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A new constitutive model describing the rate-, temperature- and strain-dependent behaviour of polymeric sealing materials

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The continuous advancement of virtual simulation platforms is a fundamental aspect of the ongoing global digitalization megatrend. To broaden the applicability of these platforms to sealing applications, there is a need for more comprehensive material models that can describe the complex and diverse behaviour exhibited by various polymeric materials used in sealing products, such as elastomers, thermoplastic polyurethanes (TPUs) and PTFE. To address this need, a new generalized modelling framework and approach is introduced in this paper. The proposed approach allows different aspects of the model to be modified, tailoring to the material being studied. The model demonstrates a remarkable potential to accurately describe the strain rate, temperature, and strain dependent behaviour of a chosen seal material, PTFE, within the relevant conditions experimentally covered.

1 Introduction

In recent years, virtual simulation platforms have become indispensable in the design and development of sealing solutions. Within these platforms, the usage of material models serve as foundational elements that accurately captures the seal's behaviour under various operating conditions. The challenge here lies in the diverse range of polymeric materials used in sealing products, displaying a spectrum of behaviours ranging from simple linear viscoelasticity to more complex and non-linear elastoviscoplasticity. Consequently, the stress levels experienced by these materials in application are significantly influenced by various factors such as deformation level, deformation rate, and operating temperature.

Over the past decades, numerous models have been proposed to predict the complex behaviour exhibited by different polymer materials, and the approach generally tailored to specific applications. These models often employ networks of springs and dashpots, such as Maxwell or Kelvin Voigt units, to establish constitutive equations that determine the material's stress response as a function of strain. However, there remains a need for a more generic and universal modelling framework, that allows for adapting the model to the material under study by selectively activating and deactivating its different components.

Therefore, this paper seeks to address the existing gap by proposing a new constitutive modelling approach, validated using Polytetrafluoroethylene (PTFE) as the material system. PTFE exhibits a complex, non-linear elastoviscoplastic behaviour compared to their rubber counterparts and hence, serves as an ideal candidate for demonstrating the complete capabilities of the proposed approach. Uniaxial tensile experiments are conducted on unfilled PTFE material at different strain rates and temperatures. The yield and post-yield kinetics are then carefully analysed to gain

insights into the underlying phenomenology and a suitable law is proposed to describe the observed behaviour. Finally, the output from the proposed model is compared against the experimental data to demonstrate its predictive capabilities.

2 Phenomenology and constitutive modelling of PTFE

Polytetrafluoroethylene (PTFE), also commonly known as Teflon, has some remarkable and unique properties that makes it suitable for sealing applications. Notably, PTFE has the ability to withstand extremely high and low temperatures, demonstrates excellent chemical resistance and a low coefficient of friction [1] resulting in enhanced wear resistance and reduced energy consumption compared to elastomers. Therefore, despite the ongoing PFAS discussions, PTFE is still being used in a wide array of applications.

Given its diverse usage, there exists a pressing need to understand its mechanical behaviour under anticipated operating conditions. However, being a semicrystalline thermoplastic polymer, PTFE exhibits a complex behaviour that is dependent on several factors including processing conditions, operating temperature, deformation rate, load and deformation levels, among others.

Figure 1 provides a schematic illustration of a typical stress-strain curve observed with PTFE materials when subjected to an uniform deformation rate. Initially, at very small strains, PTFE exhibits fairly linear viscoelastic response, which progressively transitions to a non-linear response until reaching a yield point. The Young's modulus and yield point are strongly dependent on the applied deformation rate, $\dot{\epsilon}$ and temperature, T . Post-yield, at large deformations, stress levels continue to increase exhibiting a strain hardening behaviour due to the orientation of polymer chains along the loading direction [2].

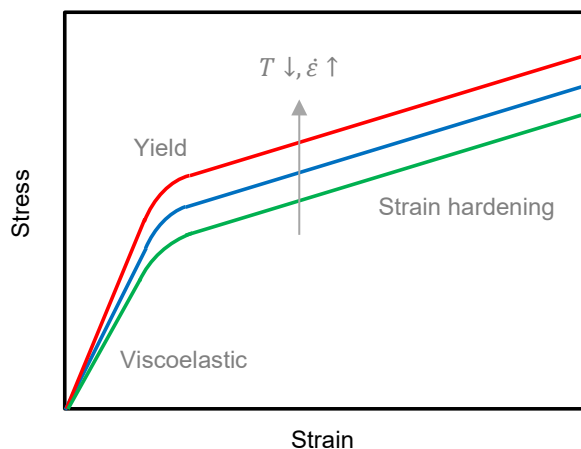


Figure 1: Schematic representation of a typical stress-strain response of PTFE

The new constitutive model proposed in this study is a phenomenological model, following the notable work of Haward and Thackray [3]. The model can account for the stress contributions arising from diverse interactions within polymer over the entire deformation range. Consequently, the total stress (σ) is decomposed into two components: a driving stress (σ_d) and a hardening stress (σ_h) contribution as depicted in equation (1).

$$\sigma = \sigma_d + \sigma_h \quad (1)$$

The hardening stress is typically characterized by the elastic response of the entanglement network at large deformations and can be represented by a linear elastic spring. The driving stress, on the other hand, is primarily governed by the intermolecular interactions and helps in characterizing the viscoelastic response of the polymer up to and including yield. This is typically represented by an elastic spring connected in series with a dashpot. The well renowned Eyring equation [4], which has been shown to effectively capture the rate and temperature dependent yield kinetics of several polymers [5-7], is used for representing the dashpot. The yield kinetics can then be expressed as shown in Equation (2).

$$\sigma_y = \frac{kT}{V^*} * \sinh^{-1} \left(\frac{\dot{\epsilon}}{\dot{\epsilon}_0} \exp \left(\frac{\Delta U}{RT} \right) \right) \quad (2)$$

where $\dot{\epsilon}_0$ is a thermodynamic constant, ΔU is the activation energy, V^* is the activation volume, k and R are Boltzmann and universal gas constant respectively. Furthermore, in order to accurately represent the behaviour of polymers across a wide range of temperatures and strain rates, the use of just a single spring-dashpot network is found to be rather insufficient. Hence, it is a common practice to include multiple branches in parallel, which is representative of a spectrum of relaxation times that is associated with the polymer.

Additionally, it is known that certain polymers also exhibit a thermorheologically complex behaviour. As a result, the driving stress contribution could potentially arise from more than one molecular process. In case of a semicrystalline thermoplastic polymer like PTFE, driving stress originates from the mobility of the main chain of the amorphous phase (α -relaxation) as well as transitions associated with the crystalline phase (β -transition) [8].

From a modelling perspective, this is easily addressed by adding multiple spring-dashpot networks in parallel, each representative of the relevant molecular process under consideration. The yield kinetics are then described using the Ree-Eyring modification to the Eyring equation [9], as shown in equation (3), where the different molecular processes are considered to act simultaneously and their stress contributions being additive in nature.

$$\sigma_y = \sum_{i=1}^n \frac{kT}{V_i^*} \sinh^{-1} \left(\frac{\dot{\epsilon}}{\dot{\epsilon}_{0,i}} \exp \left(\frac{\Delta U_i}{RT} \right) \right) \quad (3)$$

Incorporating multiple relaxation times and accounting for contributions from molecular processes having different origins are essential for effectively capturing the different transitions occurring within the material, in the range of conditions that is relevant to real-world applications.

Additionally, in the subsequent section (refer Figure 8 (b)), it will be demonstrated that the mere implementation of the conventional Ree-Eyring equation for the dashpot is rather insufficient to fully describe the complex behaviour exhibited by PTFE. In addition to the strain rate and temperature dependence, PTFE also exhibits a strong dependency on the strain level. Therefore, to address this, a new strain modified Eyring equation is proposed in this study, as expressed in equation (4).

$$\dot{\varepsilon} = \dot{\varepsilon}_0(\varepsilon) * \sinh\left(\frac{qV^*(\varepsilon)}{kT}\right) \exp\left(\frac{\Delta U(\varepsilon)}{R} \left(\frac{1}{T} - \frac{1}{T_{\text{ref}}}\right)\right) \quad (4)$$

$$V^*(\varepsilon) = f(V_{\text{initial}}^*, V_{\text{final}}^*, \varepsilon, \varepsilon_0) \quad (5)$$

$$\Delta U(\varepsilon) = f(\Delta U_{\text{initial}}, \Delta U_{\text{final}}, \varepsilon, \varepsilon_0) \quad (6)$$

$$\dot{\varepsilon}_0(\varepsilon) = f(\dot{\varepsilon}_{0,\text{initial}}, \dot{\varepsilon}_{0,\text{final}}, \varepsilon, \varepsilon_0) \quad (7)$$

Where ε is the strain accumulated in the material. $\dot{\varepsilon}_0(\varepsilon)$, $V^*(\varepsilon)$ and $\Delta U(\varepsilon)$ are the strain modified rate constant, activation volume and activation energy parameters. These parameters are defined as a function of the strain level and ε_0 , a strain based numerical constant, in such manner that they change from an initial to final value as indicated in equations (5-7).

Considering all essential aspects discussed above to describe the behaviour of PTFE, the mechanical analogue of the proposed model is constructed and illustrated in Figure 2. The model incorporates stress contributions from both α and β processes, connected in parallel. Each process has multiple branches that corresponds to the multiple relaxation times associated with them. The dashpot is modelled using the strain-modified Eyring equation, resulting in a fully non-linear elastoviscoplastic model. The final equilibrium branch, represented by a standalone elastic spring, captures the elastic strain hardening component of PTFE.

It is worth noting that, having such a modular and versatile modelling approach helps in selectively choosing the level of complexity based on the material under consideration. For instance, when applying the model to a material exhibiting predominantly linear viscoelastic behaviour, the approach can be simplified by deactivating the strain dependency in the Eyring dashpot or even using a linear Newtonian dashpot if proven sufficient. Furthermore, if the material demonstrates a thermorheologically simple response across temperatures and strain rates, a single-process version of the model may suffice. Therefore, this approach presents a universal sealing material modelling framework that can be tailored to any polymeric sealing product and application.

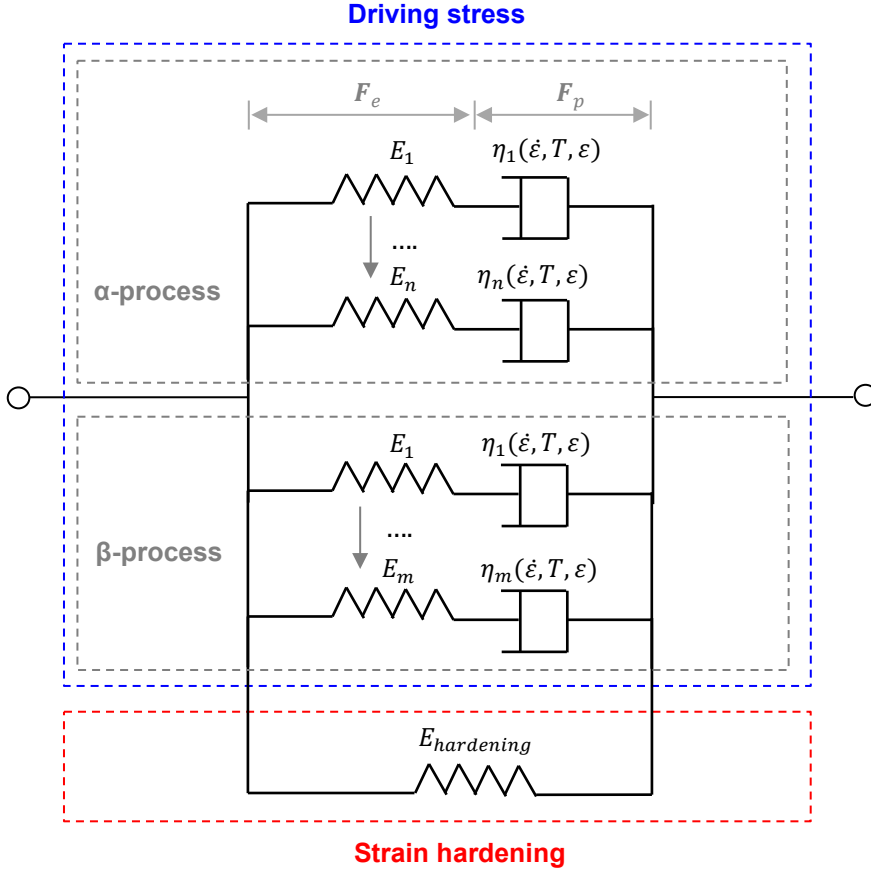


Figure 2: Mechanical analogue of a two process model for PTFE, with each process having multiple relaxation times. The modified Eyring dashpot takes into consideration the influence of strain rate, temperature as well as the strain levels.

2.1 Implementation using ABAQUS Parallel Rheological Framework

In order to implement the proposed model for PTFE, the parallel rheological framework (PRF) of ABAQUS, a finite element software package, is utilized. The complete description and formulation of PRF can be found in the work of Dalrymple *et al.* [10]. Though the constitutive equations can be found there, it is important to emphasize that, for a finite deformation, the deformation gradient ($\mathbf{F}_i$) in each branch is multiplicatively decomposed into an elastic ($\mathbf{F}_{e,i}$) and plastic part ($\mathbf{F}_{p,i}$) as depicted in equation (7).

$$\mathbf{F}_i = \mathbf{F}_{e,i} \cdot \mathbf{F}_{p,i} \quad (8)$$

Meanwhile, the total stresses are determined as a summation of the stress contributions from each individual branch. Furthermore, PRF only allows the use of a single hyperelastic model for all the springs used in the model. Therefore, In order to get a simple linear elastic response, the neo-Hookean hyperelastic model available in ABAQUS is utilized. On the other hand, although PRF has several built-in functions for describing the viscosity evolution in the dashpot, they are not representative of the strain modified Eyring equation that is necessary to describe the behaviour of PTFE. Therefore, the subroutine UCREEPNETWORK of the PRF is utilized to implement equation (4). In the following section, the experimental methodology adopted for testing the unfilled PTFE material is discussed, followed by the experimental results and a comparison of the model output against it in order to showcase its capabilities.

3 Experimental method

The material used for model development and validation as mentioned above is virgin PTFE. Experiments are conducted using a universal testing machine equipped with a 0.5 kN load cell, employing uniaxial tension. The testing procedure consists of two phases. In the initial *loading* phase, samples undergo loading at a constant crosshead displacement rate until reaching a predefined strain level. Subsequently, in the *relaxation* phase, the sample is maintained at the predefined strain level for one hour, during which the decay of stress over time is recorded. This testing protocol is designed to replicate real-world scenarios (Figure 3), such as when a seal is assembled on to a shaft (loading phase) and subjected to operational conditions where radial forces gradually diminish over time post-installation (transient relaxation phase).

During the loading phase, strain rates of 0.1 s^{-1} and 0.02 s^{-1} controlled by the crosshead displacement are chosen to understand the strain rate sensitivity of PTFE. In order to analyse the influence of temperature, tests are conducted at room temperature, 50°C , 80°C and 120°C . In the relaxation phase, the influence of strain on the relaxation rate is analysed by stopping the loading phase at three different strain levels, at each strain rate and temperature tested. The overall test matrix listing all the test conditions are presented in Table 1. Two samples are tested per test condition to ensure repeatability of the test results presented.

Table 1: Test matrix with Virgin PTFE

Temperature [$^\circ\text{C}$]	Strain rate [s^{-1}]	Strain level [%]
23	0.1	5, 16, 25
	0.02	5, 16, 26
50	0.1	6, 16, 28
	0.02	6, 16, 28
80	0.1	6, 18, 28
	0.02	6, 17, 28
120	0.1	5, 17, 28
	0.02	6, 17, 28

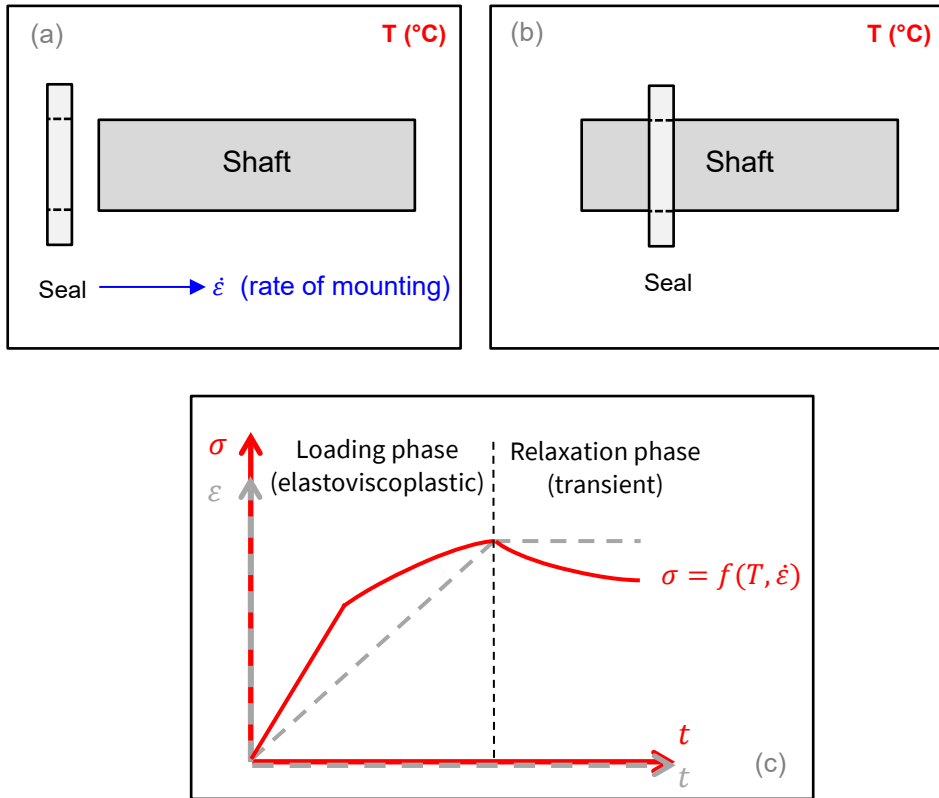


Figure 3: (a) Loading phase: Seal is assembled on to a shaft inducing strain in the material. (b) Relaxation phase: After the seal is assembled, the strain in the material is fixed while the stresses that developed during the loading phase decays over time. (c) Schematic representation of the stress and strain as a function of time during both loading and relaxation phase.

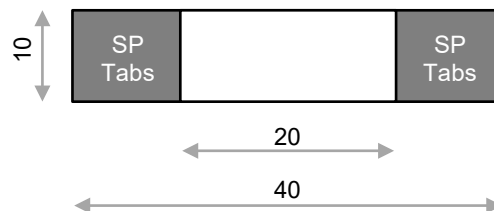


Figure 4: Schematic representation of tensile test samples. All dimensions in mm.

Tensile test samples were prepared in a stripe geometry, as depicted in Figure 4, with dimensions of 40 mm * 10 mm * 2 mm. A gauge length of 20 mm and a gripping length of 10 mm on either side of the gauge section were employed. To prevent slipping during testing, sandpaper (SP) tabs were used in the gripping region. Additionally, a clamping pressure of 7 bar was used with the help of pneumatic clamps while performing the experiments.

Digital Image Correlation (DIC) technique was employed for a precise measurement of the engineering strain values. Spray pattern was applied on the specimen in the gauge section. Considering the homogeneous deformation state of the specimen, the line strains of the middle part of the specimen is considered as the region of interest, with a gauge length of 10 mm. After the completion of an experiment, the engineering strains are calculated, which are then converted to true strain and true stress following the expression in equation (9).

$$\varepsilon_t = \ln(1 + \varepsilon) ; \sigma_t = \sigma(1 + \varepsilon) \quad (9)$$

where ε_t is the true strain, σ_t is the true stress, ε is the engineering strain and σ is the engineering stress. Furthermore, since the strains are calculated after the experiment, it is important to note that reaching the same precise true strain as intended is not feasible with each experiment.

4 Results and discussion

4.1 Uniaxial tensile test results

In general, the experimental methodology adopted in this study produces highly repeatable results and is demonstrated in Figure 12 for some of the results in Appendix 1. Therefore, in order to ensure clarity in visualizing the data in the following sections, only one set of data is presented in the figures.

In Figure 5 (a), the quasi-static data obtained during the loading phase illustrates the material's response to different strain rates and temperatures. It becomes evident that stress levels within the material are strongly influenced by these factors due to the time dependent behaviour exhibited by PTFE, with higher strain rates or lower temperatures resulting in increased stresses for a given strain level, and vice versa.

However, identification of a distinct yield point proves challenging from the true stress strain curves presented in Figure 5 (a). Instead, the yield point is determined by considering the intersection between the initial slope within the viscoelastic region and the hardening slope observed at larger strains. Subsequently, the yield kinetics of PTFE is depicted in Figure 5 (b), where the experimental markers are fit using Equation (3) and the parameters listed in Table 2. The data reveals two distinct regions: Region I having a higher rate dependence at lower temperatures, and Region II with a lower rate dependence at higher temperatures. As discussed in the previous section, this observation is attributed to the contributions of multiple molecular processes within PTFE, stemming from its amorphous (α -process) and crystalline (β -process) phases.

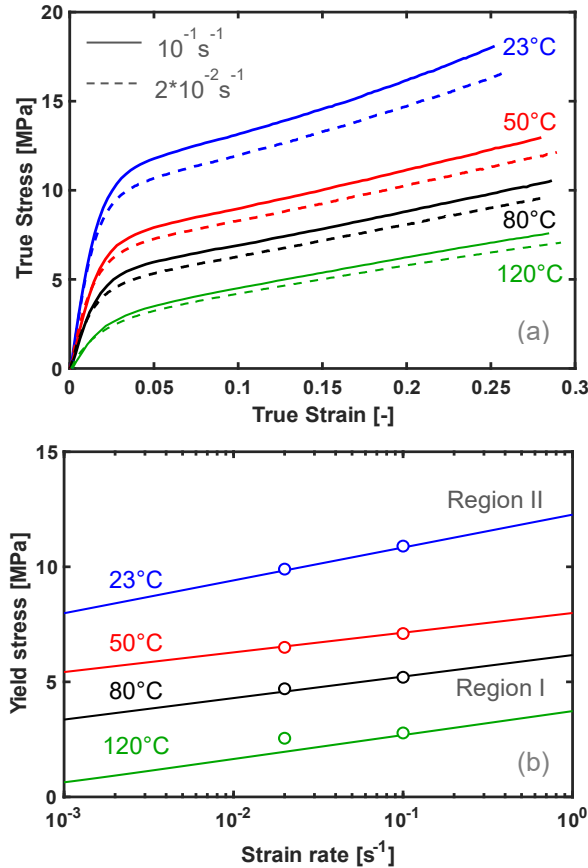


Figure 5: (a) True stress-strain plots at four different temperatures and two strain rates. (b) Yield stress dependence on strain rate and temperature visualized on a semi-log scale. Solid lines are fit using Equation (3) and parameters listed in Table 2.

Table 2: Eyring parameters needed to fit the yield kinetics of PTFE

Process	V^* [nm ³]	$\dot{\epsilon}_0$ [s ⁻¹]	ΔU [KJmol ⁻¹]
α -process	12	$2 \cdot 10^{23}$	205
β -process	14.7	$6 \cdot 10^{164}$	950

Dynamic Mechanical Analysis (DMA) data obtained internally (Figure 6), along with other literature findings [8], provide substantial support for this observation. The β -transition associated with the crystalline phase occurs between 20°C and 30°C, and the α -relaxation related to the amorphous phase, occurs at 120°C. As a result, in Region I, only the α -process is active, whereas in Region II, both α and β processes

are active, resulting in increased rate dependence. Consequently, at 120°C, where α -relaxation occurs, the strain rate dependence becomes almost negligible, and therefore depicts a deviation from the Ree-Eyring description as observed in Figure 5 (b).

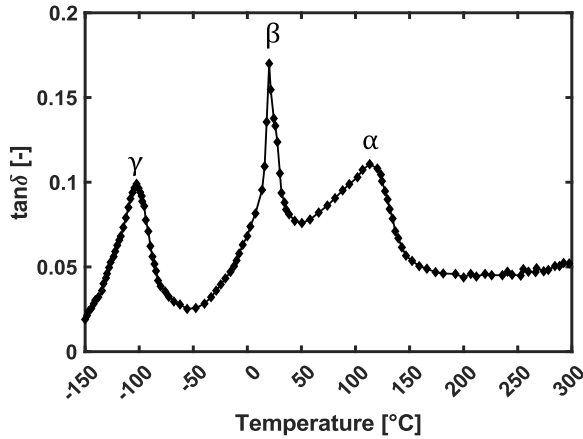


Figure 6: Tan delta versus temperature plot from DMA analysis showing the different relaxation processes occurring in PTFE.

Moreover, the quasi-static data presented in Figure 5 (a) indicates a strain hardening response that is not purely elastic, but rather dependent on both the loading rate and temperature. Specifically, the strain hardening component appears to be more pronounced at lower temperatures and higher strain rates. This observation has been shown for other polymers such as polycarbonate [11] and may be attributed to an additional viscous contribution arising from the entanglement network within the material.

Figure 7 illustrates the data in the subsequent stress relaxation phase¹, for a loading rate of 0.02 s⁻¹. The stresses in Figure 7 are normalized to highlight the differences in the relaxation behaviour as the strain level and temperature varies in the experiment. The data clearly suggests a strong dependence of the relaxation behaviour on the predefined strain level at which the relaxation phase begins and the testing temperature. This observation further highlights the need for the implementing the full non-linear elastoviscoplastic model proposed in this study. The following section will detail on the procedure adopted to obtain all the necessary parameters required for the model.

¹ Please note that the data for a loading rate of 0.1 s⁻¹ is presented in Appendix 2, Figure 13

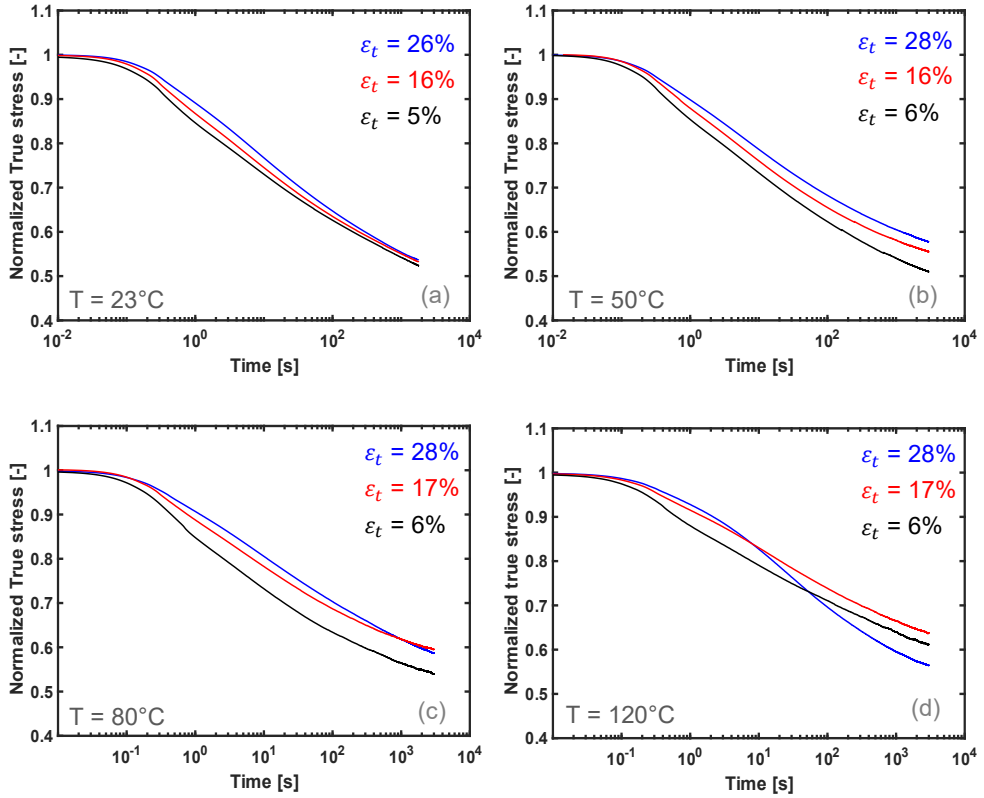


Figure 7: Stress relaxation behaviour of PTFE at three different strain levels under a strain rate of $2 \cdot 10^{-2} \text{ s}^{-1}$ and four different temperatures.

4.2 Parameter determination for the PTFE model

As briefly described in section 2.1, the PRF framework of ABAQUS is utilized to implement the model. Since the quasi-static data suggests the contribution of two molecular processes towards the yield kinetics of PTFE, a two-process model as depicted in Figure 2 is used. The initial Eyring parameters for both the processes are taken from Table 2.

The methodology presented in the work of Kadin and Schaake [12] is utilized to determine the C10 modulus of the neo-Hookean hyperelastic spring and the initial Eyring parameters of the dashpot in each branch of α -process. The true stress-strain curve at 50°C and a strain rate of 0.1 s^{-1} is used for this purpose, as at this condition only the α -process is active (refer DMA data presented in Figure 6). To achieve an accurate fit of PTFE's initial non-linear viscoelastic response up to the yield point, 14 branches for the α -process are required, as shown in Figure 8(a). It has to be noted that the strain hardening parameter is not yet included in the model and hence, once the yield point is reached, there is no additional stress contribution from the model.

The final Eyring parameters of α -process are determined by fitting the post-yield data at 50°C and 80°C under a strain rate of 0.1 s⁻¹ and the model fit is presented in Figure 8 (b) (solid lines). The figure also shows the output from the standard Eyring model (dash-dotted lines) without strain dependency. While the standard Eyring model matches the data well up to the yield point, they deviate significantly at higher strain levels. In contrast, the new model, which includes strain dependency, aligns closely with the experimental data. This observation once again reiterates the importance of implementing the strain dependent Eyring equation proposed in this study to accurately describe the mechanical behaviour exhibited by PTFE. It is worth noting that in these simulations, the strain hardening parameter was included to showcase the differences highlighted.

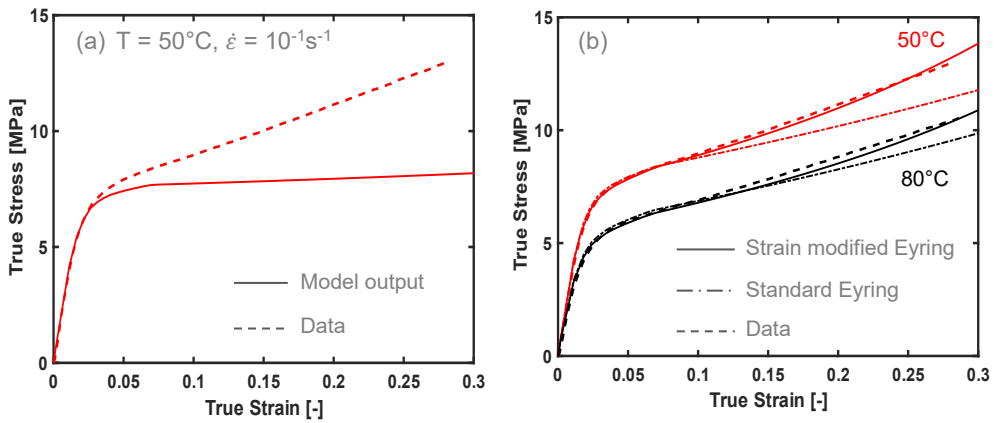


Figure 8: (a) Model fit (solid line) to tensile data obtained at 50°C under a strain rate of 0.1 s⁻¹, to determine the initial Eyring parameters for all the branches of α -process using the methodology proposed in [12]. (b) Model output compared against the experimental data. Dashed-dotted lines are output from the standard Eyring model, whereas solid lines are output from the strain modified Eyring model proposed in this study. Dashed line in both figures represent experimental data.

Subsequently, the initial and final Eyring parameters for the β -process is obtained through the best fit of the data at room temperature under a strain rate of 0.1 s⁻¹. This is illustrated in Figure 9, where the model fit is represented by blue solid line. The figure also highlights the importance of having a two process model for PTFE by displaying the model output (grey solid line) when only the contribution from the α -process is considered. Clearly, in this case, there is a substantial difference in the modulus and the stress levels predicted by the model when compared to the experimental data. Therefore, the decision to include multiple processes, supported by the DMA data, is also verified through the simulation results. It is worth noting that, since the β -process is active only at room temperature among the tested temperatures, a single branch is sufficient to achieve a good fit, as observed in Figure 9.

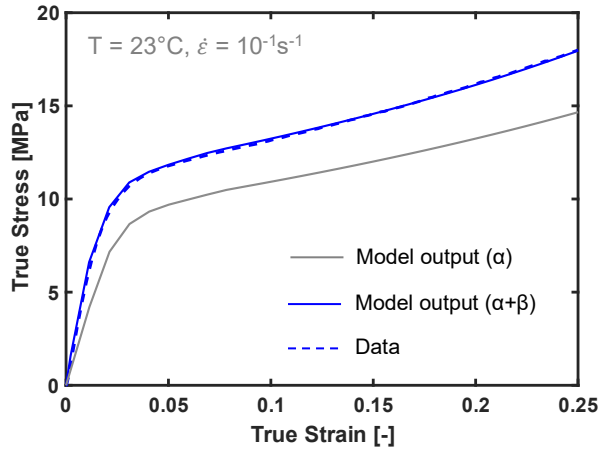


Figure 9: (a) Model fit to tensile data obtained at 23°C under a strain rate of 0.1 s^{-1} , to determine the initial and final Eyring parameters for β -process. Blue solid line is the model output when both α and β processes are included in the model. Grey solid line is the model output when only α process is included in the model.

Furthermore, based on the best fit of the data presented in both Figure 8 (b) and Figure 9, it is found that it is necessary to implement the strain dependency for the activation volume of both the processes. The activation energy parameter is made strain dependent for the α -process, whereas, it is strain independent for the β -process. The thermodynamic constant, $\dot{\epsilon}_0$, is kept constant for both the processes. Importantly, it should also be noted that all the model parameters are determined based on the *short-term* quasi-static data obtained during the loading phase. Therefore, the model output for the *long-term* stress relaxation behaviour in the subsequent phase can be considered to be predictive.

4.3 Model predictions

After acquiring all the necessary parameters as discussed in the preceding section, simulations are run in ABAQUS using a single element, 2-D axisymmetric model, considering the isotropic nature of virgin PTFE, and replicating the conditions of uniaxial tensile testing outlined in section 3.

Figure 10 illustrates the comparison between the output from these simulations, represented by solid lines, and the experimental data obtained during the loading phase, represented by dashed lines. Given that the model parameters are derived through fitting data at temperatures of 23°C, 50°C and 80°C under a strain rate of 0.1 s^{-1} , it is anticipated that the model would exhibit good agreement with these data. However, it is quite remarkable to observe that the predictions at other conditions also closely align with the data. The strain rate and temperature dependencies are aptly captured, following the Eyring equation. Importantly, the model demonstrates a remarkable potential in capturing the strain rate- and temperature-dependent hardening behaviour, due to the addition of strain dependency in the Eyring equation.

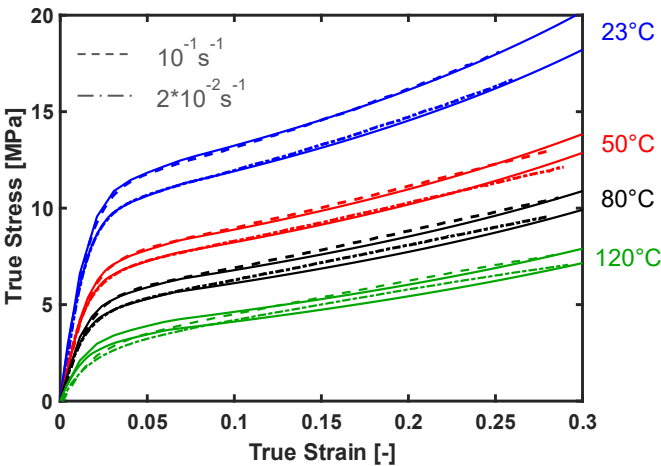


Figure 10: Model predictions, represented by solid lines, compared against the experimental data obtained at four different temperatures and two strain rates during the loading phase.

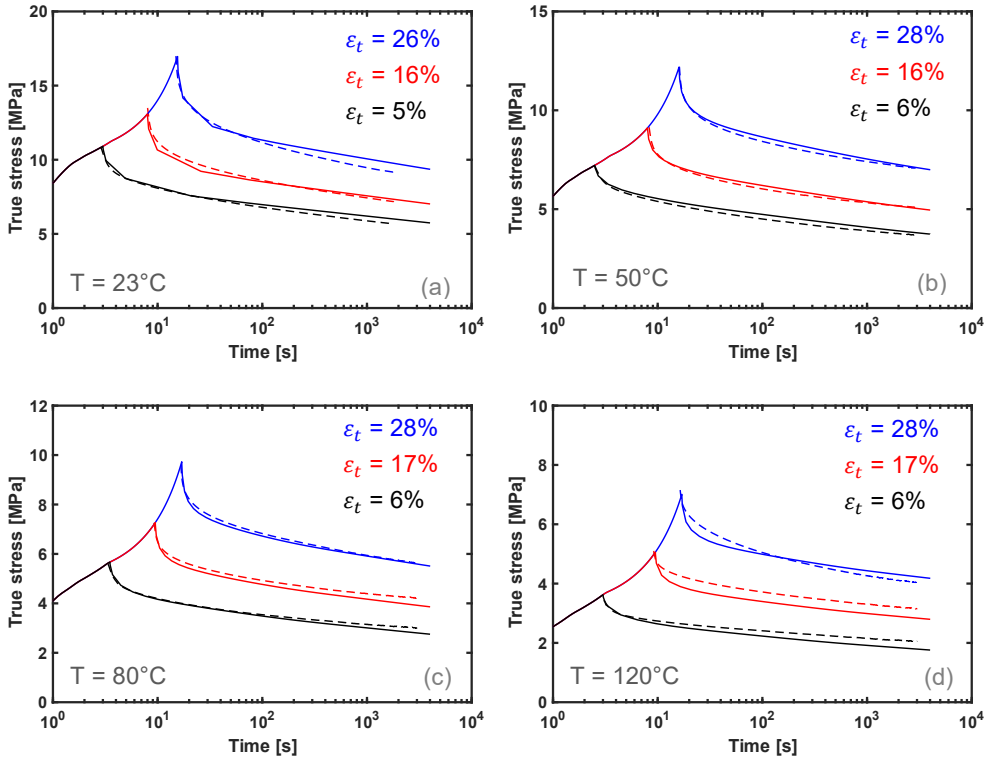


Figure 11: Stress relaxation behaviour of virgin PTFE under a strain rate of 0.02 s^{-1} . Solid lines represent model predictions and dashed lines are experimental data in all figures.

Following this, the model's capability to predict the stress relaxation behaviour is assessed. Figure 11 reveals an excellent agreement between the data and model predictions across all strain levels and temperatures tested ², under a strain rate of 0.02 s^{-1} . Similar to the observation in the loading phase, the figures strongly suggest that the strain rate, temperature and strain dependency exhibited by PTFE in the relaxation phase are also captured remarkably well. Additionally, the model is able to capture the transient relaxation response based only on quasi-static short-term stress-strain data obtained at different temperatures and strain rates during the loading phase. This clearly depicts the consistency of the model in terms of strain rate, temperature and strain level influence across different solicitations and response mechanisms (loading versus relaxation).

5 Conclusion

A novel modelling approach is proposed in this study which is found to be very effective in describing the non-linear elastoviscoplastic behaviour exhibited by virgin PTFE under uniaxial tensile loading conditions. By incorporating the newly proposed strain-modified Eyring equation, the model accurately predicts both quasi-static and transient relaxation phenomena, while taking into account the influence of strain rate, temperature and strain levels. Consequently, the model also holds the potential to predict seal behaviour under anticipated operating conditions.

Furthermore, given the versatility of the proposed approach, it is possible to tailor it to other polymeric materials such as elastomers and thermoplastic polyurethanes (TPUs), thereby enabling a universal modelling framework for materials used in sealing applications.

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² Comparison between data and model predictions for a strain rate of 0.1 s^{-1} is presented in Figure 13 under Appendix 2

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9 Additional data and simulation output

Appendix 1: Repeatability of the uniaxial tensile test data

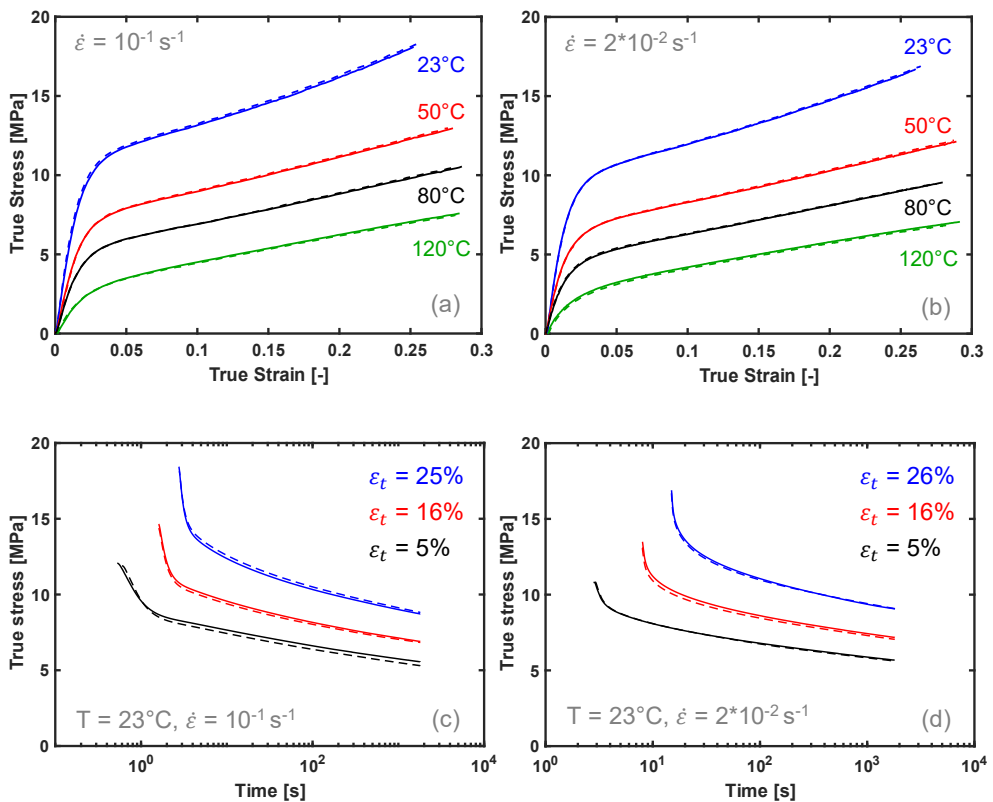


Figure 12: Quasi-static and stress relaxation data for Virgin PTFE. For stress relaxation, only the data at room temperature is presented as an example. Dashed lines indicate test that is repeated. The data at each test condition lies exactly on top of each other, indicating excellent repeatability of the methodology adopted.

Appendix 2: Stress relaxation behaviour of virgin PTFE obtained at a strain rate of 0.1 s⁻¹ and compared against model predictions

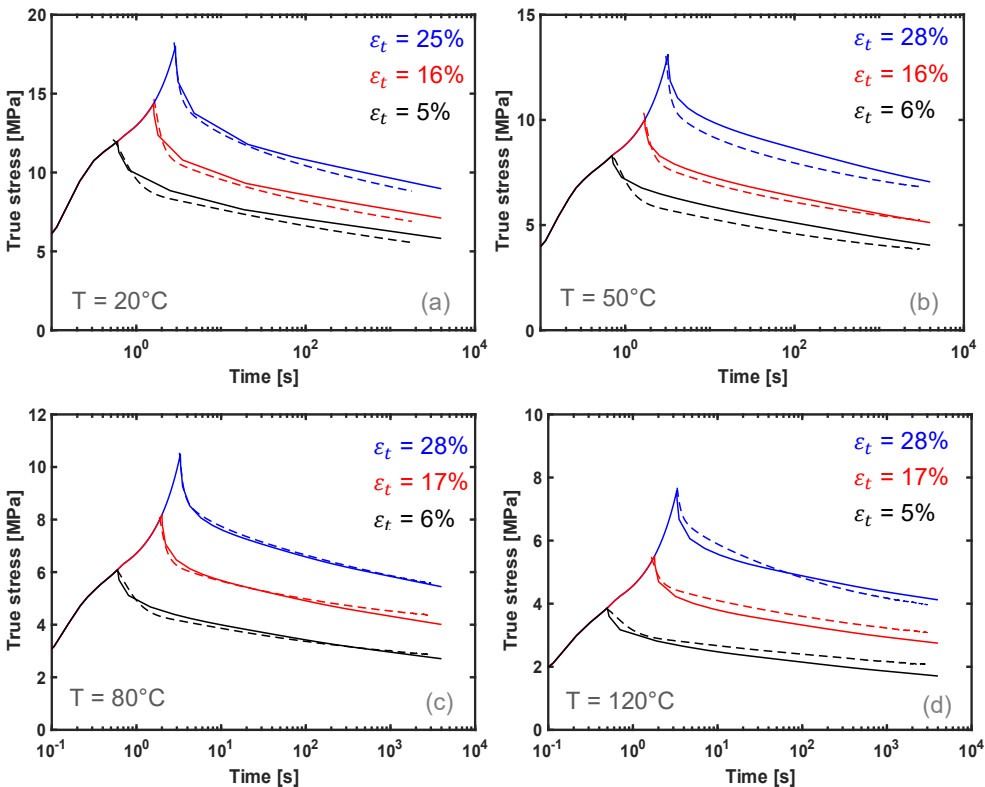


Figure 13: Stress relaxation behaviour of virgin PTFE at three different strain levels at a strain rate of 0.1 s⁻¹ and four different temperatures, namely 23°C, 50°C, 80°C and 120°C. Solid lines represent model predictions and dashed lines are experimental data.

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