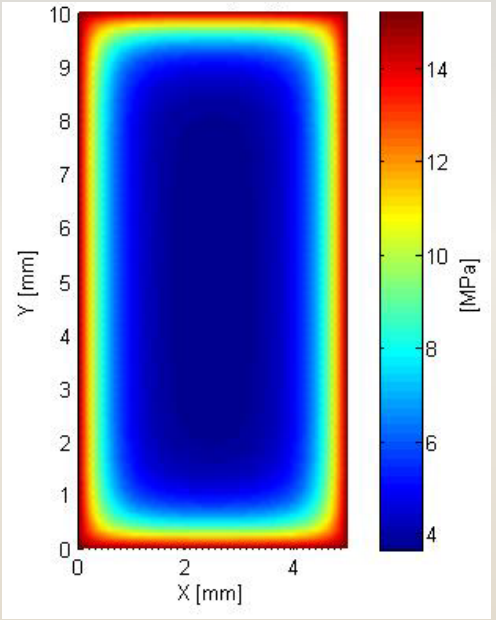
- Spatially resolved O2 concentration and oxidation rates
- For example: Aged Tensile Test Specimen
- Local oxidation levels can be correlated with local modulus







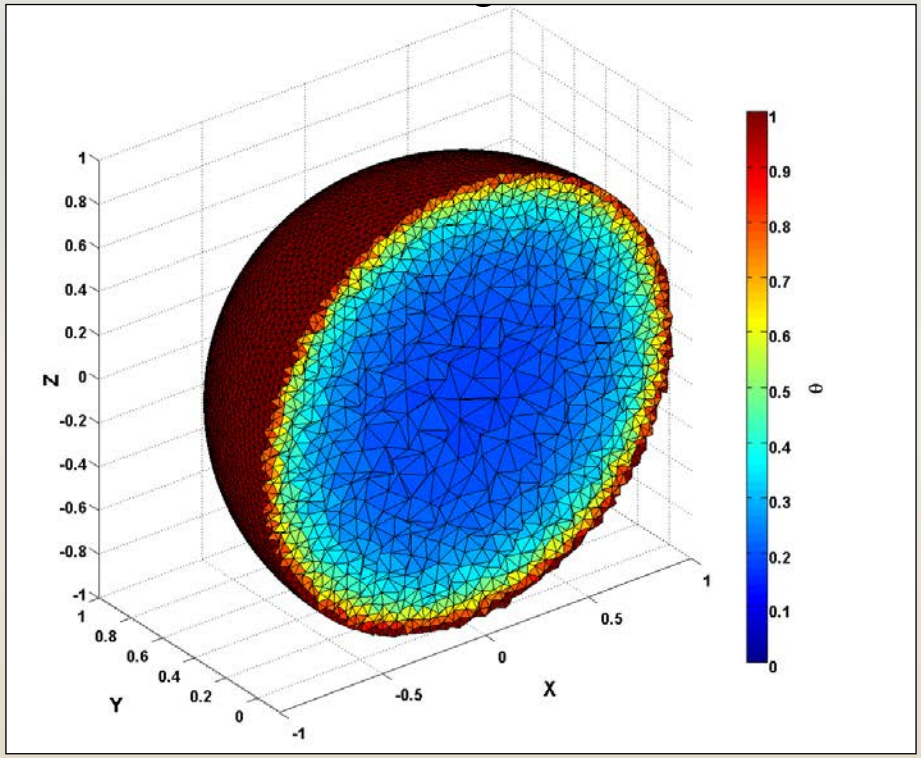
Localized modulus



Models show depth of oxidation for different geometries

DLO – 3D FE model

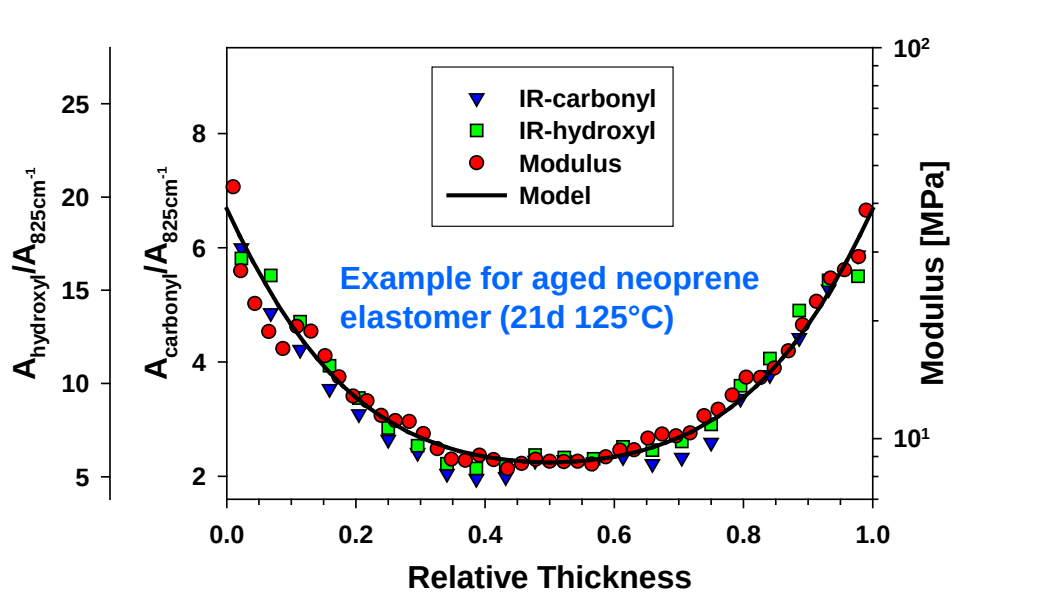
- 3D representation of reactive diffusion phenomena resulting in material surface degradation
- Mathematically very challenging for residual error minimization

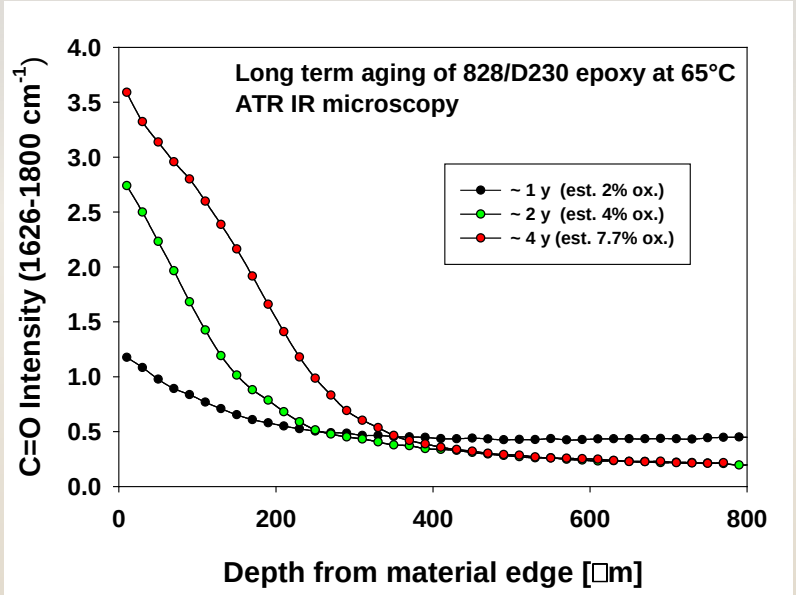


Currently pushing 3D models into complex shapes and multiple materials

Model Validation

- Cross-correlation between model, degradation chemistry (IR) and mechanical property (modulus) profiles
- Semi-analytical and validated boundary conditions from simple behaviors are embedded in higher level (2D and 3D) degradation processes
- Models are developed step by step, cross-checked with exptl. degradation





Connection between 'chemistry' and 'physical properties' is material specific

Accelerated aging – what are the limits?

- Ambient aging under oxidative environment, polymers will age
- Is accelerated aging applicable and has predictive value?
- When does accelerated aging become non-predictive?

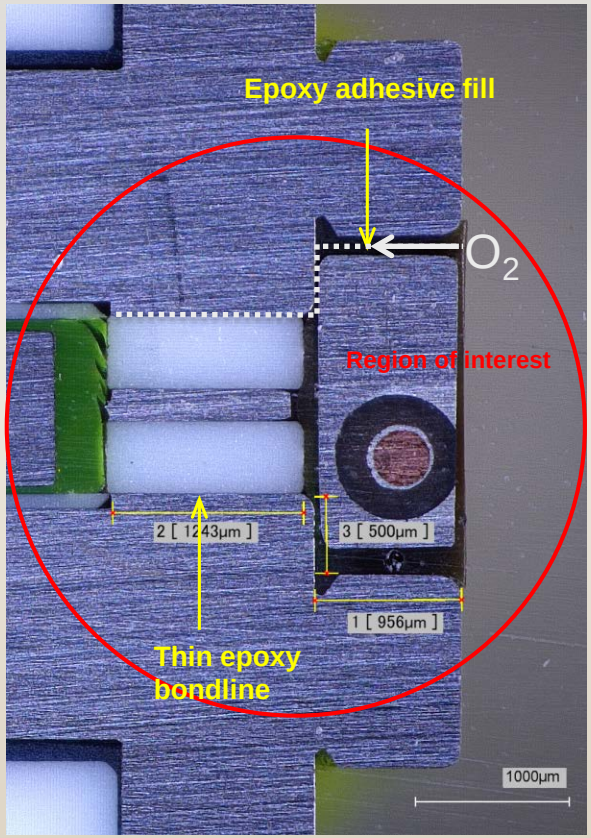
- Pass or fail under screening scenarios has limited real predictive value
- DLO is a mechanistic variation, when does it matter? Can it mask feedback from empirical testing?
- Our models enable science centered predictions based on the underlying material behavior and set boundaries for aging conditions

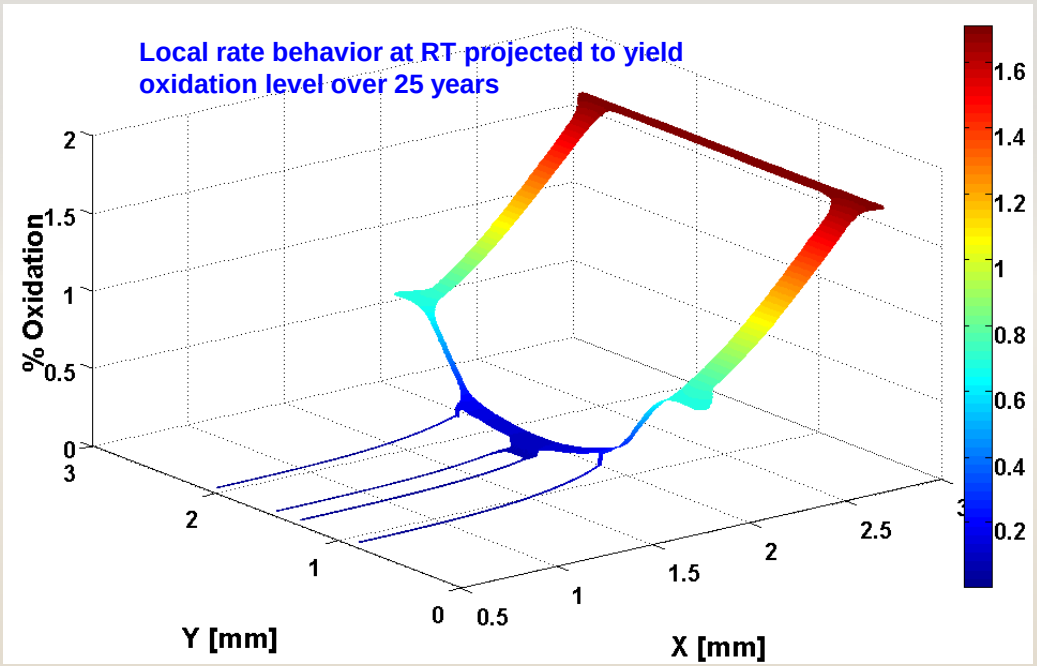
What are the predictive limits of accelerated aging?

Celina MC. Review of polymer oxidation and its relationship with materials performance and lifetime prediction. Polym Degrad Stab 2013;98:2419.

DLO Phenomenon Protects Interior Bondline

- Understanding long-term ambient aging processes is a necessity
- Requires understanding of O_2 permeation and reaction with two epoxies
- FEM DLO models predict that interior epoxy material cannot oxidize

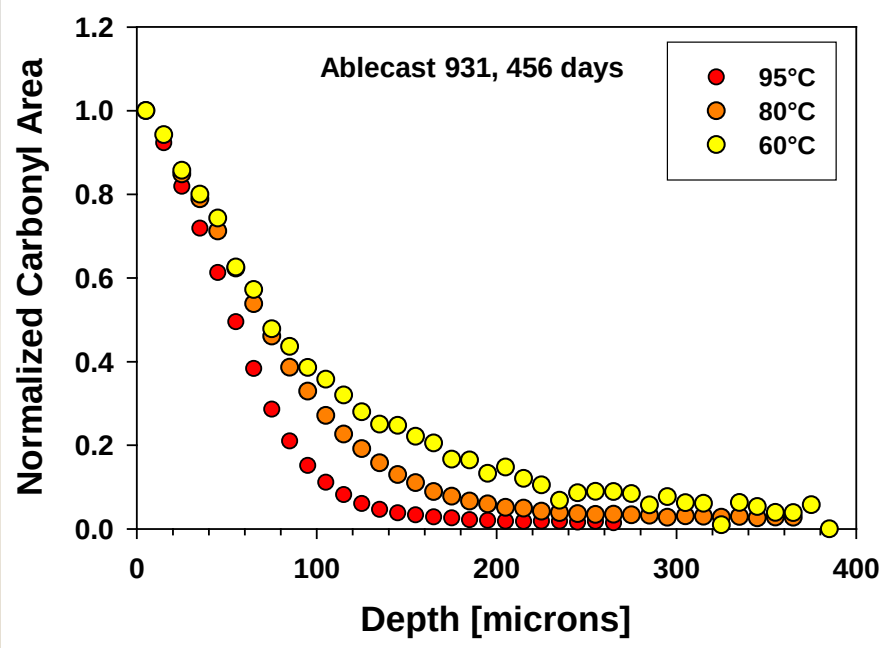




DLO phenomena validate effectiveness of a protective encapsulation material

Oxidation Depth Depends on Temperature

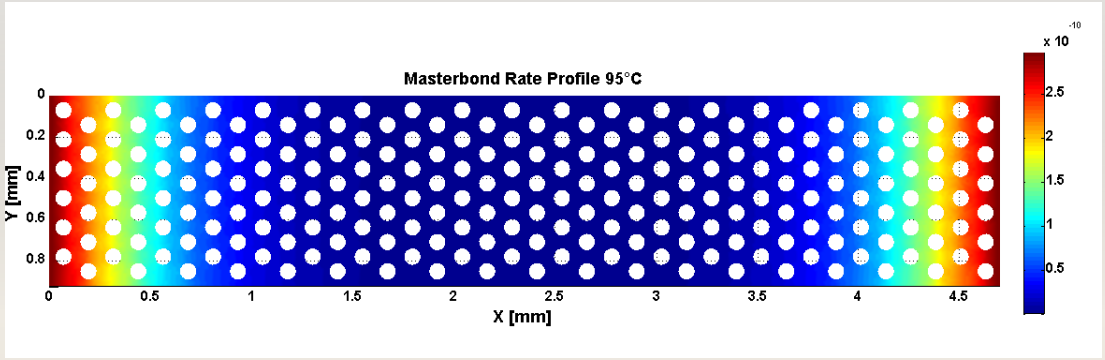
- Depth and shape of DLO profiles depends on temperature
- Oxidation is usually further into the material with lower temperature
- Normalized profiles show the effective behavior of $Ø/P_{O2}$

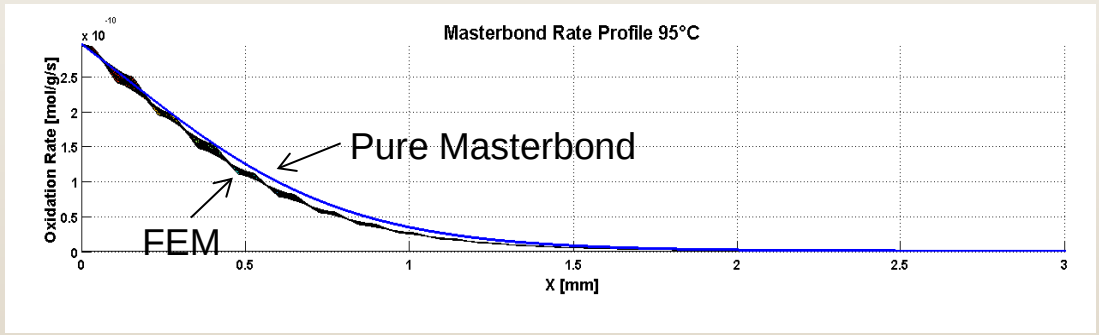


- **Systematic analysis of temperature dependent profile shapes establishes material parameters**
- **Such measurements validate our DLO concepts**

DLO Solves Degradation Issue in Application

- Problem: Composite structure appeared to consume O_2
- This affected a sealed environment in a component
- DLO modeling showed that a 'bake out' would be unsuccessful





FEM surface flux integration enabled estimation of total 'composite' reactivity in assembly: 1.17e-11 mol/cm^2/s
Pure resin: 1.72e-11 mol/cm^2/s

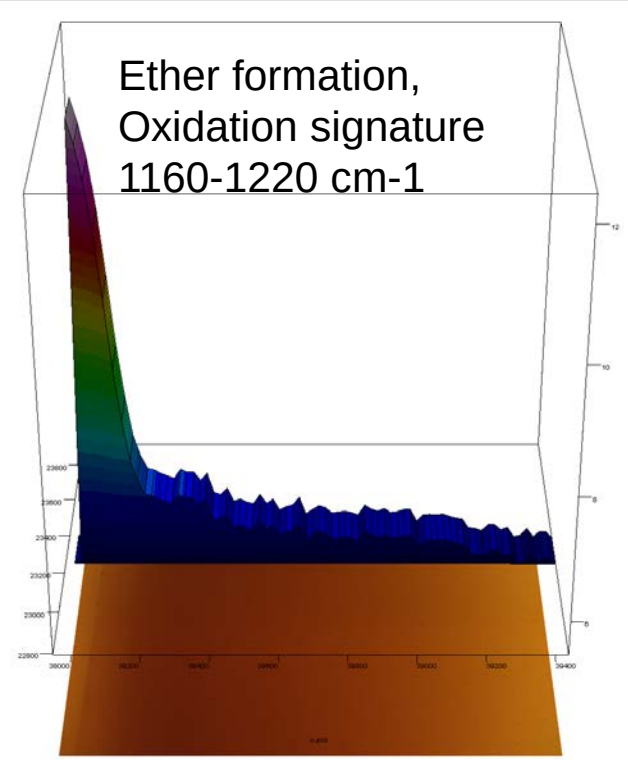
Solution: Ability to predict long-term O2 depletion kinetics through integration of DLO profile

Understanding DLO phenomena and accelerated aging behavior enabled a completely different solution to this problem

DLO Affects Materials Aging in System

- Qualification testing of cable harness encapsulation material
- PU polymer aged and showed stress cracking
- Micro ATR IR imaging shows significant DLO (surface oxidized material)

Thermally aged PU sample from aging tests supporting material selection/qualification

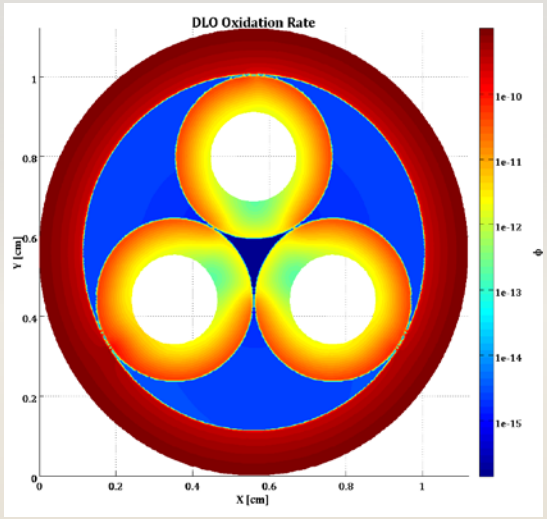


- **Understanding DLO phenomena helps to predict material and aging behavior at more moderate exposure conditions**
- **When does accelerated aging introduce mechanistic anomaly?**

DLO Connection with Nuclear Power Plants

- Requalification and accelerated testing of cable assemblies
- Is accelerated testing representative of ambient aging?
- What is the basis for condition monitoring approaches?

Brand-Rex Cable
300 kGy, 340 Gy/hr (2/20/15)
Accelerated testing



Modeled aging in XLPO cable.
Weak gradient in jacket, DLO through jacket and in cable interior

DLO phenomena are not considered in rapid qualification testing

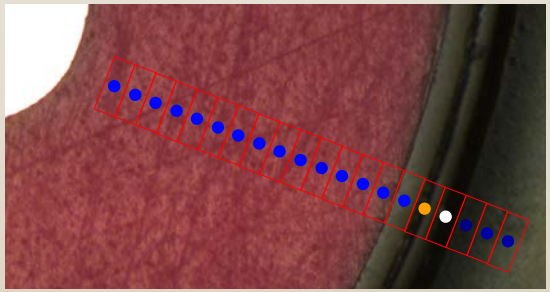
Celina MC, Gillen KT, Lindgren ER. Nuclear Power Plant Cable Materials: Review of Qualification and Currently Available Aging Data for Margin Assessments in Cable Performance, Sandia National Laboratories, 2013, SAND 2013-2388, pp 136.

DLO Governs Accelerated Aging Behavior

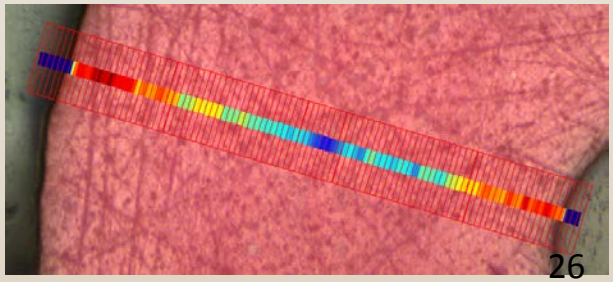
- Insulation within cable assembly does not experience oxidative aging
- Individual polymer removed from cable/conductor oxidizes
- **Fast accelerated aging DOES NOT represent ambient aging processes**

Brand-Rex Cable
300 kGy, 340 Gy/hr
Accelerated testing

Carbonyl imaging
1790-1675
with ATR IR microscopy



DLO implies major mechanistic differences

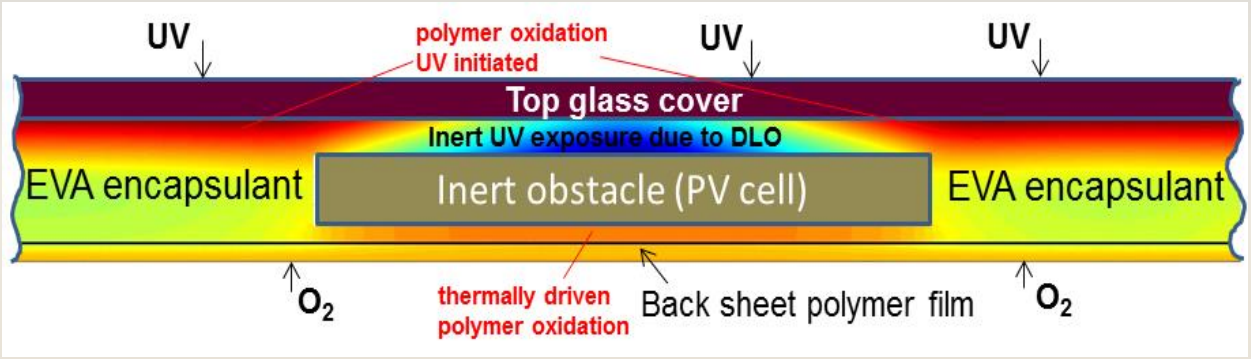


DLO and Solar Module Reliability

- Encapsulation material degrades
- Photo-thermal and oxidative versus inert processes

PV Module, ~15 years Arizona. Encapsulant is PHOTOCAP 15295

Picture of Xe Arc Aged Coupon: Glass-EVA-Glass, ~65 x 65 mm

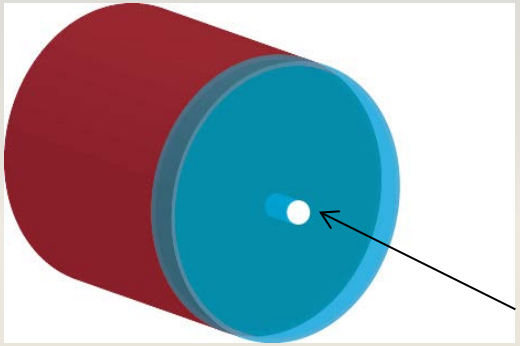


Perceived EVA heterogeneous degradation in PV applications due to DLO. Competition between surface UV induced and thermal degradation is strongly dependent on local oxygen availability.

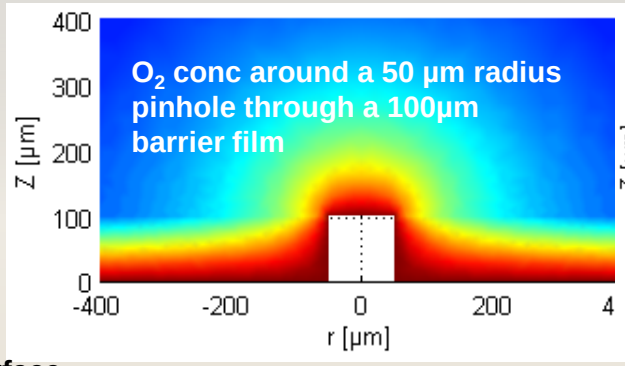
PREDICTS Proposal: Predictive Modeling and Verification of Degradation in Polymeric Encapsulants (Industry, Fraunhofer Institute, NIST, SNL)

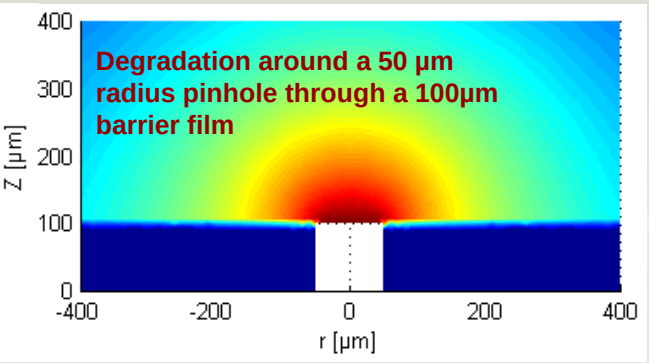
Reactive Diffusion Model Aids Material Reliability

- An application required effective barrier coatings
- Of concern was whether pinholes in a coating could 'absorb' O_2
- Geometry specific DLO modeling allowed prediction of O_2 flux into material and extent of local material degradation chemistry



Pinhole in top surface





- **Up to a pinhole size of 20 micron the flux (consumed O_2) approximately doubles**
- **Larger holes through the barrier coating will result in a noticeable total flux increase**

Our modeling tools can be applied to other aging scenarios

Similar Diffusion Processes Related to Failure

- Water diffusion phenomena, FEM codes for O2 are applicable
- Failure of immersed MV cables associated with H2O ingress
- Spot corrosion of shielding material may precede dielectric breakdown

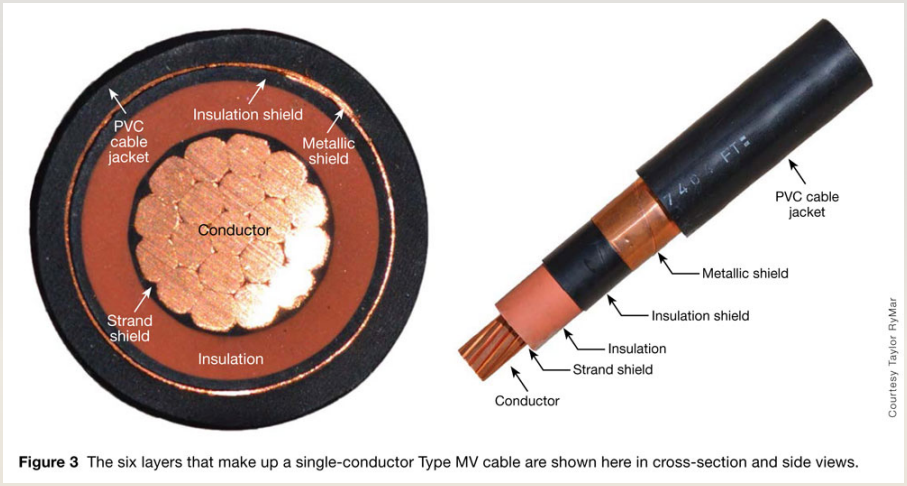


Figure 3 The six layers that make up a single-conductor Type MV cable are shown here in cross-section and side views.

Actual cable failure (aged neoprene jacket)

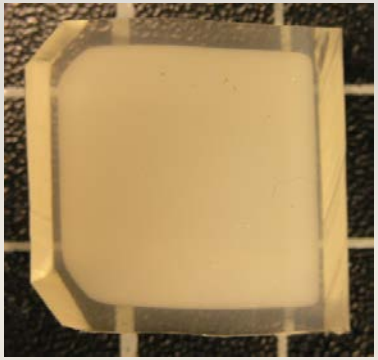
- **Inner inert core of 10mm OD, surrounded by 1" XLPE**
- **PDS estimation for XLPE: P=2e-8 ccSTP/cmHg/cm/s**
- **S= 0.046 ccSTP/cc/cmHg, D=5e-7cm²/s**

- **What about pin holes in shield between jacket and main insulation?**

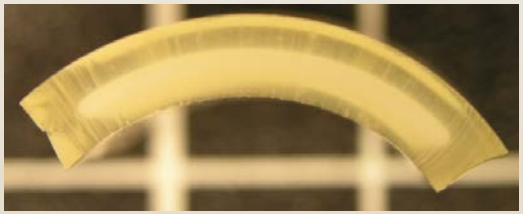
Water Diffusion Affecting Radiation Degradation

- Observed unusual discoloration for rad. degradation of vinyl in LICA pool
- Opaqueness was reversible
- Part of degradation process or intrinsic material property?

Aged vinyl after drying for 1d at RT
Opaqueness is in center of material, edges are drier



- Drying of new vinyl after 1d at RT
- Residual water in center of material

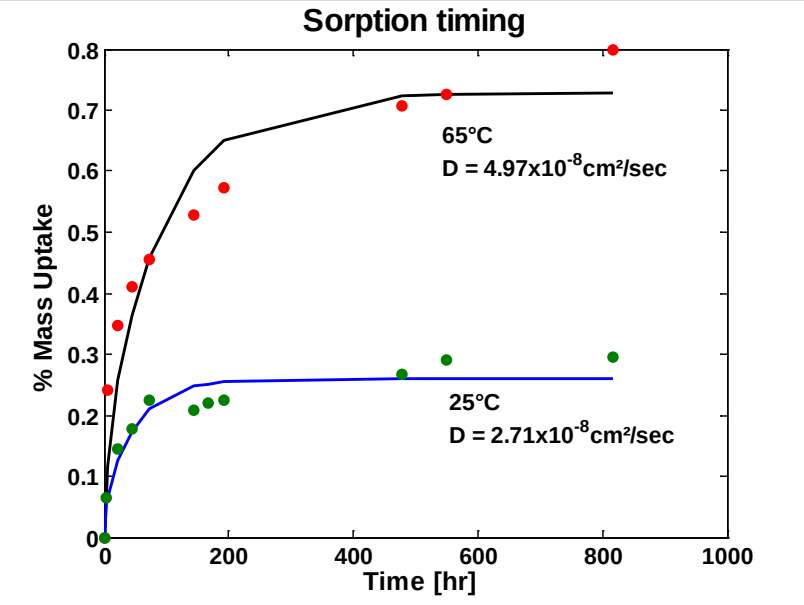


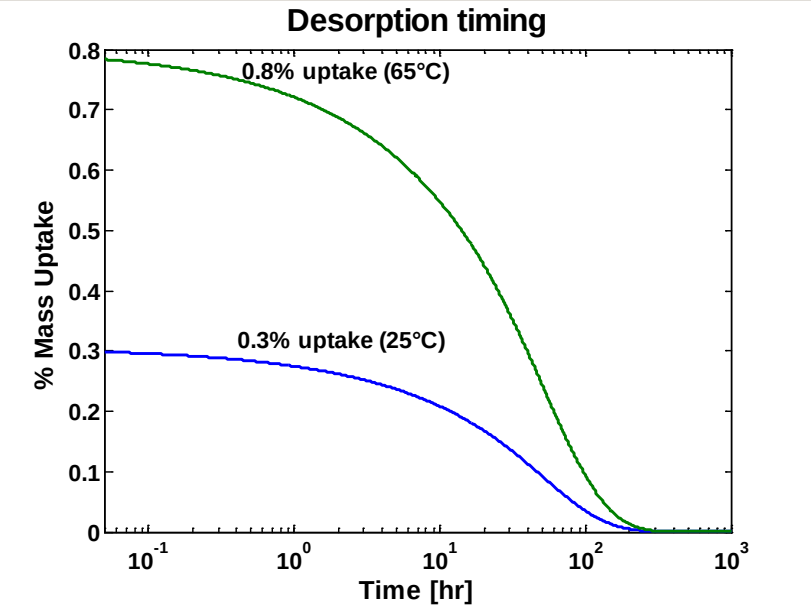
- Drying after 48 h at RT
- About 1/3 of material has water remaining
- Opaque above a specific water level

Physical absorbed water is part of the degradation process

Water Sorption/Desorption – Diffusion Physics

- Guidance for D obtained from water immersion weight increase data
- L = 3mm, D = 3 e-8 cm2/s at RT, 5 e-8 at 65˚C,
- S = 0.3% (2.0 ccSTP/cc-cmHg), S = 0.8% at 65˚C
- Drying (desorption curves) can be predicted. Timing as governed by diffusivity is identical, absorbed water and flux depend on time.

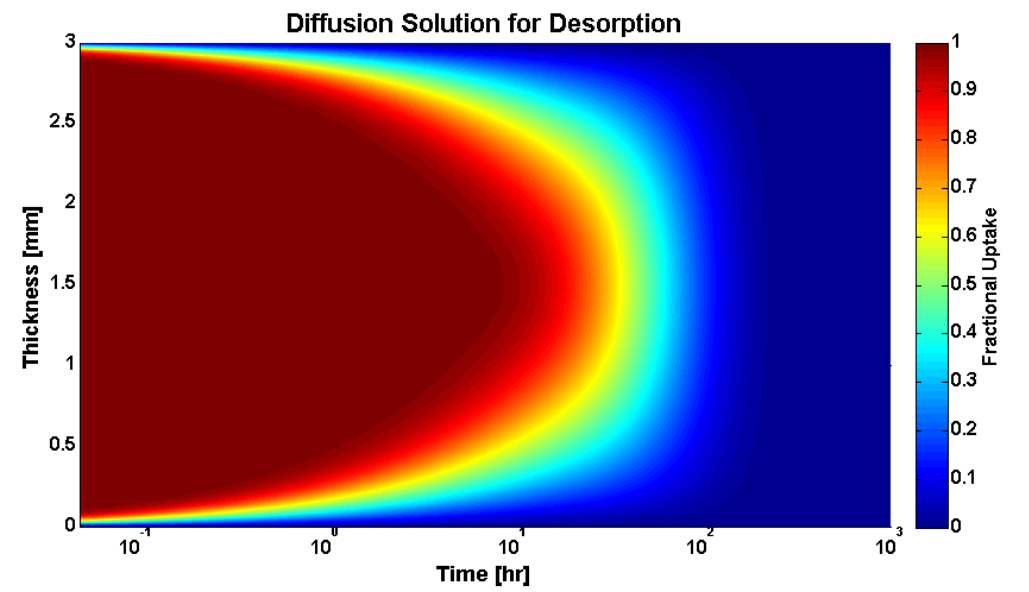


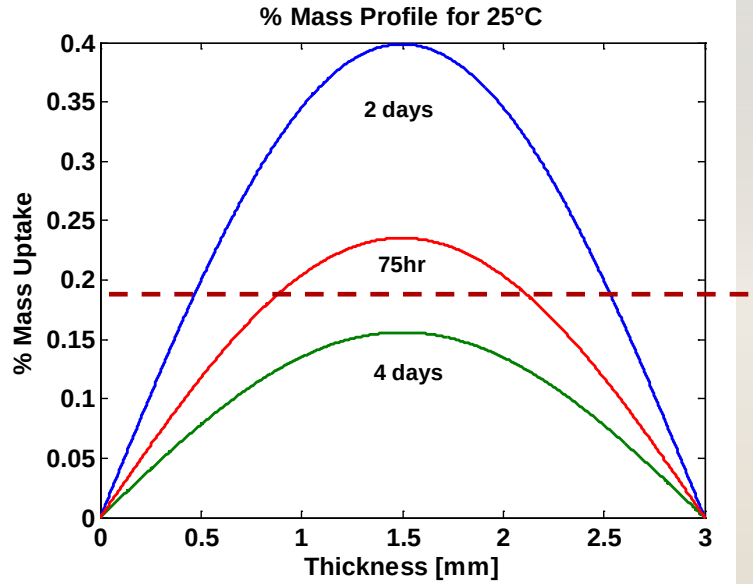


Water diffusion on the order of days

Diffusion Limited Water Profiles

- Fickian diffusion modeling can be expanded to predict momentary spatially resolved concentration profiles with time
- This is the same as the evolution of DLO profiles in polymer aging
- Here, drying commences at the edges and water is retained in center



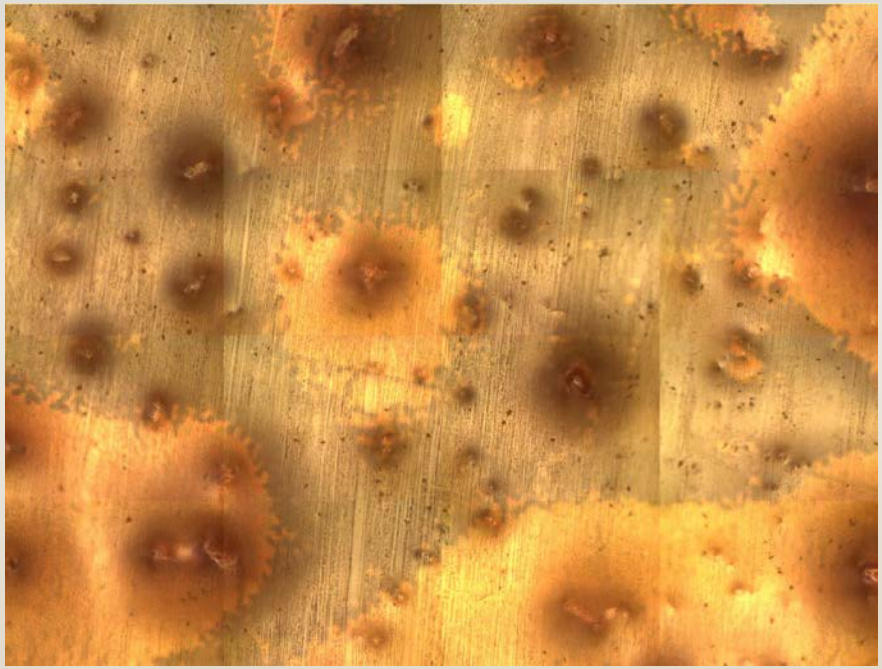


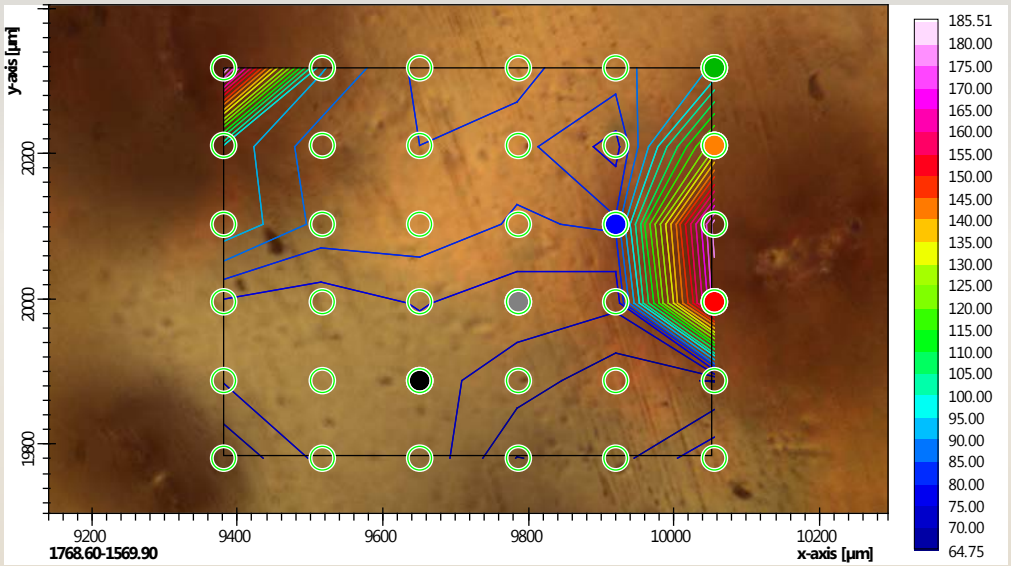
- Predicted local water concentration for drying after 2 days (0.25%), 75 h (0.15%) and 4 days (0.10%) at RT (for initial 0.8% 65°C)

Chemical conditions are affected by diffusion physics

Heterogeneity in Epoxy Oxidation

- Thin 100 um film processed/cured at RT, aged 86 days at 125˚C
- Severe oxidation of adhesive (FA51), just a demo of material behavior
- Darker regions around inorganic silica agglomerations
- Imperfect mixing and local catalytic environments



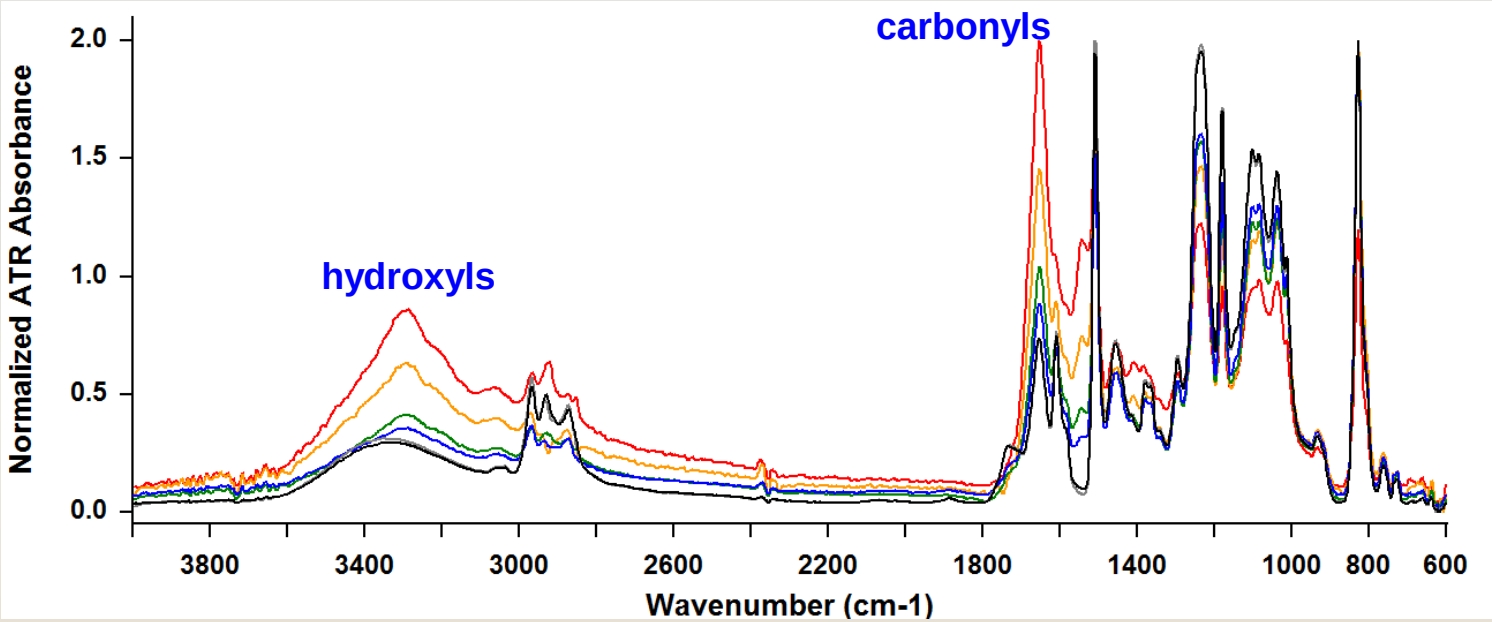


Location specific spectra are on next slide

Spatial variations in oxidation level – carbonyl imaging

Heterogeneity in Epoxy Oxidation

- Thin 100 um film processed/cured at RT, aged 86 days at 125˚C
- Carbonyl variation with x-y location, not with depth

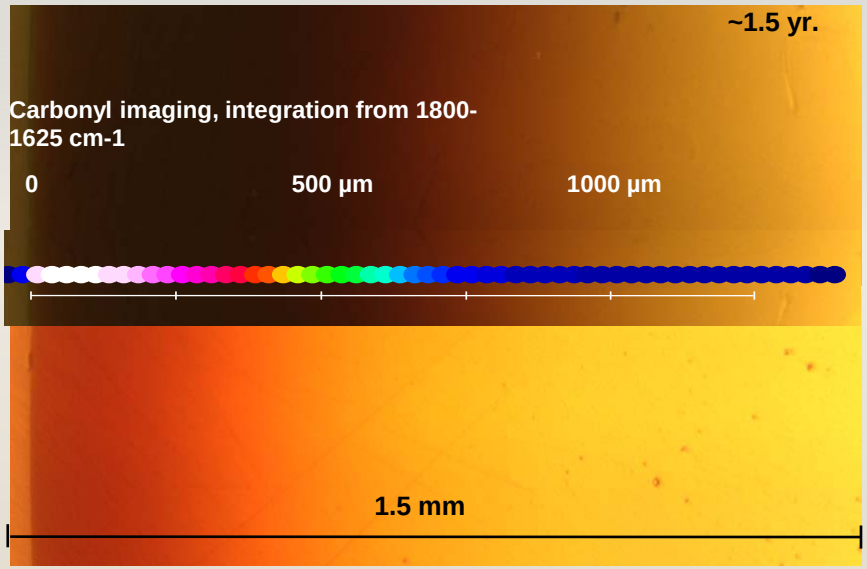


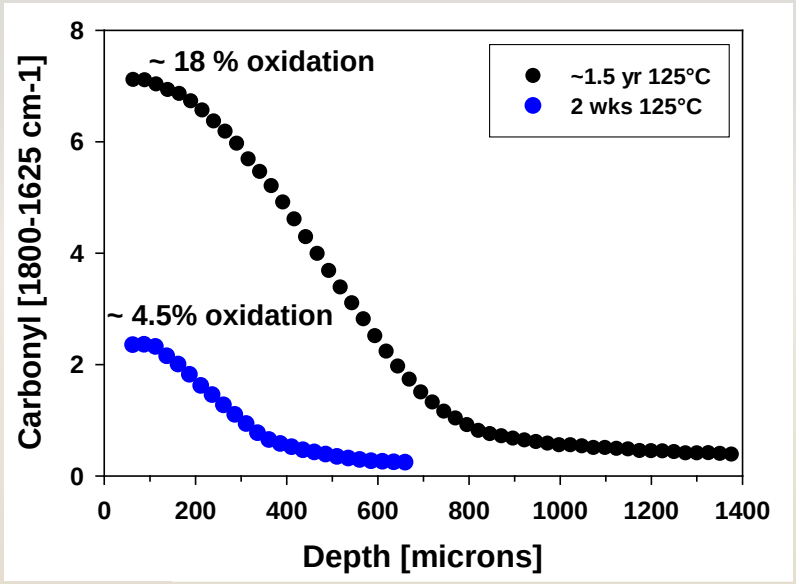
Spatial variation in epoxy film degradation has also been observed in photo-oxidation (NIST work)

DLO Stage II – Oxidized Layer Formation

- Only a fraction of the polymer can be oxidized
- Local reactivity will decrease with time (limit of oxidizable material)
- Permeation will drive oxidized zone deeper into the material
- Relevant for high temperature composite degradation phenomena

Masterbond aging at 125°C





Early degradation is classic DLO - long term an oxidized layer will push further into the material – permeation and ox-rate slows

Future work

- Move from static equilibrium to time-dependent models
- Advanced 3D models for multiple materials assemblies
- Hydrolytic aging models, very challenging due to need for 'reactivity' with local concentration in polymer
- Better parameter precision for material properties (beta factor)
- Improved permeability measurements, ongoing activity
- Couple polymer physics (relaxation) with chemically driven degradation, goal: Unified Aging Models
- Paper in preparation "Permeability of O_2 through polymers"
- Further demonstrate the challenges for 'accelerated aging' and its predictive value due to DLO

Summary & Impact

- SNL leads this field internationally
- Developed DLO and reactivity models for a range of materials and conditions with new computational approaches
- Resolved multiple material challenges for internal and external SNL customers
- Dedicated long-term research effort has led to fundamental understanding of material behavior and theoretical description
- We are the experts for O_2 permeation and reactivity with polymers within the DOE complex, our papers are widely cited

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Celina MC. Review of polymer oxidation and its relationship with materials performance and lifetime prediction. Polym Degrad Stab 2013;98:2419.