A Finite Strain Quasi-Non-Linear Thermoviscoelastic Model for Semi-Crystalline Thermoplastic Polymers subjected to Cyclic Loading

U.-K. Jinaga*, L. Noels

Computational & Multiscale Mechanics of Materials (CM3),
B52, Allée de la découverte 9, B4000 Liège, Belgium

http://www.ltas-cm3.ulg.ac.be/

K. Zulueta, A. Burgoa

Leartiker Polymer,
Xemein Hiribidea, 12 A, 48270 Markina-Xemein, Biscay, Spain

https://www.leartiker.com/polymers

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Introduction

Motivation

- Semicrystalline polymers in thermoplastic composites with remolding capabilities [1] –> High mechanical loads, large deformations and temperatures (~150 ◦C)
- Multiple phenomenologies –> Nonlinear viscoelasto-viscoplasticity

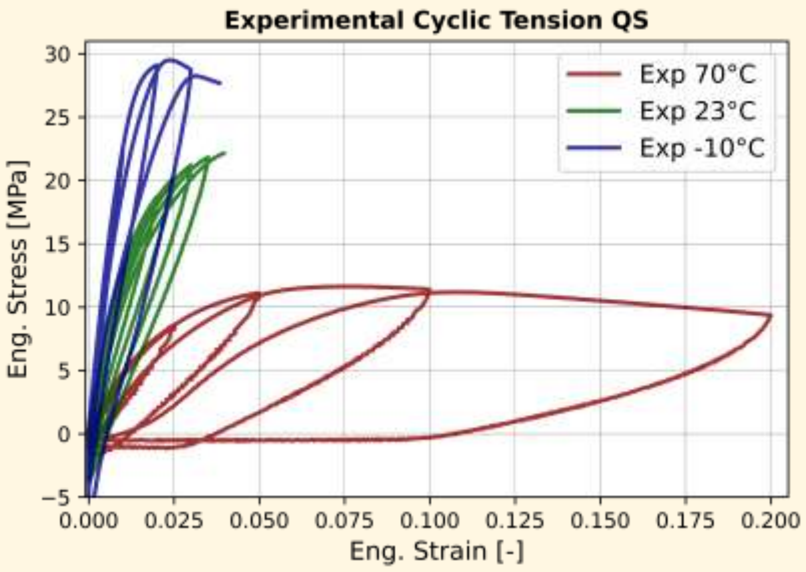
Modelling Requirements

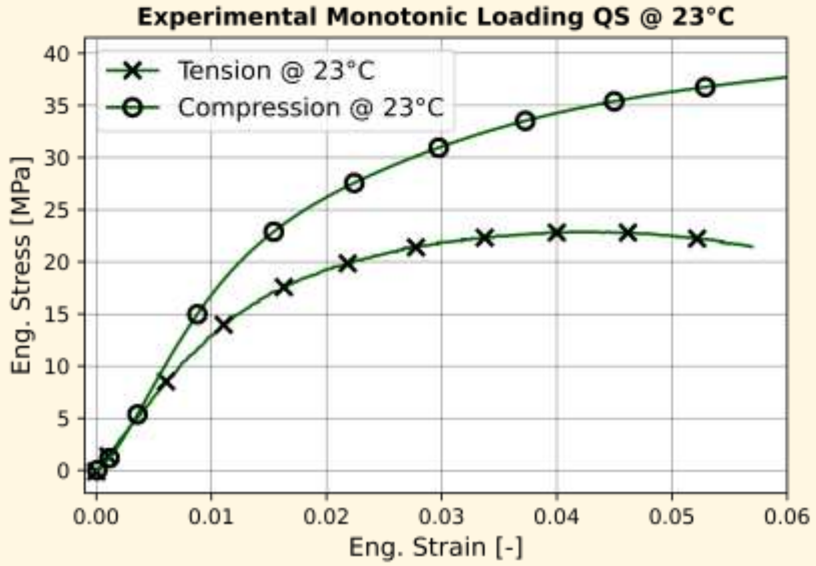
- Strain rate and temperature dependencies
- Tension-compression asymmetry in moduli and yield strengths
- Glass Transition Phenomena (Glass <-> Rubber)
- <u>Material dependency</u> - Calibration -> Experiments using Polypropylene (Borealis BJ380MO), Thermoplastic Urethane (EOS TPU 1301)

Objective: A finite strain quasi-nonlinear thermoviscoelastic model coupled with thermoviscoplasticity!

Experimental Data of PP BJ380MO - ISO 527 -1A

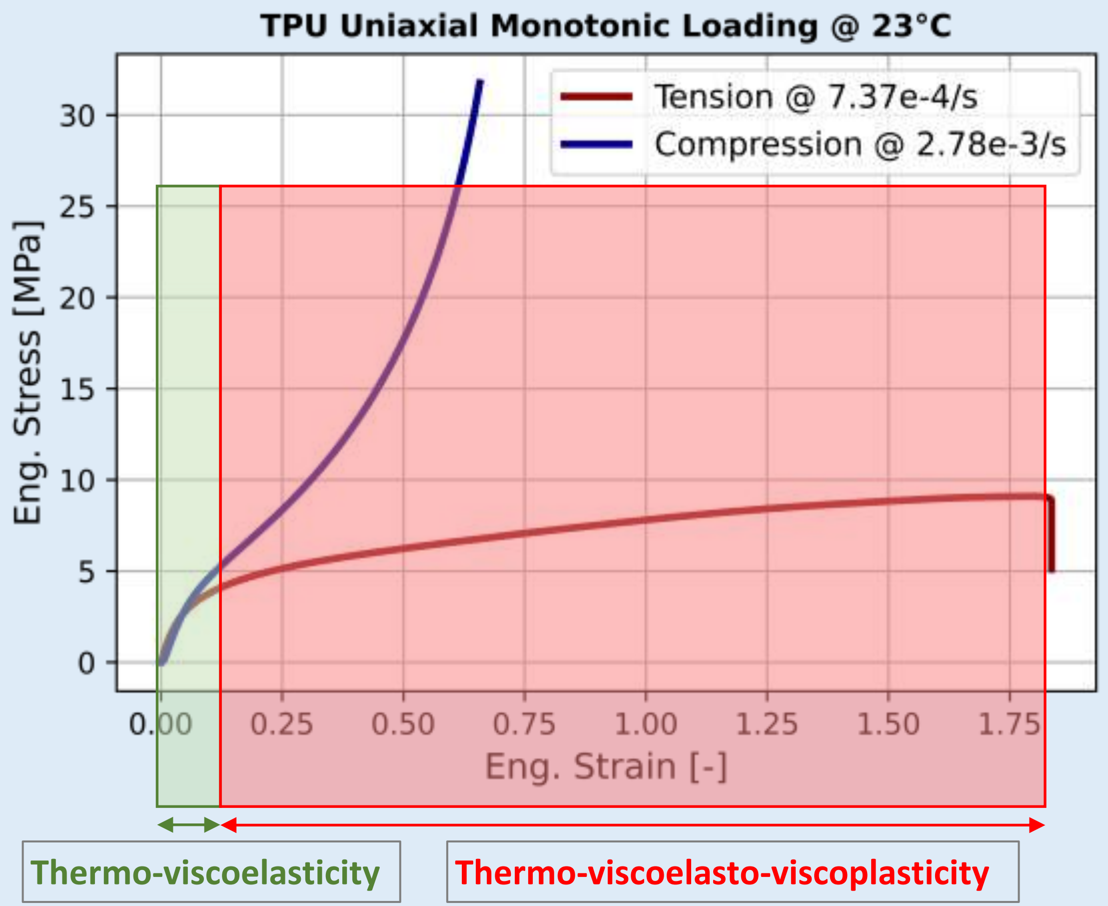
(Provided by Experimental Partner - Leartiker)



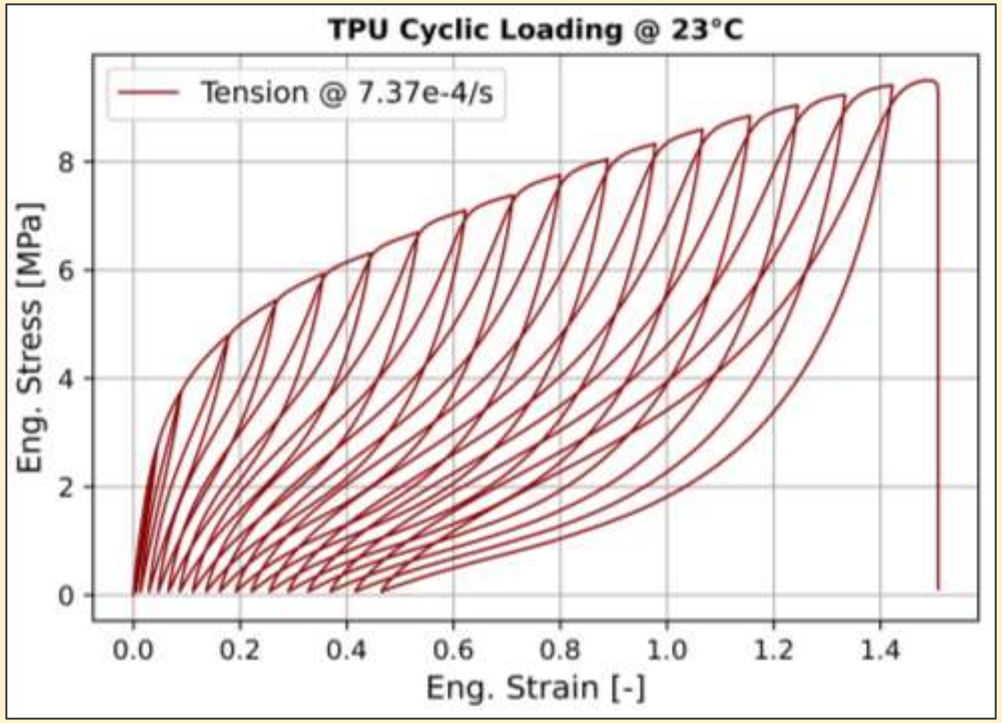


Thermo(visco)mechanics

- Perspective of Thermoplastic Urethane



- Experiments performed using ISO 527, provided by [2 - https://www.moammm.eu/ - This project has received funding from the H2020-EU.1.2.1.-FET Open Programme project MOAMMM under grant No 862015.]



Load-Unload Curves showing Mullins Effect

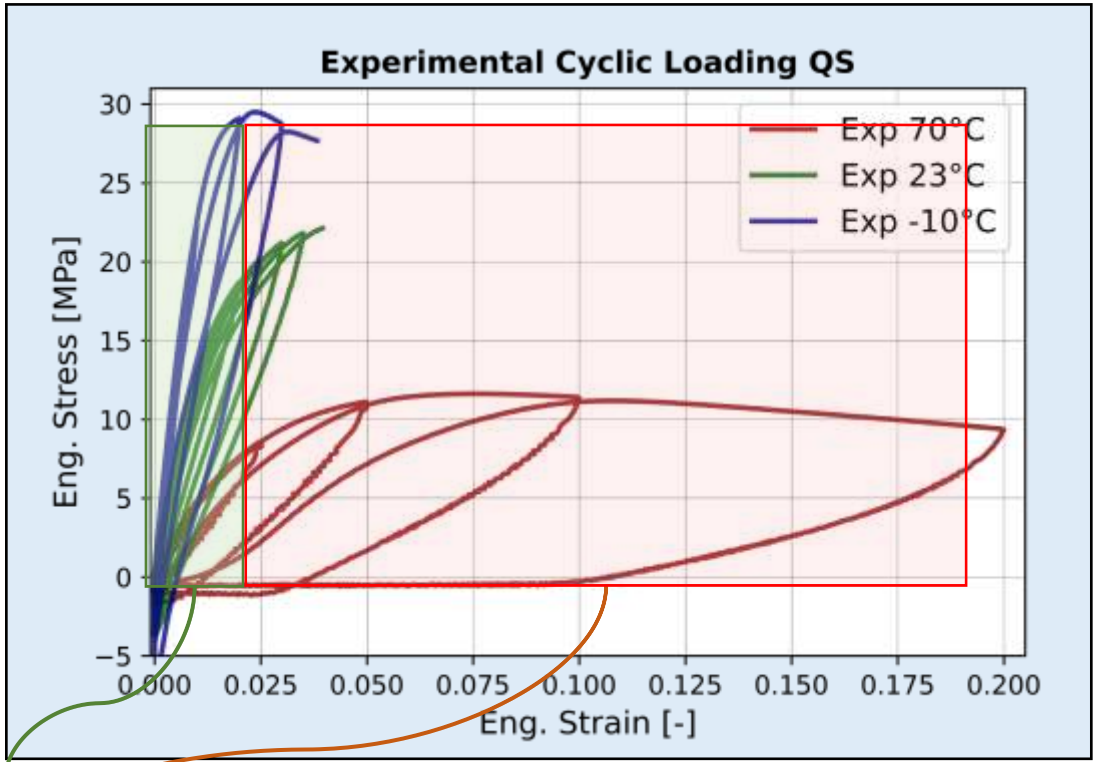
Objectives

1. **Non-linear Thermoviscoelasticity**
2. **Thermoviscoplasticity – Isotropic and Kinematic hardening**
3. **Mullin's-like Effect to capture cyclic loading**

Thermodynamically consistent Formulation!!

Thermo(visco)mechanics

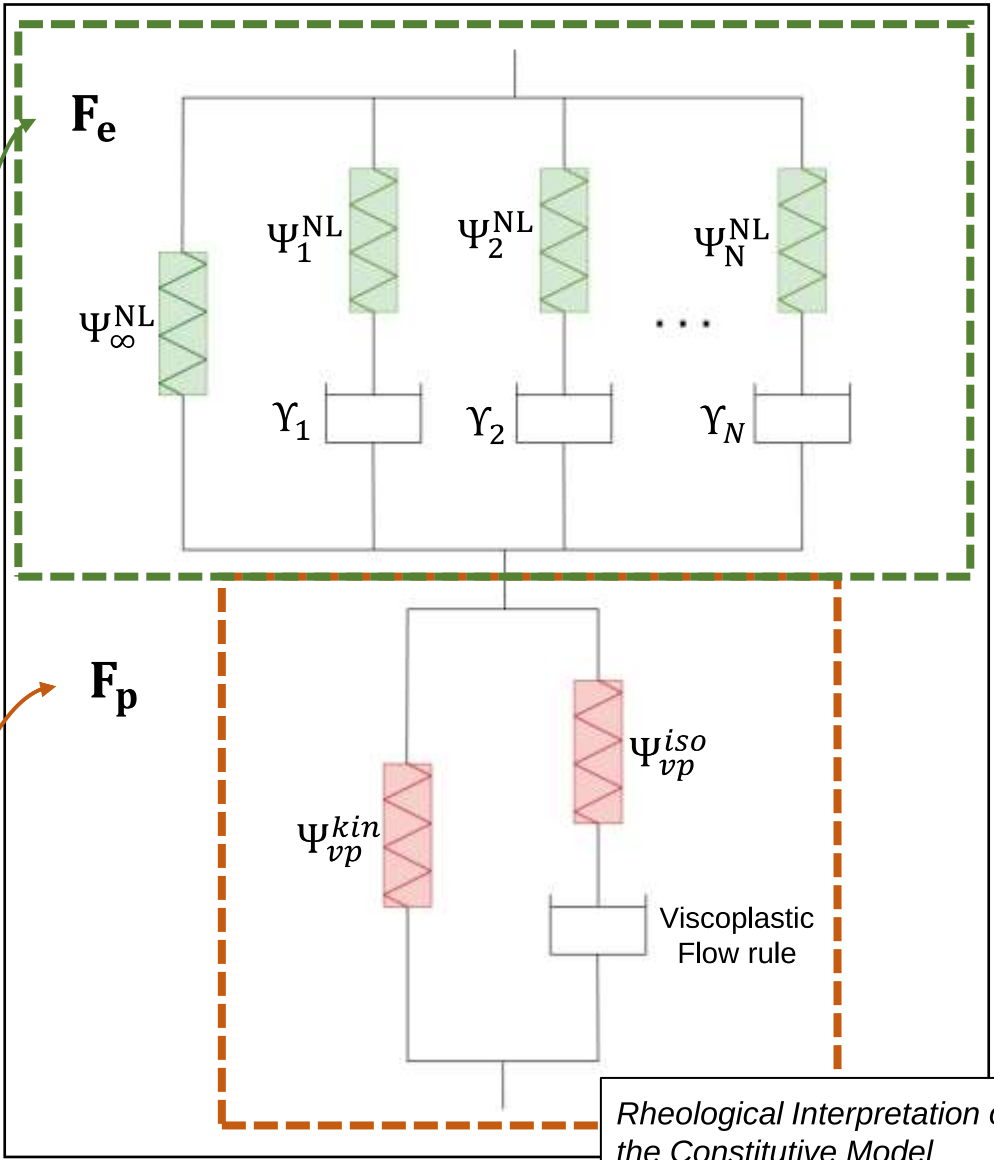
- Rheological Interpretation



Thermo(Visco)Elasticity - Quasi-nonlinear Maxwell Model

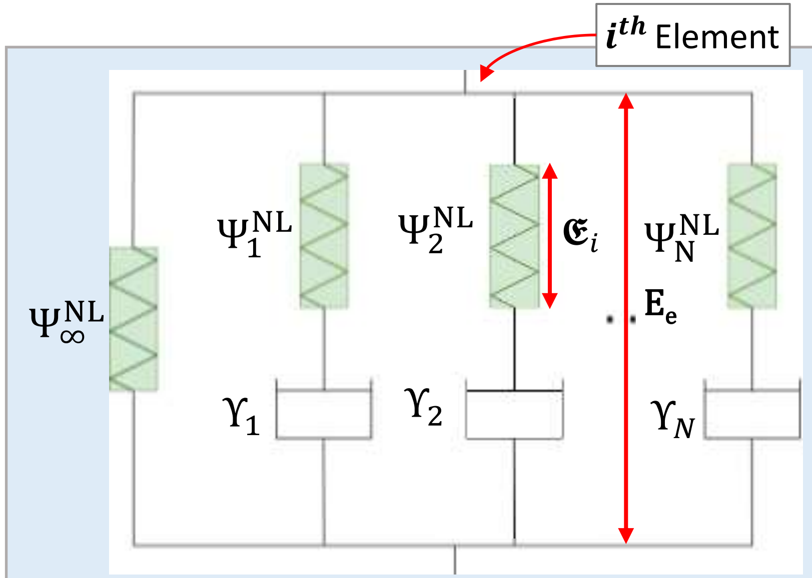
Thermo(Visco)Plasticity - Power Law YF + Perzyna Flow

Additional - Mullins Effect



Rheological Interpretation of the Constitutive Model

Quasi-Nonlinear Thermoviscoelasticity 1 (QNL TVE)



Quasi-Nonlinear Maxwell Model

"Quasi-Nonlinear Viscoelasticity"

Linear viscoelasticity -> Newtonian Dashpots and Hookean springs

+

Strain-dependent moduli -> Nonlinear functions in terms of log strains $\mathbf{E_e}$, $\mathfrak{E}_i$

Linear Viscoelasticity for the i^{th} Maxwell element:

- Rate equations in terms of the strain in the i^{th} spring ($\mathfrak{E}_i$),

$$\dot{\mathfrak{E}}_i = f(\mathbf{E}_e, \mathfrak{E}_i, k_i, g_i)$$

where, $\mathbf{E_e}$ is the logarithmic strain in the intermediate configuration, k_i and g_i are the bulk and shear relaxation times for the i^{th} branch.

- Corotational Kirchhoff stress ($\hat{\boldsymbol{\tau}}_i$) of the i^{th} element in pressure and deviatoric components,

$$\begin{aligned} \hat{p}_i &= K_i \operatorname{tr} \mathfrak{E}_i \\ \operatorname{dev} \hat{\boldsymbol{\tau}}_i &= 2G_i \operatorname{dev} \mathfrak{E}_i \end{aligned}$$

where, K_i and G_i are the bulk and shear moduli for the i^{th} branch.

Strain-dependent moduli:

- Corotational Kirchhoff stress ($\hat{\boldsymbol{\tau}}_i$) in the i^{th} branch retains its form,

$$\begin{aligned} \hat{p}_i &= K_i (\operatorname{tr} \mathfrak{E}_i, V_j) \operatorname{tr} \mathfrak{E}_i \\ \operatorname{dev} \hat{\boldsymbol{\tau}}_i &= 2G_i (||\operatorname{dev} \mathfrak{E}_i||, D_j) \operatorname{dev} \mathfrak{E}_i \end{aligned}$$

where, K_i and G_i functions contain the non-linearity, V_j and D_j are treated as material parameters.

Quasi-Nonlinear Thermoviscoelasticity 2 (QNL TVE)

Strain-dependent moduli:

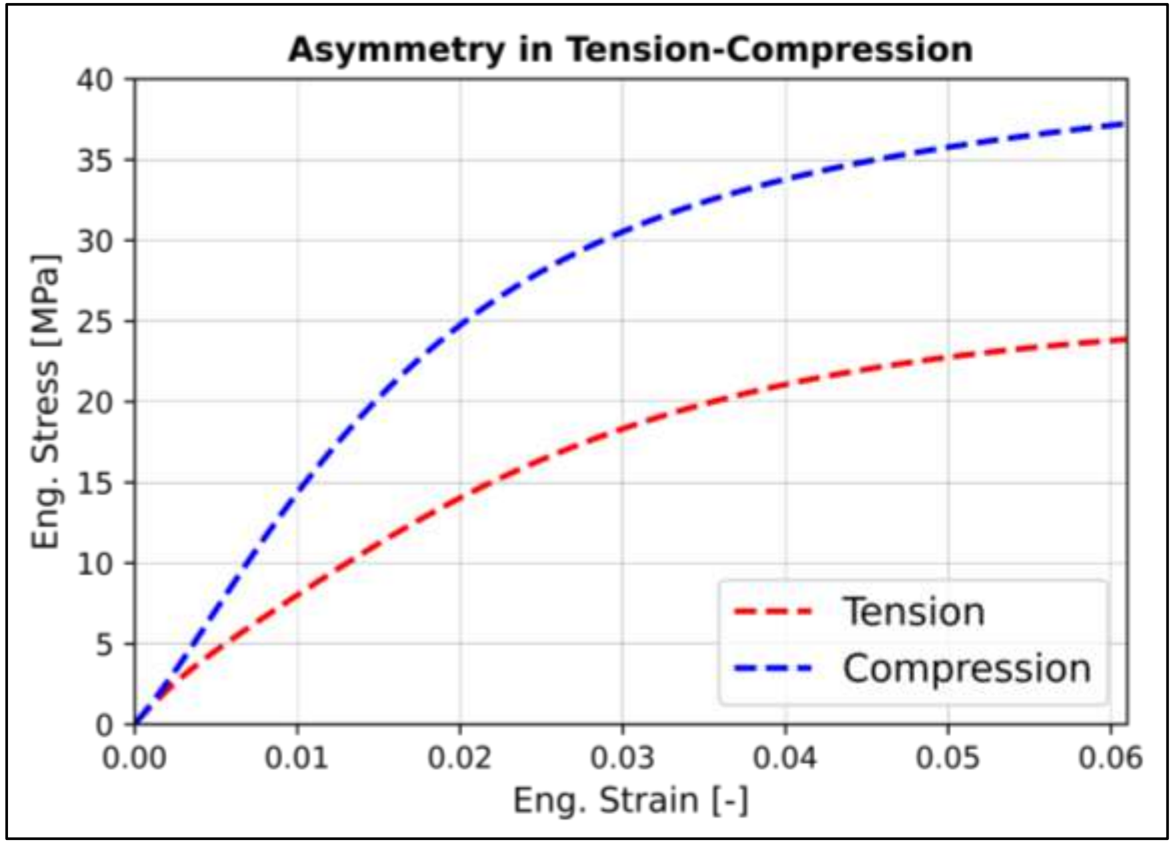
- Corotational Kirchhoff stress ($\hat{\boldsymbol{\tau}}_i$) in the $\boldsymbol{i^{th}}$ branch retains its form,

$$\begin{aligned} \widehat{p}_i &= K_i \left(\operatorname{tr} \mathfrak{E}_i, V_j\right) \operatorname{tr} \mathfrak{E}_i \\ \operatorname{dev} \widehat{\boldsymbol{\tau}}_i &= 2G_i \left(||\operatorname{dev} \mathfrak{E}_i||, D_j\right) \operatorname{dev} \mathfrak{E}_i \end{aligned}$$

where, K_i and G_i functions contain the non-linearity, V_j and D_j are treated as material parameters.

- **Features -** Non-linearity in viscoelastic regime at low strains, large tension-compression asymmetry in stiffness.

- **To calibrate:** V_j and D_j are calibrated from uniaxial tension and compression tests at different temperatures.



Numerical Setup: Uniaxial stress and quasi-steady tension-compression sweeps using quasi-nonlinear elasticity.

Temperature Dependency of QNL TVE

Convolution Integrals in shifted time

- Deviatoric integral shown below

$$\mathrm{dev}\,\mathfrak{E}_i = \int_{-\infty}^{t} \exp\left(-\frac{t^* - s^*}{g_i}\right) \frac{d}{ds} \mathrm{dev}\,\mathbf{E}_e(s)\,\mathrm{ds}$$

where, the WLF shift factor (a_T) is used to evaluate the shifted time-step.

$$\Delta t_n^* = \int_{t_n}^{t_{n+1}} \frac{1}{a_T(\epsilon)} d\epsilon$$

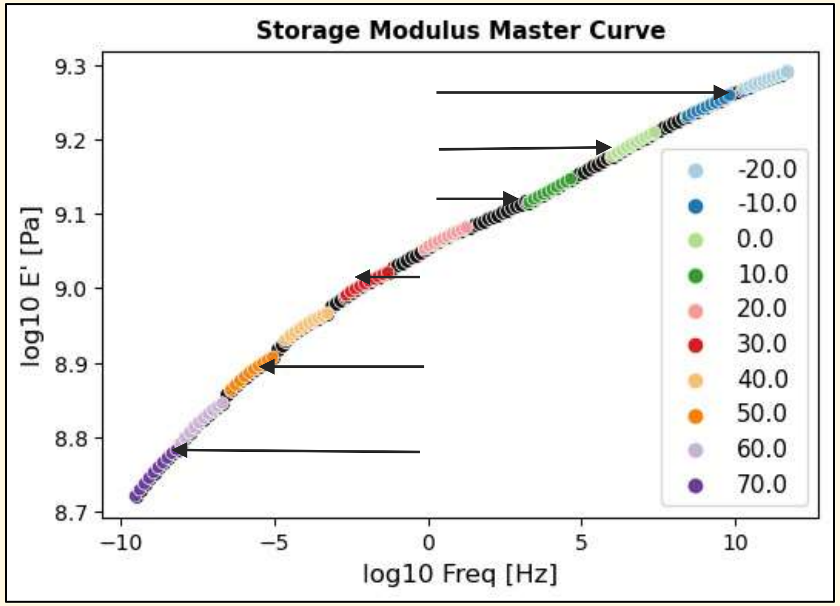
To calibrate: Dynamic moduli and shift factor.

- Dynamic Mechanical Analyses (DMA) in tension.

Construction of Tension Master Curve

$$E'(\omega, \mathrm{T}) = E'(\mathrm{a_T}\,\omega\,, \mathrm{T_{ref}})$$

where, T_{ref} is the reference temperature (a bit lower than Tg). The isotherms are rigidly shifted in the log-log plot.



- Rigid shifts give the shift factor:

$$a_T = exp\left(-\frac{C_1(T - T_{ref})}{C_2 + T - T_{ref}}\right)$$

- Curve fitting gives the relaxation spectrum:

$$E(t) = E_\infty + \sum_{(i)} E_i \exp(-\frac{t}{\tau_i})$$

Thermo(Visco)Plasticity (TVP)

Elements

- Extended Drucker-Prager Power Yield Function ($\bar{F}$) - pressure dependency in $\boldsymbol{\phi}_p$ and $\mathbf{a_2}$, $\mathbf{a_1}$, $\mathbf{a_0}$ coefficients as functions of plastic strain ($\Delta\gamma$) and temperature-dependent tensile and compressive yield strength, Γ is the flow parameter.

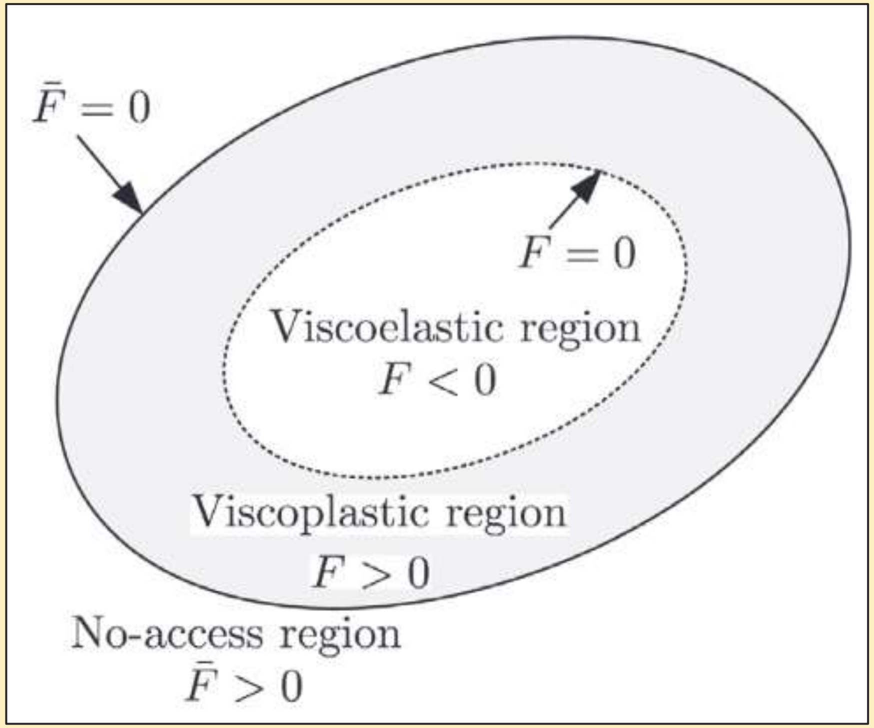
$$\bar{F}(\Gamma, \Delta\gamma, T) = a_2 \boldsymbol{\phi}_{\boldsymbol{e}}^{\alpha} - a_1 \boldsymbol{\phi}_{\boldsymbol{p}} - a_0 - \left(\eta \frac{\Gamma}{\Delta t}\right)^p$$

- Perzyna Flow Rule (non-associative): with temperature-dependent viscosity (η), quadratic plastic potential (P)

$$\mathbf{D}_p = \frac{1}{\eta(T)} \langle \boldsymbol{F} \rangle^{\frac{1}{p}} \frac{\partial P}{\partial \mathbf{M}_e}$$

- Chaboche NLKH - $\mathbf{B}$ is the backstress and $H_{\mathbf{B}}$ is the kinematic hardening modulus expressed as a function of plastic strain

$$\dot{\mathbf{B}} = k^2 H_{\mathbf{B}} \mathbf{D}_p + \frac{1}{H_{\mathbf{B}}} \frac{\partial H_{\mathbf{B}}}{\partial \gamma} \dot{\gamma} \mathbf{B} + \frac{1}{H_{\mathbf{B}}} \frac{\partial H_{\mathbf{B}}}{\partial T} \dot{T} \mathbf{B}$$

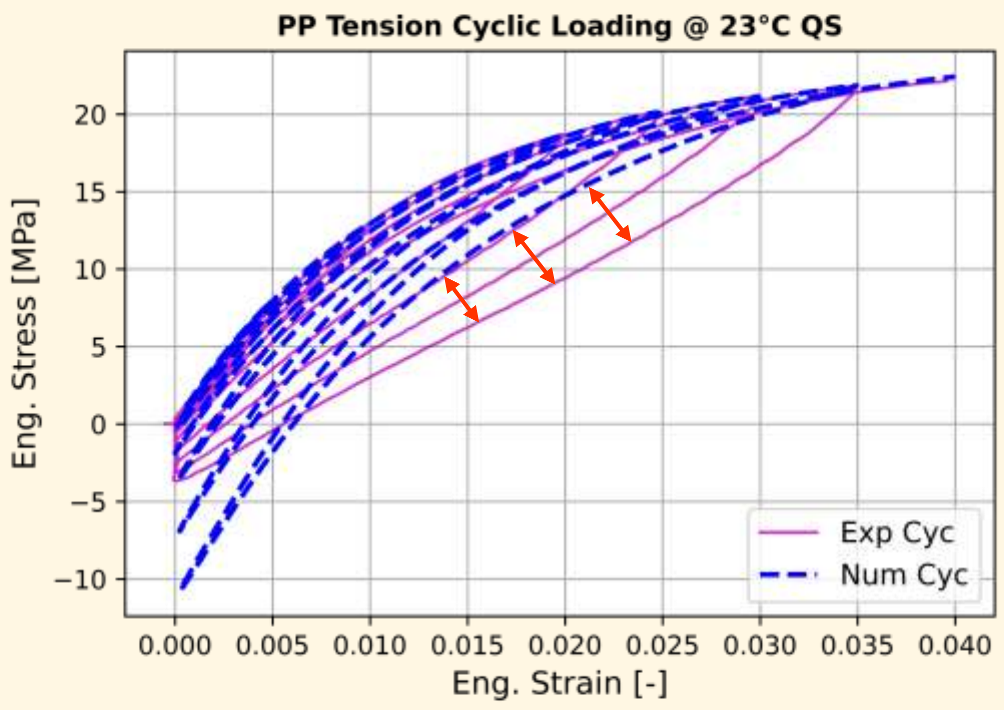


Extended yield surface with various regimes (Nguyen et al. 2016 [3])

To calibrate: Yield strengths, kinematic hardening modulus, viscosity and power yield parameters

- Uniaxial tensile and compressive tests at different temperatures.

Mullin's Effect



Viscoelasto-viscoplastic simulation without Mullin's-like effect

Concept

- Mullin's effect to lower the stress in unloading using a damage variable (ζ -> 1 to ζ_{min}) functional of maximum deformation energy encountered in load history [4]

$$\zeta = \zeta(\widehat{\psi}, \widehat{\psi}_{max}) \Rightarrow \boldsymbol{\tau}_e = \zeta \widehat{\boldsymbol{\tau}}_e$$

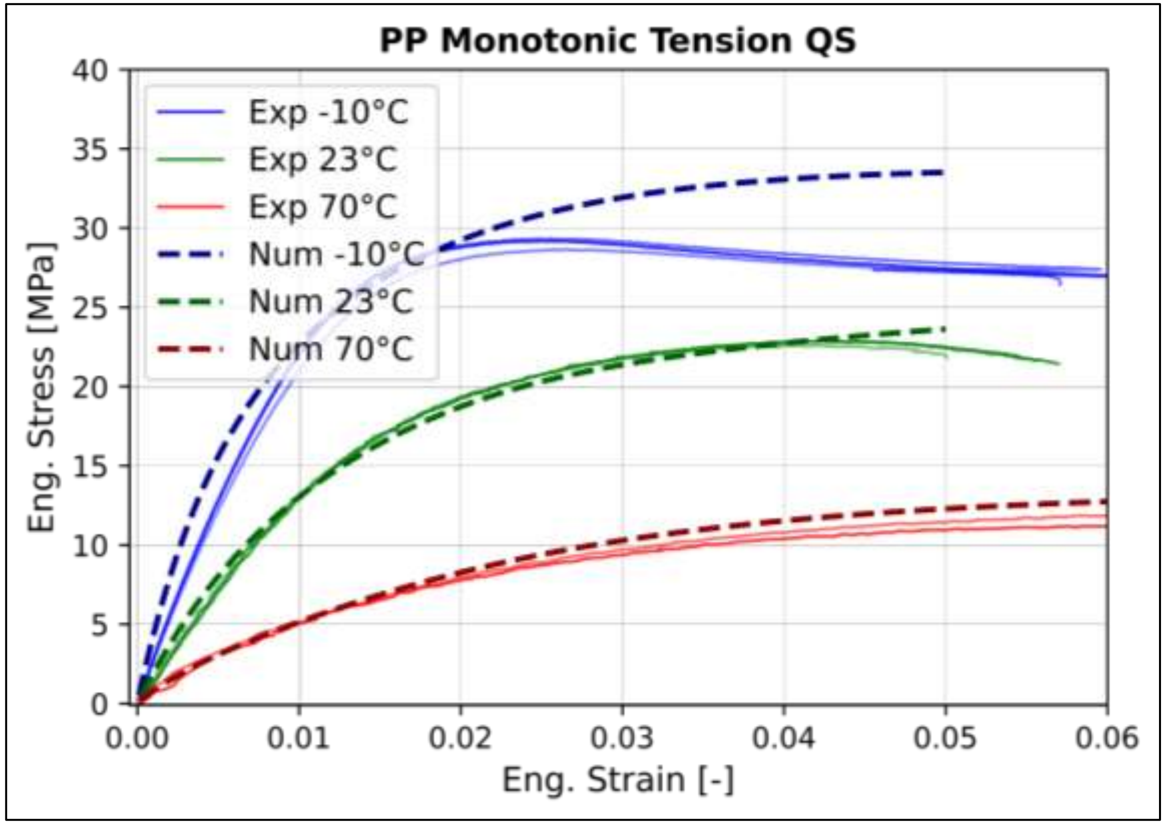
- **Linear Mullin's Scaler**

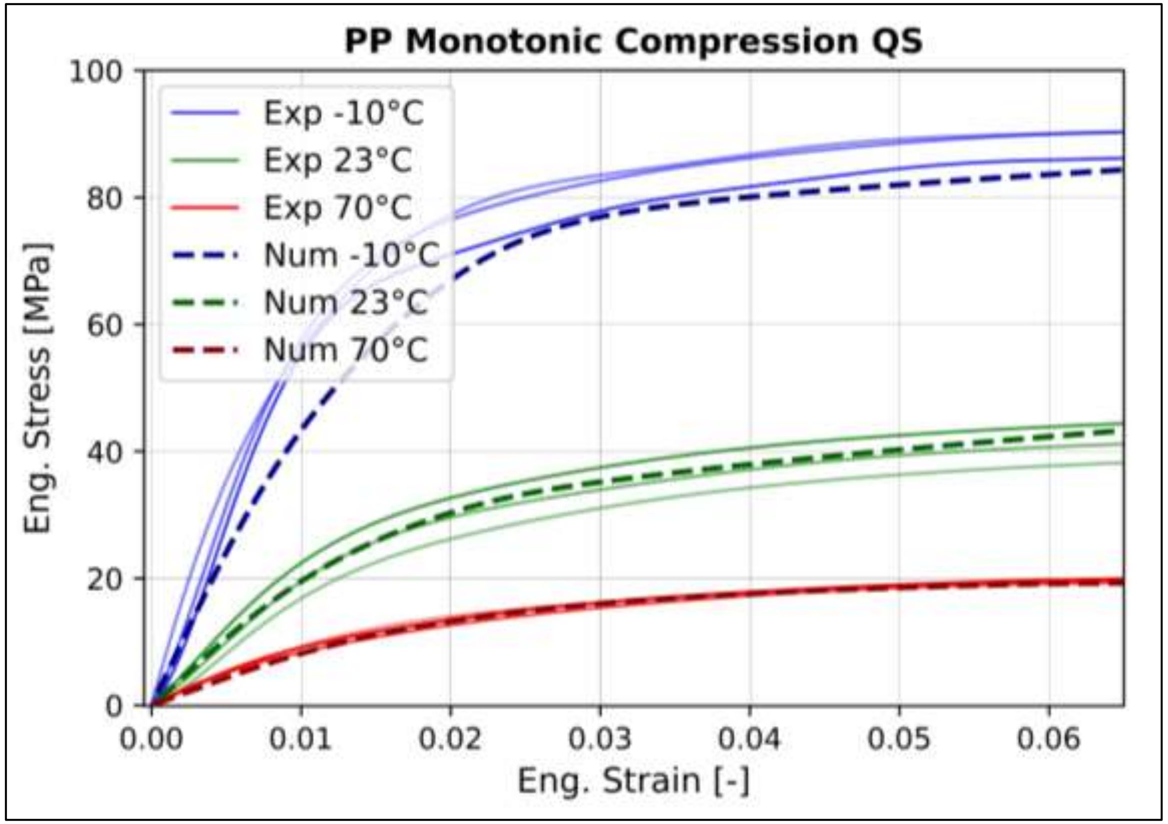
$$\zeta = 1 - z\left(1 - \frac{\widehat{\psi}}{\widehat{\psi}_{max}}\right)$$

Where, the **material parameter (z)** has the limits $0 \leq z < 1$ for consistency and is temperature dependent.

Assumption: Approximated solution of the viscoelastic deformation energy ($\widehat{\psi}_{ve}$) is assumed to be sufficient, viscoplastic energy ($\widehat{\psi}_{vp}$) is not considered here.

Results for Polypropylene BJ380MO
- Uniaxial Monotonic Loading

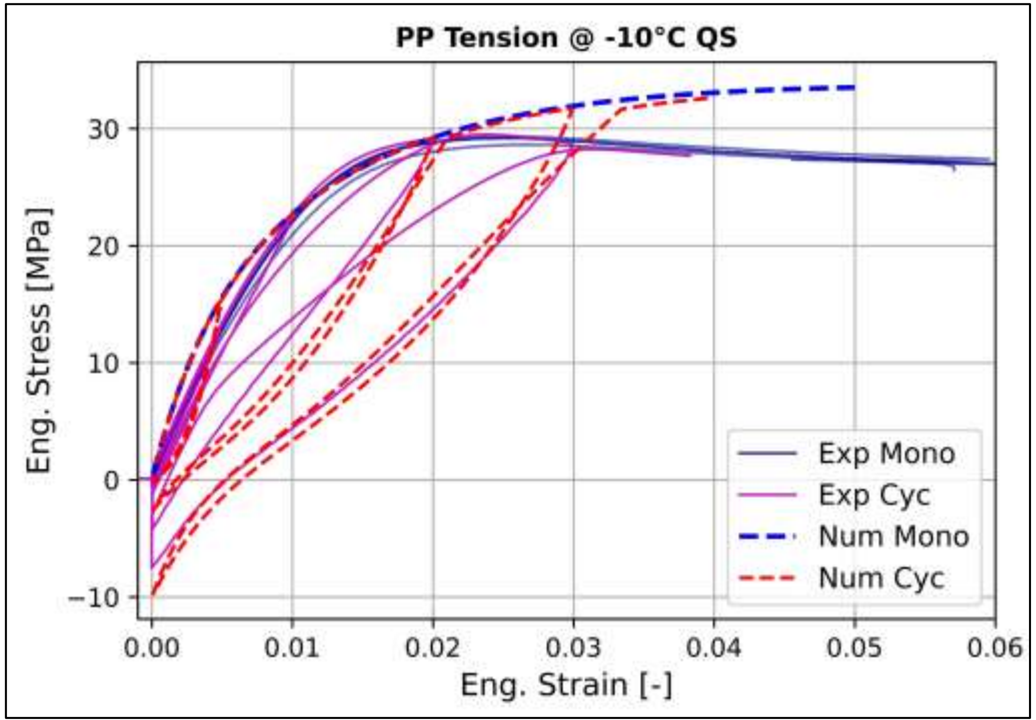




Numerical Setup - Uniaxial Stress with QNL TVE + linear exponential isotropic hardening TVP

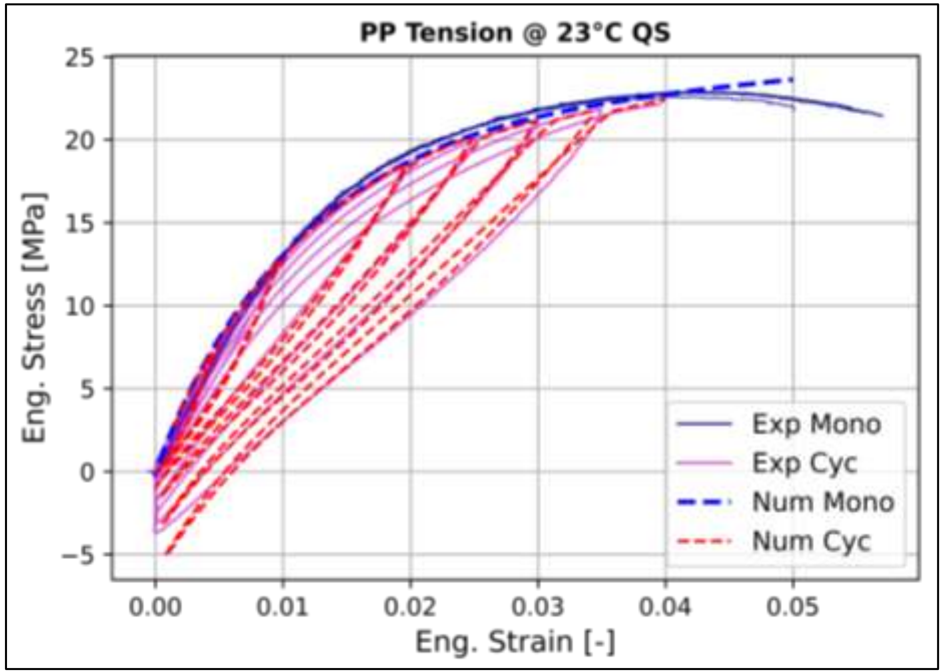
- QNL TVE: Linearly decreasing V_j and D_j with temperature
- Good match with the experimental results -> Large asymmetry in elastic moduli and yield strengths captured at all temperatures

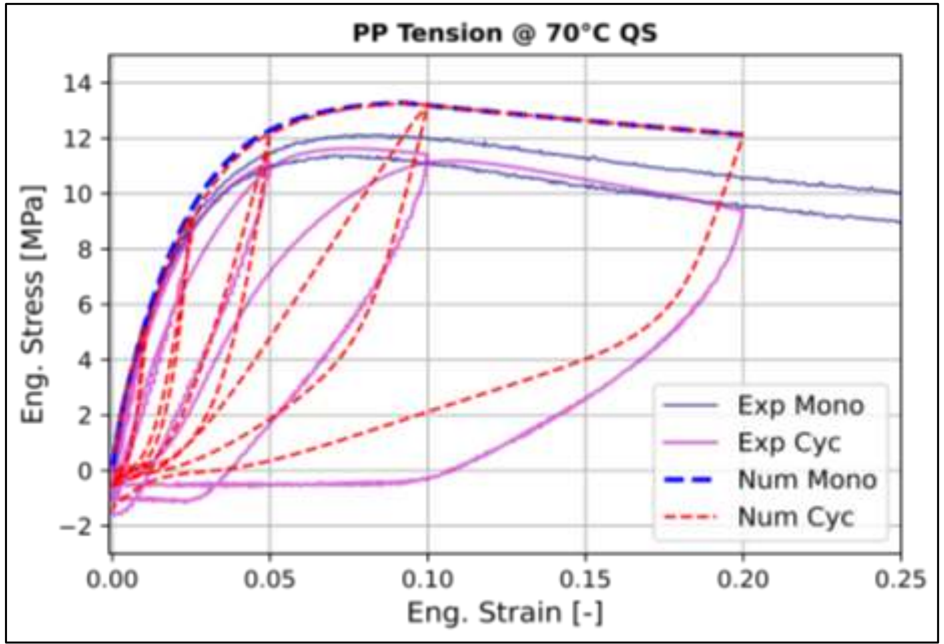
Results for Polypropylene BJ380MO
- Uniaxial Cyclic loading



Numerical Setup - Uniaxial Stress with QNL TVE linear exponential isotropic hardening + Mullin's effect

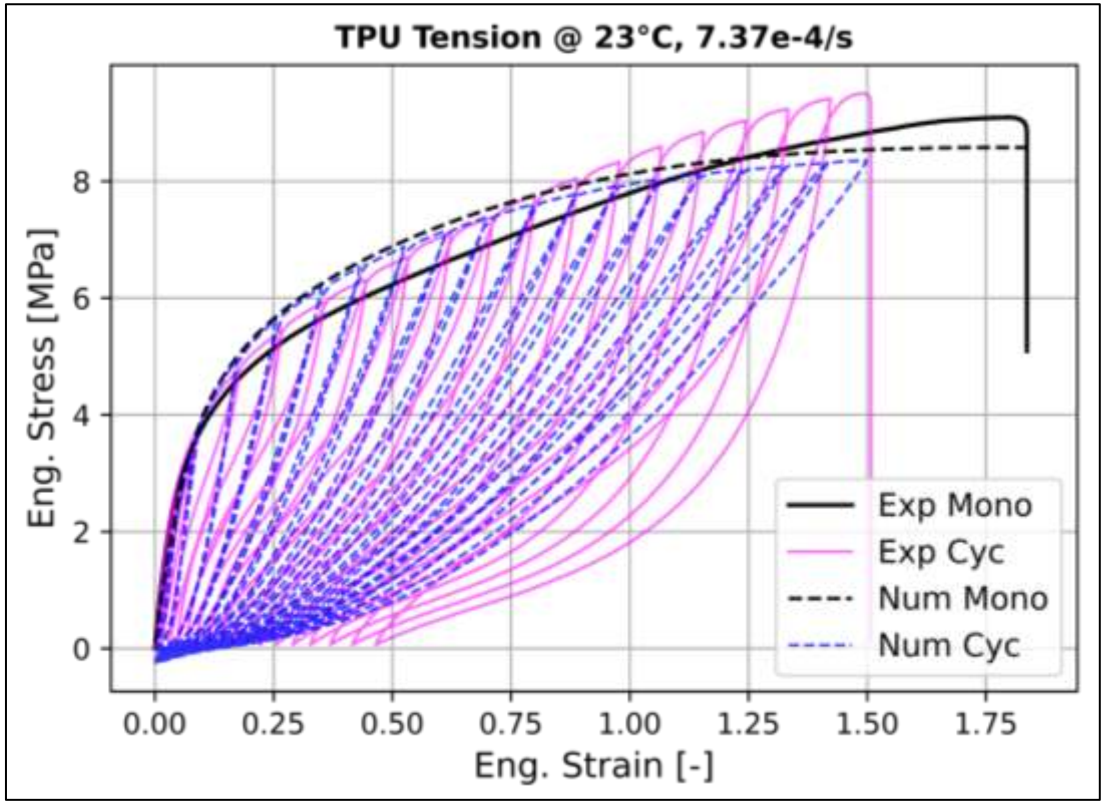
- Good agreement with experimental results in cyclic loading

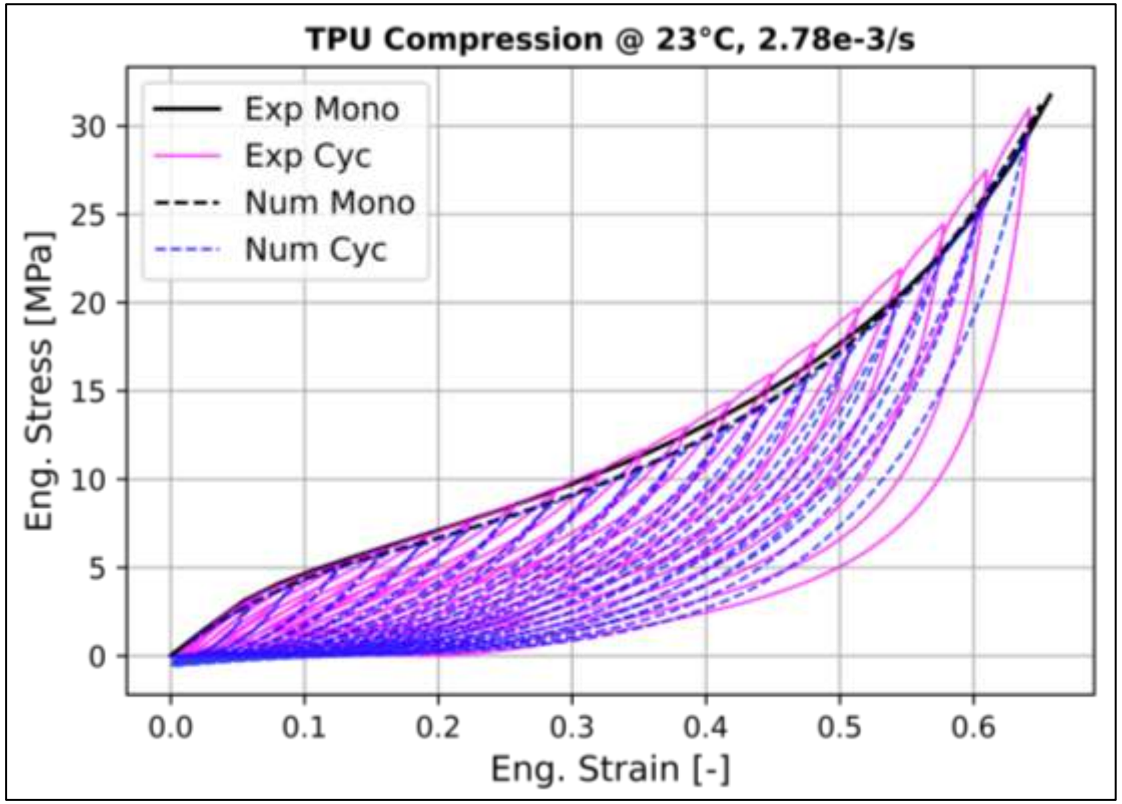




Results for Thermoplastic Polyurethane EOS 1301

– Monotonic and Cyclic Loading

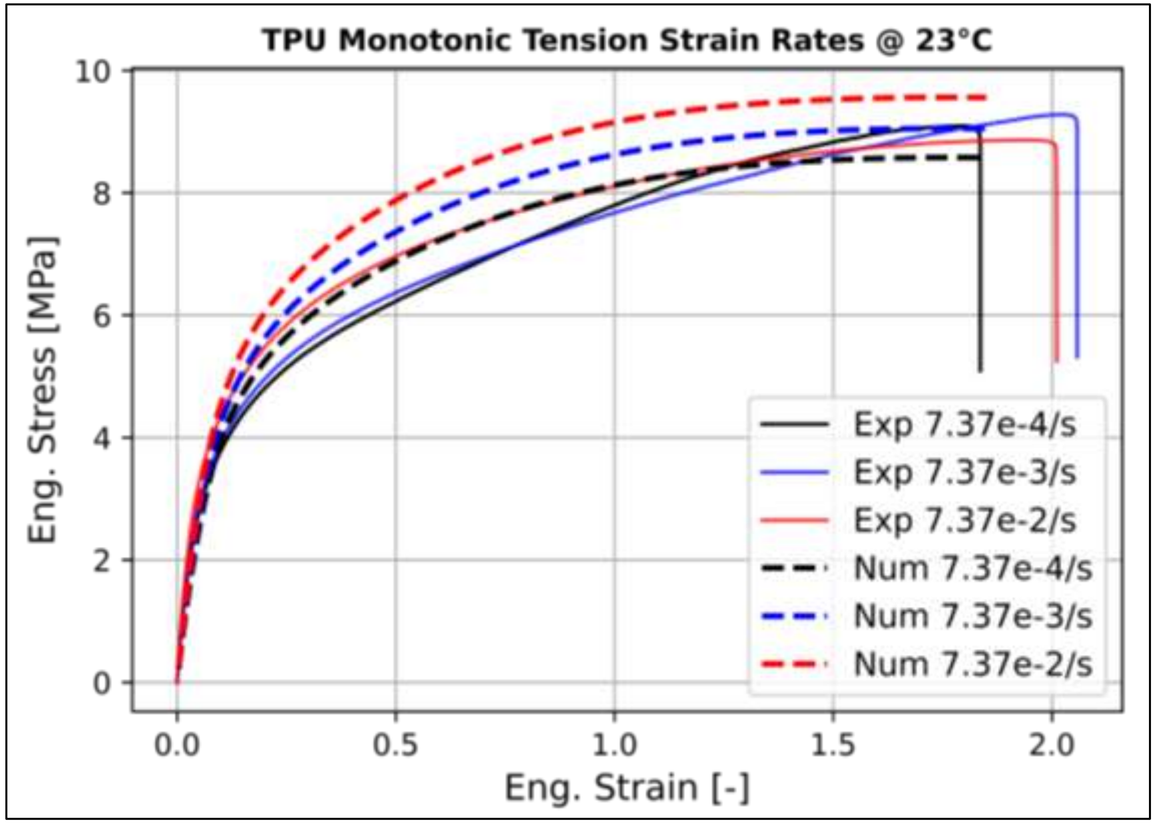


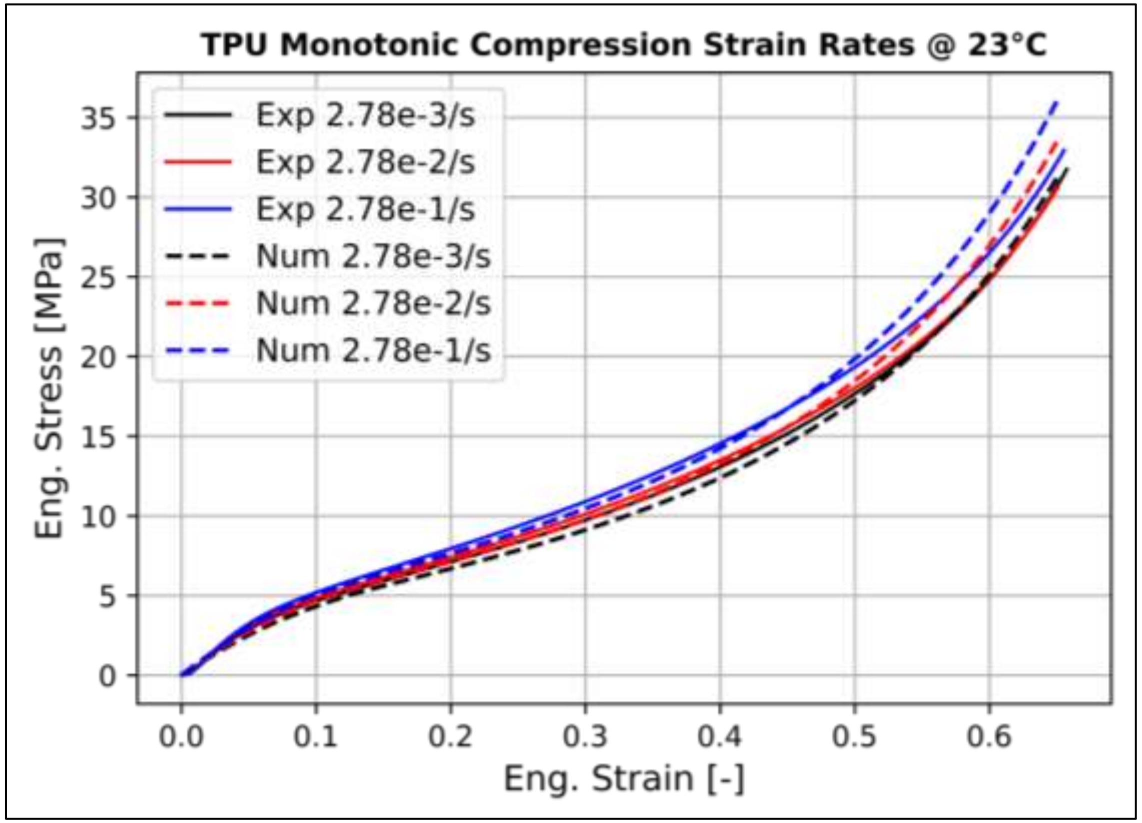


Numerical Setup - Uniaxial Stress with QNL TVE + linear exponential isotropic hardening + polynomial kinematic hardening TVP + Mullins effect

- QNL TVE: All TVE branches with equal parameters
- Good agreement with experimental results in compression, mismatch in tension attributed to less viscoplasticity

Results for Thermoplastic Polyurethane EOS 1301
– Variable Strain Rate





Numerical Setup - Uniaxial Stress with QNL TVE + linear exponential isotropic + polynomial kinematic hardening TVP

- QNL TVE: All TVE branches with equal parameters

Takeaway: Sensitivity to strain rates is overpredicted by the model

Final Remarks

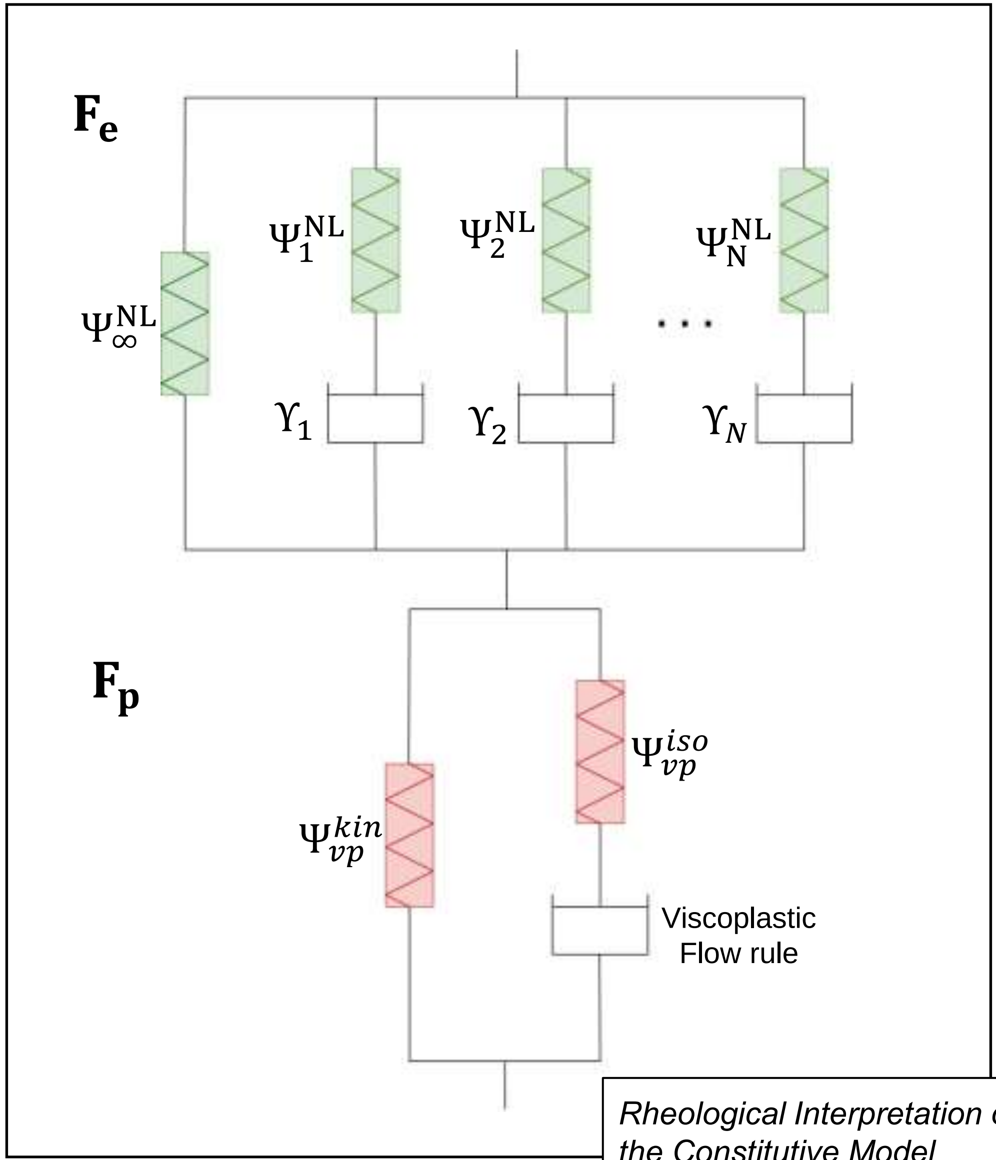
Conclusions

- Captured large strain and temperature-dependent non-linearities in the viscoelastic regime.
- Addressed tension-compression asymmetry in moduli and yielding.

- For semi-crystalline co-polymer Polypropylene (Borealis BJ380MO) and elastomeric Thermoplastic Urethane (EOS TPU 1301).

Remarks

- Pure validation simulations in 3D using ISO 527 -1A (dogbone) and ASTM D638 (dumbbell) in tension/compression at variable strain rates and temperatures.



Rheological Interpretation of the Constitutive Model

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Thank You for your attention

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References

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2. https://www.moammm.eu/ - This project has received funding from the H2020-EU.1.2.1.-FET Open Programme project MOAMMM under grant No 862015.
3. *Nguyen, V-D., F. Lani, T. Pardoen, X. P. Morelle, and L. Noels. "A large strain hyperelastic viscoelastic-viscoplastic-damage constitutive model based on a multi-mechanism non-local damage continuum for amorphous glassy polymers." International Journal of Solids and Structures 96 (2016): 192-216.*
4. Naumann, Christoph, and Jörn Ihlemann. "On the thermodynamics of pseudo-elastic material models which reproduce the Mullins effect." *International Journal of Solids and Structures* 69 (2015): 360-369.
5. Krairi, Anouar, I. Doghri, Joanna Schalnat, G. Robert, and Wim Van Paepegem. "Thermo-mechanical coupling of a viscoelastic-viscoplastic model for thermoplastic polymers: Thermodynamical derivation and experimental assessment." *International Journal of Plasticity* 115 (2019): 154-177.

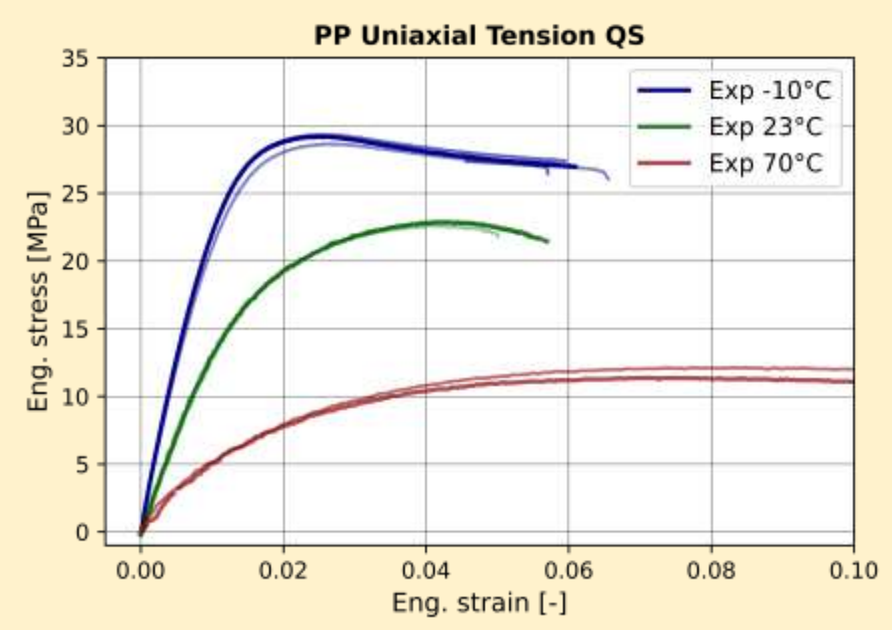
Appendix: Experimental Campaign Details
(with Leartiker, JKU)

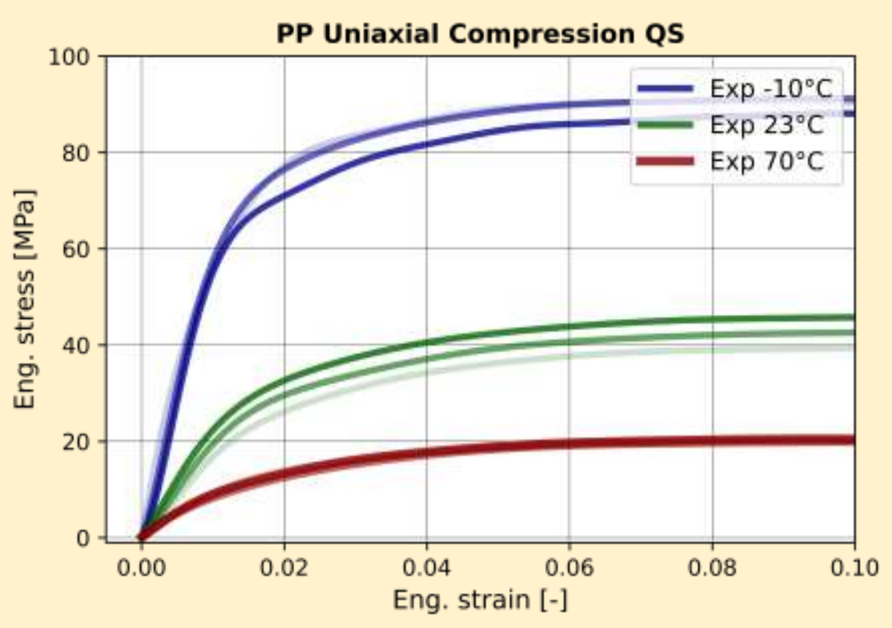
Choice of Polymers

- Thermoplastics - reversible glass transition, recyclable.
- **Polypropylene** – TC asymmetry in elasticity, varied performance with temperature
- **Thermoplastic Polyurethane** – Wide strain range of (visco)elasticity and prominent Mullins effect

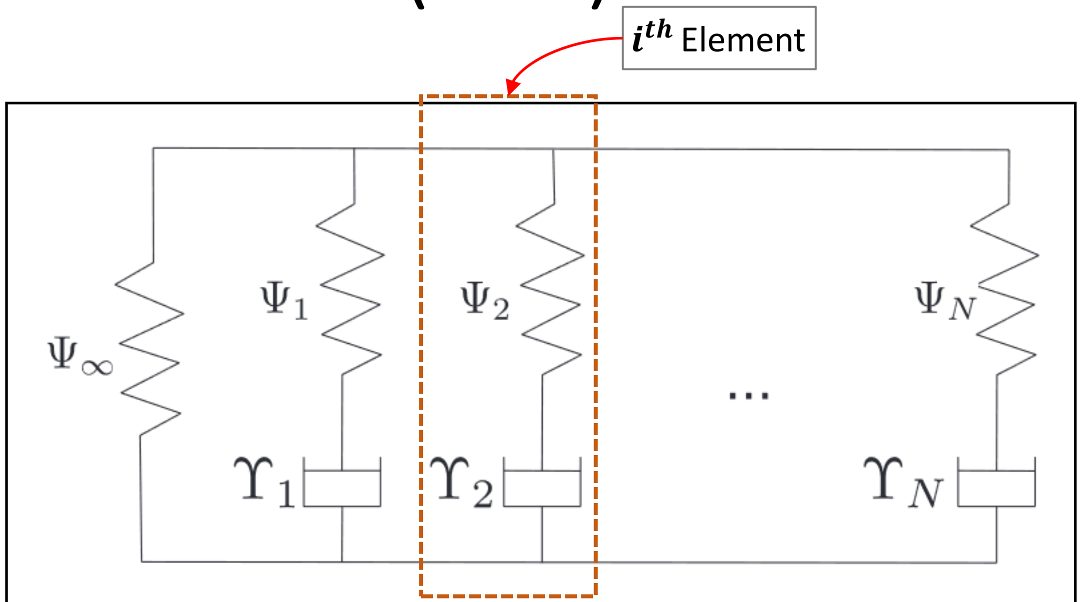
Performed Experiments

1. DMA Tension Mode – Glass transition range, TVE shift factor and relaxation spectrum.
2. QS Tension-Compression Tests - Stress strain isotherms for TVP calibration.
3. Thermal Property Tests : DSC -> Specific Heat (Cp), TMA -> Coefficient of Thermal Expansion (CTE) – zero/small force
4. Cyclic Loading Tests - Stress strain isothermal cycles at different temperatures.





Appendix:
Linear Thermo(Visco)Elastic Model



Generalised Maxwell model

ODEs for internal variable ($\mathfrak{E}_i$). To solve for $\mathfrak{E}_i$ using **TTSP**!

$$\mathrm{tr}\,\dot{\mathfrak{E}}_i + \frac{\mathrm{tr}\,\mathfrak{E}_i}{k_i} = \mathrm{tr}\,\dot{\mathbf{E}}_e$$
$$\mathrm{dev}\,\dot{\mathfrak{E}}_i + \frac{\mathrm{dev}\,\mathfrak{E}_i}{g_i} = \mathrm{dev}\,\dot{\mathbf{E}}_e$$

where, $\mathbf{E_e}$ is the logarithmic strain in intermediate configuration.

$$\mathbf{E}_e = \frac{1}{2}\ln \mathbf{C}_e$$

For the i^{th} Maxwell element:

- Elastic (log) Strain in each Maxwell spring, ($\mathfrak{E}_i$ = $\mathbf{E_e}$ $-\boldsymbol{\Gamma}_i$)
- Hookean Viscoelastic Free Energy ($\hat{\psi}_{ve}$) is quadratic in $\mathfrak{E}_i$

$$\hat{\psi}_{ve} = \hat{\psi}_{ve}\left(\mathrm{tr}\,\mathfrak{E}_i^2, \mathrm{dev}\,\mathfrak{E}_i : \mathrm{dev}\,\mathfrak{E}_i\right)$$

- Consistent Corotational Kirchhoff stress ($\hat{\tau}_i$)

$$\hat{p}_i = K_{i0}\,\mathrm{tr}\,\mathfrak{E}_i$$
$$\mathrm{dev}\,\hat{\boldsymbol{\tau}}_i = 2G_{i0}\,\mathrm{dev}\,\mathfrak{E}_i$$

where, K_{i0} and G_{i0} are the initial moduli.

- Rate equations solved for i^{th} spring strain ($\mathfrak{E}_i$), in shifted time using time-temperature superposition (TTSP) , (deviatoric shown below)

$$\mathrm{dev}\,\mathfrak{E}_i = \exp\left(-\frac{\Delta t^*|_{rec}}{g_i}\right)\mathrm{dev}\,\mathfrak{E}_i^n + \exp\left(-\frac{\Delta t^*|_{mid}}{g_i}\right)\left(\mathrm{dev}\,\mathbf{E}_e - \mathrm{dev}\,\mathbf{E}_e^n\right)$$

- Stress relaxation in shifted time using WLF shift factor.

$$\Delta t_n^* = \int_{t_n}^{t_{n+1}} \frac{1}{a_T(\epsilon)} d\epsilon \quad \Rightarrow \quad a_T = exp\left(-\frac{C_1(T - T_{ref})}{C_2 + T - T_{ref}}\right)$$

<u>**To Calibrate:**</u> Dynamic Modulus (E(t)) – Tension/Compression DMA

$$E(t) = E_\infty + \sum_{(i)} E_i\,\exp\left(-\frac{t}{\tau_i}\right)$$

Appendix: Thermo(Visco)Plastic Model: Unknowns

Elements

- Extended Drucker-Prager Power Yield Function ($\bar{F}$) - pressure dependency in $\boldsymbol{\phi}_p$ and $\mathbf{a_2, a_1, a_0}$ coefficients as functions of plastic strain ($\Delta\gamma$) and temperature-dependent tensile and compressive yield strength, Γ is the flow parameter.

$$\bar{F}(\Gamma, \Delta\gamma, T) = a_2 \boldsymbol{\phi}_{\boldsymbol{e}}^{\alpha} - a_1 \boldsymbol{\phi}_{\boldsymbol{p}} - a_0 - \left(\eta \frac{\Gamma}{\Delta t}\right)^p$$

- Perzyna Flow Rule (non-associative): with temperature-dependent viscosity (η), quadratic plastic potential (P)

$$\mathbf{D}_p = \frac{1}{\eta(T)} \langle \boldsymbol{F} \rangle^{\frac{1}{p}} \frac{\partial P}{\partial \mathbf{M}_e}$$

- Chaboche NLKH - $\mathbf{B}$ is the backstress and $H_\mathbf{B}$ is the kinematic hardening modulus expressed as a function of plastic strain

$$\dot{\mathbf{B}} = k^2 H_\mathbf{B} \mathbf{D}_p + \frac{1}{H_\mathbf{B}} \frac{\partial H_\mathbf{B}}{\partial \gamma} \dot{\gamma} \mathbf{B} + \frac{1}{H_\mathbf{B}} \frac{\partial H_\mathbf{B}}{\partial T} \dot{T} \mathbf{B}$$

TVP Internal Variables:

- Equivalent plastic strain ($\Delta\gamma$): Solved using a rate equation. (k is a material parameter depending on the initial plastic Poisson's ratio)

$$\dot{\gamma} = k\sqrt{\mathbf{D}_p : \mathbf{D}_p}$$

- Flow parameter (Γ): Solved through Newton-Raphson on $\bar{F}$.

- **Two Equations:** Yield function ($\bar{F}$=0) and $\Delta\gamma$ rate equation**!**

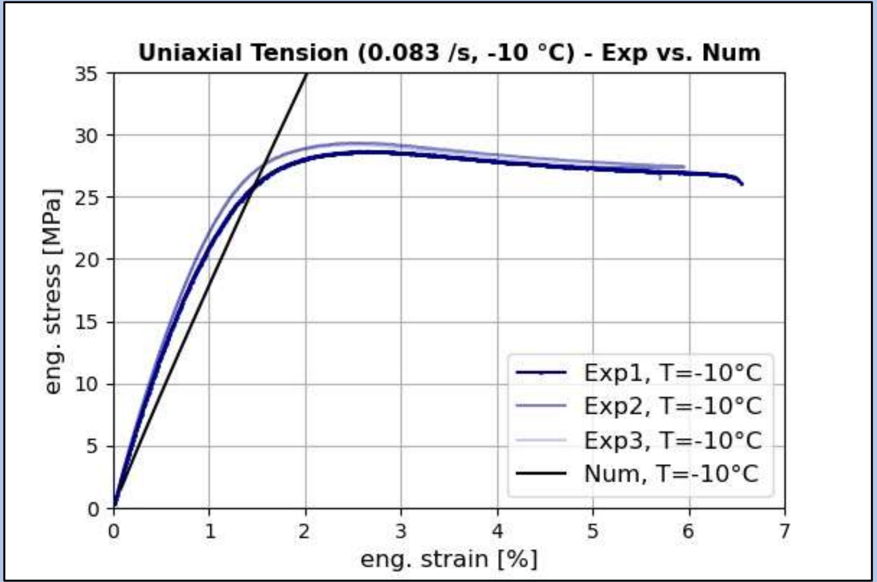
  Two unknowns: Γ and $\Delta\gamma$**!**

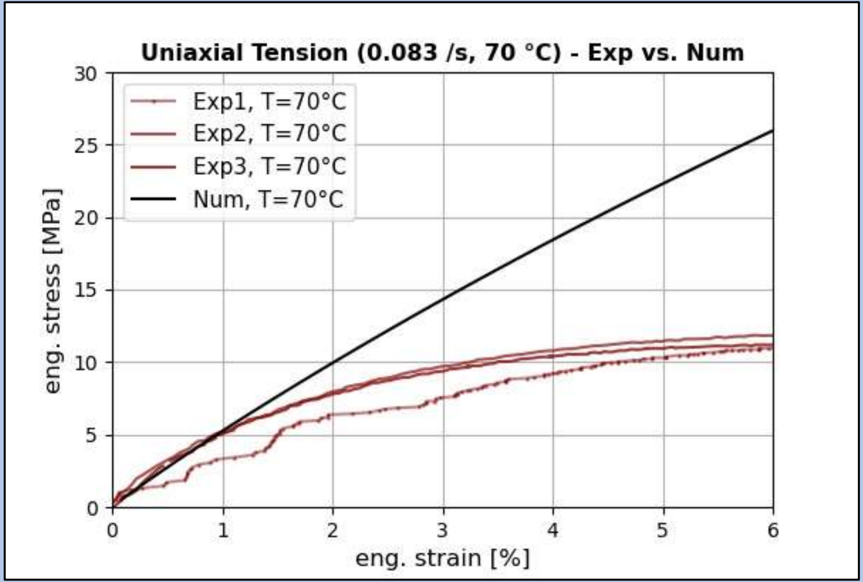
Temperature Dependency:

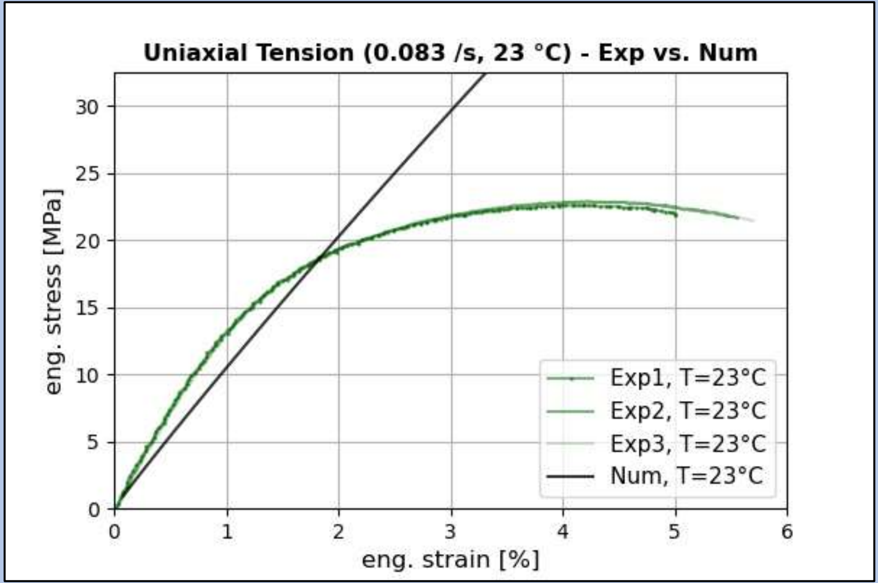
Yield strengths, kinematic hardening modulus ($H_\mathbf{B}$) and viscosity (η) are scaled with temperature dependent negative exponential functions like the WLF shift factor (a_T). For a parameter, g = g($\Delta\gamma$,T), [Krairi, 2019]

$$g(\gamma, T) = g(\gamma)\, a_g(T)$$

Appendix: TVE Uniaxial Numerical Results

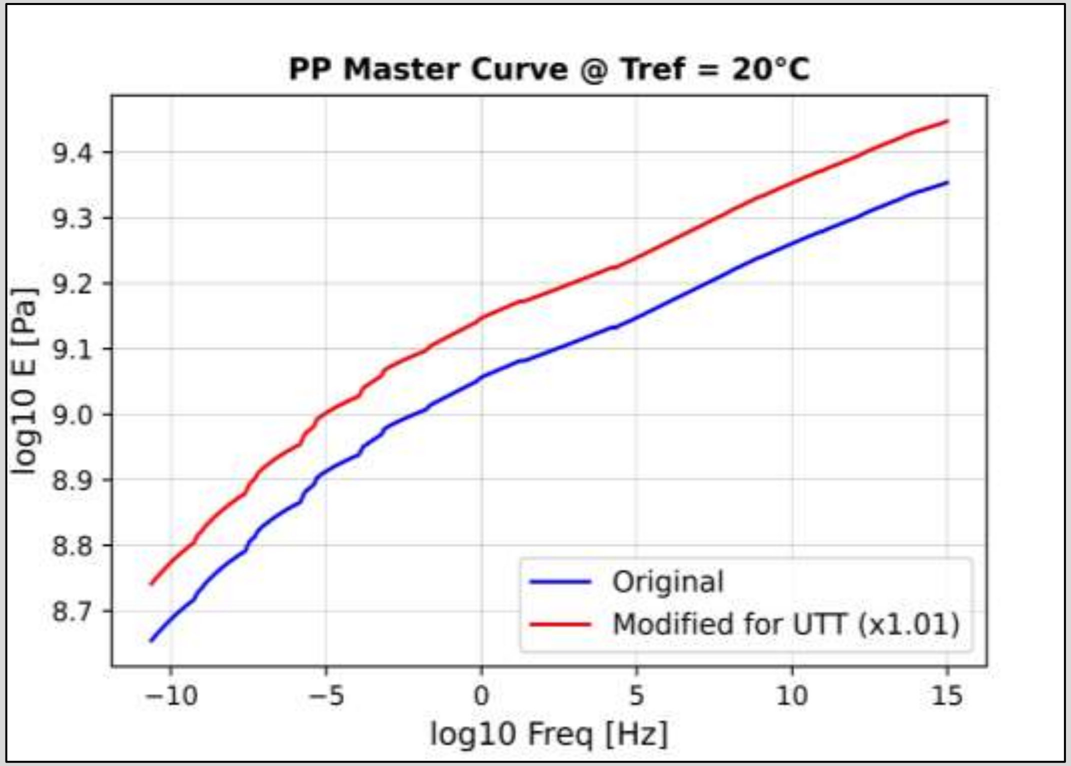




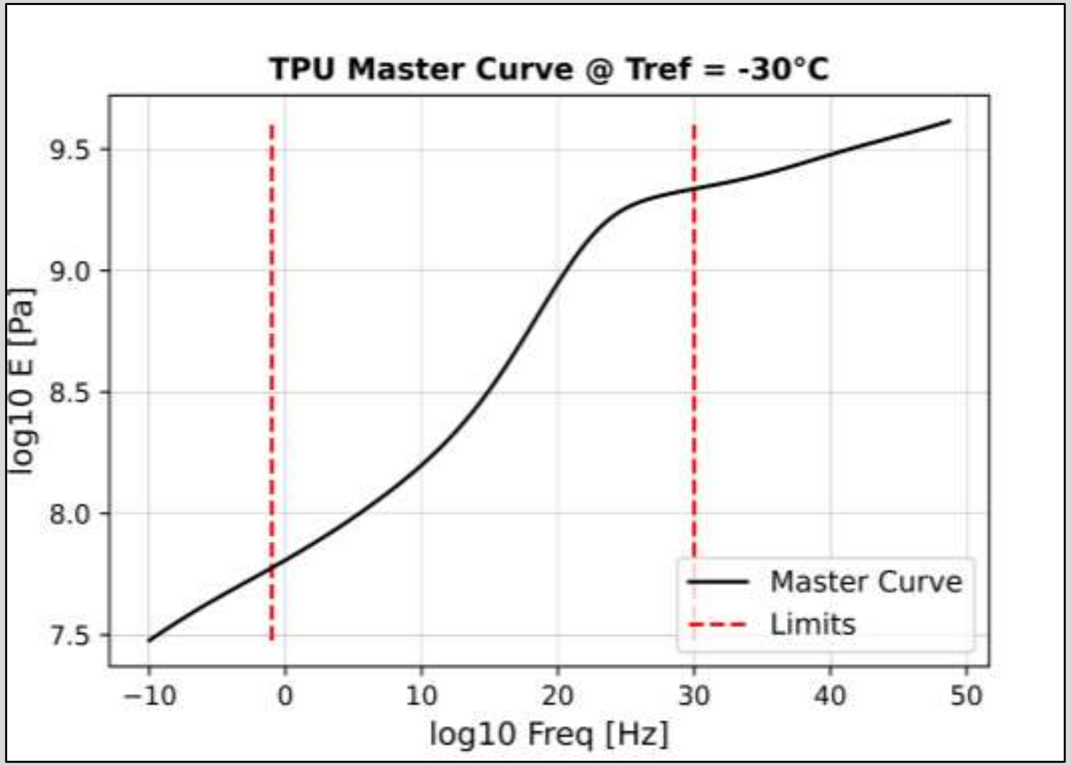


Tg assumed to be ~293 K. The relaxation spectrum from Tension DMA is found to be sufficient in predicting the tensile slope in the thermo(visco)elastic region using N = 27 terms.

Appendix: Master Curves

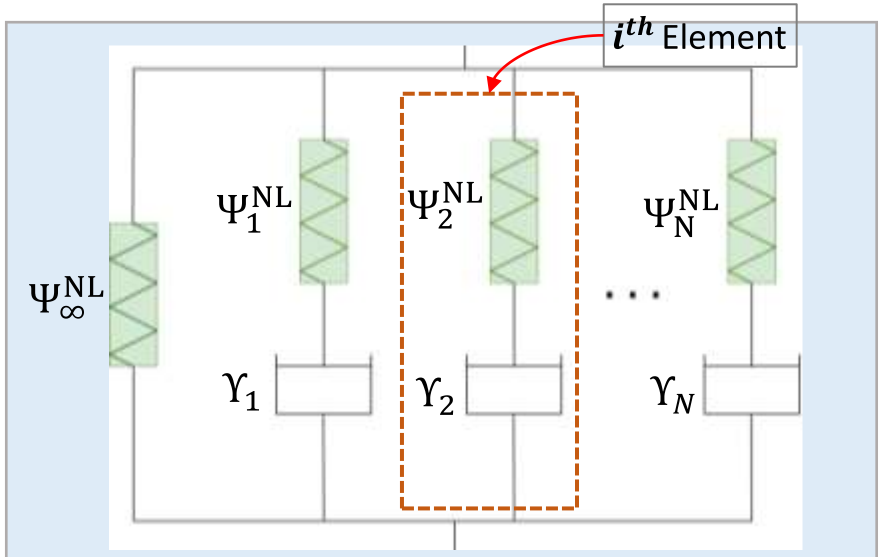


Vertically shifted master curve in -10 to 70˚C to match Uniaxial Tensile Test experiments



Master curve obtained for a wide range in -120 to 120˚C – limited terms for uniaxial tests at 23˚C.

Appendix: QNL TVE Abridged



Quasi-Nonlinear Maxwell Model

For the i^{th} Maxwell element, in Linear Viscoelasticity:

- Rate equations solved for i^{th} spring strain ($\mathfrak{E}_i$), in shifted time using time-temperature superposition (TTSP) , (deviatoric shown below)

$$\text{dev}\,\mathfrak{E}_i = \exp\left(-\frac{\Delta t^*|_{rec}}{g_i}\right)\text{dev}\,\mathfrak{E}_i^n + \exp\left(-\frac{\Delta t^*|_{mid}}{g_i}\right)(\text{dev}\,\mathbf{E}_e - \text{dev}\,\mathbf{E}_e^n)$$

- Relaxation in shifted time using WLF shift factor from DMA Master Curves.

$$\Delta t_n^* = \int_{t_n}^{t_{n+1}} \frac{1}{a_T(\epsilon)} d\epsilon \;\Rightarrow\; a_T = exp\left(-\frac{C_1(T - T_{ref})}{C_2 + T - T_{ref}}\right)$$

Quasi Non-linearity in stress:

- Form of Corotational Kirchhoff stress ($\hat{\boldsymbol{\tau}}_i$) is preserved,

$$\begin{aligned}\hat{p}_i &= K_{i0}\,\text{tr}\,\mathfrak{E}_i \\ \text{dev}\,\hat{\boldsymbol{\tau}}_i &= 2G_{i0}\,\text{dev}\,\mathfrak{E}_i\end{aligned} \;\Rightarrow\; \begin{aligned}\hat{p}_i &= K_{i0}\,A_{v_c}\,\text{tr}\,\mathfrak{E}_i \\ \text{dev}\,\hat{\boldsymbol{\tau}}_i &= 2G_{i0}\,B_{d_c}\,\text{dev}\,\mathfrak{E}_i\end{aligned}$$

induced non-linearity using strain-dependent scalars $\boldsymbol{A}_{v_c}$ and $\boldsymbol{B}_{d_c}$.

Here, $K_{\infty 0}, K_{i0}$ and $G_{\infty 0}, G_{i0}$ are the initial moduli from DMA Master Curves.

"Quasi-Nonlinear Viscoelasticity"

Linear viscoelasticity -> Newtonian Dashpots with Separable rate equations

+

Strain-dependent moduli -> Nonlinear functions in terms of log strains $\mathbf{E_e}$, $\mathfrak{E}_i$

Appendix: Strain-Dependent Functions in QNL TVE

General Form

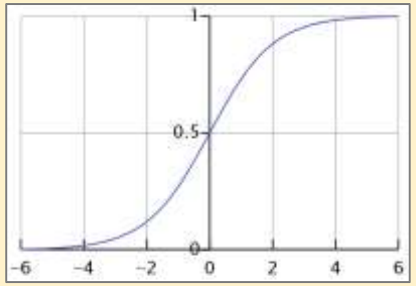
- Moduli expressed as $K_i = K_{i0} A_{vc}$ and $G_i = G_{i0} B_{dc}$
- Large tension-compression asymmetry in moduli at low strains without a slope discontinuity

$$A_{v_c} = f(\mathrm{tr}\,\mathfrak{E}_i)\, A_{v_i 1} + C_i \left(1 - f(\mathrm{tr}\,\mathfrak{E}_i)\right) A_{v_i 2}$$
$$B_{d_c} = f(\mathrm{tr}\,\mathfrak{E}_i)\, B_{d_i 1} + C_i \left(1 - f(\mathrm{tr}\,\mathfrak{E}_i)\right) B_{d_i 2}$$

Where, $\boldsymbol{A_{vi\,1}}, \boldsymbol{B_{di\,1}}$ activate in tension and $\boldsymbol{A_{vi\,2}}, \boldsymbol{B_{di\,2}}$ in compression; $\boldsymbol{C_i}$ is the compression scaler.

- $\boldsymbol{f}$ is the logistic function

$$f = \frac{1}{1 + \exp\left(-m\,\mathrm{tr}\,\mathfrak{E}_i\right)}$$



Where, ***m*** (>100) is a numerical regularisation parameter.

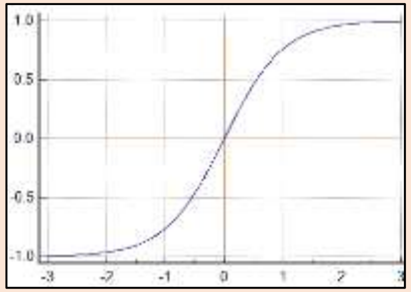
Explicit Form

- Strain-dependent scalars for every branch -> Scalable sigmoid shape of moduli

$$A_{v_i} = \frac{1}{\sqrt{V_1 \mathrm{tr}\,\mathfrak{E}_i^{\,2} + V_2}} + V_0 \left(1 + V_3 \tanh\left(\mathrm{tr}\,\mathfrak{E}_i^{\,2}\right)\right)$$
$$B_{d_i} = \frac{1}{\sqrt{D_1 \mathrm{dev}\,\mathfrak{E}_i : \mathrm{dev}\,\mathfrak{E}_i + D_2}} + D_0 \left(1 + D_3 \tanh\left(\mathrm{dev}\,\mathfrak{E}_i : \mathrm{dev}\,\mathfrak{E}_i\right)\right)$$

- The first terms are sigmoidal in “$A_{vi}\mathrm{tr}\,\mathfrak{E}_i$” and “$B_d \mathrm{dev}\,\mathfrak{E}_i$”

$$A_{v_i}\,\mathrm{tr}\,\mathfrak{E}_i = \frac{\mathrm{tr}\,\mathfrak{E}_i}{\sqrt{V_1 \mathrm{tr}\,\mathfrak{E}_i^{\,2} + V_2}}$$
$$B_{d_i}\,\mathrm{dev}\,\mathfrak{E}_i = \frac{\mathrm{dev}\,\mathfrak{E}_i}{\sqrt{D_1 \mathrm{dev}\,\mathfrak{E}_i : \mathrm{dev}\,\mathfrak{E}_i + D_2}}$$



and the second terms with tanh allow additional stiffness at higher strains.

- V_j and D_j are treated as material parameters ≥ 0.