A Hybrid Physics-Based, Data-Driven Approach to Model Damage Accumulation in Corrosion of Polymeric Adhesives

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2021 U.S. DOE Vehicle Technologies Office
Annual Merit Review

Project ID #: MAT152 May 11, 2021

This presentation does not contain any proprietary, confidential or otherwise restricted information.

Project overview

Partners

- Michigan State University(MSU) (Lead)
- Robert Bosch LLC.
- Endurica LLC.
- JDV Lightweight, LLC. (consultant)
- Composite Center@MSU (subcontract)
- Dow Chemicals, Parker Lord (suppliers)

Budget

- DOE Share: $ 967,662
- Collaborators Share: $ 474,526
- Cost Share: 32.9%
- FY 2020 DOE Share: $ 612,311

Timeline

Start: January 2019
End: December 2022
Completion: 75%

Barriers*

1. Lack of reliable joining technology for dissimilar materials
2. Lack of cost-effective tests for evaluation of corrosion
3. Lack of constitutive model capable of predicting corrosion
4. Predictive modeling tools
   - Prediction error <10%
   - Lack of validated test protocols

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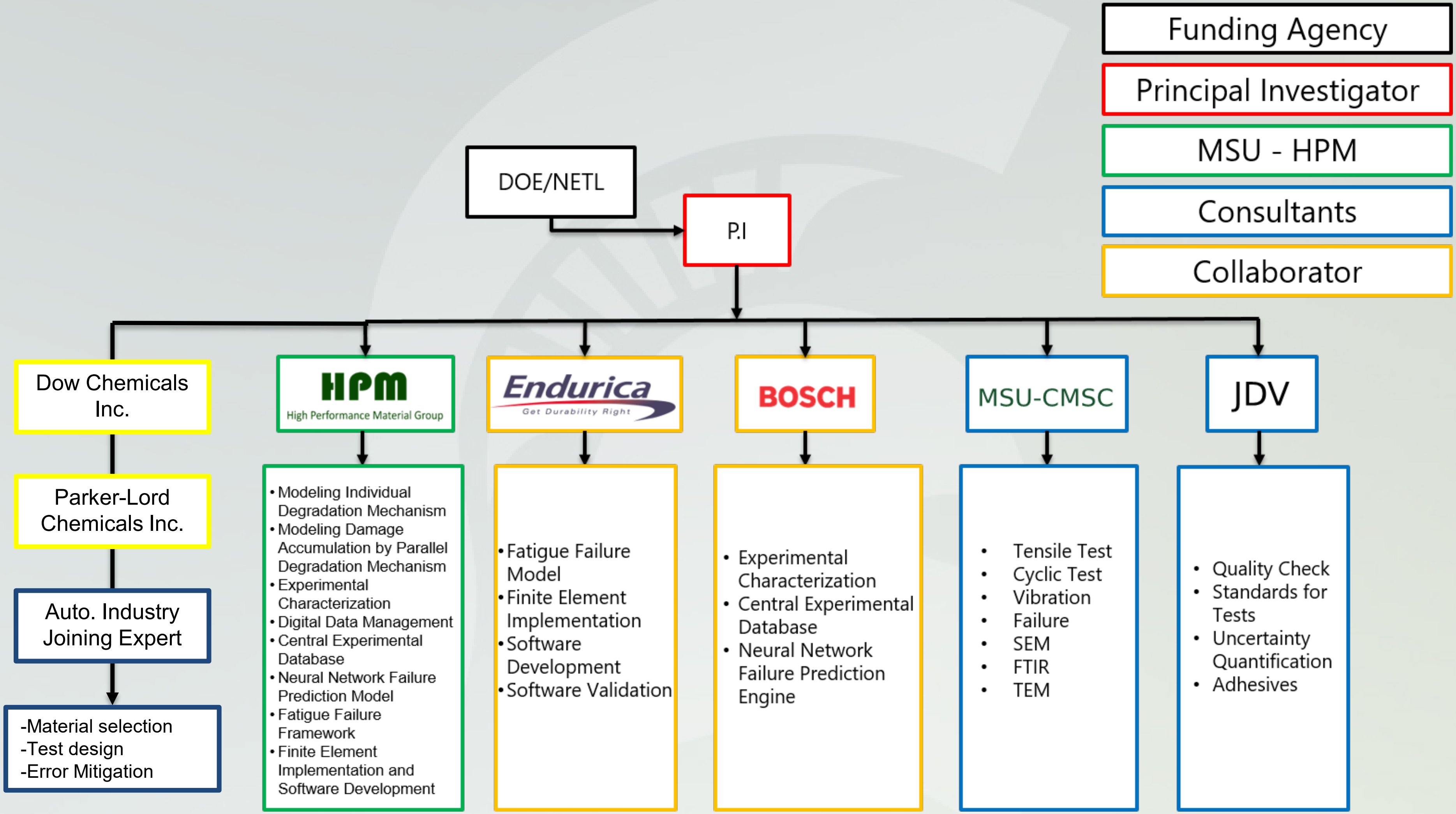
Read *Light Duty Workshop Final Report and U.S. DRIVE MTT Roadmap, section 5.1.

Collaboration and Coordination



Relevance & Objectives

Overall Objectives:

- A **theoretical model** to describe damage accumulation in constitutive behavior with respect to (1) deformation, (2) vibration, (3) hydrolysis, (4) thermo-oxidation and (5) photo-oxidation.

- A **software** to predict failure of cross-linked polymeric adhesives with respect to damage accumulated by environmental and mechanical loads with a **10% error**.

Impact/Relevance to DOE

Predicting failure in adhesives of dissimilar materials is necessary to

- facilitate use of lightweight material for vehicle mass reduction
- **Speed up the design** of composite joints in vehicle structures for light-weighting to address DOE 2030 targets
- **reduce time/cost** required for testing corrosion failure which makes the use of lightweight materials more attractive for OEM
- Improve CAE prediction capability to achieve a **reliable service-life** of joints

Critical segments

Headlamps, Brake lights and reflector housing

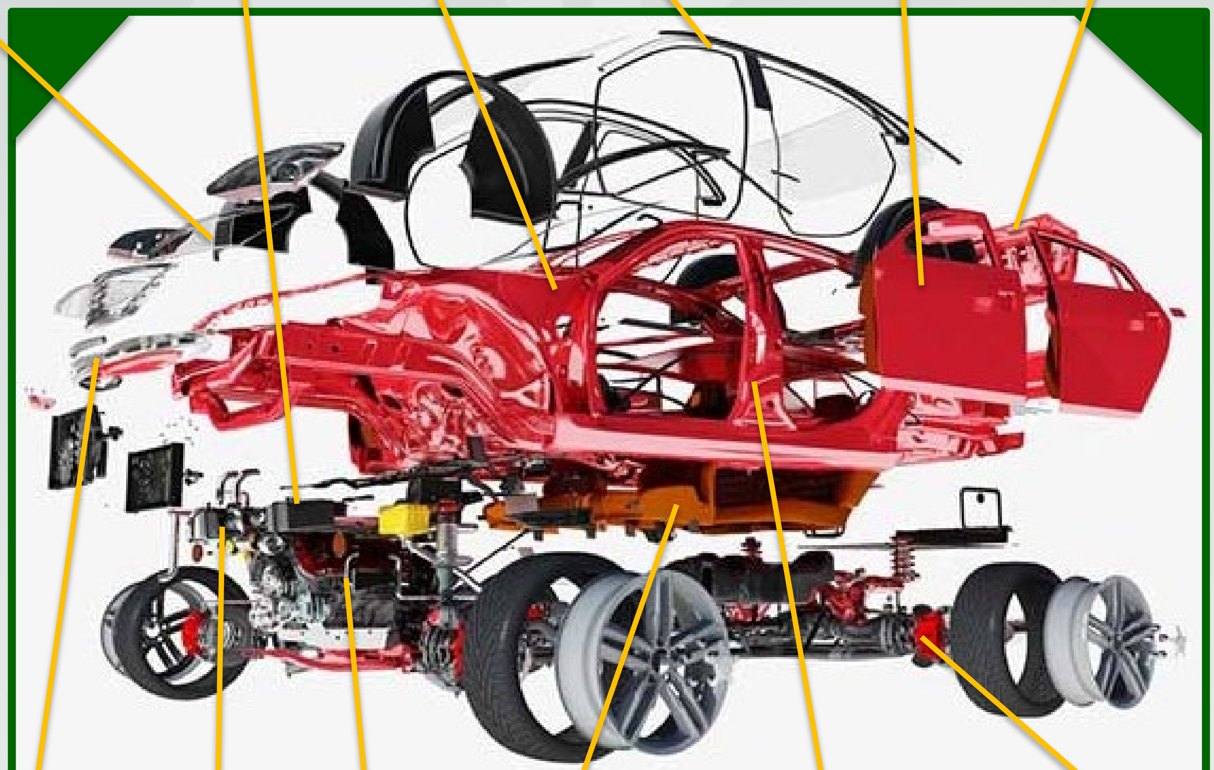
Electronics

Cockpit

Exterior Trim

Door Capping and Modules

Spoiler



Bumper

Filters

Motor Parts

Seats

Structural modules

Brake shoes and pads friction material

Degradation	
Thermo	
Hydro	
Hygro	RH%
UV	UV

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Approach & Milestones

In Progress	+	
Finished	✓	

FY-2019		FY-2020		FY-2021		FY-2022
Development	Individual Damage Mechanism	Platform Assembly	Recursive Neural Network	Experiment Database	Framework Assembly	UMAT software design
Development of a theoretical platform for constitutive modeling of adhesives ✓	Quasi-Static deformation ✓ Vibration ✓ Thermo-oxidative ✓ Photo-oxidative ✓ Hydrolysis ✓	Hydro+ Thermo ✓ Photo+ Thermo ✓ Photo + Thermo + Hydro +	Predict corrosion-induced failure with multi-vector input parameters +	1. Effect of environmental factor ✓ 2. Effect of Mech. factors ✓ 3. Database Completion by virtual points ✓	Validation of hybrid platform on combined mechanism in the lab Endurica Get Durability Right	Software predictions under lab condition against sample adhesives exposed to all combination mechanisms Endurica Get Durability Right
	BOSCH	BOSCH	BOSCH	+	BOSCH	
FY-2019	FY-2020	FY-2020	FY-2021	FY-2021		FY-2022

Micro- to macro- scale modeling, Central Experimental Database

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Milestones

Completed	
In-progress	
Shifted to next year	

Finished FY19	Derivation & Validation of the quasi-static model		Milestone
	Derivation & Validation of the vibration induced damage model		Milestone
	Derivation of the Hydrolysis model		Milestone
	Derivation & Validation of Thermo-oxidation model with multiple adhesives		Go/No-Go
Finished FY20	Validation of Hydrolysis model with multiple adhesives		Go/No-Go
	Validation of the modular platform concept		Milestone
	Derivation of Accumulative Damage Failure Model	Validation (2021)	Milestone
	Derivation of photo-oxidation model with multiple adhesives	Validation (2021)	Go/No-Go
	Derivation of coupled Thermo- & photo-oxidative model	Validation (2021)	Milestone
	Derivation of coupled Thermo-oxidative & Hydrolysis model	Validation (2021)	Milestone
Ongoing FY21	Training/Fitting Neural network engine	Validation (2022)	Milestone
	Hybrid platform on combined degradation	Validation (2022)	Milestone
Plann ed FY22	Software predictions against sample adhesives exposed to all combination mechanisms for all degradation mechanisms		Milestone

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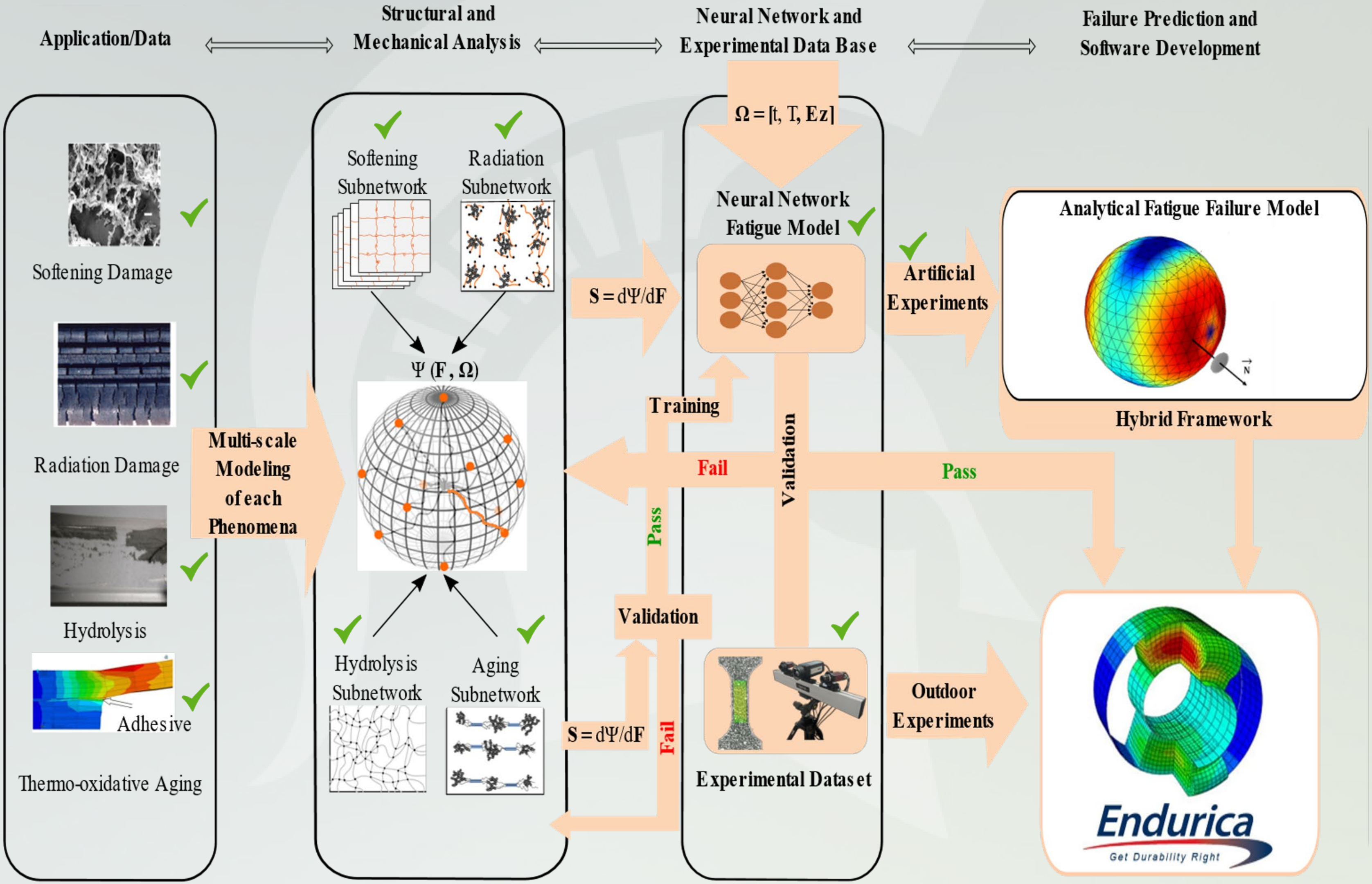
Any proposed future work is subject to change based on funding levels.

Approach- Modeling

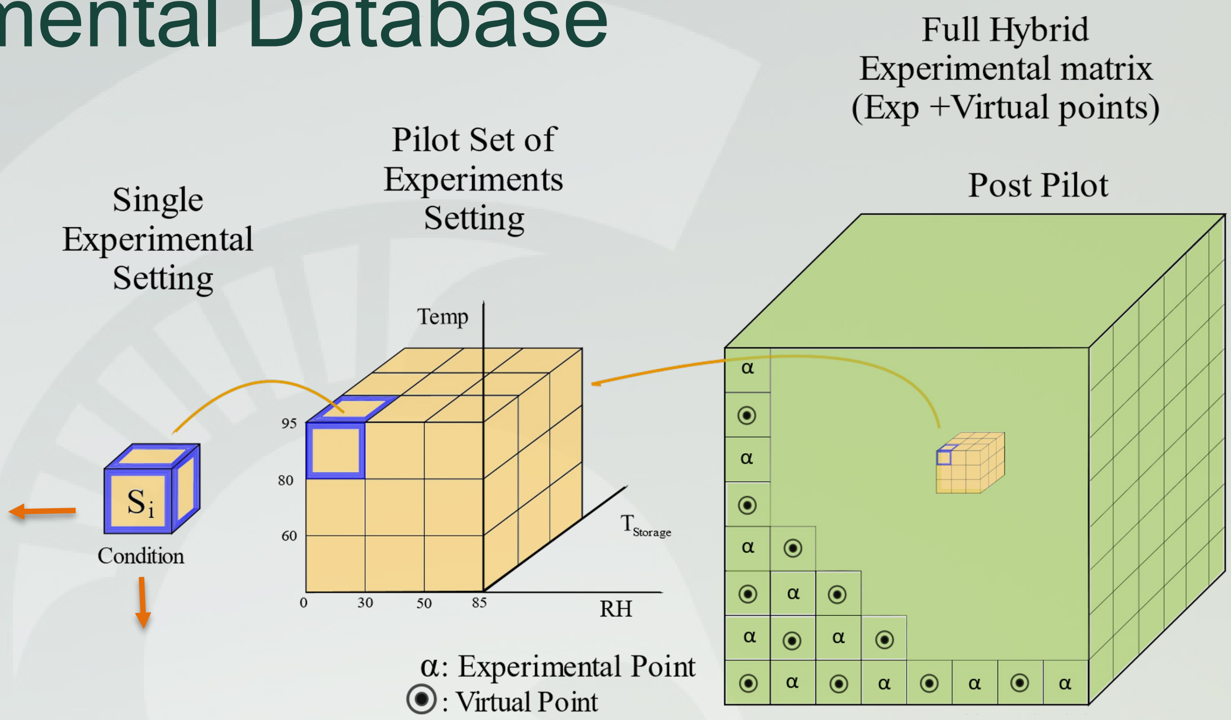
In Progress	+	
Finished	✓	



Central Experimental Database (Pilot test)

S_i

Condition	Range
$T_{storage}$	1 day – 2 years
Relative Humidity	0 – 80 %
Temp.	-5 – 200 C
UV	1 – 2 kW/m²/nm



Test Type \ Material		ACR	DC	PUB	PUG
Mech. Characht. tests	Failure	✔	✔	✔	✔
	Cyclic	✔	✔	✔	✔
	Permanent Set	✔	✔	✔	✔

Test Type \ Material		ACR	DC	PUB	PUG
Chemical. tests	FTIR	100%	100 %	100 %	100%
	DSC	100%	100 %	100 %	100%
	Cross link Density Measurement	100%	100 %	100 %	100%

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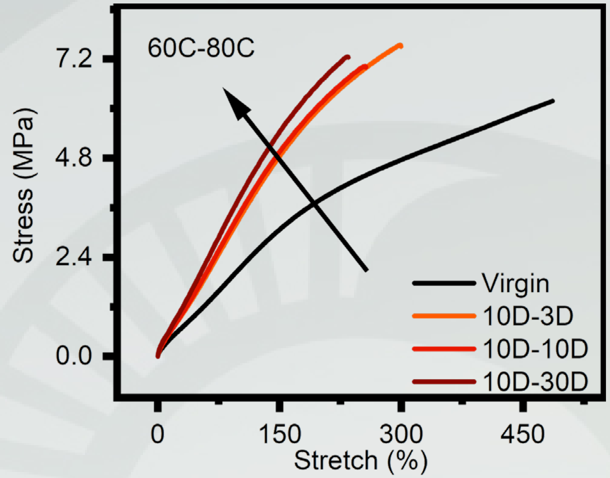
Thermo-Oxidation Analysis

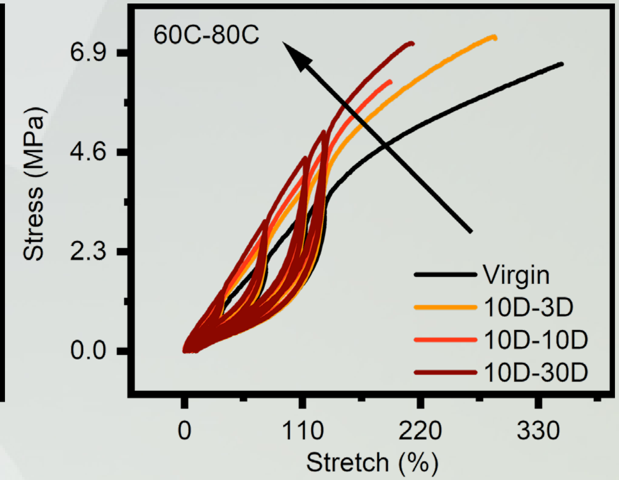
Experiments

1. Thermal-oxidation
2. Photo-thermal oxidation (Photo + Thermal)
3. Hydrolysis (Hydro + Thermal)
4. Hygrothermal (%RH + Thermal)

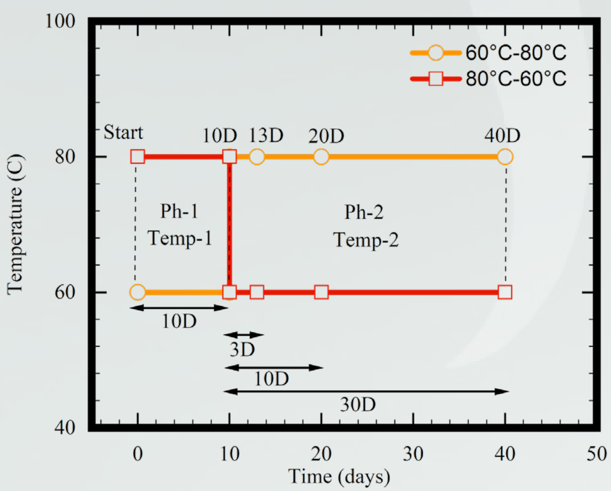
Polyurethane: Temperature-jump

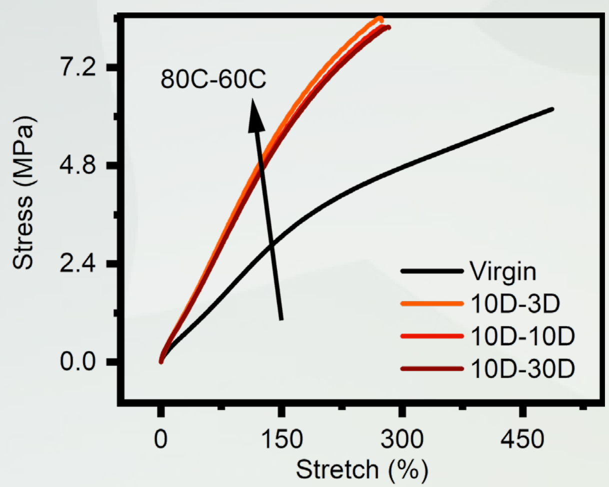
- Adhesive was aged at 60℃ to 80℃ and vice versa.
- Degradation due to higher temperature.
- High temperature causing crosslink formation.

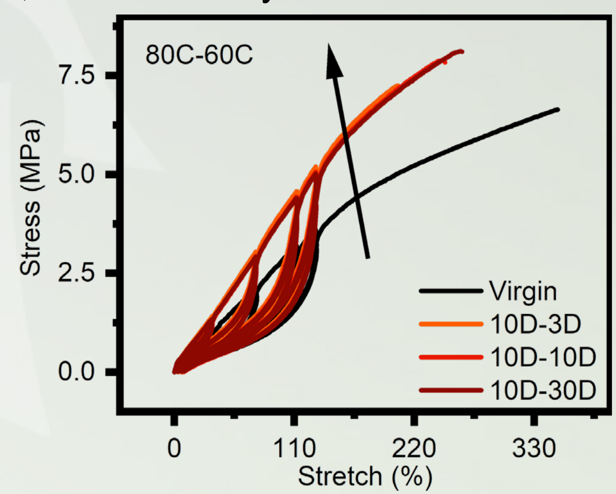




Specimens aged in 60℃ for 10-days and then aged in 80℃ for 3, 10 and 30-days







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Specimens aged in 80℃ for 10-days and then aged in 60℃ for 3, 10 and 30-days

Experiments

1. Thermal-oxidation
2. Photo-thermal oxidation (Photo + Thermal)
3. Hydrolysis (Hydro + Thermal)
4. Hygrothermal (%RH + Thermal)

Single condition aging

To determine short and long-term aging effects:

Intensity of 1 W/m²/nm

Aging periods: 1 – 270 days

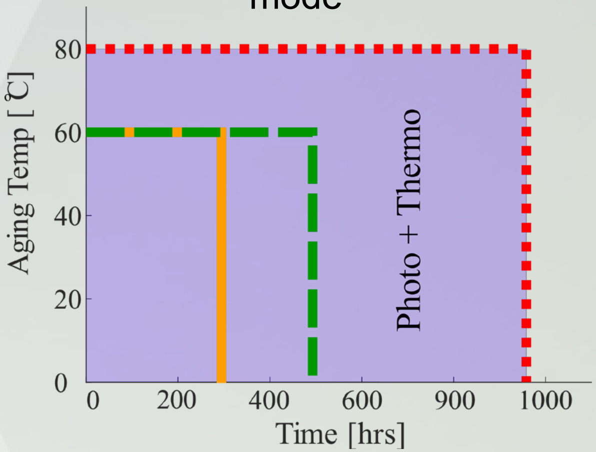
To determine high and low temperature effects:

Aging Temperatures: 45- 80 C

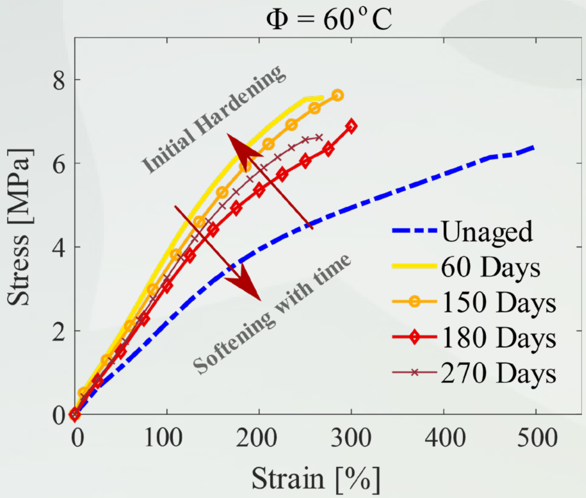
Materials break down in extended periods and high temperatures

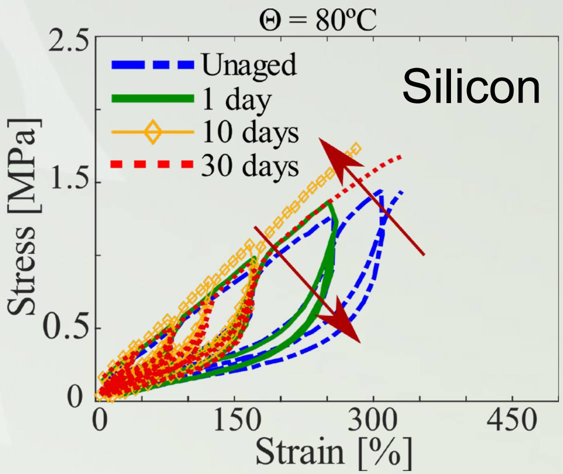
UV Machine

Single aging mode



Poly-Urethane





Temperature jump / Dual condition aging

To further investigate the effects of Temp + Irradiation, Temperature Jump & Dual aging was conducted

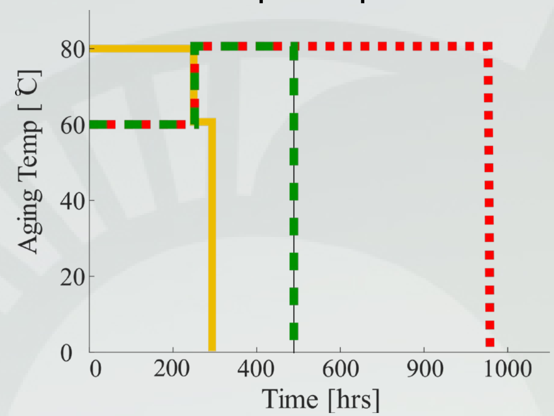
Material ages to a limit, and further deterioration is limited.

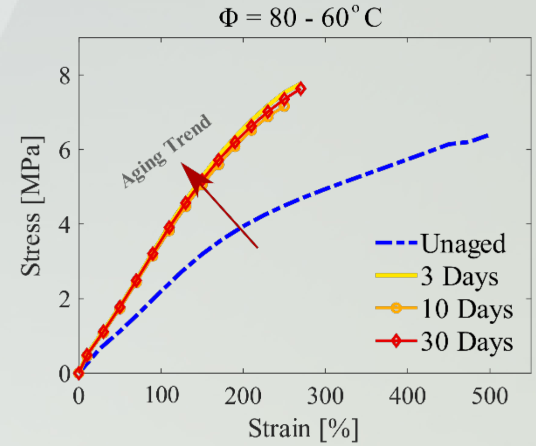
Independent of aging period and temperature regime.

We conclude that photo-oxidation dominates material behavior more than thermo.

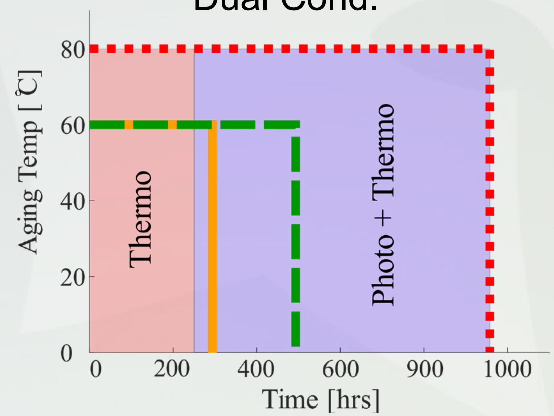
Poly-Urethane

Temp Jump





Dual Cond.



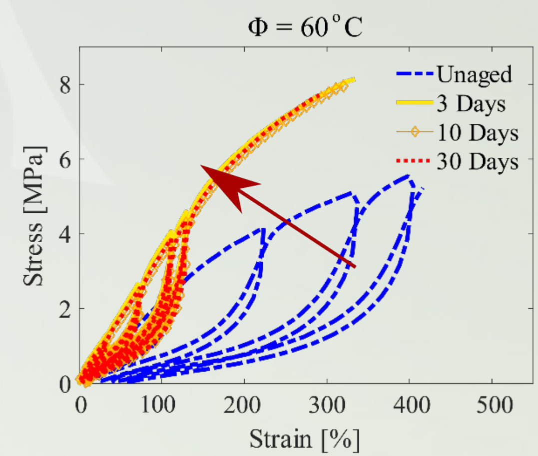
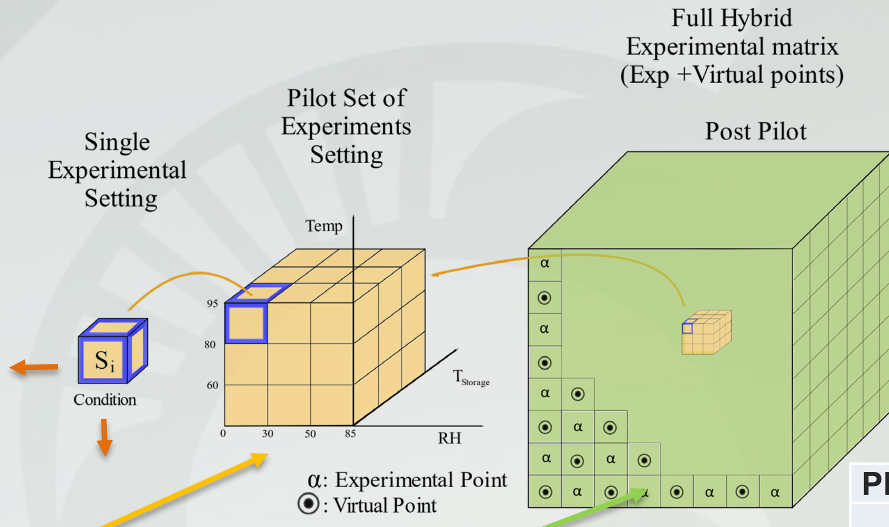


Photo-Experimental Database

Condition	Range S_i
$T_{storage}$	1 day – 5 months
Relative Humidity	0% RH



LEGEND	symbol
Completed	✅
On-going Aging	☑
Test in-progress	✔
Discontinued	✗

	Material \ Mech Test Type		Pilot			Post Pilot				
			1D	10D	30D	3M	6M	9M	1.5Y	2
Photo TESTS	DC	Failure Cyclic	✅	✅	✅	✅	✅	✅	✗	✗
	PUB	Failure Cyclic	✅	✅	✅	✅	✅	✅	✗	✗
	PUG	Failure Cyclic	✅	✅	✅	✗	✗	✗	✗	✗
Dual Effect		DC	✔	✔	☑					
		PUB	✅	✅	✅					
Temp Jump		DC	✔	✔	☑					
		PUB	✅	✅	✅					

PHOTO GROUP PROGRESS		
Material \ Mech Test Type		Pilot
DC	FTIR	✅
	DSC	✅
	Cross-Link density	✅
PUB	FTIR	✅
	DSC	✅
	Cross-Link density	✅
PUG	FTIR	✅
	DSC	✗
	Cross-Link density	✗

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Photo-Thermo Relaxation set

- Designed and Created in-house
- Material used was poly-urethane.
- Test was conducted in thermo-oxidation and photo-oxidation
- Greater relaxation at higher temperatures
- UV Irradiation counteracts relaxation caused by temperature

Thermo-oxidation

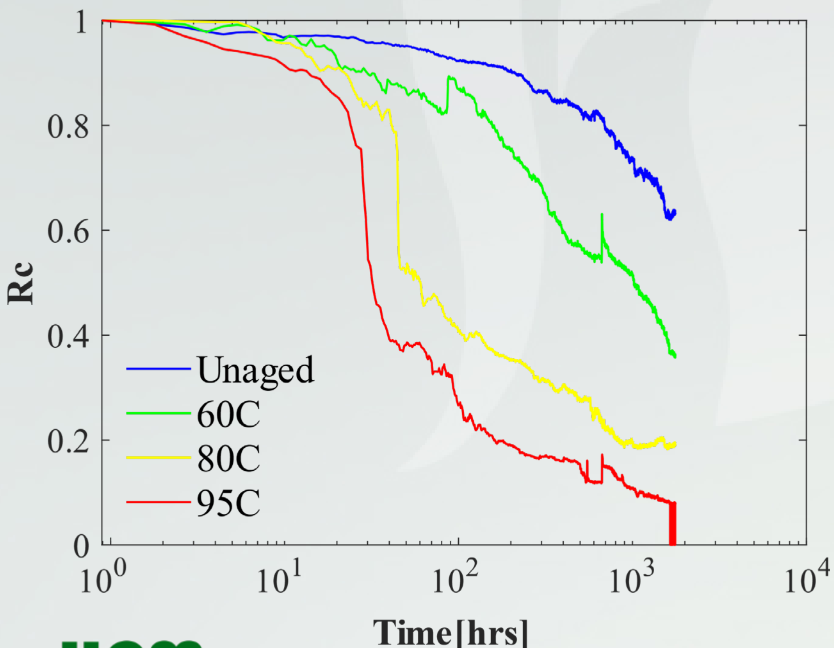
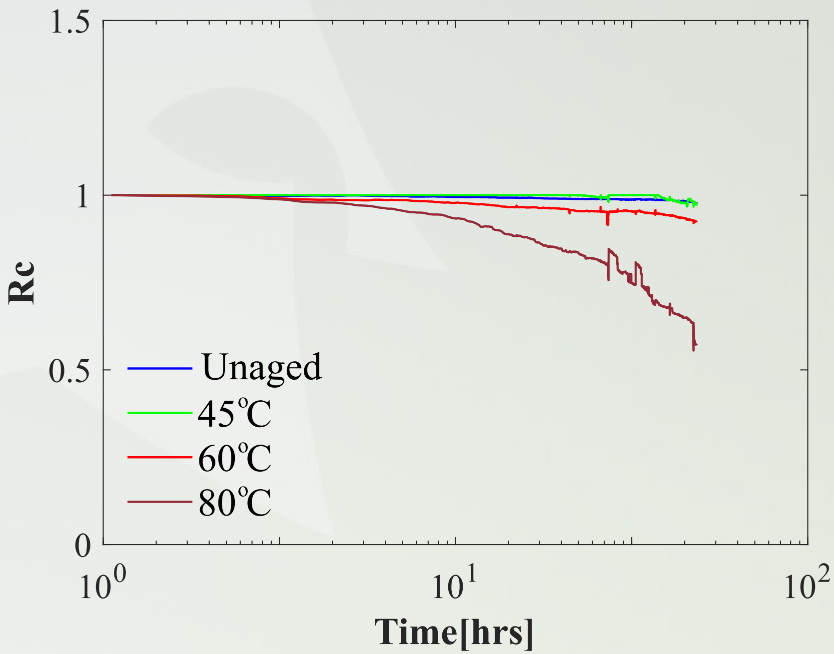


Photo-oxidation



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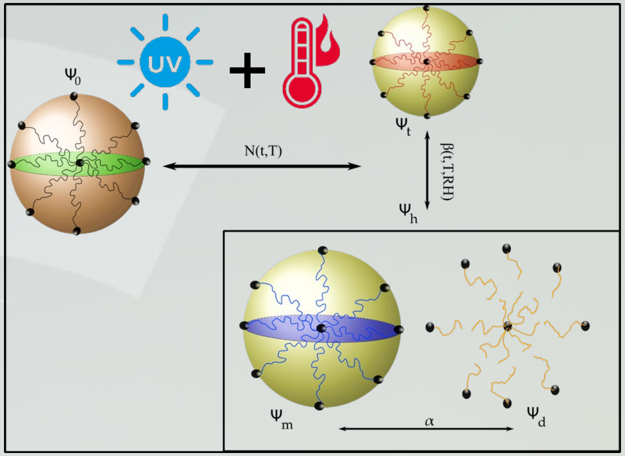
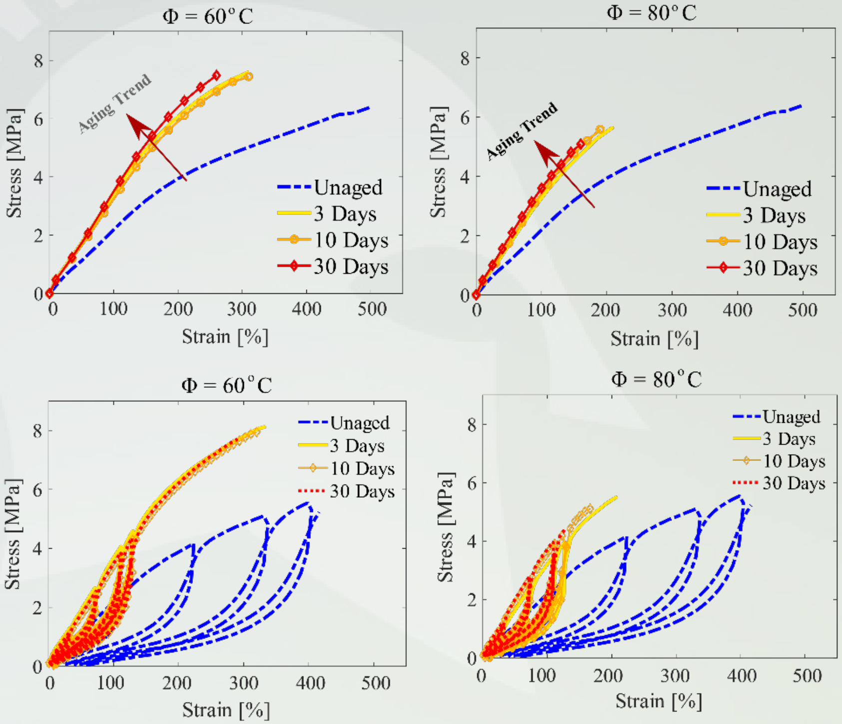
Dual condition: Photo –Thermo aging



Dual Condition: PUB

- Was aged for 10 days in either 60 or 80° C.in thermo-oxidative conditions
- Was then aged in the same temperature in photo-oxidative conditions for durations 3, 10 & 30 days
- Failure and cyclic tests were conducted.
- Material degrades from higher temperature aging mode.



Specimens aged in Dual aging at 60 & 80℃ for 30 days for thermo-oxidation at 0%RH for then moved to photo-oxidation for 3, 10, and 30 days.

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Hygrothermal / Hydrolytic Analysis

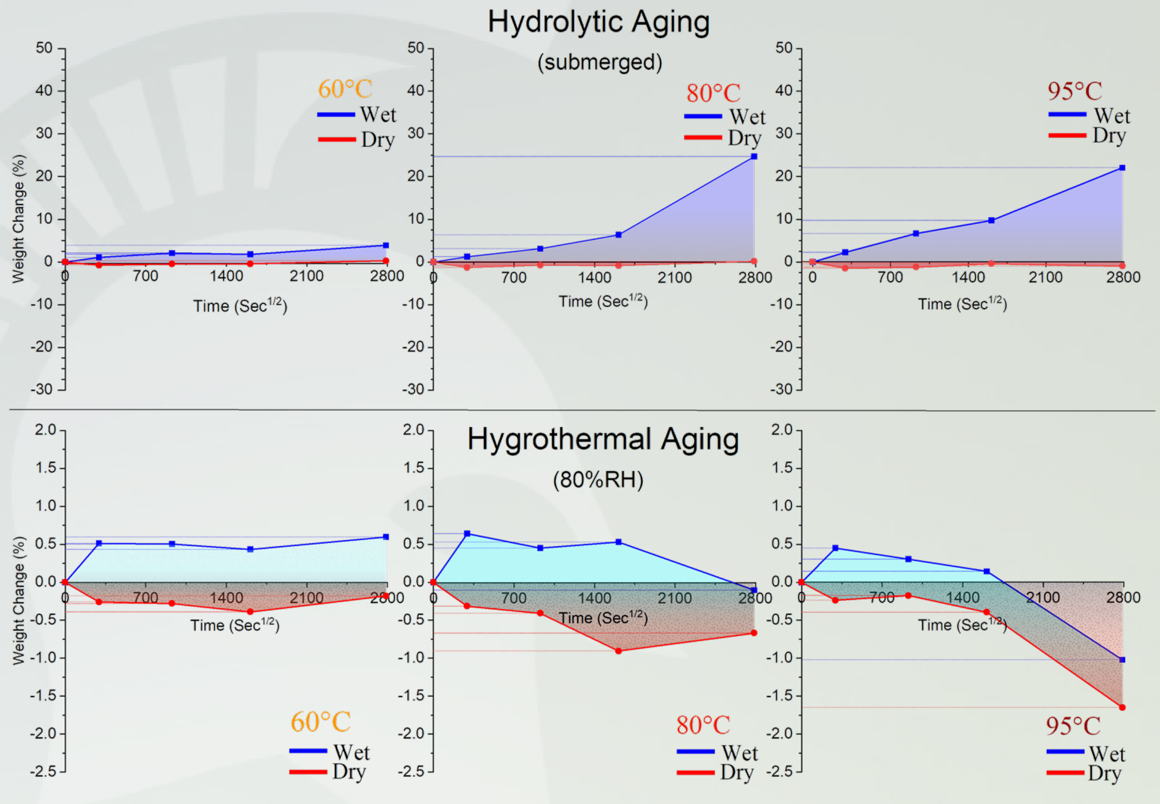
Polyurethane

Water uptake as a damage precursor

- Hydrolytic aging
  - High water uptake and swelling
  - Severe damage and reduction in crosslink density
  - Linear trend with square root of time

- Hygrothermal aging
  - Low moisture uptake (0.5-1%)
  - Mass loss α T

PU specimens after aging in hydrolytic condition after 90 days. Maximum swelling observed in 80C.



Water uptake and mass-loss

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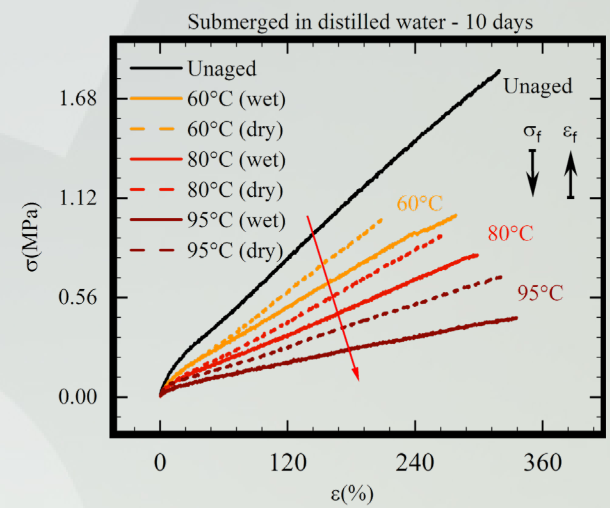
Experiments

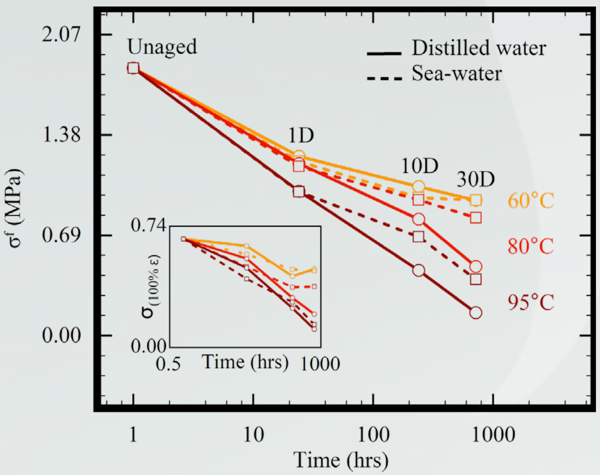
1. Thermal-oxidation
2. Photo-thermal oxidation (Photo + Thermal)
3. Hydrolysis (Hydro + Thermal)
4. Hygrothermal (%RH + Thermal)

Damage reversibility in hydrolytic aging

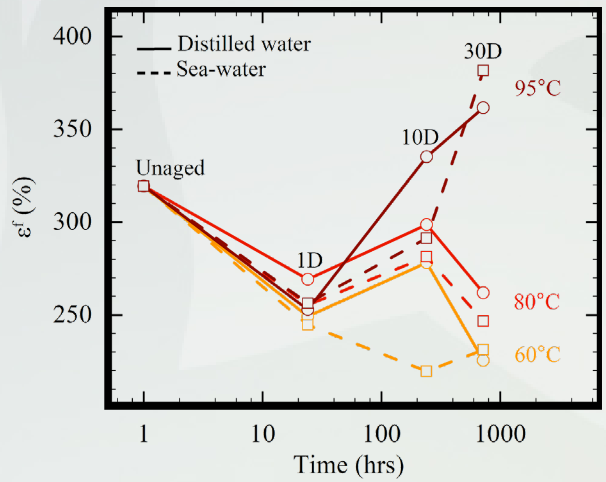
Silicone

- Mechanical testing in wet state vs dry state
- Softening behavior due to plasticization
- Better mechanical properties after removing water
- Higher damage in distilled water than sea-water (salt barrier)
- Stress-strain behavior indicates severe chain scission during hydrolytic aging
    - Stress decreased with increase in time (t) and temperature (T) i.e. σ α 1/T, 1/t
    - Strain increases with the increasing temperature but decreases with time i.e. ε α T, 1/t

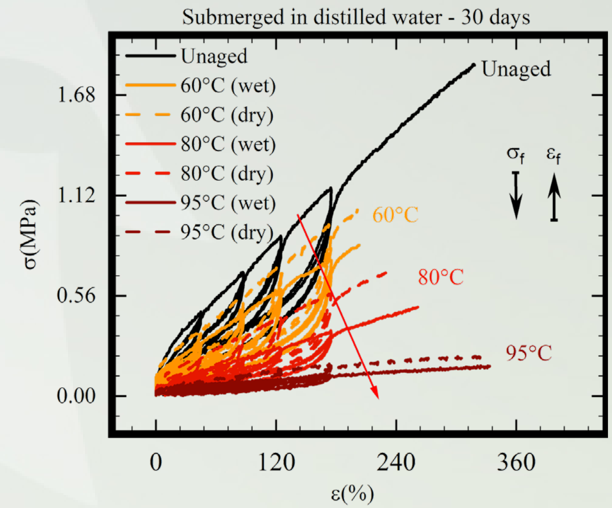




σ variation
distilled water vs sea-water



ε variation
distilled water vs sea-water



Failure stress (σ) and strain (ε)
Wet vs Dry silicone adhesive samples

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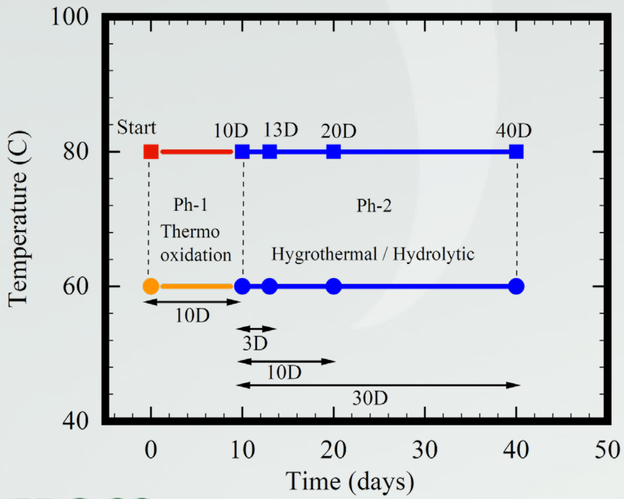
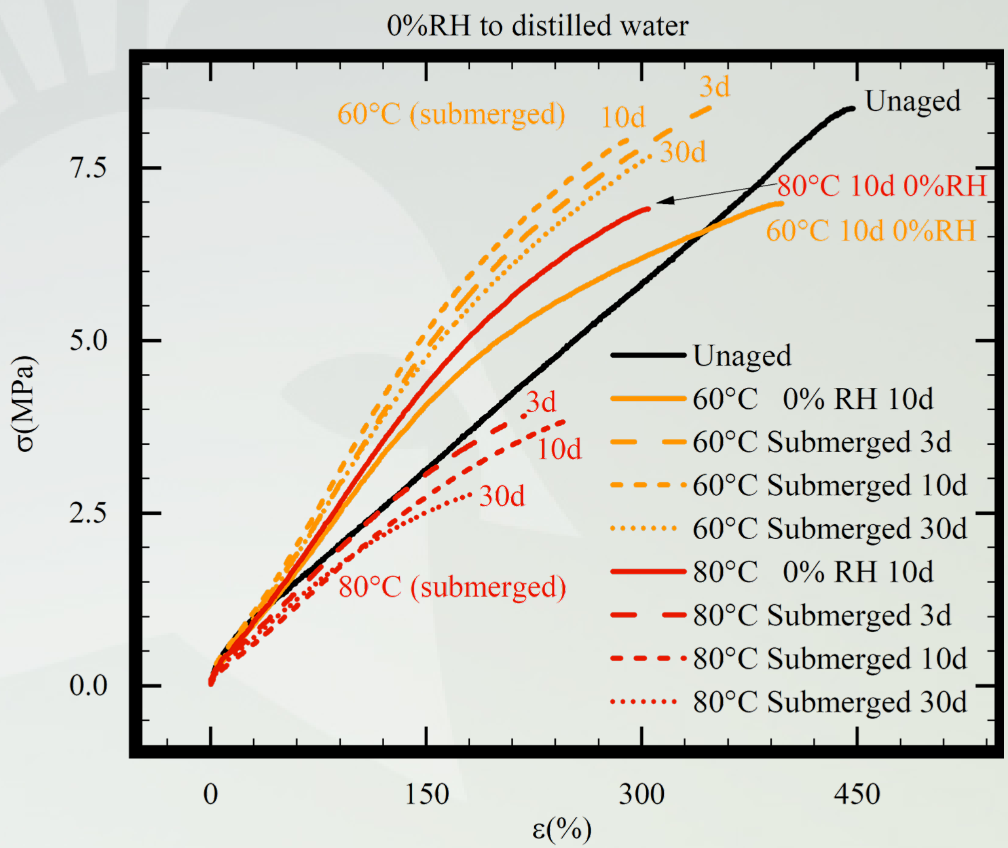
Polyurethane

Dual-effect Aging: 0%RH to Submerged

- Aging environments:
  - Thermo-oxidation (10d) to submerged (3,10,30days)
- Temperature (T) : 60℃ and 80℃
- Cross-linking (CLD) and chain scission (CS):
  - Ph-1 (0%RH): T α CLD (hardening)
  - Ph-2 (submerged):
    - 60℃ : CLD dominates (hardening)
    - 80℃ : CS dominates (softening)





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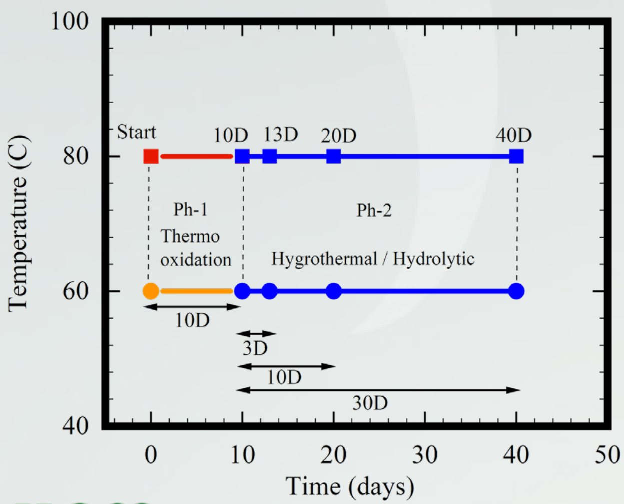
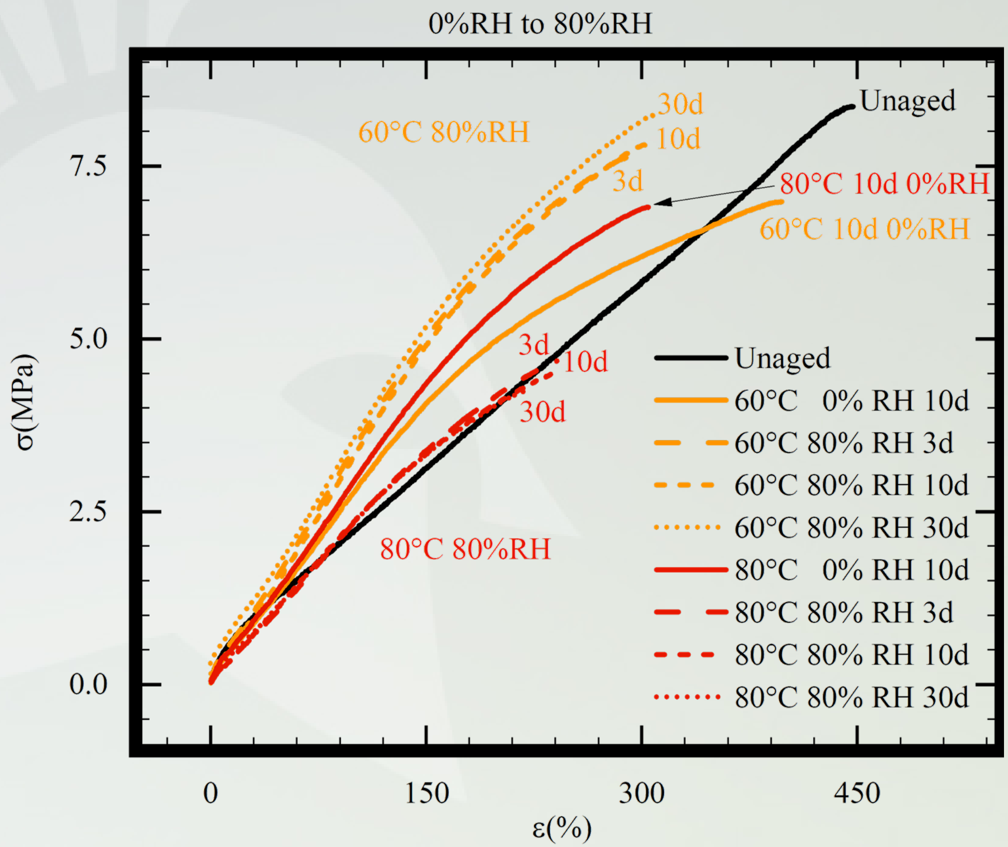
Dual-effect Aging:0%RH to 80%RH

- Aging environments:
  - Thermo-oxidation (10d) to hygrothermal 80%RH (3,10,30days)
- Temperature (T) : 60℃ and 80℃
- Cross-linking (CLD) and chain scission (CS):
  - Ph-1 (0%RH): T α CLD (hardening)
  - Ph-2 (80%RH):
    - 60℃ : CLD dominates (hardening)
    - 80℃ : CS (heat + water molecules)





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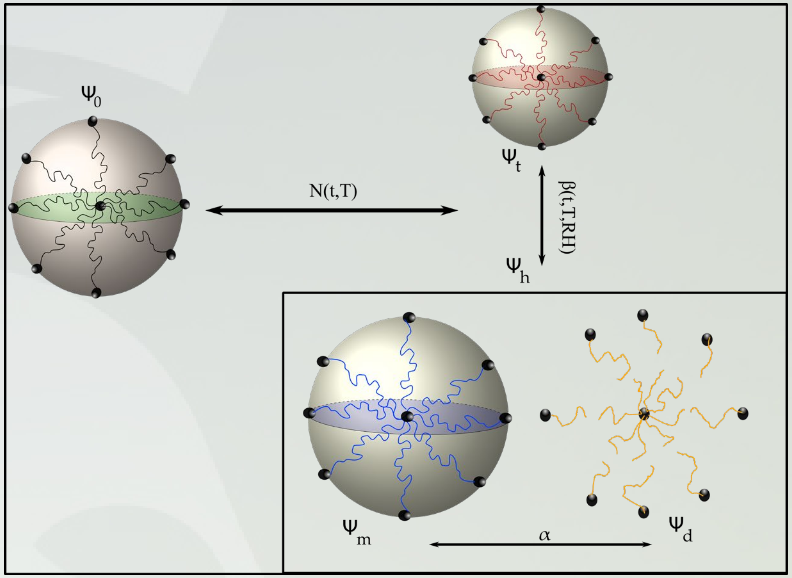
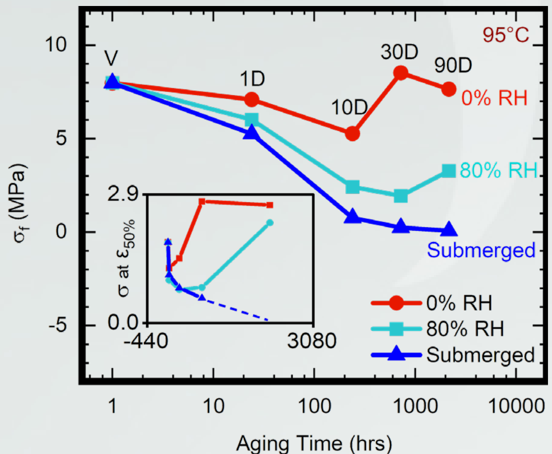
Hygrothermal aging: Competing sub-aging phenomenon

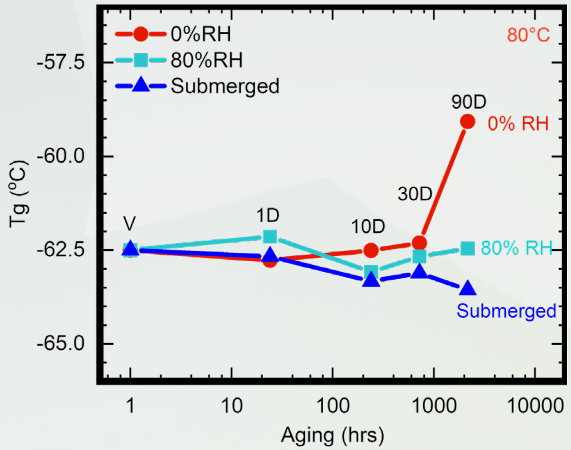
- Thermo-oxidation: Chain scission + Increase in crosslink = Stress hardening + Tg increase
- Hydrolytic aging: Chain scission + reduction of crosslink = Stress softening + Tg decrease
- Hypothesis:

  Hygrothermal aging is a competitive environment between two sub-aging phenomena i.e. hydrolysis and thermo-oxidation aging

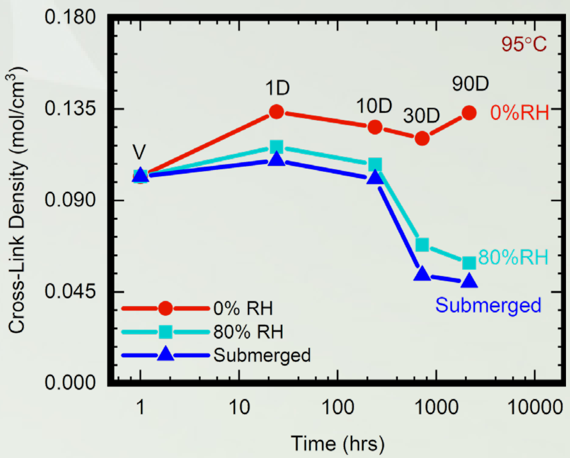




Stress Variation



Tg changes



Cross-link Density

Technical Accomplishments

Modular Platform publication

- Khalili et al. (2019), Rubber Chem. & Tech. 92(1), 51-68
- Morovati & Dargazany (2019), SoftwareX 100229
- Morovati et al. (2019), ', Math. Mech. Solids
- Morovati & Dargazany (2019), Phys Rev. E. 100229

Vibration

- Moravati & Dargazany IMECE2020
- Morovati et al., Int. J. Plasticity (2021)

Thermo

- Mohammadi & Dargazany, Int J. Plasticity, 118 (2019)
- Mohammadi et al. ECCMR 2019
- Morovati & Dargazany, IEC 2019

Hydro

- Bahrololoumi et al., Int. J. Plasticity 1. (2020)
- Bahrololoumi & Dargazany IEC 2019

Hygro = Hydro + thermo

- Wanru et al. IMECE2020
- Bahroulouloumi et al. IMECE 2020
- Bahroulouloumi et al., Int. J. Mechanical Science

Photo+Thermo

- Mohammadi & Dargazany, Polymer Deg. & Stability

Vibration + thermo

Vibration + Hydro

Vibration + Hygro

In Progress

Non-cooperating Multi-agent model

Machine learned Agent

Experimentalist Classifier

Model Free approaches

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Modeling

1. Vibration (Finished in 2020)
2. Thermal-oxidation
3. Hydrolysis
4. Hygrothermal (Hydro + Thermo)
5. Photo-thermal oxidation (Photo+Thermal)
6. Vibration + thermal-oxidation (on going)
7. Machine-learned engine (on going)

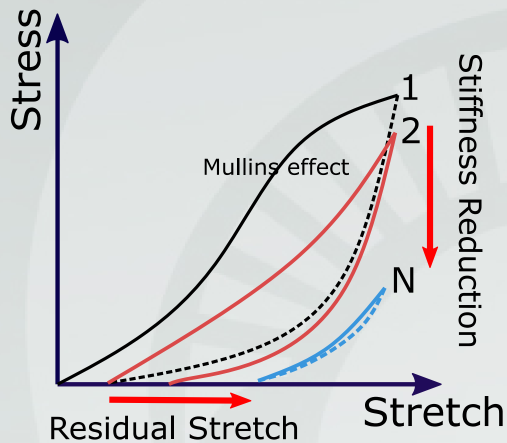
Vibration-induced damage

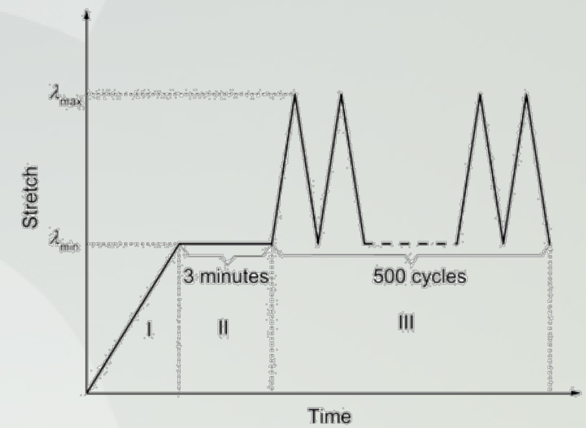
Softening of the material due to large time usage

To model the constitutive behavior of adhesives through **vibration**

Approach

Experiment :



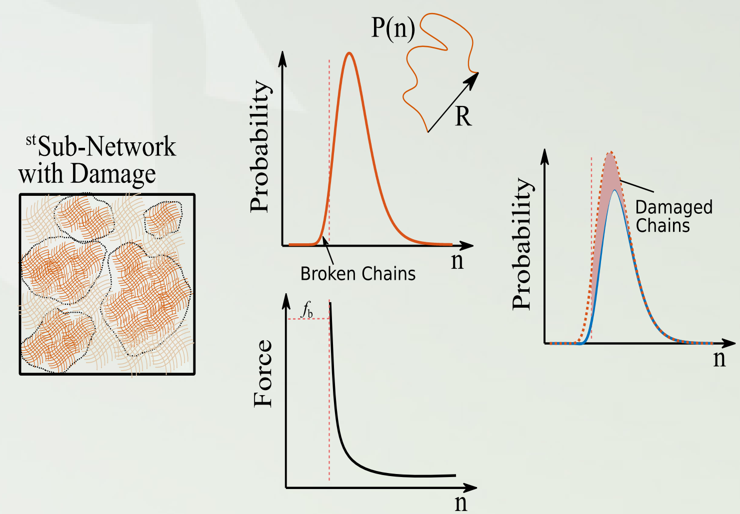


Constitutive model :

Using kinetics of irreversible chain scission

$$\overset{d_i}{\tilde{P}}(n) = P_0(n)\, e^{-C_s(n)\, j}$$

$$C_s(n) = \int_{cycle} \exp\left[\frac{\alpha}{k_B T}\left(\mathcal{L}^{-1}\left(\frac{R\lambda^{d_i}}{n}\right) - f_a\right)\right] dt$$



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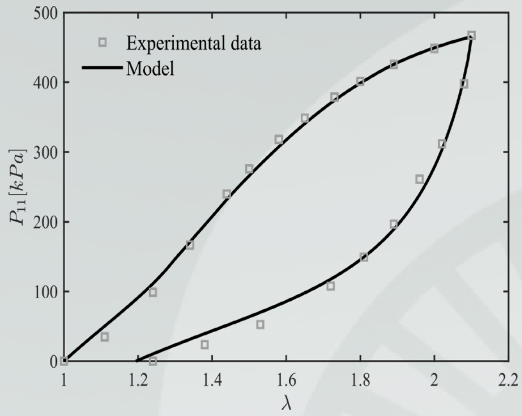
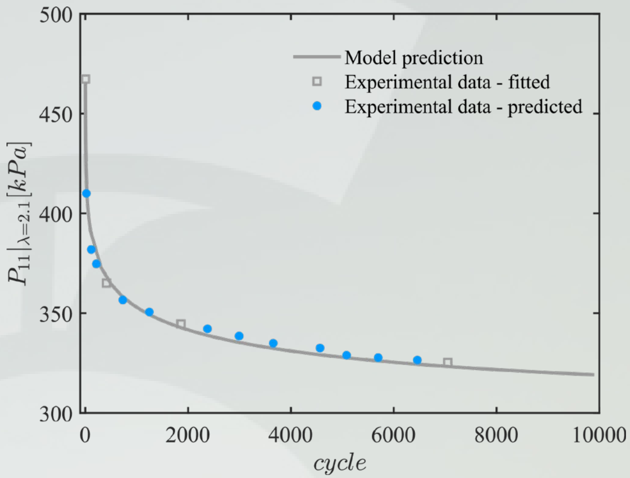
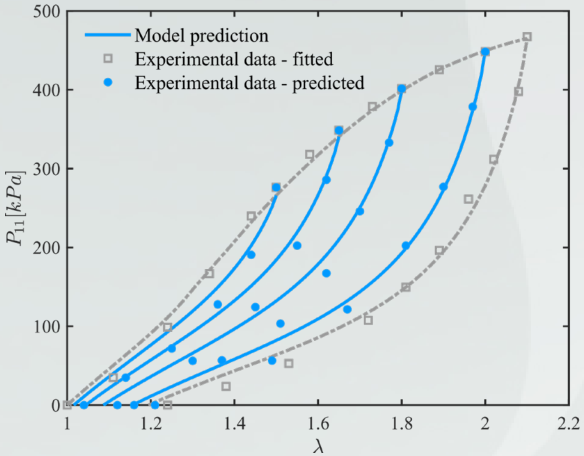
Vibration-induced damage

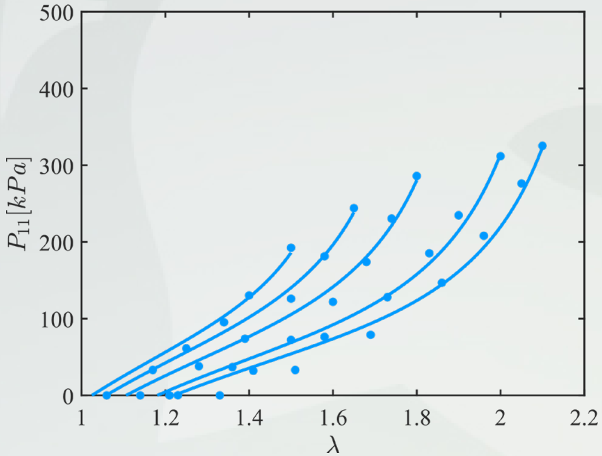
Fitting

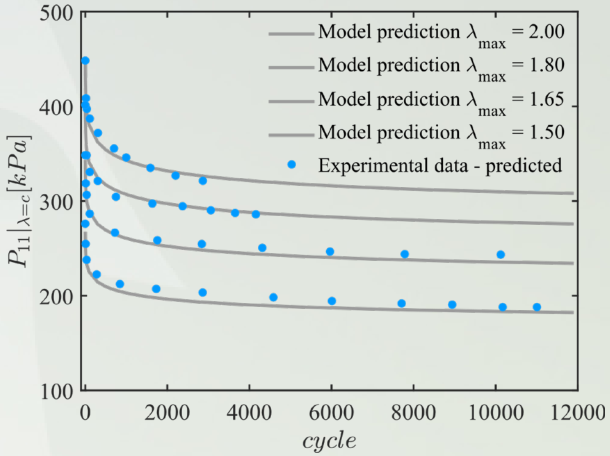




Prediction







N_1KT	$\bar{R}$	p	n_{max}	λ_p	α	ξ	k_s	θ	f_0	N_2KT	n_2
18.51	20.4	0.38	500	1.3	0.95	0.977	0.45×10^{-4}	0.98	10.4	$0.6N_1$	91

HPM High Performance Material Group

Modeling

1. Vibration
2. Thermal-oxidation (Finished in 2020)
3. Hydrolysis
4. Hygrothermal (Hydro + Thermo)
5. Photo-thermal oxidation (Photo+Thermal)
6. Vibration + thermal-oxidation (on going)
7. Machine-learned engine (on going)

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Thermo-Oxidative Aging

Goal: To model the constitutive behavior of adhesives through thermo-oxidative aging

Challenge

Finding the correct decay function

Approach

Dual network hypothesis

Arrhenius functions as decay function

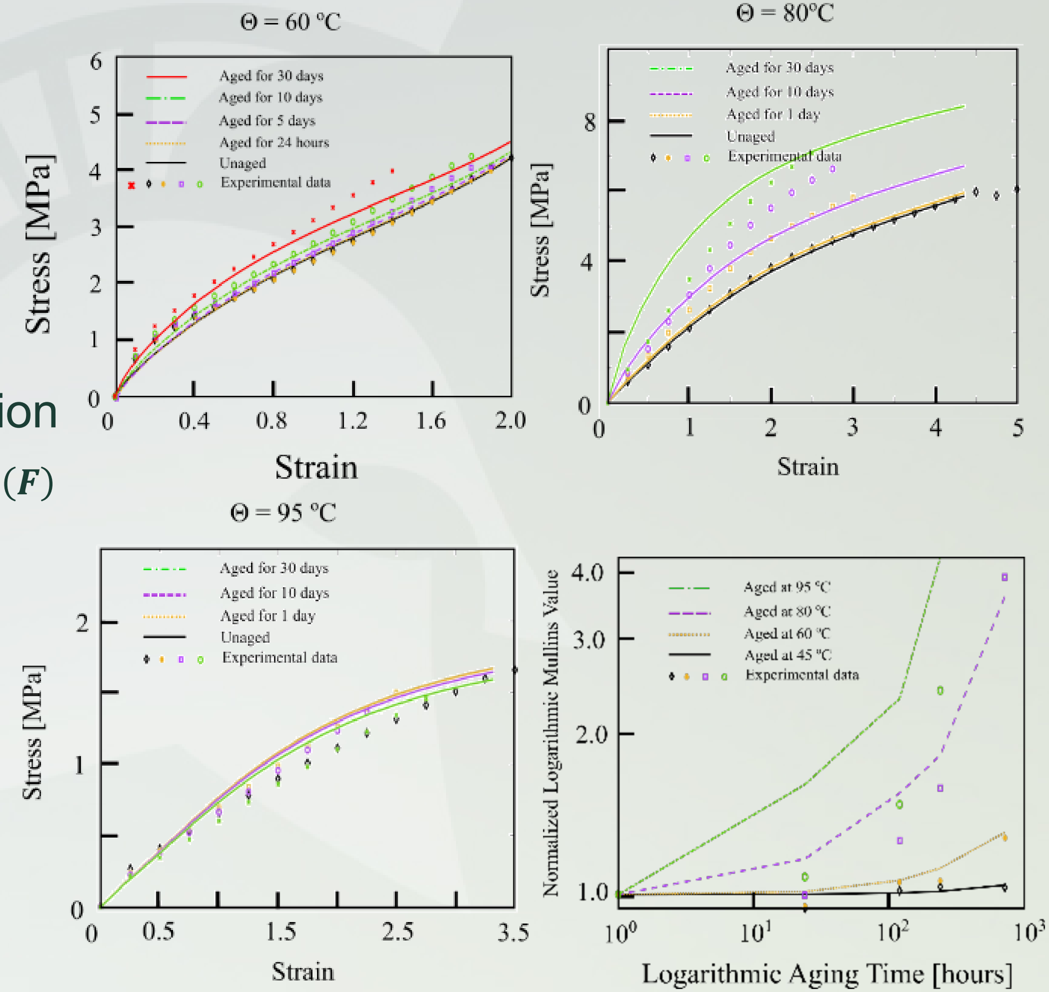
$$\Psi_M(t,T,\boldsymbol{F}) = \rho\,(t,T)\Psi_0(\boldsymbol{F}) + \big(1-\rho(t,T)\big)\Psi_\infty(\boldsymbol{F})$$

$$\rho\,(t,T) = A_1\ exp(-\alpha t) + A_2\,exp(-\beta t)$$

Time-temperature superposition

$$a_T = exp\left(\frac{E_a}{R}\left(\frac{1}{T_{ref}} - \frac{1}{T}\right)\right)$$

Result



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Hydrolysis Model

$$\Psi_M(t,T,\mathbf{F}) = N(t,T)\Psi_0(\mathbf{F}) + N'(t,T)\Psi_\infty(\mathbf{F})$$

$$N(t,T) = \exp\left(-\gamma \exp(-\frac{E_a}{\mathcal{R}T})t\right)$$

- Strain energy of a single chain

$$\hat{\psi}_c\,(n,\bar{r}_\bullet) = nK_bT\int_0^{\varphi}\hat{\beta}d\varphi,\qquad \hat{\beta} = \left[1-\frac{1+\varphi^2}{n}\right]\beta$$

- Probability Distribution Function of a Polymer Chain

$$\mathcal{P}_\bullet\,(n) = \frac{1}{2\sqrt{\pi\sigma^2}}\exp(\frac{(n-\mu_\bullet)^2}{-2\sigma^2})$$

- Networks and Subnetworks

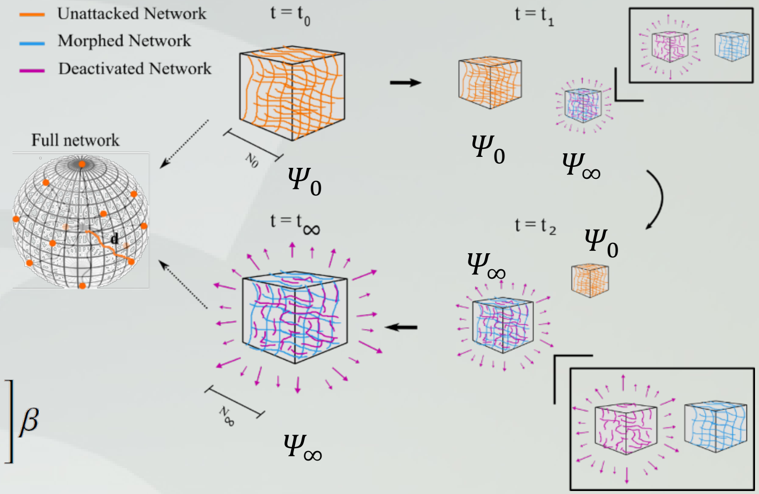
$$\Psi_\bullet = \frac{1}{A_s}\int_S \overset{d}{\psi}_\bullet \overset{d}{du} \cong \sum_{i=1}^{k}\overset{d_i}{\psi}_\bullet w_i$$

- Inverse Langevinge Function approximation

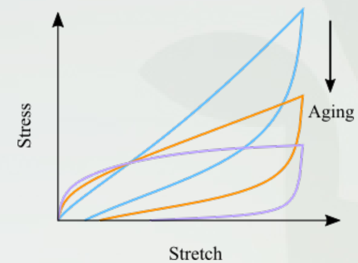
$$\mathcal{L}^{-1}\,(x) \cong \frac{1}{1-x} + x - \frac{8}{9}x^2$$

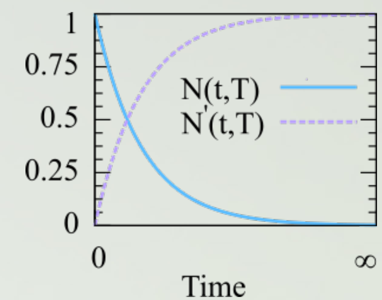
- Kinetics (Esters, Amide, Carbonate)

$$-\frac{d[COOH]}{dt} = \xi[Ester][Water][COOH] = \kappa[COOH]$$



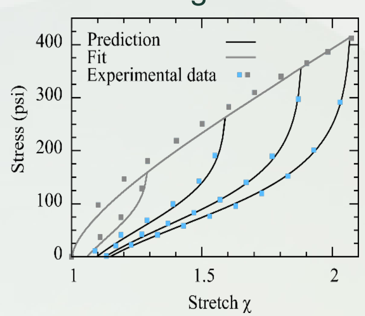
$$\Psi_M = N(t,T)\Psi_0 + \alpha\, N'(t,T)\Psi_m + (1-\alpha)N'(t,T)\Psi_d$$



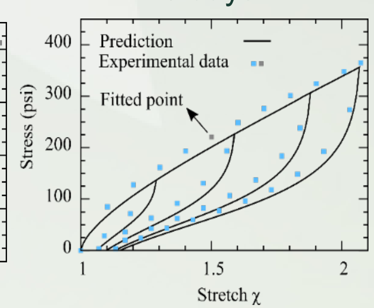


Θ= 60 ° C

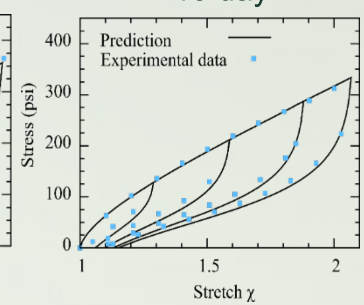
Unaged



6 days



10 day



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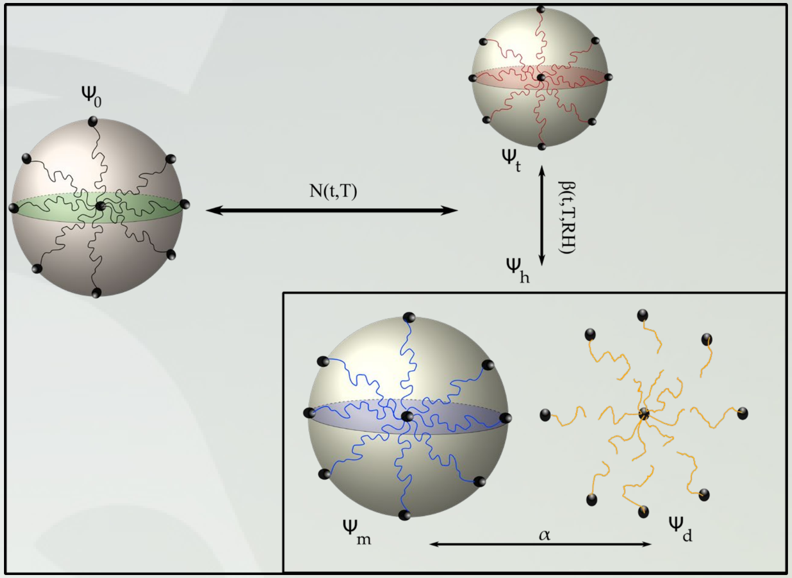
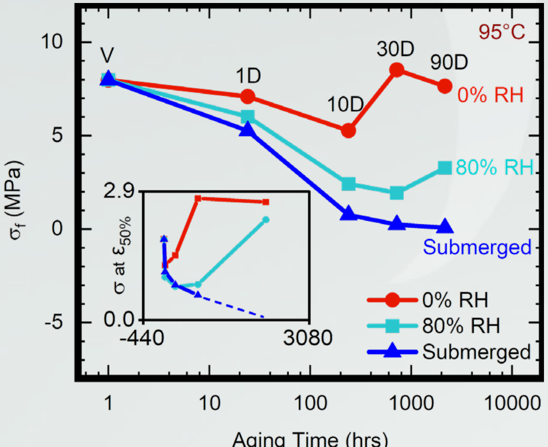
Polyurethane

Hygrothermal aging: Competing sub-aging phenomenon

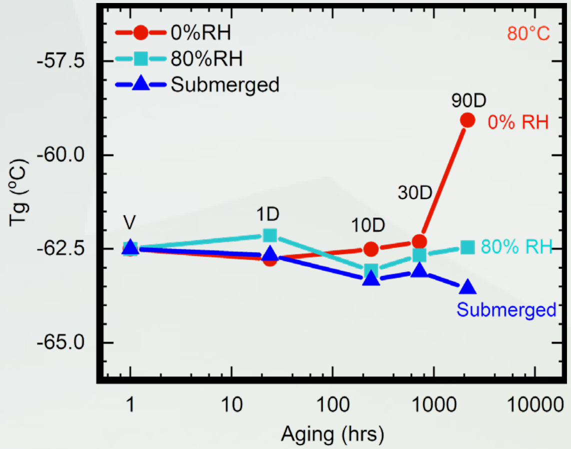
- Thermo-oxidation: Chain scission + Increase in crosslink = Stress hardening + Tg increase
- Hydrolytic aging: Chain scission + reduction of crosslink = Stress softening + Tg decrease
- Hypothesis:

  Hygrothermal aging is a competitive environment between two sub-aging phenomena i.e. hydrolysis and thermo-oxidation aging

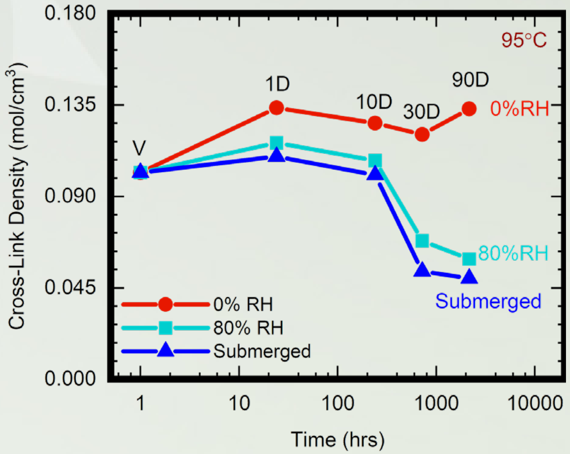




Stress Variation



Tg changes



Cross-link Density

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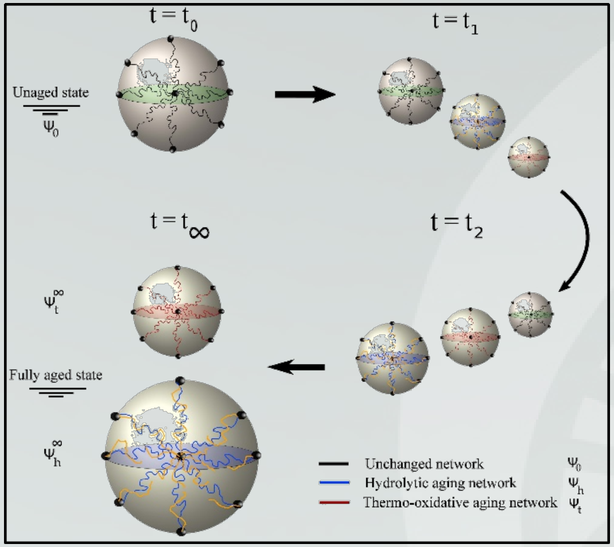
Hygrothermal model



The strain energy of the material in all states of aging,

$$\Psi_M(t, T, RH, \boldsymbol{F}) = N(t, T)\Psi_0 + N'(t, T)\Psi_0$$

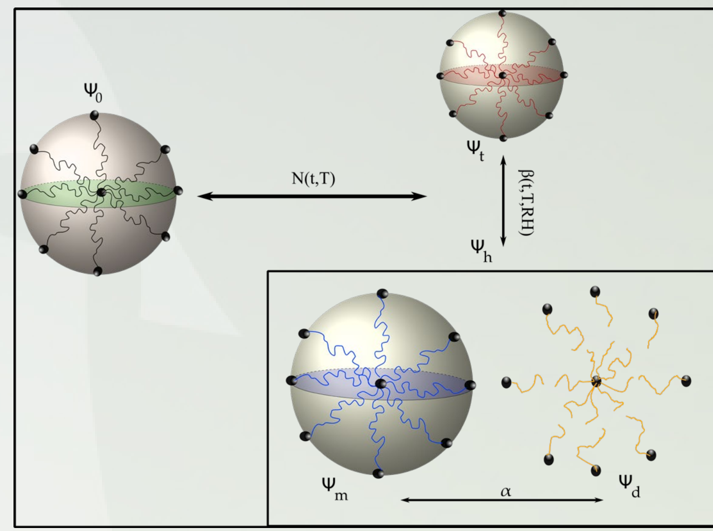
$$N(t, T) = \exp(-\gamma exp\left(-\frac{E_a}{RT}\right) t)$$

$N(t, T) = 1 - N'(t, T)$ are predefined shape function

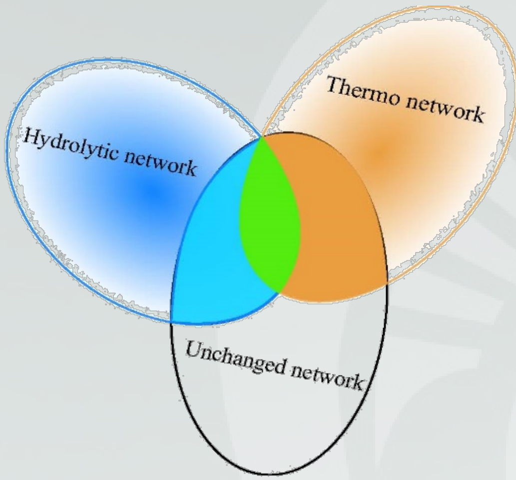
We defined two end-state of the material as the state of polymer matrix at initial state Ψ_0 and fully aged state at time infinity Ψ_∞.

The hydrolysis network decomposes into morphed Ψ_m^∞ and deactivated network Ψ_d^∞

$$\Psi_h^\infty = \alpha\Psi_m^\infty + (1 - \alpha)\Psi_d^\infty \qquad 0 \leq \alpha \leq 1$$



Summary



Strain energy function	Material parameters: Unchanged network	Hydrolytic network	Thermo network	Coupling parameters
$\Psi = \Psi_0$	$N_0 k_b T$ $\bar{R}_0$ μ_0 σ ν			
$\Psi = N(t,T)\Psi_0 + N'(t,T)\Psi_h$	$N_0 k_b T$ $\bar{R}_0$ μ_0 σ ν	$\bar{R}_m$ μ_m α $\frac{E_a}{R}$ γ		
$\Psi = N(t,T)\Psi_0 + N'(t,T)\Psi_t$	$N_0 k_b T$ $\bar{R}_0$ μ_0 σ ν		$\bar{R}_T$ μ_T $\frac{E_a}{R}$ γ	
$\Psi = N(t,T)\Psi_0 + N'(t,T)(1-\beta)\Psi_t + N'(t,T)\beta\Psi_h$	$N_0 k_b T$ $\bar{R}_0$ μ_0 σ ν	$\bar{R}_m$ μ_m α $\frac{E_a}{R}$ γ	$\bar{R}_T$ μ_T	$\frac{E_b}{R}$ Q θ

$\mathcal{N}_0$	Number of chains of virgin network per unit volume
μ_0	Mean value of unchanged network chain length distribution
σ	Standard deviation of chain length distribution in all networks
$\overline{R_0}$	Normalized end-to-end distance of reference chains
ν	Sliding ratio w.r.t. bond strength
$\overline{R_m}$	The normalized end-to-end distance of chains in the morphed network
μ_m	The mean value of morphed network chain length
α	The percentage of active chains in the hydrolytical network
μ_t	The mean value of thermo network chain length distribution
γ	Arrhenius rate factor
Q, θ	Adjusting parameters to keep β between zero and one
E_a, E_b	Activation energies

Final Equation:

$$\boldsymbol{T} = \frac{\partial \Psi_M}{\partial \boldsymbol{F}} - p\boldsymbol{F}^{-T} = N(t,T)\frac{\partial \Psi_0}{\partial \boldsymbol{F}} + N'(t,T)\left[\beta(t,T,RH)\frac{\partial \Psi_t^\infty}{\partial \boldsymbol{F}} + (\boldsymbol{1} - \beta(t,T,RH))\frac{\partial \Psi_h^\infty}{\partial \boldsymbol{F}}\right] - p\boldsymbol{F}^{-T}$$

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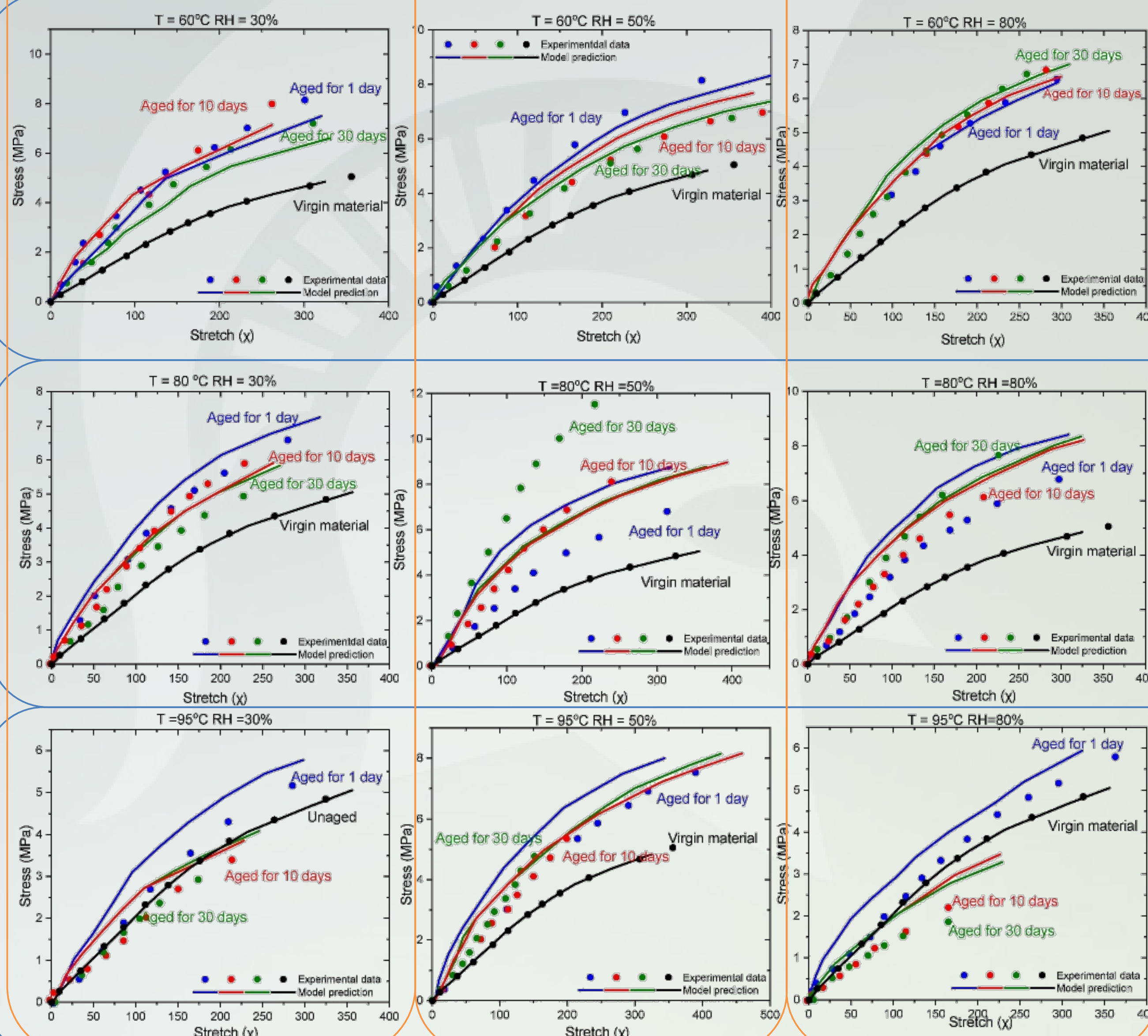
Model prediction against Rubric of Env. Condition

30% RH | 50% RH | 80% RH

60°C

80°C

100°C



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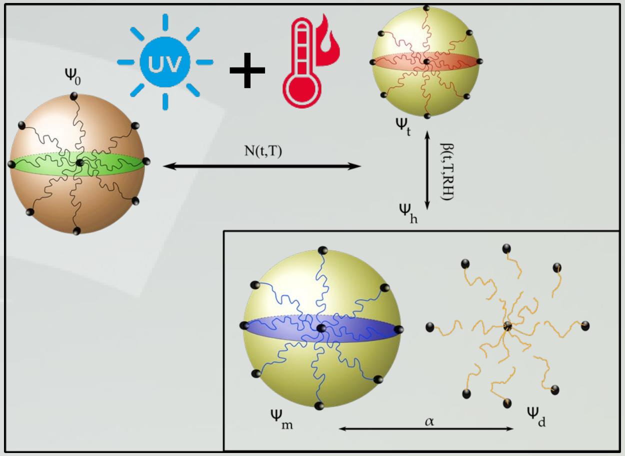
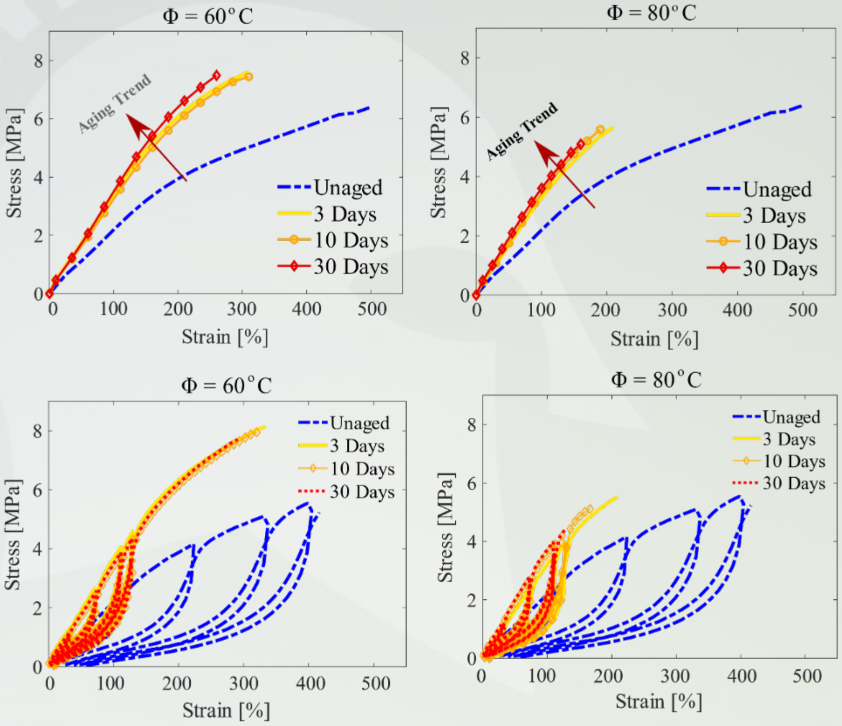
Dual condition: Photo – Thermo aging



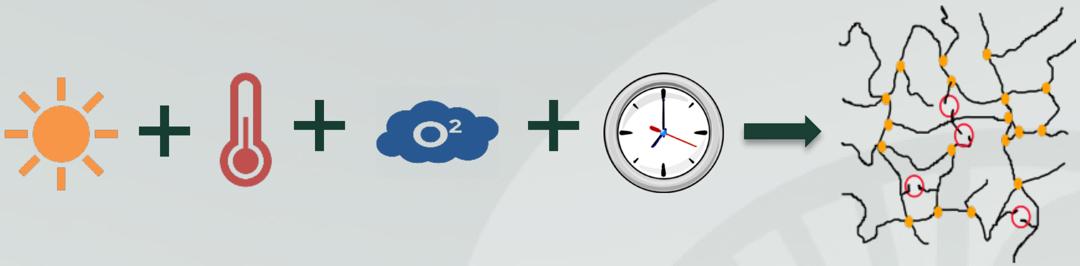
Dual Condition: PUB

- Was aged for 10 days in either 60 or 80̊ C.in thermo-oxidative conditions
- Was then aged in the same temperature in photo-oxidative conditions for durations 3, 10 & 30 days
- Failure and cyclic tests were conducted.
- Material degrades from higher temperature aging mode.



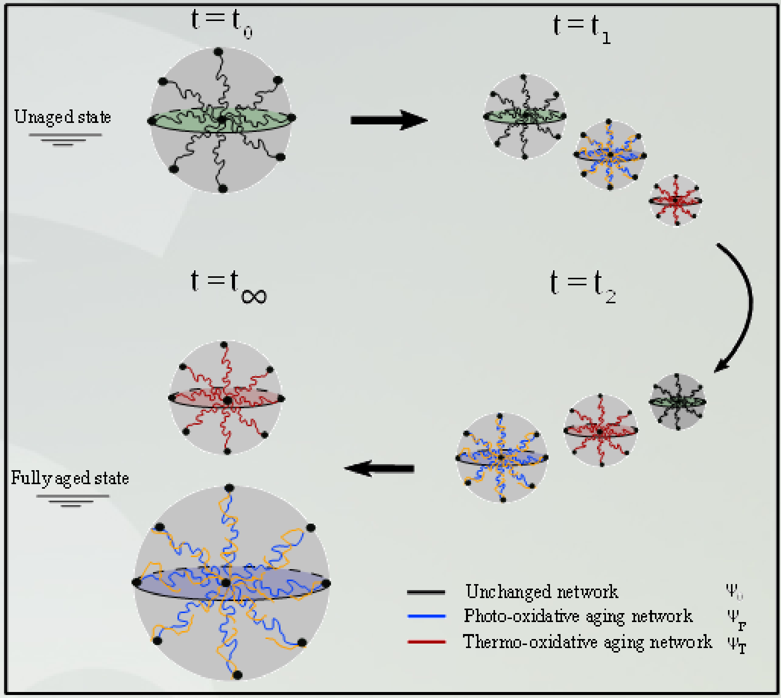
Specimens aged in Dual aging at 60 & 80̊C for 30 days for thermo-oxidation at 0%RH for then moved to photo-oxidation for 3, 10, and 30 days.

Kinetic Model



- The general scheme of photo-oxidation reactions can be written as:
  - Initiation: $POOH \rightarrow \alpha P^*$
  - Propagation: $P^* + O_2 \rightarrow PO_2^*,\ PO_2^* + PH \rightarrow POOH + P^*$
  - Termination: $PO_2^* + PO_2^* \rightarrow Product$

- P is a symbol for polymeric chemical compounds. Defining the rate of oxidation in the course of aging:
  - $-\frac{d[P]}{dt} = k[P]^q$
  - $[P]$ is the concentration of chemical compound.
  - k is the reaction rate coefficient, it is a function of temperature T and the light intensity I. $k = \tau I^{\alpha} e^{-E'_a}$, $E'_a = \frac{E_a}{RT}$
  - $[P] = A\exp\left(-\tau I^{\alpha}(e^{-E'_{a_{ref}}})\alpha_T t\right) \xrightarrow{\textbf{decay function}\ for\ photo-oxidative\ aging} \rho_o(t) = \exp\left(-\tau \Gamma^{\gamma} e^{-E'_{a_{ref}}} \alpha_T t\right)$, Γ reflects the effect of UV.
  - γ is a function of UV radiation.



Unaged network $(t = t_0)$ continues to change and got replaced by two new-subnetwork at the fully aged state $(t = t_\infty)$.

$$\varphi_{photo+thermo} = \rho_{thermo}\varphi_0 + (1 - \rho_{thermo})\big(\rho_{photo}\varphi_{thermo} + (1 - \rho_{photo})\varphi_{photo}\big)$$

$$\rho_{thermo} = A_1 \exp(-e^{\frac{-E_a}{RT}} t)$$

$$\rho_{photo} = A_2 \exp(-I^{\alpha} t)$$

- A_1, A_2, α : Constants
- I : Radiation intensity
- E_a : Activation energy

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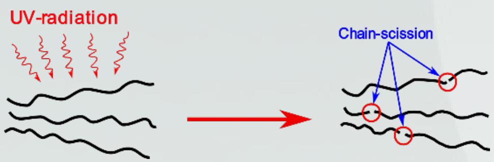
Continuous Network Hypothesis



γ is a material parameter to show the effect of light intensity

$\gamma = 1$ means we only have thermo-oxidation

$$R(t) = \boldsymbol{R_0} - \overbrace{\left(R_c - \frac{R_I}{2}\right)\left[1 - \exp\left(-\tau_1 e^{-\frac{E_{a2}}{RT_{ref}}} \gamma^{\alpha} (\alpha_{T_1}\, t)\right)\right]}^{\text{Crosslinking process}} + \underbrace{\left(R_s - \frac{R_I}{2}\right)\left[1 - \exp\left(-\tau_2 e^{-\frac{E_{a2}}{RT_{ref}}} \gamma^{\beta} (\alpha_{T_2}\, t)\right)\right]}_{\text{Chain scission process}}$$

$$cr(t) = cr_0 - \overbrace{\left(cr_c - \frac{cr_I}{2}\right)\left[1 - \exp\left(-\tau_1 e^{-\frac{E_{a2}}{RT_{ref}}} \gamma^{\alpha} (\alpha_{T_1}\, t)\right)\right]}^{\text{Crosslinking process}} + \underbrace{\left(cr_s - \frac{cr_I}{2}\right)\left[1 - \exp\left(-\tau_2 e^{-\frac{E_{a2}}{RT_{ref}}} \gamma^{\beta} (\alpha_{T_2}\, t)\right)\right]}_{\text{Chain scission process}}$$

R	average end-to-end distance of chains
$\boldsymbol{R_0}$	end-to-end distance of unaged material
R_c, R_s	effect on R from crosslinking and chain scission respectively
R_I	effect of crosslinking and chain scission on R from radiation
E_{a2}	activation energy
τ_1, τ_2	defines the speed of aging as a function of t, T

cr_0	crosslink density at virgin state
cr_c, cr_s	crosslink density from thermo-oxidative data
γ^{α}, γ^{β}	effect of radiation intensity on crosslinking and chain scission
cr_I	crosslink density from photo-oxidative data

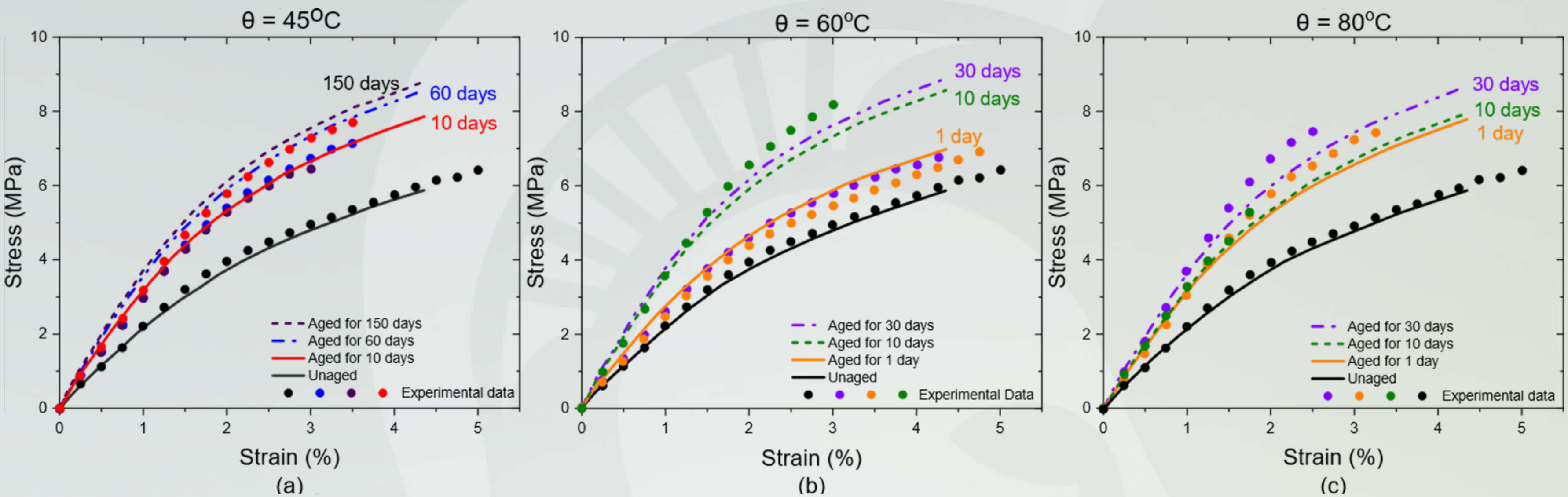
Constraints to make sure

$0 < cr(t) < 1 cr_0 > 0$

$cr_2 < \frac{cr_I}{2} < cr_1$

Model validation

Material: Black Polyurethane



Fitting Parameters

N_0CT	C_{r0}	R_0	υ	C_{rs}	C_{rc}	R_c	R_s	E_{a1}	C_{rI}	R_I	α	β	γ	E_{a2}
Material parameters for virgin state				Thermo-oxidative material parameters					Photo-oxidative material parameters					

Parameters are independent to each other between 3 states: virgin, thermo, and photo

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Vibration with thermo-oxidative model

- Material:
  - Ph-1: Development (SBR)
  - Ph-2: Validation (DOWSIL-7091 and 3M-590)
- Aging environments:
  - Ph-1: Thermo-oxidation
  - Ph-2: Thermo-oxidation, UV, hygrothermal / hydrolytic aging
- Temperature (T) : 60℃ and 80℃
- Model is based on toughness comparisons between:
  - Unaged specimen
  - Fatigued unaged specimen (mechanical damage)

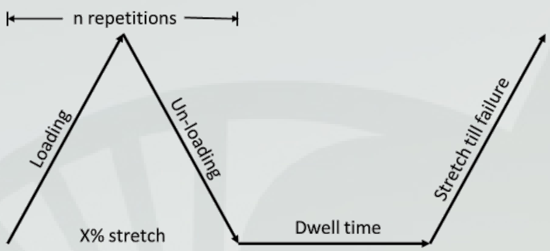
$$\mathscr{D}_{me} = 1 - \Theta j^{-\zeta}$$

  - Aged specimens (environmental damage)

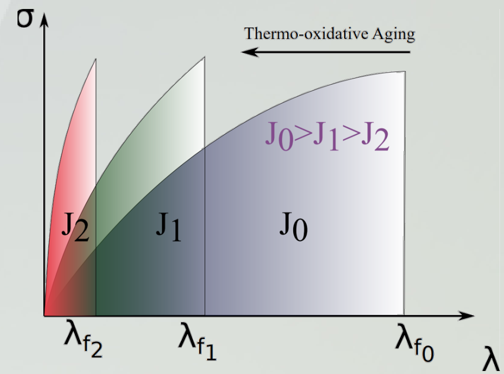
$$\mathscr{D}_{ev} = 1 - exp\left(-\gamma exp\left(-\frac{E_a}{\mathscr{R}T}\right)t\right)$$

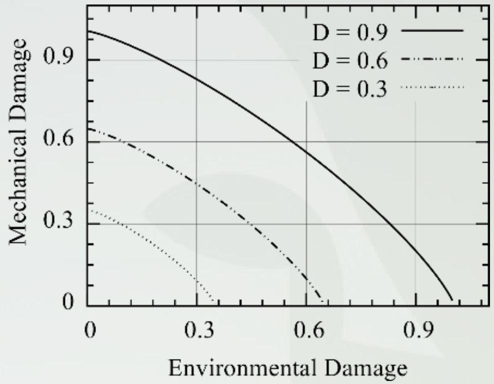
  - Aged + fatigued specimens (environmental + mechanical damage)

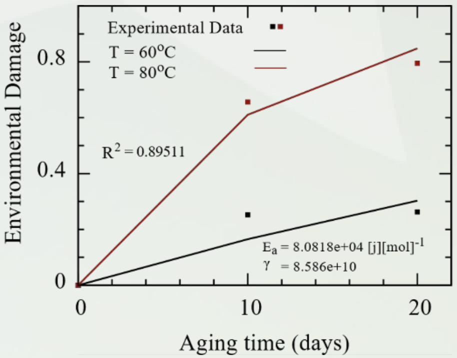
$$\mathscr{D} = d_1 \tanh\left(\mathscr{D}_{me}{}^{d_2} + \mathscr{D}_{ev}^{d_2}\right)$$

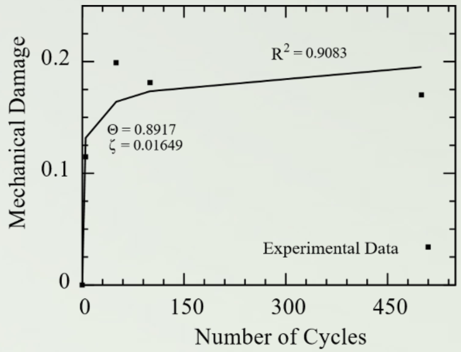


Vibration test profile









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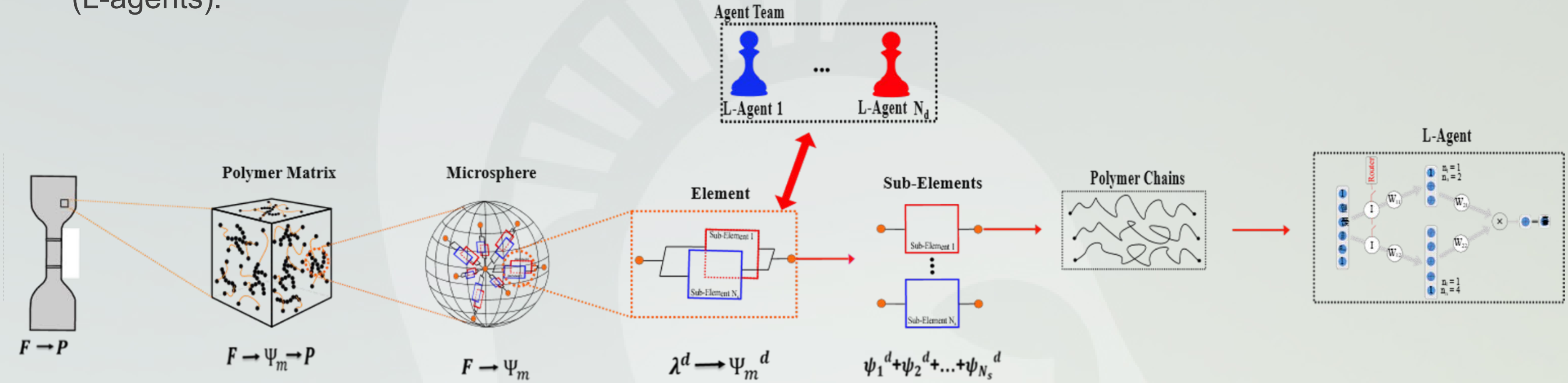
Machine Learned models: Hybrid physics induced data-driven framework

- The proposed model is based on the concept of a cooperative multi-agent system $\mathcal{A}_j^i$, $i \in \{1, n\}, j \in \{1, m\}$ to describe different features in the material behavior with $n \times m$ different Neural Network learning agents (L-agents).



$$\text{Two} - \text{point}\,\frac{\text{strain}}{\text{stress}}\,\text{tensors}(\boldsymbol{F}{:}\,\boldsymbol{P}) \rightarrow \boldsymbol{E}{:}\,deformation\ gradients, \boldsymbol{P}{:}\,first\ Piola - Kirchhoff\ stress$$

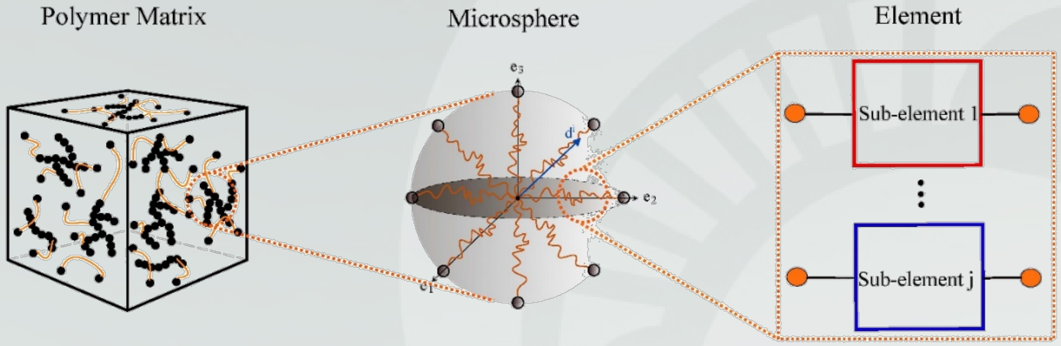
$$Material\ \frac{\text{strain}}{\text{stress}}\,\text{tensors}(\boldsymbol{E}{:}\,\boldsymbol{S}) \rightarrow \boldsymbol{E}{:}\,Lagrange\ strain, \boldsymbol{S}{:}\,second\ Piola - Kirchhoff\ stress$$

$$Spatial\ \frac{\text{strain}}{\text{stress}}\,\text{tensors}(\boldsymbol{L}{:}\,\boldsymbol{\tau}) \rightarrow \boldsymbol{L}{:}\,Hencky\ strain, \boldsymbol{\tau}{:}\,Kirchhoff\ stress$$

The model is constrained at multiple steps:
1. Model defined based on strain energy
2. Hiring micro-sphere for 3D to 1D order reduction
3. Network decomposition to separate different inelastic effects
4. Defining learning agents to represent each 1D subnetwork

Conditional Neural Network (CondNN) L-Agent

For which the outputs are not only dependent on past occurrences e.g deformation effects on the matrix, but also on external actions e.g. temperature and time of aging effects on the polymer matrix



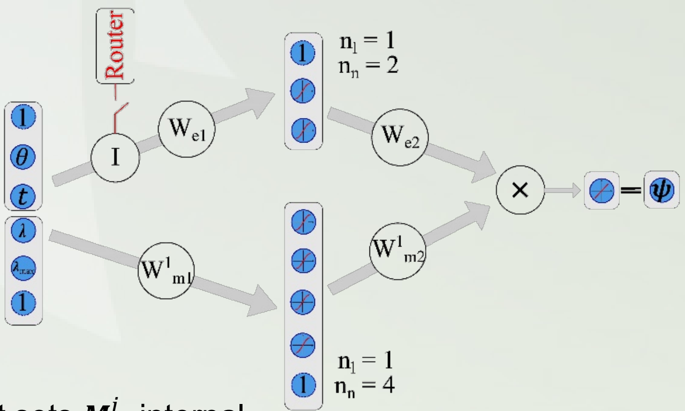
$\Psi_m^{d_i}$ is the element energy represented by team of L-Agents $\mathcal{B}^{d_i}$ reflecting an additive cooperation between multiple L-Agents $\mathcal{B}^{d_i} = \sum_{i=1}^{N_s} \mathcal{A}_j^i$ based on microsphere concept.

We assume all teams to be identical in the virgin state $\mathcal{B}^{d_i} = \mathcal{B}^{d_j}$.

During matrix deformation, different teams will be exposed to different deformations based on their directions, and the matrix become anisotropic.

$$\Psi_m = \frac{1}{4\pi}\int_S \Psi_m^d \, dS^d \cong \sum_{i=1}^{N_d} \omega_i \Psi_m^{d_i} \qquad \Psi_m^{d_i} = \mathcal{B}^{d_i} = \sum_{i=1}^{N_s} \mathcal{A}_j^i$$

For the CondNNs structure of L-agents, we considered one input layer, one hidden layer with 4 neurons, and 3 activation functions (soft plus, sinusoid, hyperbolic tangent).



L-agent response is computed using a feed-forward algorithm for a given set of hyper-parameters (n_l, n_n). Each L-agent can be represented by a CondNNs $\mathcal{A}_j^i := \mathcal{D}^{d_i}(\mathrm{E}^{d_i})\psi_j^d(\boldsymbol{M}_j^i, S_j^i)$

Where $\mathcal{D}_j^i := CNN_e\,(\boldsymbol{W_e}, \boldsymbol{E}^i)$, $\psi_j^d = CNN_m(\boldsymbol{W}_m^{\boldsymbol{j}}, \boldsymbol{M}_j^i, S_j^i)$

$\psi_j^d(\boldsymbol{M}_j^i, S_j^i)$ is trained on the basis of a non-kinematic input sets $\boldsymbol{M}_j^i$, internal parameters S_j^i for the mechanical damage CondNN. $\boldsymbol{W_e}$ and $\boldsymbol{W}_m^{\boldsymbol{j}}$ are related to weight matrices of environmental and mechanical damage.

L-agent response

For the CondNNs structure of L-agents, we considered one input layer, one hidden layer with 4 neurons, and 3 activation functions (soft plus, sinusoid, hyperbolic tangent).

Internal parameter λ_{j-max} to capture the deformation of the rubbers with full memory

$$\boldsymbol{M}_1^{d_i} = [\lambda_1^{d_i}],\ \boldsymbol{S}_1^{d_i} = [\lambda_{1-max}^{d_i}],\ \boldsymbol{E}^{d_i} = [t, \theta],\ \boldsymbol{M}_2^{d_i} = [\lambda_2^{d_i}],\ \boldsymbol{S}_2^{d_i} = [\lambda_{2-max}^{d_i}],\ \boldsymbol{E}^{d_i} = [t, \theta]$$

Where $\lambda_1^{d_i} = \sqrt{\boldsymbol{d_i}\mathbf{C}\boldsymbol{d_i}}$, $\lambda_2^{d_i} = \sqrt{\boldsymbol{d_i}\mathbf{C}^{-1}\boldsymbol{d_i}}$, $\mathbf{C} = \mathbf{F}^T\mathbf{F}$

$\lambda_1^{d_i}$, $\lambda_2^{d_i}$ are related to I_1, I_2, as the first and second invariants of $\mathbf{C}$

We used identical engines, a relatively simple engine built by $N_d = 21$ teams, where each team has $N_s = 2$ agents

The final cost function: $\mathcal{L}(\boldsymbol{W}_m^1, \boldsymbol{W}_m^2, \boldsymbol{W_e}) = \frac{1}{2}\sum_{n=1}[\mathfrak{g}_1\left(\sum_{i=1}^{21}\sum_{j=1}^{2}\omega_i\frac{\partial\mathcal{A}_j^i}{\partial\mathbf{F}} - p\mathbf{F}^{-T}\right)\mathfrak{g}_1 - P_n^{11}]^2$

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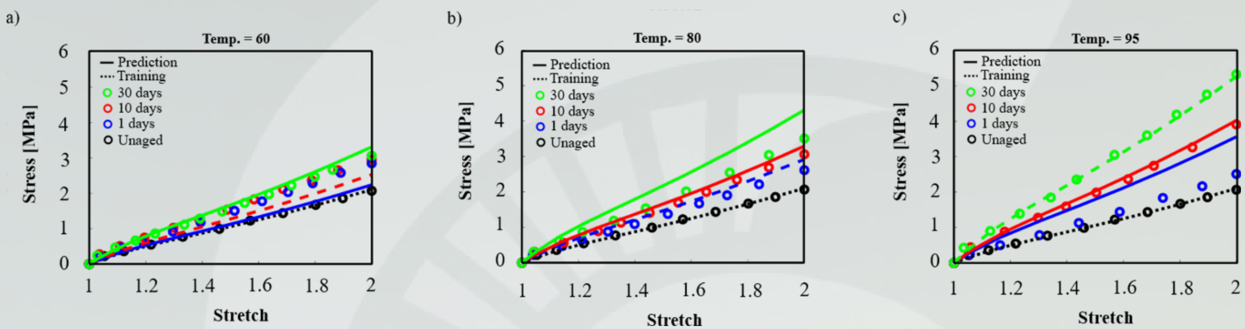
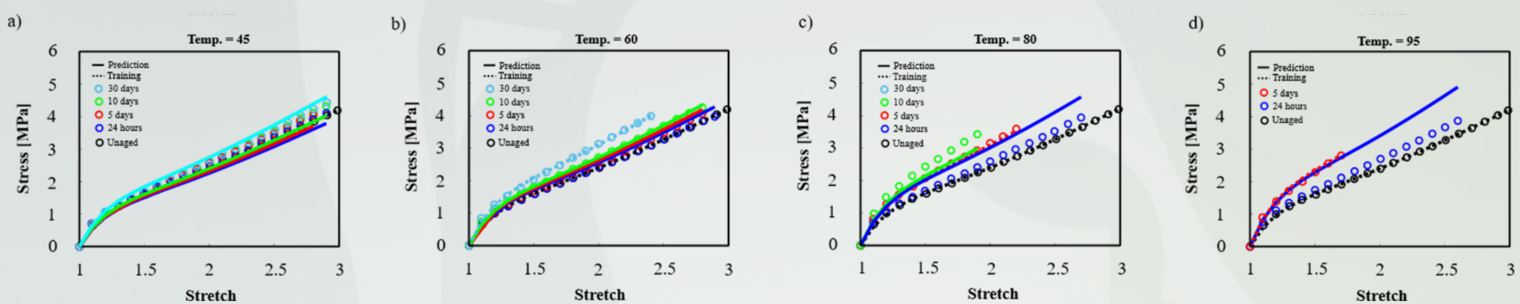
Training and model prediction

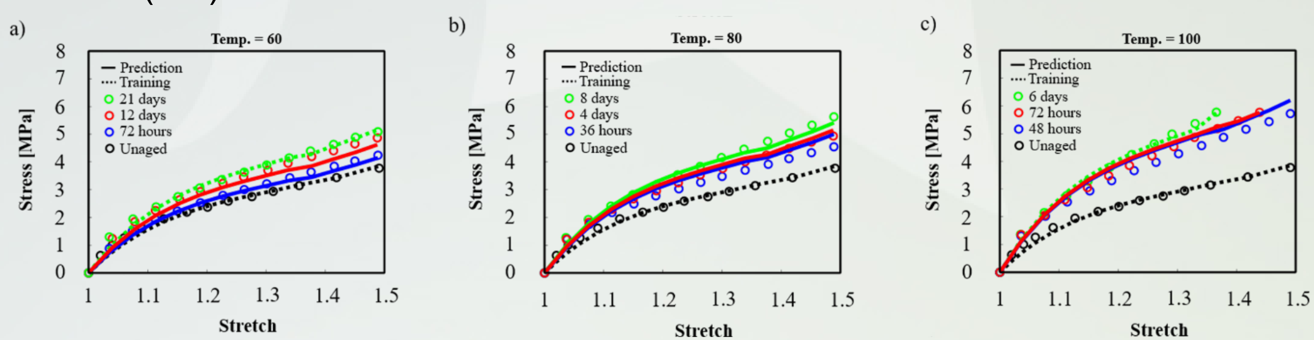
Material: Black polyurethane



Material: Styrene-butadiene rubber(SBR)



Material: Natural Rubber (NR)



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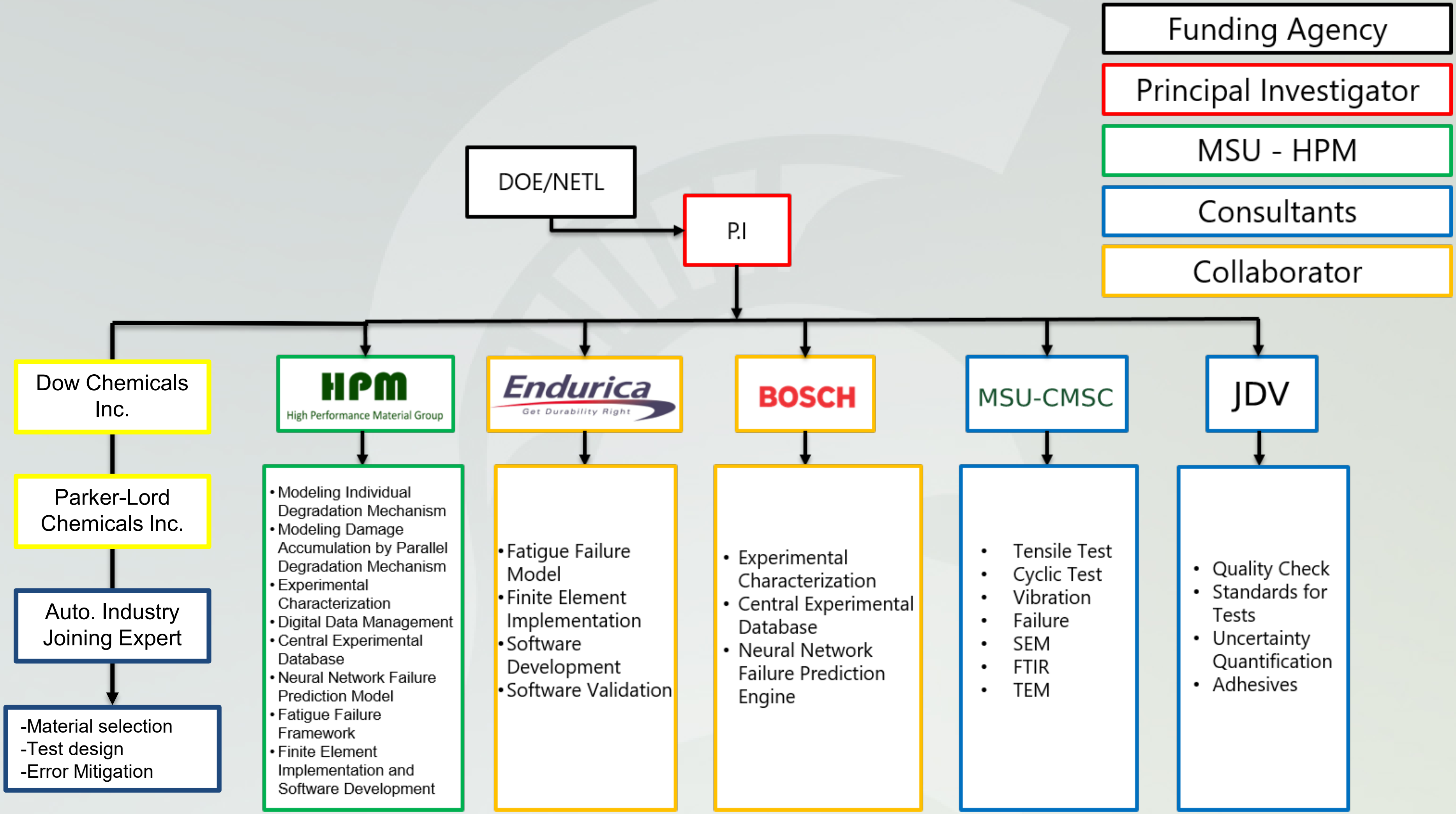
Response to Previous Year Review Comments

- **Risk Management**: *"There were no risks or risk mitigation strategies identified by the presenter: however, challenges were identified by the project team that indicate an awareness of where risks may occur"*

COVID-19 shut-down consequences:
- Forced shut down of all Aging Tests
- Removal of all ultra-long natural aging samples
- Capacity shift by industrial collaborators (uncertainty on resource allocation)

Risks	Mitigation Plan
Supply chain issue to obtain all the adhesives	Fatigue +Environmental damage validation with accelerated aging at short and Mid-range level aging Ultra-long agin only with Polyurethane adhesive
Lack of time/resources to redo ultra-long tests:	Reducing validation set of materials used for ultra-long aging
Non-uniform damage mechanism in the material	-> use of 5 reliability samples for each test -> collaboration with suppliers to use same batch
Complicated and inseparable sources of degradations mechanism	Multi-path aging tests to define synergy

Collaboration and Coordination



Summary

Accomplishments

- Finished Vibration, Thermo-oxidative and hydrolysis model.
- Developed vibration & thermo-oxidative damage model and machine-learned engine.
- Developed hygrothermal model and verified against rubric of Env. Condition.
- Developed photo-thermo oxidation model with multiple adhesives.
- All pilot tests (mechanical and chemical) finished for all single-aging condition.
- Dual effects (hygrothermal, photo-thermo oxidative) for Polyurethane is finished.
- Temperature jump test for Polyurethane is finished.
- Relaxation test finished for thermo-oxidative aging on Polyurethane.

Future Research

- Correlating ultra-long & Accelerated characterization
- Degradation of adhesion properties
- Data minimization for training/validation of Multi-agent simulators

Any proposed future work is subject to change based on funding levels.

U.S. DEPARTMENT OF ENERGY

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Thank you

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