Thermo-mechanically coupled modelling of a single-asperity scratch on an isotropic isotactic polypropylene surface

Citation for published version (APA):

Document license:
CC BY-NC-ND

DOI:
10.1016/j.triboint.2019.105946

Document status and date:
Published: 01/01/2020

Document Version:
Publisher’s PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:

• A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
• The final author version and the galley proof are versions of the publication after peer review.
• The final published version features the final layout of the paper including the volume, issue and page numbers.

Link to publication

General rights
Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

• Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
• You may not further distribute the material or use it for any profit-making activity or commercial gain
• You may freely distribute the URL identifying the publication in the public portal.

If the publication is distributed under the terms of Article 25fa of the Dutch Copyright Act, indicated by the “Taverne” license above, please follow below link for the End User Agreement:
www.tue.nl/taverne

Take down policy
If you believe that this document breaches copyright please contact us at:
openaccess@tue.nl
providing details and we will investigate your claim.

Download date: 15. Jan. 2020
Thermo-mechanically coupled modelling of a single-asperity scratch on an isotropic isotactic polypropylene surface

Tarek Kershah\textsuperscript{a,b}, Stan F.S.P. Looijmans\textsuperscript{a,b}, Patrick D. Anderson\textsuperscript{a}, Lambert C.A. van Breemen\textsuperscript{a,*}

\textsuperscript{a} Polymer Technology, Department of Mechanical Engineering, Materials Technology Institute, Eindhoven University of Technology, P.O. Box 513, 5600 MB, Eindhoven, the Netherlands
\textsuperscript{b} Dutch Polymer Institute (DPI), P.O. Box 902, 5600 AX, Eindhoven, the Netherlands

Keywords:
Contact mechanics
Single-asperity sliding friction
Finite element modelling
Thermomechanical analysis

\textbf{A B S T R A C T}

Isotactic polypropylene (iPP) is a low cost semi-crystalline polymer that is easy to process, has a wide variety in properties and is, therefore, used in many applications. Many of these applications require enhanced wear-resistance to prolong the lifetime of the product. Essential is to first investigate the intrinsic response of the material in order to describe its friction and wear response. In this respect, a hybrid experimental-numerical approach is used to couple the intrinsic response to the single-asperity scratch response. The numerical model used is a 3D elasto-viscoplastic model based on the Eindhoven Glassy Polymer (EGP) model. For the first time a coupled thermo-mechanical EGP model is implemented in a Finite Element Method (FEM)-framework. The model is capable of accurately describing the intrinsic response of the material, which opens the door to qualitatively and quantitatively describe its frictional response and understand the damage formation mechanism (i.e. the initiation of wear). In this study, \(\alpha\)- and \(\beta\)-phase iPP are studied. We show that the difference in the intrinsic response between the two phases has a significant influence on the friction and wear response. Moreover, a stick-slip phenomenon is proven to be the main responsible for the damage mechanism observed. The observed periodic “fish-scale” damage pattern results from periodic changes in resistance during the tip movement. A relation between the polymer intrinsic response and the damage formation mechanism is established. The influence of the applied load and scratch speed on damage formation is investigated as well.

1. Introduction

Nowadays, semi-crystalline polymers are used in many applications where moving parts are in contact. The reason is that semi-crystalline polymers possess excellent physical properties such as a light weight and a high wear resistance. Polymers are favoured above their metal counterparts in many applications such as artificial joints, gears, and bearings.

The background of this study is the methodology developed in our group over last two decades where numerical simulations are combined with experiments in order to link the intrinsic behaviour of the polymer to its frictional response. The constitutive model framework used is the Eindhoven Glassy Polymer (EGP) model \cite{Boyce1997, Parks1997, Argon1999, Krop2017, Anderson2018}, which, like the Boyce-Parks-Argon model \cite{Boyce1997, Parks1997} and the Oxford Glass-Rubber (OGR) model \cite{Wilson1999, Yip2004, Uo2010, Krop2017} accurately describes the deformation kinetics of glassy polymers. The constitutive model was extended to better capture the non-linear visco-elastic response (multi-mode) \cite{VanBreemen2014, Krop2017} and the thermorheological complex systems (multi-process) \cite{VanBreemen2014}. Recently, Van Breemen et al. \cite{Breemen2018} and Krop et al. \cite{Krop2017} used the EGP model to couple intrinsic material properties to the observed frictional response on both unfilled and particle-filled polycarbonate. In practice however, for example in bearing cages and medical implants such as knee or hip replacements, semi-crystalline polymers are used \cite{Bennett2003, Tarek2014, Kershah2019, Breemen2018, Krop2017}. Generally speaking, semi-crystalline polymers exhibit high strength, better wear and chemical resistance than glassy polymers.

Semi-crystalline polymers have a large variety of morphological structures. Therefore, contrary to glassy polymers, semi-crystalline systems display a large variation in mechanical properties and lifetime depending on for instance the cooling rate, applied pressure, and presence of anisotropy \cite{Kershah2019}. We chose isotactic polypropylene (iPP) as a model material since it is one of the widely used semi-crystalline polymers, in addition to its well-defined mechanical properties \cite{Tijssen2008, Kershah2019, Breemen2018}.

In this study we aim to link the intrinsic material behaviour to the observed friction and wear response, and obtain a comprehensive...
understanding of the influence of intrinsic response on macroscopic deformation, frictional energy loss, and damage mechanism using single-asperity scratching simulations. Single-asperity scratch simulations have been previously used to simulate the damage formation on polypropylene surface [27]. In addition, Looijmans et al. [28] experimentally investigated the effect of pre-stretch on the frictional response of iPP using single-asperity scratch with a rigid diamond indenter. However, a solid link between the intrinsic response of the polymer and its scratch and frictional response has never been established. A key aspect in this study is the thermo-mechanically coupled modelling that is introduced into the EGP model and implemented in a Finite Element Method (FEM) framework for the first time. The reason is, like metals, a percentage of the mechanical work of polymers has been found to be dissipative, in other words, heat is generated due to plastic deformation [29,30]. The thermal model is analogue to the work of Boyce et al. [30] and Klompen et al. [31]. It is used to first quantitatively investigate the effect of pre-stretch on the frictional response of polymers and Klompen et al. [31].

2.2. Sliding friction experiments

An MTS Nano Indenter XP is used to perform scratch test experiments by sliding a conical, diamond indenter tip (cone angle 90°, top radius 50 μm) over the smooth polymer surface. The normal load applied to indenter geometry, as well as the sliding velocity, are kept constant during a single sliding friction experiment. Normal loads are varied between 200 and 400 mN at sliding velocities ranging from 1 till 100 μm/s. The penetration depth and lateral force are recorded as function of sliding distance. To ensure steady-state measurements, scratch tests of 1 mm in length are performed. At each combination of applied load and sliding velocity, the scratch response is measured three times to check for reproducibility.

3. Constitutive modelling

3.1. The EGP model for thermorheologically simple polymers

The 3D elasto-viscoplastic constitutive model used, see Van Breemen et al. [13] for more details, consists of multiple Maxwell elements connected in parallel to a neo-Hookean spring. In the model the total stress $\sigma$ is split into the driving stress $\sigma_i$ and the hardening stress $\sigma_s$:

$$\sigma = \sigma_i + \sigma_s.$$  \hfill (1)

Physically, the hardening stress is interpreted as a rubber elastic contribution of the entangled network. Mathematically, it is described with a simple neo-Hookean relation:

$$\sigma_s = \frac{G_0 \varepsilon_s^p}{J_0}.$$  \hfill (2)

where $G_0$ denotes the hardening modulus, $\varepsilon_s^p$ is the deviatoric part of the isochoric left Cauchy-Green strain tensor, and $J_0$ is the volume change ratio. The driving stress is attributed to intermolecular interactions [3, 12] and is split into a hydrostatic and a deviatoric part [1]:

$$\sigma_i = \sigma_i^h + \sigma_i^d = \kappa (J - 1) |I| + \sum_{\alpha = 1}^{n} G_{\alpha} \tilde{B}_{\alpha,i}^d,$$  \hfill (3)

where $\kappa$ is the bulk modulus, $G$ is the shear modulus, $\tilde{B}_{\alpha,i}^d$ is the elastic deviatoric part of the isochoric left Cauchy-Green strain tensor. The subscript $i$ refers to a specific mode, and $n$ denotes the number of modes [13]. The plastic deformation-rate tensor $D_{\alpha,i}$ is related to the deviatoric stress $\sigma_i^d$ via a non-Newtonian flow rule:

$$D_{\alpha,i} = \frac{\sigma_i^d}{2 \eta_i},$$  \hfill (4)

where $\eta_i$ are the viscosities of each Maxwell element which are described by the extended Eyring flow rule. This flow rule was extended [2, 38–40] to take the pressure dependence and strain softening into account:

$$\eta_i = \eta_0,ref \cdot \tau / \tau_0 \cdot \exp \left[ \frac{p \mu}{n_p} \exp \left[ S, R_{\gamma}(\tilde{\gamma}) \right] \right].$$  \hfill (5)

where $\eta_0,ref,i$ is the reference viscosity of each Maxwell element, $\tau$ is the total equivalent stress, $\tau_0$ defines the characteristic shear stress, $p$ is the hydrostatic pressure, the pressure dependency is governed by the parameter $\mu$, the physical ageing is contained in the state parameter $S$. The softening function $R_{\gamma}(\tilde{\gamma})$ describes the strain-softening process, i.e. the erasure of thermal history upon the inception of plastic deformation. Klompen et al. [3] expressed $R_{\gamma}(\tilde{\gamma})$ as a function of $\tilde{\gamma}$ using a modified Carreau-Yasuda relation:

$$R_{\gamma}(\tilde{\gamma}) = \left[ \frac{1 + (\tau_0 \exp(\phi \tilde{\gamma}))^{(n_2 - 1)/n_2}}{1 + \tau_0^{n_2 - 1}} \right]^{n_2 / (n_2 - 1)},$$  \hfill (6)

2. Experimental

2.1. Materials and sample preparation

An injection moulding grade iPP homopolymer with weight-averaged molar mass $M_w$ of 320 kg/mol and polydispersity $M_w/M_n$ of 5.4 is kindly provided by SABIC (Riyad, Saudi Arabia). This particular grade was selected because the intrinsic deformation kinetics of its main crystal phases are well-characterized by Caelers et al. [26]. Compression response data is adopted from this work. Experimental data regarding the scratch response of α-phase iPP is adopted from a previous work [35]. Polymer granules are molten at 230 °C and manually compressed between glass slides to ensure a smooth surface with respect to the indenter geometry. Polymer films of approximately 500 μm in thickness, are kept isothermal in the melt for 10 min to erase thermomechanical history and subsequently cooled to room temperature. Using this procedure, α-phase samples with a crystallinity of about 61% are obtained. Analogously, β-phase samples are prepared from granules containing 0.1 wt% β-nucleating agent (NISTAR NU100 New Japan Chemical Group). The exact compounding procedure of this material can be found elsewhere [25]. Samples having a volumetric crystallinity of 64% were obtained, using the thermal protocol described above.
where \( \eta \) is the equivalent plastic strain, and \( r_0, r_1, \) and \( r_2 \) are the fitting parameters.

When temperature is considered, a temperature-dependent pre-exponential factor is added to the Eyring equation:

\[
\eta_i = \frac{\tau_i}{r_0} \exp \left[ \frac{\rho \mu p}{r_0} \exp \left[ \frac{\Delta U}{k T} \right] \right],
\]

where \( \Delta U = \frac{k T}{V} \) is the activation energy, \( R \) is the universal gas constant, \( T \) represents the temperature, and \( T_{ref} \) is the room temperature. In addition, \( t_0 \) is determined using the following equation:

\[
\tau_0 = k \frac{T}{V^2},
\]

where \( k \) is the Boltzmann’s constant and \( V^* \) is the activation volume.

### 3.2. Extension to thermorheologically complex behaviour

The experimental findings of Van Breemen et al. [5] clearly show the existence of a secondary molecular process in iPP as it has been tested over broad range of strain rates. This thermorheologically complex behaviour contributes to an increased strain-rate dependency of the polymer at high strain rates. To capture this complex behaviour of the polymer we use a straightforward extension of Equation (1) based on the Ree-Eyring equation [41]:

\[
\sigma = \sigma_{i,1} + \sigma_{i,2} + \sigma_c,
\]

where \( \sigma_{i,1} \) is the driving stress of the primary process (\( \sigma_i \) in Equation (3)), and \( \sigma_{i,2} \) is the driving stress for the secondary process. Both stresses have their own temperature and rate dependencies. Therefore, each process has its own characteristic values of the activation volume \( V^* \) and activation energy \( \Delta U \). In comparison to experimental data, Equation (9) has been shown to be effective in capturing the response of many polymers including iPP [42].

### 3.3. Extension to thermo-mechanically coupled model

A thermo-mechanically coupled model must be used if deformation converts mechanical work into heat through an irreversible process which cannot be neglected relative to other heat sources [43–45]. For metals the amount of dissipated energy is approximately 90–95% [30]. Adams et al. [29] showed that around 50%–80% of the work of deformation is dissipated into heat during cold drawing of polycarbonate. Therefore, in case of applying relatively high deformation rates, it becomes a necessity to use a thermo-mechanically coupled model in order to capture the intrinsic response of the polymer. The model is formulated analogous to Boyce et al. [30]. The general energy balance equation of a 3D system is given by:

\[
\rho c_v \frac{\partial T}{\partial t} = -\nabla q + \dot{Q}_b,
\]

where the left hand side represents the rate of change in the internal heat energy per unit volume in the material with \( \rho \) being the material density and \( c_v \) the specific heat capacity. In the first part of the right hand side, the operator \( \nabla \) acts as the divergence operator, and the vector \( q = q(x, t) \) is a vector field that represents the magnitude and the direction of the heat flow at the point \( x \) of space and time \( t \). This vector is given by Fourier’s law for an isotropic homogeneous medium where the rate of flow of heat energy per unit volume through a surface is proportional to the negative temperature gradient across it:

\[
q = -k_p \nabla T,
\]

where \( k_p \) is the proportionality coefficient which represents the polymer thermal conductivity. The second part of the right hand side, \( \dot{Q}_b \), is the rate of change in the dissipative work done by the driving stress given by:

\[
\dot{Q}_b = \Gamma \sigma : D_{pp},
\]

where \( \Gamma \) is the percentage of mechanical work dissipated into heat. This percentage gradually decreases as the material continues to be strained due to extensive molecular orientation [30].

Next, the boundary conditions should be considered. In compression experiments, steel compression plates are used to compress the sample, while in scratch experiments, the indenter in contact with the polymer. In that respect, the change in the heat flux due to the conduction boundary condition is given by:

\[
\dot{Q} = A_k (k_b/\tau_b) (T - T_{ref}),
\]

where \( A_k \) is the contact area between the sample and the steel compression plates in the compression test or the indenter in the scratch test, \( k_b \) and \( \tau_b \) are the thermal conductivity and the thickness of the body in contact with the polymer. Similarly, heat convection to the surrounding air must be considered, it is given by the following relation:

\[
\dot{Q} = A_k h_k (T - T_{air}),
\]

where \( A_k \) is the contact area between the sample and the air, \( h_k \) is the heat transfer coefficient of the air. Radiation effects are very small and, therefore, neglected. Since the thermal conductivity of polymers \( k_p \) is relatively small and the volume of the surrounding contact bodies are relatively large with respect to the sample, the surrounding mediums are considered as heat sinks. Therefore, the heat transfer within the polymer will be neglected. Similar to Ref. [31], the boundary conditions parts are added to Equation (10). The equation of evolution can be written as follows:

\[
\frac{\partial T}{\partial t} = \frac{1}{\rho c_p} \left[ \Gamma (\sigma : D_{pp}) - k (T - T_{ref}) \right].
\]

It is difficult to quantitatively determine the boundary conditions parameters, they are also different for different tests, i.e. compression or scratch. In that respect, a boundary condition parameter \( k \) is used instead and is determined by fitting the simulation results to the experimental compression data. For simplicity the same value of \( k \) will be used in the scratch simulations. This parameter encompasses all the boundary condition parameters in addition to the thermal conductivity and heat transfer coefficient of the mediums which are in contact with the polymer. The final evolution equation is given by:

\[
\frac{\partial T}{\partial t} = \frac{1}{\rho c_p} \left[ \Gamma (\sigma : D_{pp}) - k (T - T_{ref}) \right].
\]

The evolution equation is then solved using the forward Euler method:

\[
T_c = T_b + \Delta T,
\]

where \( T_c \) is the nodal temperature of the current increment, \( T_b \) is the temperature of the previous increment, and \( \Delta T \) is the incremental time step.

### 3.4. Friction modelling and FEM mesh

The constitutive model is implemented in the FEM package MSC. Marc in order to simulate the single-asperity scratch test. The Coulomb model is adopted as a friction model. The arctangent model is used to avoid the numerical singularities by smoothening the stick-slip transition, similar to Refs. [14, 15]:

\[
f_f = -\mu_s \frac{2}{\pi} \arctan \left( \frac{\mu_1}{\mu_s} \right) t.
\]

where \( f_f \) and \( f_n \) are the friction and normal forces respectively, \( \mu_1 \) the
local friction coefficient, $v_s$ is the relative sliding velocity, and $r$ is the tangential vector. The value of $\delta$ determines the value of the relative velocity below which sticking occurs. A large value of $\delta$ will guarantee quick convergence but poor estimation of the friction force. A small value will result in better estimation of the friction force at the cost of more computational power, and, if too small, it will be impossible to reach convergence. Many simulations have been used to optimize this parameter. The friction coefficient $\mu_t$ is determined by fitting the simulations frictional response to experimental data in a fashion similar to Van Breezen et al. [14]. A local friction coefficient value of $\mu_t = 0.28$ is found to give the best description of the experimental scratching data of both iPP phases.

The single-asperity scratch mesh is shown in Fig. 1a. Only half of the polymer surface is meshed since the simulation is symmetric. The meshed volume is $0.2 \times 0.2 \times 0.8 \text{mm}^3$. The symmetry plane is fixed in y-direction and the sides are restrained in x- and z-direction. Room temperature is used as an initial condition at each node. An indenter with a tip radius $50 \mu m$ is used, and is modelled as a rigid body. Mesh refinement is applied to the area of greatest interest, i.e. largest deformation. Fig. 1b shows the FEM mesh during scratching. First, a normal force $F_n$ is gradually applied to the indenter for 25 s, then, the scratch cycle takes place at constant scratch speed $v_s$.

### 3.5. Intrinsic response and model parameters

To investigate the intrinsic response of the polymer, single-element FEM compression simulations are performed and fitted to experimental data, obtained from Ref. [26]. The resulting reference spectra of $\alpha$- and $\beta$-iPP are shown in Tables 1 and 2 in Appendix A. The thermodynamic state parameter of the material, $S_\alpha$, is determined by using indentation simulations and fitting them to the experiments, similar to Ref. [14]. The material parameters for $\alpha$- and $\beta$-iPP are obtained from Ref. [5] and tabulated in Tables 3 and 4 respectively. The pressure dependency parameter $\mu$ is determined from the compressive and tensile data obtained from Refs. [25,26]. The thermal parameters are adopted from Ref. [31], and presented in Table 5. It should be noted that $V_1$ and $V_2$ are the activation volumes for the primary and secondary molecular processes respectively, the same holds for the activation energies $\Delta U_1$ and $\Delta U_2$.

The experimental intrinsic response of iPP is plotted in Fig. 2 by solid lines. The data show the response of $\alpha$-iPP, Fig. 2a, and $\beta$-iPP, Fig. 2b. Due to the strain-rate dependence of the polymer, a higher yield stress is observed at higher strain rates for both phases. Fig. 3 shows the upper- and lower-yield stress for each phase versus the applied strain rate. The upper-yield stress displays a higher strain-rate dependence than the lower-yield stress. This implies that the upper-yield stress is controlled by a secondary molecular process in addition to the primary one, while only the primary process contributes to the kinetics of the lower-yield stress [5].

The simulation results are fitted to experimental data in order to obtain the model parameters as can be seen in Fig. 2. The first set of simulations “sim1” plotted by dotted lines clearly shows, at high strain rates, that the post-yield response is not captured by the model. This is due to the internal heat generation at these high rates. Due to high plastic deformation-rates, the material starts to soften as a result of heat dissipation. In addition, there is less time for conduction and convection of the generated heat. This leads to a significant effect on the post-yield response [5,30]. It should be noted that the existence of a secondary process controlling the upper-yield stress only manifests itself in a higher yield drop as the deformation rates increase within the assessed range of strain rates. The heat generation due to plastic deformation only appears at relatively high strain rates and alters the entire post-yield response. This explains the decrease in slope of the lower-yield stress at the highest strain rate, see Fig. 3. In this respect it is necessary to use a thermo-mechanically coupled model to accurately predict the post-yield response of the polymer. The other set of simulations “sim2” plotted by dashed lines shows the simulation results fitted to the experimental data after implementing the thermo-mechanically coupled model, Equations (16) and (17), and fitting the parameters $\Gamma$ and $k$. Fig. 4 shows the temperature evolution due to plasticity-induced heating during the intrinsic simulations of iPP at different strain rates. The results reveal that the model is qualitatively able of determining the temperature evolution due to heat generation resulting from plastic deformation. Higher deformation rates result in more mechanical work dissipated into heat, and less time for cooling and temperature rises. The slight difference in the maximum temperatures between $\alpha$-iPP and $\beta$-iPP can be related to the slightly higher yield stress of $\beta$-iPP at high strain rates.

### 4. Results and discussion

#### 4.1. Influence of the thermo-mechanical model

The use of the thermo-mechanical model is proven to be necessary to capture the intrinsic response of the polymer at high strain rates. The model is expected to have a significant influence on the deformation kinetics during scratch. In order to show this influence, two single-asperity scratch simulations are performed using a mechanical model and a thermo-mechanical model respectively. In these simulations, indentation is first performed for 25 s where a normal load of $F_n = 200 \text{mN}$ is gradually built-up, followed by constant-speed scratching of $v_s = 100 \mu m/s$ for 250 $\mu m$. During this study, several elements are considered, see Fig. 1b. First, element 1 is an element on the surface of the polymer that is chosen to analyse the influence of the thermo-mechanical model on deformation kinetics. Steady-state response is
already present when element 1 is reached. The analysis of element 1 only considers the scratch part which takes 2.5 s, while the indentation part which lasts for 25 s is not included. By definition, no mechanical work is dissipated into heat using the mechanical model, and therefore, no change in temperature can be observed, see Fig. 5a and b. Due to plastic heat generation using the thermo-mechanical model, material softens and deformation becomes slightly faster, see Fig. 5c. The equivalent stress is dependent on both deformation rate and temperature. Lower deformation rates and higher temperatures result in lower stresses. The values of deformation rate are almost the same in both cases. However, temperature is significantly higher when using the thermo-mechanical model. This leads to lower value of equivalent stress due to thermal softening, see Fig. 5d.

4.2. Effect of friction on heat generation

Friction is a crucial aspect in contact mechanics. Generally, more friction implies more adhesion between bodies in contact. This leads to more deformation in case of scratching, which is manifested in more distorted elements in simulations, compare Fig. 7a and b. Since we use a
thermo-mechanical model, more plastic deformation will lead to an increased heat generation. It should be noted however, that the we investigate how friction affects heat generation via deformation, and that heat generated due to friction itself is not accounted for in the used coulomb model. In order to investigate the influence of friction quantitatively, two single-asperity scratch simulations are performed. In the first one, the frictionless case is assumed, i.e. friction coefficient \( \mu_f = 0 \). In the second simulation a friction coefficient of \( \mu_f = 0.28 \) is used.

Fig. 5. The effect of using a thermo-mechanical model for scratch simulation and its influence on the deformation kinetics of element 1; (a) rate of mechanical work dissipated into heat, (b) the resulting rise in temperature, (c) the equivalent plastic strain, (d) the equivalent stress.

Fig. 6. (a) Plastic deformation of each element, (b) the resulting rise in temperature due to plastic deformation.

Fig. 7. Single-asperity scratch; (a) using friction coefficient \( \mu_f = 0 \), (b) using friction coefficient \( \mu_f = 0.28 \). Friction implies more polymer-tip adhesion, and thus, elements are more deformed.
Both simulations are performed using the thermo-mechanical model, \( F_n = 200 \text{ mN} \), and \( v_s = 100 \mu \text{m/s} \). Element 1 in Fig. 1b is chosen for this analysis. Frictional scratch leads to polymer-tip sticking, leading to more deformation and a higher deformation rate, see Fig. 8c. Therefore, more mechanical work is dissipated into heat, increasing the temperature of the polymer, see Fig. 8a and b. Fig. 8d shows the value of the equivalent stress. As mentioned earlier, the equivalent stress is dependent on both deformation rate and temperature. Before any plastic deformation of element 1, \( t = 25 \text{ s} \) till \( t = 25.7 \text{ s} \), there is no heat generation, and thus, stress is only dependent on deformation rate (elastic) which is lower in the frictionless case, leading to a lower stress level. As plastic deformation takes place, \( t = 25.7 \text{ s} \) till \( t = 26.5 \text{ s} \), and temperature starts to rise, the stress becomes dependent on both deformation rate and temperature. Finally, when the indenter continues scratching the surface and element 1 stops being deformed, \( t = 26.5 \text{ s} \) till \( t = 27.5 \text{ s} \), stress becomes only dependent on temperature which is higher when friction is accounted for. Therefore, during this period the resulting equivalent stress from frictional scratch becomes lower than the frictionless case, leading to a lower stress level. As plastic deformation the higher the amount of mechanical work dissipated into heat parameter, \( \Gamma \). This agrees with Equation (16).

### 4.3. Analysis of heat generation at various scratch velocities

As shown before, the equivalent plastic strain \( \gamma_p \) dictates the trend of temperature evolution. However, the time span in which this plastic strain takes place is critical. In another words, the faster the plastic deformation the higher the amount of mechanical work dissipated into heat according to Equation (16). In this respect, we test the influence of scratch velocities on heat generation. Three simulations are performed at \( v_s = 1 \mu \text{m/s}, 10 \mu \text{m/s}, \) and \( 100 \mu \text{m/s} \). The applied load is \( F_n = 200 \text{ mN} \) and the friction coefficient is \( \mu_f = 0.28 \). The scratch distance is \( 250 \mu \text{m} \) for all three simulations. Element 1 from Fig. 1b is again analysed for the three cases. Fig. 9a shows the evolution of the equivalent plastic strain along the entire scratch distance. Higher equivalent plastic strain is observed at lower scratch speeds due to the viscoelasticity of the polymer. However, at this lower speed, the time span to complete the entire scratch cycle is one order of magnitude lower than at \( v_s = 10 \mu \text{m/s} \), and two orders of magnitude lower than at \( v_s = 100 \mu \text{m/s} \). Thus, the plastic strain-rate is the lowest at this speed, leading to the lowest temperature increase observed, see Fig. 9b. Since there is enough time for conduction and convection of the generated heat, the temperature goes back to room temperature before the scratch cycle ends. As the scratch speed increases, plastic strain-rate also increases leading to higher maximum temperature generated, as seen in Fig. 9. It is clear how increasing the scratch speed by one order of magnitude has a significant effect on the temperature profile along the polymer surface (see Fig. 10).

### 4.4. Scratch and frictional response of \( \alpha \)-IPP and \( \beta \)-IPP

Scratch tests are performed at three different scratch velocities; \( v_s = 1 \mu \text{m/s}, 10 \mu \text{m/s}, \) and \( 100 \mu \text{m/s} \) and at applied normal load \( F_n = 200 \text{ mN} \) on both \( \alpha \)-IPP and \( \beta \)-IPP. Indentation is first performed for \( 25 \text{ s} \), followed by a constant-speed scratch for \( 600 \mu \text{m} \). The single-asperity scratch response for \( \alpha \)-IPP and \( \beta \)-IPP is shown in Fig. 11. Fig. 11a and b show the value of indenter penetration into the polymer after indentation till it reaches the steady state for \( \alpha \)-IPP and \( \beta \)-IPP respectively. Fig. 11c and d display the corresponding frictional response. One observation is the higher bumps in the beginning of the scratch cycle of \( \alpha \)-IPP, which comes from the relatively low strain hardening that allows the tip to initially penetrate deeper before the frictional shear stress pushes the indenter back up and as a result the friction force drops. This observation is even more pronounced at the lowest speed due to the rate dependence of the polymer which allows more initial tip penetration. A second observation is the wavy response in case of \( \alpha \)-IPP which is also related to the relatively low strain hardening that enhances the build-up of the bow wave in front of the tip, resulting in a more abrupt stick-slip cycle. Whereas in case of \( \beta \)-IPP, the higher strain hardening resists the build-up of the bow wave, resulting in

![Fig. 8. The effect of friction during scratch simulation and its influence on the deformation kinetics of element 1; (a) rate of mechanical work dissipated into heat, (b) the resulting rise in temperature, (c) the equivalent plastic strain, (d) the equivalent stress.](image-url)
a smoother stick-slip cycle. Fig. 12a presents the values of the steady-state tip penetration into the polymer at each scratch speed. Fig. 12b displays the corresponding friction force values. The plots show more deformation-rate dependence for $\beta$-iPP which resembles its intrinsic response. As the scratch speed increases, the penetration depth and friction force values decrease at higher rate in case of $\beta$-iPP. This suggests that, in terms of friction and wear resistance, $\beta$-iPP performs better at higher scratch speeds.

4.5. Damage formation

The intrinsic response of the polymer has a crucial influence on its friction response. In this respect, it is necessary to quantitatively investigate and link between the intrinsic response and the resulting friction response, and come up with a criterion for damage formation (i.e. initiation of wear). It has been reported that the so-called fish-scale damage pattern is the dominant damage mechanism that appears on the surface of polypropylene when scratched. This damage mechanism is controlled by the stick-slip phenomenon which occurs when the indenter experiences periodic changes in resistance. This periodic change comes from the periodic build-up of material in front of the tip, which in turn leads to the build-up of friction force to keep the constant velocity of the indenter. When the frictional shear stress is high enough, the indenter is suddenly pushed away, and slipping occurs. During the sticking cycle the material is plastically drawn along the scratch direction. This damage mechanism for polypropylene has been reported by many researchers [34–37]. Based on this, we select the equivalent plastic strain $\gamma_p$ as a physical criterion to qualitatively assess and predict the formation of the fish-scale damage pattern. This criterion has been previously used to qualitatively assess damage formation on polymer and metal surfaces [46, 47].

First, we are interested in investigating the influence of boundary conditions, i.e. normal load and scratch speed on damage formation. Fig. 13a shows the polymer surface of $\alpha$-iPP after single-asperity scratch at various normal loads. At higher applied normal load, the tip penetrates more into the surface, creating more material build-up in front of the tip, making the stick-slip more abrupt and severe. This leads to material being more plastically drawn along the scratch direction. Fig. 13b displays the simulation results, in which the equivalent plastic strain $\gamma_p$ contour profile is shown. Due to the increased normal load, more material is plastically deformed giving rise to the equivalent plastic strain. This is confirmed from the values of the width of the scratch which are quantitatively comparable to the experimental values.

Due to rate dependency of the polymer, lowering scratch speed has an effect on the scratch response similar to that of a higher applied normal load. As scratch speed decreases, the indenter penetrates more into the polymer creating a more abrupt stick-slip cycles in which material experiences more plastic deformation, see Fig. 14.

The intrinsic response of the polymer is a critical factor, and plays a decisive role in its damage formation mechanism. Fig. 15a shows the polymer surface of $\alpha$-iPP and $\beta$-iPP after single-asperity scratch. Fish-scale damage pattern is clearly visible in case of $\alpha$-iPP. Simulation results, Fig. 15b, show that the maximum value of $\gamma_p$ is significantly higher in case of $\alpha$-iPP although penetration depth and friction force values at this speed are almost identical. However, looking at the intrinsic response of both phases, Fig. 15c, a significant difference in strain hardening is observed. Strain hardening is believed to be the reason behind damage formation differences between the two phases. A material with low strain hardening experiences more localized plastic deformation, and more material build-up, creating a more severe stick-slip cycle, leading to higher plastic deformation and fish-scale damage pattern becoming more visible.
Fig. 11. Single-asperity scratch results of iPP; penetration into the surface versus sliding distance (a) α-iPP, (b) β-iPP, and friction force versus sliding distance (c) α-iPP, (d) β-iPP.

Fig. 12. (a) Steady-state penetration depth values at various scratch speeds, (b) steady-state friction force values at various scratch speeds. Data points are experiments, lines are fitting of simulation data points. The plots show more deformation-rate dependency for β-iPP which resembles its intrinsic reponse.

Fig. 13. Polymer surface of α-iPP after single-asperity scratch at various normal loads; (a) experimental, (b) simulations showing maximum value of γp. Wider scratch profile appears at high normal loads. Higher equivalent plastic strain γp in simulations at high normal loads resembles the appearance of fish-scale damage pattern in experiments at these loads.
Due to the crystalline nature of iPP and other semi-crystalline polymers, material orientation has an effect on its intrinsic response [22,48]. During scratch, the material is oriented along the scratch direction. It is possible that the resulting material orientation has a critical role in damage formation. To test this hypothesis, the three principle strains of element 1 from Fig. 1b are obtained during scratching at a speed of 1 μm/s and 100 μm/s, see Fig. 16. Their values are comparable to the values of equivalent plastic strain $\gamma_p$ in Fig. 14b. The equivalent plastic strain is only a summation of the magnitude of the plastic part of all principle strains. Scratching at 1 μm/s results in a 10% increase in all three principle directions when compared to scratching at 100 μm/s. Previous studies have shown that orientation, i.e. pre-stretch, results in less scratch depth and friction force and reduced damage [28,49]. However, in our case, the lowest scratch speed results in the highest penetration depth and friction force which leads to more apparent damage. This suggests that the viscoelasticity of the material, and not orientation, is the decisive factor in damage formation when comparing the scratch response at different scratch speeds. Similar observations hold when comparing different loads and phases. Although the current

Fig. 14. Polymer surface of α-iPP after single-asperity scratch at various scratch speeds; (a) experimental, (b) simulations showing maximum value of $\gamma_p$. Higher equivalent plastic strain $\gamma_p$ appears in simulations at lower speeds due to the rate dependency of polymers. This explains the formation of fish-scale damage pattern at these speeds in experiments.

Fig. 15. Polymer surface of α-iPP and β-iPP after single-asperity scratch; (a) experimental, (b) simulations showing maximum value of $\gamma_p$. Fish-scale damage pattern is clearly visible in case of α-iPP, and simulation results show higher equivalent plastic strain $\gamma_p$ for α-iPP. (c) Intrinsic response of α-iPP and β-iPP show a significant difference in strain hardening, which is believed to be reason behind the damage formation differences between the two phases.

Fig. 16. The three principle strains of element 1 during scratching at a speed of 1 μm/s and 100 μm/s. Small difference in element stretching suggests that material orientation is not a critical factor in damage formation.
model does not account for material anisotropy, it is plausible for future work to include it in order to quantitatively investigate its effect on the scratch and frictional response. Senden et al. [48] used a 3D viscoelastic model based on Hill theory in order to predict the yield of drawn polypropylene tapes at various orientation angles with respect to the drawing direction. It is possible to simulate single-asperity scratch of an anisotropic polypropylene surface by implementing such a model in FEM.

5. Conclusions

A 3D elasto-viscoplastic constitutive model has been implemented in a FEM-framework to simulate a single-asperity scratch of two morphological structures; α- and β-IPP at different normal loads and scratch velocities. The physics-based thermo-mechanical model along with the experimental work formed the base of a hybrid experimental-numerical approach that was used to investigate the polymer scratch and frictional response based on its intrinsic response. From this study we conclude that:

1. The thermo-mechanical model is necessary to quantitatively predict the intrinsic response of the material especially at higher strain rates where heat is produced due the plastic deformation.
2. At higher scratch velocities, the polymer becomes more resistant to deformation due to its strain-rate dependence. This results in a lower scratch depth and friction force.
3. β-IPP exhibits a slightly higher strain-rate dependence than α-IPP. This leads to lower penetration depth and friction force at relatively high deformation rates. In this respect, β-IPP is a good choice for high-speed applications.
4. The stick-slip phenomenon is mainly responsible for the damage mechanism observed. During the sticking cycle the material is plastically drawn along the scratch direction creating the damage pattern. For this reason, the equivalent plastic strain $\gamma_p$ has been chosen as a physical criterion to qualitatively assess and predict the formation of the fish-scale damage pattern.
5. The stick-slip phenomenon becomes more pronounced when the tip penetrates deep into the polymer, which introduces additional material build-up in front of the tip. A high normal load and low scratch speed lead to more tip penetration, thus, stick-slip becomes more severe. The material is more plastically deformed along the scratch direction and the fish-scale damage pattern is clearly observed.
6. A significant intrinsic difference between α-IPP and β-IPP is the strain hardening. In case of β-IPP the higher strain hardening resists the build-up of the bow wave in front of the tip resulting in less sticking and more slipping. Less material is plastically drawn along the scratch direction, preventing the formation of the fish-scale damage pattern. This makes β-IPP, again, a favourable choice over α-IPP for applications that require wear resistance.
7. The induced material orientation due to scratching is relatively low and does not have a significant role in damage formation.

Acknowledgements

This research forms part of the research programme of Dutch Polymer Institute (DPI), project 783t. The authors wish to thank DPI for their financial support.

Appendix A. Material parameters

<table>
<thead>
<tr>
<th>Table 1</th>
<th>Reference spectrum of α-IPP.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Process 1</td>
<td>Mode</td>
</tr>
<tr>
<td>1</td>
<td>1.6 $\times 10^5$</td>
</tr>
<tr>
<td>2</td>
<td>3.5 $\times 10^7$</td>
</tr>
<tr>
<td>3</td>
<td>2.3 $\times 10^6$</td>
</tr>
<tr>
<td>4</td>
<td>3.3 $\times 10^5$</td>
</tr>
<tr>
<td>5</td>
<td>3.3 $\times 10^4$</td>
</tr>
<tr>
<td>6</td>
<td>6.7 $\times 10^3$</td>
</tr>
</tbody>
</table>

| Process 2 | Mode | $\eta_{0,ref}$ [MPa.s] | $G_i$ [MPa] |
| 1 | 2.4 $\times 10^{-2}$ | 80 |

<table>
<thead>
<tr>
<th>Table 2</th>
<th>Reference spectrum β-IPP.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Process 1</td>
<td>Mode</td>
</tr>
<tr>
<td>1</td>
<td>1.6 $\times 10^5$</td>
</tr>
<tr>
<td>2</td>
<td>3.5 $\times 10^7$</td>
</tr>
<tr>
<td>3</td>
<td>2.3 $\times 10^6$</td>
</tr>
<tr>
<td>4</td>
<td>3.3 $\times 10^5$</td>
</tr>
<tr>
<td>5</td>
<td>3.3 $\times 10^4$</td>
</tr>
<tr>
<td>6</td>
<td>6.7 $\times 10^3$</td>
</tr>
</tbody>
</table>

| Process 2 | Mode | $\eta_{0,ref}$ [MPa.s] | $G_i$ [MPa] |
| 1 | 2.4 $\times 10^{-2}$ | 80 |
Table 3
Material parameters of α-iPP, adopted from Ref. [5].

<table>
<thead>
<tr>
<th>G [MPa]</th>
<th>ϵ [MPa]</th>
<th>S₀ [μ]</th>
<th>ρ [-]</th>
<th>V₁ [mm²]</th>
<th>ρ [g/m³]</th>
<th>r₀ [-]</th>
<th>r₁ [-]</th>
<th>r₂ [-]</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.0</td>
<td>1650</td>
<td>5.0</td>
<td>0.12</td>
<td>3.10</td>
<td>3.0</td>
<td>0.90</td>
<td>0.95</td>
<td>2.0</td>
</tr>
</tbody>
</table>

Table 4
Material parameters of β-iPP, adopted from Ref. [5].

<table>
<thead>
<tr>
<th>G [MPa]</th>
<th>ϵ [MPa]</th>
<th>S₀ [μ]</th>
<th>ρ [-]</th>
<th>V₁ [mm²]</th>
<th>ρ [g/m³]</th>
<th>r₀ [-]</th>
<th>r₁ [-]</th>
<th>r₂ [-]</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.0</td>
<td>1650</td>
<td>5.0</td>
<td>0.18</td>
<td>2.90</td>
<td>3.0</td>
<td>0.90</td>
<td>0.95</td>
<td>2.0</td>
</tr>
</tbody>
</table>

Table 5
Thermal parameters of α- and β-iPP, activation energy and specific heat capacity obtained from Ref. [31].

<table>
<thead>
<tr>
<th>Γ [-]</th>
<th>k [MPa/K s]</th>
<th>ΔU₁ [kJ/mole]</th>
<th>ΔU₂ [kJ/mole]</th>
<th>cₚ (kJ/kg K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.6</td>
<td>0.10</td>
<td>274</td>
<td>251</td>
<td>1.92</td>
</tr>
</tbody>
</table>

References
