A numerical investigation of the potential of rubber and mineral particles for toughening of semicrystalline polymers

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Abstract

The impact strength of many semicrystalline polymers can be improved by the dispersion of second-phase rubber particles. A criterion for the effect of this practice is based on the average interparticle matrix ligament thickness. The critical interparticle distance, below which a substantial toughness increase can be observed, is considered to be an intrinsic material property of the matrix. A toughening mechanism has recently been suggested which considers a layer of transcrysallized material around well-dispersed particles, having a reduced yield strength in certain preferentially oriented directions, thereby opening the possibility of using mineral fillers. In this work, the potential of toughening of semicrystalline polymeric material by local anisotropy in combination with soft rubber and hard mineral filler particles is investigated. The matrix material is modeled within the framework of anisotropic Hill plasticity with a rate dependent and hardening yield stress. Various particle/matrix interface conditions are used to study the role of debonding and cavitation. The presence of debonded moderately stiff or hard fillers is found to affect the shear yielding effect of local anisotropy that was found for voided material.

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1. Introduction

Semicrystalline polymeric materials are widely used in a range of engineering applications. An important point of concern that limits their application is their often occurring brittle response. The toughness of these materials may be enhanced by blending them with rubber particles. The present-day notion of the toughening mechanism in these materials is founded on the criterion proposed by Wu [1], which states that a sharp brittle/tough transition occurs for nylon/rubber blends when the average interparticle matrix ligament thickness $L$ is reduced below a critical value $L_c$. 

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which appears to be independent of the rubber volume fraction and particle size.

A physical explanation of the absolute length parameter was offered by Muratoglu et al. [2,3], who recognized the brittle/tough transition as a true material feature. It was attributed to thin layers of preferentially oriented material, with a reduced plastic resistance, appearing in the microstructural morphology of particle-modified semicrystalline material. Effectively, the crystallization behavior of the matrix is influenced by the rubber/matrix interface, leading to a layer of parallel crystalline lamellae, growing from the interface, with the crystalline planes having the lowest plastic resistance parallel to the interface. It is experimentally established that these transcrystalline layers have a well-defined thickness of approximately $A_c/2$ [3,4]. When the average matrix ligament thickness $A$ is below the critical value $A_c$, the favorably oriented material percolates through the system, bridging between the second-phase particles. A blended system consists of (i) rubber particles having a low modulus, (ii) preferentially oriented anisotropic matrix material enveloping the particles and (iii) the bulk matrix material having a randomly oriented structure and effectively having isotropic material properties. In the toughening mechanism postulated by Muratoglu et al. [3], after cavitation of the second-phase rubber particles, the regions with a lowered yield resistance will facilitate large plastic deformation and thereby improve toughness. Tzika et al. [5] used a micromechanical numerical model, with a staggered array of particles, to study the influence of preferentially oriented anisotropic layers, modeled with anisotropic Hill plasticity, on the deformation mechanisms under high triaxiality conditions. They observed plastic deformation in the matrix to occur diagonally away from particles (i.e. in the matrix material between particles, parallel to the interfaces) for $A \leq A_c$, rather than bridging between particles for $A > A_c$.

Bartczak et al. [4,6] generalized the Wu criterion to other material (HDPE) and showed the critical interparticle distance to be an intrinsic property of the matrix material, thereby opening the possibility of using mineral fillers for the toughening of semicrystalline polymers, the advantage of which would be an improved modulus of the blend. They argued that intraparticle cavitation is not necessary for toughening and therefore that debonding of hard filler particles could be an alternative for the cavitation of the rubbery phase. However, by using calcium carbonate filler particles in a Nylon-6 matrix, Wilbrink et al. [7] did not obtain the tough response of rubber/Nylon-6 blends, as was reported by Muratoglu et al. [3,8], and attributed this to the development of triaxial stresses. A four times increase of the Izod impact energy was obtained by Thio et al. [9] by incorporation of calcium carbonate particles in isotactic polypropylene, resulting from combined mechanisms of crack deflection and local plastic deformation of interparticle ligaments.

In van Dommelen et al. [10], an idealized, polymeric matrix material is modeled by anisotropic Hill plasticity, and various representative volume elements are used to model the system containing dispersed voids. It is shown that a local plastic anisotropy of matrix material around the voids can very effectively replace localization by dispersed shear yielding and change the occurring hydrostatic stresses, potentially leading to toughened material behavior. However, to achieve these improvements, a morphology must be pursued that has a radially oriented structure around the dispersed voids and provides a sufficiently large amount of anisotropy.

In the present work, the influence of using moderately stiff rubber fillers or hard mineral particles for the toughening of semicrystalline polymers is investigated. For this purpose, again the anisotropic Hill model is used, with a rate dependent and hardening yield stress. The system contains a length parameter, which is the ratio of the average distance between particles and a critical distance. This length parameter is represented in the calculations by the thickness of an anisotropic layer around the particles. Large and small scale configurations are modeled by entirely isotropic or anisotropic matrix material, respectively. The work differs from the earlier work of Tzika et al. [5] by using a plane strain multiparticle model to account for the important influence of the interparticle interactions in an irregular microstructure on the local modes of deformation. However, three-dimensional
stress fields are considered for the effect of anisotropy on the hydrostatic pressure. Furthermore, the presence of either rubber or hard particles is included into the micromechanical models. Debonding particles with various interface strengths, precavitated rubber shell structures, and fully bonded hard particles are used. The combination of local anisotropy and precavitated rubber shell structures is found to promote matrix shear yielding. The presence of easily debonding hard particles is found to partly disturb the anisotropy-based toughening mechanism, whereas fully bonded particles induce large tensile hydrostatic stresses.

2. Constitutive model

In this work, the potential of plastic anisotropy, in combination with moderate or high stiffness filler particles for enhancing the toughness of a particle-modified semicrystalline polymeric material is investigated. For this purpose, an idealized polymeric material is modeled by isotropic elasticity, characterized by the Young’s modulus \( E^m \) and Poisson’s ratio \( \nu^m \), and anisotropic plasticity. For the yield behavior, the anisotropic Hill yield criterion [11] is used:

\[
F(\sigma_{22} - \sigma_{33})^2 + G(\sigma_{33} - \sigma_{11})^2 + H(\sigma_{11} - \sigma_{22})^2 + 2L\sigma_{23}^2 + 2M\sigma_{13}^2 + 2N\sigma_{12}^2 = \sigma_y^2,
\]

where \( \sigma_{ij} \) are stress components with respect to a local material vector basis and the anisotropic constants \( F, G, H, L, M \) and \( N \) are given by

\[
F = \frac{1}{2} \left( \frac{1}{R_{22}^2} + \frac{1}{R_{33}^2} - \frac{1}{R_{11}^2} \right);
\]

\[
G = \frac{1}{2} \left( \frac{1}{R_{11}^2} + \frac{1}{R_{33}^2} - \frac{1}{R_{22}^2} \right);
\]

\[
H = \frac{1}{2} \left( \frac{1}{R_{11}^2} + \frac{1}{R_{22}^2} - \frac{1}{R_{33}^2} \right);
\]

\[
L = \frac{3}{2R_{23}^2}; \quad M = \frac{3}{2R_{13}^2}; \quad N = \frac{3}{2R_{12}^2}.
\]

The constants \( R_{11}, R_{22} \) and \( R_{33} \) are the ratios of tensile yield strength of the anisotropic material to the virtual bulk tensile yield strength, \( \sigma_y \). The constants \( R_{12}, R_{13} \) and \( R_{23} \) are the ratios of yield strength in shear to the shear yield strength \( \tau_y \) of the virtual bulk material, with \( \tau_y = \sigma_y / \sqrt{3} \). A linear dependency of the yield strength \( \sigma_y \) on the effective plastic deformation measure \( \tilde{\epsilon}_p \) and a power law dependency of \( \sigma_y \) on the corresponding rate \( \dot{\epsilon}_p \) are assumed for the polymeric material:

\[
\sigma_y = \sigma_{y_0} \left\{ h\tilde{\epsilon}_p + q^\delta \left[ 1 + \left( \frac{\dot{\tilde{\epsilon}}_p}{q\tau_0} \right)^{p/n} \right] \right\}^{1/2n}
\]

where \( \sigma_{y_0} \) is the reference yield strength, \( h \) is the linear hardening parameter and \( n \) is the stress exponent of the strain rate. A rate independent contribution is introduced for strain rate values which are considerably smaller than the reference strain rate \( \tau_0 \), and is controlled by the parameter \( q \). The plastic strain measure \( \tilde{\epsilon}_p \) and the corresponding rate are, for anisotropic plasticity, assumed to be given by:

\[
\tilde{\epsilon}_p = \int_0^t \dot{\tilde{\epsilon}}_p \, dt; \quad \dot{\tilde{\epsilon}}_p = \frac{\text{tr}(\sigma \cdot \tilde{\epsilon}_p)}{\sigma_y},
\]

where \( \sigma \) is the Cauchy stress tensor and \( \tilde{\epsilon}_p \) is the plastic rate of deformation tensor. The constitutive behavior is implemented in the finite element package ABAQUS [12] to study the potential of local anisotropy for the toughening of semicrystalline polymers by the dispersion of particles. The material parameters of the fictitious polymer matrix are summarized in Table 1. As mentioned earlier, Eqs. (1)–(5), with the anisotropic stress ratios \( R_{ij} \), are applied in a local coordinate system. The transcry stallized material around the particles is assumed to have a reduced plastic resistance in the local 12 and 13 shear directions (at the particle/matrix interface, the 1-direction is perpendicular to the interface), and the reduction is controlled by the adjustable parameter \( \zeta \). Rubber fillers are modeled with neo-Hookean hyperelasticity [12], characterized by the shear modulus \( G^p = 30 \) MPa and the bulk modulus \( K^p = 1 \) GPa. The mineral filler particles are modeled as linearly elastic, with Young’s modulus \( E^p = 80 \) GPa and Poisson’s ratio \( \nu^p = 0.3 \).
3. Micromechanical model

For particle-toughened materials, a structure of dispersed particles and matrix material can be identified. The system is modeled by a finite element model of a representative volume element (RVE). The particle-modified system, having a three-dimensional nature, is simplified to a two-dimensional RVE, for which two different approaches are used. A comparison with fully three-dimensional calculations is presented elsewhere [10]. In order to capture the important effects of the essentially irregular nature of a system of dispersed particles, a multiparticle plane strain RVE [13] is used. For a representation of the triaxial stress state around a particle, an axisymmetric RVE is used, as suggested by Socrate and Boyce [14] and Tzika et al. [5].

3.1. Multiparticle plane strain RVE

To account for the irregular nature of particle-dispersed systems, a plane strain RVE with irregularly dispersed (ID model) particles is used. In Fig. 1(a), a schematic illustration of this RVE is shown. The periodicity assumption requires full compatibility of each opposite boundary pair. The corresponding kinematic and natural boundary tyings [15] for related points on opposite boundaries are given by:

\[ u|_{r_{14}} - u|_{c_4} = u|_{r_{12}} - u|_{c_1}; \]
\[ u|_{r_{14}} - u|_{c_1} = u|_{r_{23}} - u|_{c_2}; \]
\[ \sigma \cdot n|_{r_{12}} = -\sigma \cdot n|_{r_{14}}; \]
\[ \sigma \cdot n|_{r_{14}} = -\sigma \cdot n|_{r_{23}}; \]

where \( n \) denotes the outward normal of the boundary. A tensile loading condition is prescribed on vertex \( C_2 \), and is given by

\[ u_x|_{c_2} = L_0 [\exp(\dot{\varepsilon}t) - 1], \]

where \( \dot{\varepsilon} \) is set equal to the reference strain rate \( \dot{\varepsilon}_0 \) of the material. Furthermore, rotations are prevented by the following condition for the vertices \( C_1 \) and \( C_2 \):

\[ u_y|_{c_1} = u_y|_{c_2}. \]

The displacements of \( C_4 \) are unspecified and follow from equilibrium, whereas the displacements of \( C_3 \) are tied to the other vertices.

A structure with 20 volume percent irregularly dispersed particles is generated using a procedure from Hall [16] and Smit et al. [13]. In order to obtain initially straight boundaries, no particle is allowed to cross a boundary. A local orientation field for the matrix material is generated by taking the local 1-direction perpendicular to the closest particle/matrix interface, taking into account the periodicity of the structure, and is shown in Fig. 1(b). The meshes with 2622 four-noded bilinear plane strain matrix elements and either 324 elements for the precavitated rubber shell or 796 particle elements, are shown in Fig. 1(c) and (d), respectively.

3.2. Axisymmetric RVE

An axisymmetric RVE model of a staggered array (SA model) of particles is considered, which was previously used for the investigation of the micromechanics of particle-toughened polymers by Socrate and Boyce [14] and by Tzika et al. [5] and which corresponds to a body centered
tetragonal stacking of particles. A schematic visualization of the RVE, with $L_0 = R_0$, and a representation of the staggered arrangement, are shown in Fig. 2(a) and (b), respectively. The RVE is subjected to anti-symmetry conditions (with respect to point M) along the outer radius, which were introduced by Tvergaard [17,18]. Axial compatibility along the radial boundary $\Gamma_{34}$ is written as
\[
 u_z(z_0|_M - \eta) + u_z(z_0|_M + \eta) = 2u_z|_M. 
\]  
(14)

Symmetry conditions along the right and left boundaries are written as
\[
 u_z|_{\Gamma_{23}} = u_z|_{C_2}; 
\]  
(15)

The combined cross-sectional area of neighboring cells is assumed to remain constant along the axial coordinate:
\[
 [R_0 + u_r(z_0|_M - \eta)]^2 + [R_0 + u_r(z_0|_M + \eta)]^2 = 2[R_0 + u_r|_M]^2. 
\]  
(16)

Fig. 1. (a) Schematic visualization of an irregular plane strain RVE [15], (b) local material orientations, and (c), (d) finite element mesh including precavitated and uncavitated particles, respectively.
respectively. Since the axis of rotational symmetry coincides with boundary $C_{12}$, the following condition is imposed on this boundary:

$$u_r|_{C_{12}} = 0.$$  \hspace{1cm} (18)

The axisymmetric RVE is subjected to tension at a macroscopically constant strain rate:

$$u_z|_{C_{12}} - u_z|_{C_{1}} = L_0[\exp(\dot{\varepsilon}t) - 1],$$  \hspace{1cm} (19)

where the deformation rate $\dot{\varepsilon}$ is set equal to the material reference shear rate $\dot{\gamma}_0$.

The local 1-directions are assumed to be perpendicular to the closest particle/matrix interface, as is shown in Fig. 2(c). The finite element meshes of the axisymmetric SA model with 20 volume percent particles are shown in Fig. 2(c) and (d). The matrix material is represented by 196 four-noded bilinear matrix elements, whereas the precavitated rubber shell and the uncavitated particle regions are modeled by 40 and 136 elements, respectively.

### 3.3. Interface conditions

In the following, the influence of dispersed rubber (i.e. soft) inclusions versus mineral (i.e. hard) filler particles in semicrystalline polymeric material is investigated, and particularly the effect of these fillers on toughening by locally induced anisotropy. As a reference situation, voided matrix material will be used. A distinction is made between fully bonded particles, for which a tied particle/matrix interface is used, and debonding particles. For the latter, a contact algorithm [12] with a relatively low maximum tensile strength $r_i = r_{y0} = 0$ is used for the particle/matrix interface.

### 4. Rubber particles

For rubber inclusions with a relatively low stiffness, the influence of the presence of either debonded or cavitated particles on the stress and deformation fields is negligible. These systems may be modeled by voided matrix material. For voided polymeric material, the effect of a local, radially
oriented, anisotropy [10] is (i) a transition from localized deformation for isotropic material to dispersed shear yielding; and (ii) a relocation of hydrostatic stresses from the equator region (the particle equator is defined as the location where the interface normal is perpendicular to the loading direction) for isotropic material to the polar region (the pole denotes the region where the interface normal is in line with the loading direction).

Rubber filler particles with a moderately high stiffness have an effect on the deformation mechanisms. To investigate the effect of anisotropy in the presence of rubber filler particles on the triaxial stress field and local principal stresses, the SA model is used. The effect of rubber particles on the observed hydrostatic pressure, $p = -(1/3)\text{tr}(\sigma)$ is shown in Fig. 3 for both isotropic ($\zeta = 1$, i.e. large scale) and locally anisotropic ($\zeta = 3$, i.e. small scale) material. Results are shown for voided (i.e. low modulus fillers), debonded, precavitated, and well-bonded uncavitated fillers. For the latter, either debonding or cavitation may occur. This is due to tensile triaxial stresses in the rubber particle region and depends on the interface strength and cavitation resistance. The cavitation process itself is not modeled; instead, precavitated particles are represented by a rubber shell, as was previously done by Smit et al. [19]. For the isotropic systems, little difference in tensile triaxial pressure between the voided, debonded and cavitated RVEs is observed in the matrix. For the anisotropic systems, maximum negative pressures are found in the polar area; however these are slightly higher when a relatively stiff rubber particle is included. Crazelike features, such as interlamellar separation and voiding of amorphous regions [20], may be initiated in the semicrystalline matrix material under high tensile triaxial stress. For the anisotropic systems with both debonded and cavitated relatively stiff rubber inclusions, the maximum principal stresses are directed parallel to the matrix interface (i.e. perpendicular to the loading direction) in the area of maximum tensile triaxial stress. Therefore, crazelike events are expected to occur parallel to this loading direction, rather than perpendicular, as for the isotropic systems.

The mechanism of shear yielding is captured by the irregular plane strain RVE, as discussed in Section 3.1. For the voided systems (i.e. low modulus fillers), a distinct transition from localization in isotropic matrix material to massive dispersed shear yielding in the anisotropic system is observed [10]. This effect is shown in Fig. 4, where the obtained magnitude of plastic deformation, $\varepsilon_{\text{m}}^p = (\frac{2}{3} \varepsilon_p : \varepsilon_p)^{1/2}$, with $\varepsilon_p$ the plastic strain tensor [12], is displayed. Moderately stiff rubber inclusions were found to have only a small

![Fig. 3. Normalized hydrostatic pressure, $p/\sigma_{0}$, for the SA model with (a), (e) voids, (b), (f) rubber particles with interface strength $\sigma/\sigma_{0} = 0.4$, (c), (g) precavitated rubber particles, and (d), (h) fully bonded rubber particles, at $\Delta t = 0.1$, with (a)–(d) $\zeta = 1$ and (e)–(h) $\zeta = 3$.](image)
influence on the matrix hydrostatic pressure. Smit et al. [19] found precavitated load-bearing particles to stabilize local yield zones and promote matrix shear yielding for polystyrene/rubber blends. In Fig. 5, the consequences of either debonded or cavitated rubber inclusions on the plastic deformation, as predicted by the multiparticle ID model, are displayed for both isotropic and anisotropic systems. Prior to cavitation or debonding, plastic shearing remains dispersed through the matrix material for both the isotropic and the anisotropic system. However, fully bonded and uncavitated inclusions induce relatively large tensile triaxial stresses. Either debonding or cavitation of rubber particles is required to relieve these stresses, especially for the anisotropic system. With debonded moderately stiff rubber particles included, the effect of anisotropy is reduced. For this system, both matrix shear yielding and localized deformation in relatively thin ligaments are observed. The presence of well-bonded precavitated inclusions has a stabilizing effect on matrix yielding in thin ligaments, as was reported by Smit et al. [19]. Therefore, for polymer toughening by local anisotropy with moderately stiff rubber inclusions, cavitation of particles is preferred over debonding. Finally, the combined effect of moderate anisotropy ($\zeta = 1.5$) and included rubber shell structures is shown in Fig. 6. For this system, localized yielding in thin interparticle ligaments is entirely replaced by dispersed matrix shear yielding.

5. Hard particles

In Fig. 7, the obtained magnitudes of plastic deformation as obtained by the ID model are shown for systems containing both debonded and fully bonded hard filler particles, for both isotropic ($\zeta = 1$, i.e. large scale) and anisotropic ($\zeta = 3$, i.e. small scale) matrix material, respectively. For the voided isotropic matrix material (Fig. 4(a)), the macroscopic contraction in $y$-direction is small, corresponding to the growth of voids, due to stretching of relatively thin ligaments. Therefore,
for this matrix material, the inclusion of easily debonding hard particle fillers has no significant effect on the observed deformation, as can be seen by comparison with Fig. 7(a), where the interface strength $\sigma^i$ is negligibly small. For radially oriented anisotropic voided material, a dispersed mode of massive shear yielding is observed, with double shear bands at each side of a particle. As a result of matrix shearing, for the voided anisotropic system however, the voids become thinner in the macroscopically free direction (see Fig. 4(b)). Consequently, the presence of hard mineral fillers interferes with the mechanism of matrix shearing, as can be observed in Fig. 7(c). Therefore, although there is some effect of anisotropy, the mechanism of toughening by locally induced anisotropy is expected to be considerably less efficient for nonadhering hard fillers than for low modulus rubber particles. For material filled with well-bonded stiff particles, which is shown in Fig. 7(b) and (d), massive shear yielding is found for both isotropic and anisotropic matrix behavior.

In Fig. 8, the effect of hard filler particles on the normalized hydrostatic pressure, $p/\sigma_{\gamma_0}$, as predicted with the SA model, is displayed for (large scale) isotropic and (small scale) anisotropic matrix material. For voided systems, the effect of anisotropy on the triaxial stress field is a change of the position of maximum tensile values. The highest negative hydrostatic pressures are found at
the particle equators for the isotropic material (Fig. 8(a)). For the anisotropic material however, large tensile pressures are found in the polar regions (Fig. 8(d)). Therefore, the initiation of craze-like features may, for the voided system with radially oriented anisotropy, be expected to occur at the particle poles, rather than in the equator region. For easily debonding hard particles, a similar effect of local anisotropy on the tensile triaxial stresses is observed, with an increase of the peak value for the anisotropic situation. The growth of initiated microvoids is likely to occur along planes which are perpendicular to the direction of the maximum principal stress. In Fig. 9, the normalized maximum in-plane principal stress, $\sigma_{\text{max}}/\sigma_{y0}$, is depicted for the SA model, for both isotropic ($\zeta = 1$) and anisotropic ($\zeta = 3$) material, with either a void, or easily debonding or adhering hard particles. Moreover, in Fig. 10, the direction of the maximum in-plane principal stress is given for the systems containing a hard particle. For both nonadhering situations, the maximum in-plane principal stresses in the region of large tensile triaxial stress are parallel with the matrix/rubber interface. Therefore, growth of craze-like features is expected to occur perpendicular to the interface, i.e. perpendicular to the loading direction for the isotropic material and parallel to the loading direction for the anisotropic system. However, for the well-bonded systems, the maximum principal stresses in the polar region (where the largest tensile triaxial stresses are observed) are directed approximately in the loading direction. Consequently, for these systems the growth of microvoids or microcracks may be expected to occur perpendicular to the loading direction, thereby leading to macroscopic failure.

A secondary potential effect of hard mineral filler particles is an increase of the modulus of the particle-modified system [6]. In Fig. 11, the influence of hard fillers, with various interface conditions, on the normalized equivalent volume-averaged stress

$$\langle \sigma \rangle^\text{eq} = \sqrt{\frac{3}{2} \left\langle S \right\rangle : \left\langle S \right\rangle};$$  \hspace{1cm} (20)

$$\left\langle S \right\rangle = \left\langle \sigma \right\rangle - \frac{1}{3} \text{tr}(\left\langle \sigma \right\rangle) I,$$  \hspace{1cm} (21)

with

$$\left\langle \sigma \right\rangle = \frac{1}{V} \int_{x \in V} \sigma(x) \, dV$$  \hspace{1cm} (22)

the volume-averaged Cauchy stress [15], versus the imposed deformation is represented for the ID model. For both the isotropic and the anisotropic material with a reduced shear yield strength, an increase of the elastic modulus is observed for the fully bonded systems with respect to the voided situation. The particles with low interface strength

![Image of Fig. 9](image-url)

Fig. 9. The normalized maximum in-plane principal stress, $\sigma_{\text{max}}/\sigma_{y0}$, for the SA model, with (a), (d) voids, (b), (e) easily debonding hard particles, and (c), (f) fully bonded hard particles, at $\dot{\epsilon}_t = 0.1$, with (a)-(c) $\zeta = 1$, and (d)-(f) $\zeta = 3$. 

For an increase of modulus in the entire elastic region, the interface strength must be sufficiently high, for particles to remain bonded prior to macroscopic yielding. The volume-averaged response is also shown for two situations with elevated interface strength. The stress drops for these curves correspond to individual interface debonding events. The local anisotropy, which is achieved by a reduction of shear yield resistances, trivially results in a reduction of the overall yield stress.

\[ \left( \frac{\sigma'}{\sigma_{y0}} = 0.4 \right) \text{ are debonding early in the elastic region. For an increase of modulus in the entire elastic region, the interface strength must be sufficiently high, for particles to remain bonded prior to macroscopic yielding. The volume-averaged response is also shown for two situations with elevated interface strength. The stress drops for these curves correspond to individual interface debonding events. The local anisotropy, which is achieved by a reduction of shear yield resistances, trivially results in a reduction of the overall yield stress.} \]

6. Conclusions

A physically based mechanism for the toughening of semicrystalline polymeric materials due to the dispersion of particles is based on the presence of a layer of anisotropic transcrysallized material enveloping the particles [3,4,6]. In another communication [10], where hypothetical, idealized, polymeric matrix material is modeled by anisotropic Hill plasticity, and various representative volume elements are used to model a system containing dispersed voids, it is shown that local plastic anisotropy of matrix material around the voids can very effectively replace localization by dispersed shear yielding and change the occurring hydrostatic stresses, potentially leading to toughened material behavior. In this work, a similar modeling approach is used to investigate the influence of rubber and mineral (moderate and high modulus, respectively) particles on this toughening mechanism.

Moderately stiff inclusions, which are either debonded or precavitated, are found to have little effect on the triaxial stresses. Rubber shell inclusions however, stabilize local deformation zones and promote matrix shear yielding, whereas debonded rubber particles have a disturbing effect on the anisotropy-induced shear yielding mechanism. The use of mineral filler particles for toughening of polymeric materials requires debonding in order to prevent excessive negative hydrostatic pressures. These debonded hard particles show a relocation of tensile triaxial stresses to the particle polar areas by local anisotropy, similarly to anisotropic voided systems, with the maximum

![Fig. 10](image1.png)

Fig. 10. The direction (and magnitude) of the maximum in-plane principal stress, \( \sigma_{\text{max}} \), for (a), (b) easily debonding hard fillers, and (c), (d) well-bonded particles, at \( \dot{\epsilon} = 0.1 \), with (a), (c) \( \zeta = 1 \) and (b), (d) \( \zeta = 3 \).

![Fig. 11](image2.png)

Fig. 11. The normalized equivalent volume-averaged stress, \( \langle \sigma \rangle_{\text{eq}} \), vs. the imposed deformation, \( \dot{\epsilon} \), for the ID model, with (a) \( \zeta = 1 \) and (b) \( \zeta = 3 \), for both voids and hard particles, with variable interface conditions.
principal stresses directed such that crazelike features are expected parallel to the loading direction. However, the anisotropy-induced shear yielding mechanism is affected by the presence of stiff inclusions.

The potential of particle toughening of semicrystalline polymeric materials by local anisotropy is schematically indicated in Fig. 12. This figure is a further refinement of the experimentally based notion of the toughening of semicrystalline polymers by rubber and mineral fillers as was presented by Bartczak et al. [6], which showed a similar toughening potential for blends containing hard particles, than for rubber-modified systems. However, based on the simulations, the mechanism of toughening by local anisotropy is concluded to be less effective for nonadhering hard particles, which have the advantage of increasing the blend modulus, than for low stiffness rubber fillers.

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