

Prediction of the large strain mechanical behaviour of heterogeneous polymer systems by a multi-level approach

Citation for published version (APA):

Smit, R. J. M., Brekelmans, W. A. M., & Meijer, H. E. H. (1997). Prediction of the large strain mechanical behaviour of heterogeneous polymer systems by a multi-level approach. In M. L. Scott (Ed.), *Eleventh International Conference on Composite Materials, Gold Coast, Australia, July 14-18, 1997* (Vol. 5, pp. V585-V594). Woodhead.

Document status and date:

Published: 01/01/1997

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.
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**ELEVENTH INTERNATIONAL CONFERENCE
ON COMPOSITE MATERIALS**

**Gold Coast, Queensland, Australia
14th - 18th July 1997**

PROCEEDINGS

VOLUME V

**TEXTILE COMPOSITES AND
CHARACTERISATION**

**Editor
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**AUSTRALIAN COMPOSITE STRUCTURES SOCIETY
WOODHEAD PUBLISHING LIMITED**

PREDICTION OF THE LARGE STRAIN MECHANICAL BEHAVIOUR OF HETEROGENEOUS POLYMER SYSTEMS BY A MULTI-LEVEL APPROACH

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SUMMARY: Shear band formation in heterogeneous tensile bars is investigated by using an accurate homogenisation method that allows for a numerical coupling between the microscopic and macroscopic stress-strain behaviour. The procedure is based on a classical homogenisation theory, assuming local spatial periodicity of the microstructure, and supplies a consistent objective relationship between the local macroscopic deformation and the microstructural deformation of a spatially periodic representative volume element (RVE), representing the local microstructure. The method was used to predict the influence of the microstructure on localisation phenomena in plane strain hour-glass-shaped polycarbonate specimen with different volume fractions non-adhering low-modulus rubbery particles. One important result is that an irregular particle distribution seems to promote the widespread of deformation over the sample. This is believed to have an important contribution to the toughness enhancement of heterogeneous polymer systems by the addition of easily cavitating rubbery particles.

KEYWORDS: homogenisation, global-local analysis, multi-level finite elements, heterogeneous polymers

INTRODUCTION

Although heterogeneous polymer materials form a large part of the commercially important commodity plastics, a reliable constitutive model that describes their typical time-dependent mechanical behaviour accurately is still lacking. Recently, we proposed a new homogenisation technique that provides an unambiguous objective relationship between macroscopic and microscopic properties, which can be used to predict the overall mechanical behaviour of heterogeneous polymeric solids from their microstructure [1].

The homogenisation method is based on the classical concept of a local spatially periodic representative volume element (RVE) representing the local microstructure [2, 3]. The RVE enables the (numerical) decoupling of the macrostructure and microstructure. In a finite element context a unique representative volume element is assigned to each integration point of the discretised homogenised macrostructure. The macro-micro relationship

is obtained by the assumption that the local macroscopic (integration point) deformations and stresses equal the (discretised) RVE averaged deformations and stresses.

In the presented research the homogenisation method was applied to predict the shear band formation in plane strain hour-glass-shaped polycarbonate specimen with a fine dispersion of different volume fractions non-adhering low-modulus rubbery particles. A recently developed constitutive model [4, 5] was used to predict the large-strain time-dependent behaviour of the glassy polycarbonate. Finite element models of plane strain RVEs with an irregular stack of voids, representing debonded or cavitated rubbery particles, are introduced to model the microstructure. Results of the multi-level finite element simulations on the voided hour-glass-shaped polycarbonate specimen are discussed. The paper ends with some conclusions and recommendations for further research.

HOMOGENISATION STRATEGY

Consider a heterogeneous body consisting of a composite of two materials, e.g. a glassy polymer matrix with rubbery inclusions, and assume that the size of a heterogeneity is very small compared to the body dimensions. When the body is subjected to external loads, the real deformation and stress fields show large local gradients because of the inclusions. Since the heterogeneities are orders of magnitude smaller than the total body, the deformation field in the vicinity of one inclusion will be approximately the same as the deformation field near neighbouring inclusions. Such repetitive deformations strongly suggest that the macroscopic stress-strain relation at point \mathcal{P} can be determined by (i) applying the local macroscopic strain to a microstructural spatially periodic representative volume element belonging to \mathcal{P} in such a way that the RVE averaged strains equal the local macroscopic strains, and by (ii) averaging the non-uniform RVE stress field. This homogenisation strategy implies that a heterogeneous macroscopic body can be replaced by an equivalent homogeneous body with an effective macroscopic constitutive description.

Spatially periodic representative volume element

According to the aforementioned arguments, it is assumed that the representative cell deforms in a repetitive way identical to its neighbours. In an RVE, the spatial periodicity conditions follow from compatibility demands with respect to the opposite edges. The demands enforce two adjacent RVEs to show identical deformations, while neither overlapping nor separation may occur [6]. For the two-dimensional RVE depicted in Fig. 1 the compatibility of two opposite boundaries can be summarised as follows:

- (1) The shape of both boundaries is and remains identical. This can be enforced by (i) tying point positions on the left boundary Γ_{14} to their associated opposite points on the right boundary Γ_{23} and to the vertex points 1 and 2, and (ii) tying points on the upper boundary Γ_{43} to the opposite points on the lower boundary Γ_{23} and to the vertex points 1 and 4, according to

$$y_{\Gamma_{14}} = y_{\Gamma_{23}} - y_2 + y_1 \quad (1)$$

$$y_{\Gamma_{43}} = y_{\Gamma_{12}} - y_1 + y_4 \quad (2)$$

where $y_{\Gamma_{ij}}$ indicates the position vector of points on edge Γ_{ij} .

- (2) The stress vectors acting on two opposite boundaries are opposite in sign. If the stress vector acting on a boundary with outward normal \mathbf{n} is given by $\boldsymbol{\sigma} \cdot \mathbf{n}$, this

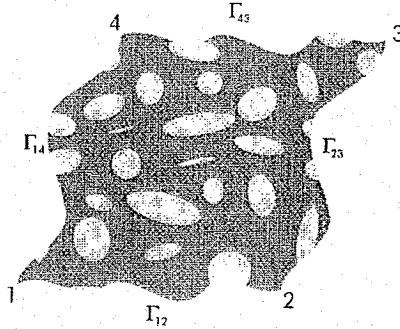


Figure 1: Plot of a typical two-dimensional (deformed) representative volume element (RVE), composed of a matrix with inclusions.

condition can be written as

$$\begin{aligned}\sigma \cdot \mathbf{n}_{\Gamma_{14}} &= -\sigma \cdot \mathbf{n}_{\Gamma_{23}} \\ \sigma \cdot \mathbf{n}_{\Gamma_{12}} &= -\sigma \cdot \mathbf{n}_{\Gamma_{43}}\end{aligned}\quad (3)$$

where s_{ij} represents a local material coordinate along boundary Γ_{ij} with point i as its origin.

The above combination of kinematical and natural boundary conditions enforces the edge shapes to deform such that the spatial periodicity conditions are preserved. The distance between the edges is prescribed by the positions of the retained vertices 1, 2, and 4. A change of retained vertex positions results in a change of edge positions and, consequently, in a change of RVE dimensions and/or orientation. Hence, only the positions of the retained vertices determine the average deformation of the RVE.

Macro-micro relationship

Macroscopic and microscopic levels are related by the assumption that the local macroscopic deformation and stress tensors at point \mathcal{P} equal the average deformation and stress tensors in the RVE assigned to \mathcal{P} . If possible cavities in the RVE are considered as relatively very soft inclusions with finite strains and negligible stresses, the deformation tensor at an arbitrary point \mathcal{P} with reference position $\mathbf{y}_0(\mathcal{P})$ is defined by

$$\mathbf{F}(\mathbf{y}_0) = (\nabla_0 \mathbf{y})^c \quad (4)$$

where the label \mathcal{P} is omitted and where \mathbf{y} denotes the actual position of \mathcal{P} in the deformed RVE, \mathbf{y}_0 is the reference position of \mathcal{P} in the undeformed RVE, and ∇_0 is the gradient operator in \mathcal{P} with respect to the reference configuration. Since the deformation tensor refers to the initially undeformed configuration at reference position \mathbf{y}_0 , the volume averaging is performed over the initial volume of the unit cell $V_{0\text{RVE}}$:

$$\bar{\mathbf{F}}_{\text{RVE}} = \frac{1}{V_{0\text{RVE}}} \int_{\mathbf{y}_0 \in V_{0\text{RVE}}} \mathbf{F}(\mathbf{y}_0) \, dV_0 \quad (5)$$

A straightforward definition of the average stress in the RVE is the volume averaged Cauchy stress tensor according to

$$\bar{\sigma}_{RVE} = \frac{1}{V_{RVE}} \int_{y \in V_{RVE}} \sigma(y) dV \quad (6)$$

where $\sigma(y)$ represents the stress tensor at position y . Notice that this averaging occurs over the current volume of the RVE because the Cauchy stress is defined in the current deformed configuration. It can be proved that the RVE averaged deformation gradient tensor can be prescribed by imposing the displacements at the angular points of the unit cell according to:

$$[u_1 \ u_2 \ u_3] = [\bar{F}_{RVE} - I] \cdot [y_{01} \ y_{02} \ y_{03}] \quad (7)$$

where u_i is defined as the displacement of angular point i .

Based on these considerations, the relationship between stresses and strains on macroscopic and microscopic levels can be obtained by the following procedure:

- (1) Replace the macroscopic heterogeneous structure by a (mesh of a) homogeneous one with an identical geometry.
- (2) Assign to each relevant macroscopic point \mathcal{P} (in fact the integration points in the macroscopic finite element mesh) a unique periodic RVE(\mathcal{P}).
- (3) Compute the local macroscopic deformation gradient tensor $F_{macro}(\mathcal{P})$ from an estimate of the macroscopic nodal displacements, and impose appropriate displacements at the angular points of the unit cell according to relation (7) with $\bar{F}_{RVE} = F_{macro}(\mathcal{P})$, so that the RVE averaged deformation tensor equals the macroscopic deformation tensor.
- (4) Compute the resulting non-uniform stress and deformation fields in the RVE(\mathcal{P}) and compute the average stress in the RVE with relation (6). The RVE averaged stress is returned to \mathcal{P} as the local macroscopic stress.

This procedure is repeated for each macroscopic (integration) point in order to compute an estimation of the macroscopic stress field. Finite element procedures are applied to converge to a deformed macroscopic equilibrium state.

CONSTITUTIVE MODELLING OF HOMOGENEOUS GLASSY POLYMERS

For the constitutive modelling of the large-strain time-dependent mechanical behaviour of the individual polymeric components, a constitutive model was applied that was recently developed by Tervoort [4] and Timmermans [5]. It concerns a so-called "Leonov model with hardening", a Maxwell model with an Eyring viscosity describing the typical visco-elastic polymeric behaviour combined with a neo-Hookean model describing strain hardening behaviour due to molecular orientation. The resulting elasto-viscoplastic constitutive model is assumed to predict the strain rate, temperature and history dependent yield, intrinsic strain softening and subsequent strain hardening of glassy polymers. In the following the Leonov model with hardening is summarised.

In the Leonov model with hardening, the Cauchy stress σ is additively decomposed in an effective stress s and a hardening stress r , according to:

$$\sigma = s + r \quad (8)$$

The hardening stress is described by a simple neo-Hookean model:

$$\mathbf{r} = H\tilde{\mathbf{B}}^d \quad (9)$$

where H is the strain hardening modulus, and $\tilde{\mathbf{B}}$ represents the isochoric left Cauchy-Green strain tensor, defined as $\tilde{\mathbf{B}} = J^{-2/3}\mathbf{F}\cdot\mathbf{F}^c$, where \mathbf{F} is the actual deformation gradient tensor and $J = \det(\mathbf{F})$ represents the relative volume change of the material.

The effective stress, determining the visco-elastic behaviour of the material, is defined by the Leonov model, originally proposed by Leonov [7] as an incompressible model to describe the rheological behaviour of polymer melts, later extended by Baaijens [8] to its current compressible form. The model is based on the multiplicative decomposition of the actual deformation gradient tensor \mathbf{F} in an isochoric elastic part $\tilde{\mathbf{F}}_e$, an isochoric plastic part \mathbf{F}_p and a volumetric part $J = \det(\mathbf{F})$, according to

$$\mathbf{F} = J^{1/3}\tilde{\mathbf{F}}_e \cdot \mathbf{F}_p \quad (10)$$

Here, \mathbf{F}_p defines the state that would instantaneously be recovered if all loads were removed from the sample. If it is assumed that the total spin tensor equals the elastic spin tensor, it can be proved that the elastic shape deformation, defined by the isochoric elastic left Cauchy-Green deformation tensor $\mathbf{B}_e = \tilde{\mathbf{F}}_e \cdot \tilde{\mathbf{F}}_e^c$, is given by the following rate equation

$$\overset{\nabla}{\mathbf{B}}_e = D_p \cdot \mathbf{B}_e + \mathbf{B}_e \cdot D_p - \frac{2}{3}\text{tr}(\mathbf{B}_e)\mathbf{I} \quad (11)$$

Here, D_p and \mathbf{I} represent the plastic strain rate tensor and the unit tensor, respectively, and the upper triangle defines the Truesdell objective rate as

$$\overset{\nabla}{\mathbf{B}}_e = \dot{\mathbf{B}}_e - \mathbf{L} \cdot \mathbf{B}_e - \mathbf{B}_e \cdot \mathbf{L} \quad (12)$$

where \mathbf{L} denotes the velocity gradient tensor. Assuming only small volumetric deformations, the deviatoric part of the effective stress \mathbf{s}^d is related to \mathbf{B}_e^d through the generalised Hookean relation $\mathbf{s}^d = G\mathbf{B}_e^d$, with G representing the shear modulus, while the hydrostatic part of the effective stress is coupled to the volume deformations by $\mathbf{s}^h = \kappa(J-1)\mathbf{I}$, with κ the bulk modulus.

With this system of equations, defining the compressible Leonov model, the effective stress \mathbf{s} is determined once the dissipative plastic strain rate D_p is known. Tervoort *et al.* [4] proposed to apply the Leonov model with hardening to predict the large-strain deformation behaviour of glassy polymers. They introduced a generalised Newtonian flow rule with a stress dependent Eyring viscosity η to relate the deviatoric effective stress to the plastic deformation rate as $D_p = \mathbf{s}^d/(2\eta)$. Timmermans [5] added pressure dependence (p) and intrinsic softening effects (D) to the viscosity equation, using a softening evolution equation originally proposed by Hasan *et al.* [9]:

$$\eta = \frac{A_0 s}{e^{D\sinh((s-\mu p)/\tau_0)}} \quad , \quad s = \sqrt{(\text{tr}(\mathbf{s}^d \cdot \mathbf{s}^d)/2)} \quad , \quad p = -\frac{1}{3}\text{tr}(\sigma) \quad (13)$$

$$\dot{D} = (1 - D/D_{inf})\frac{hs}{\eta\sqrt{2}} \quad (14)$$

with the material parameters A_0 as a time constant, τ_0 as a characteristic stress, μ as a pressure coefficient, h as the softening rate and D_{inf} as the softening limit.

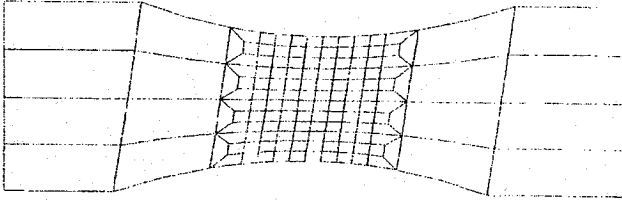


Figure 2: Geometry and mesh of undeformed plane strain hour-glass-shaped tensile specimen.

RESULTS

Numerical simulations of shear band formation in heterogeneous plane strain hour-glass-shaped tensile specimen have been performed, using the homogenisation method and Leonov model introduced in the previous sections. The heterogeneous systems considered are polycarbonate with 0, 10 and 30 vol.% dispersed non-adhering low-modulus rubbery particles.

The initial geometry and mesh of the macroscopic plane-strain tensile specimen is shown in Fig. 2. The specimen is chosen to be asymmetrical in order to obtain a preferred and thus controlled shear direction (the curved edges, with identical radii, are shifted with respect to each other). The geometries and meshes of the plane strain periodic RVEs, representing the voided microstructures near the macroscopic integration points, are visualised in Fig. 3. The rubbery particles are considered as being voids, since the properties of the non-adhering low-modulus particles are expected not to influence the mechanical behaviour of the unit cells under the expected positive hydrostatic stress states. The time-dependent mechanical behaviour of the glassy polycarbonate is modelled by the Leonov model with hardening, using the following set of material parameters adopted from Timmermans [5]: $G = 884$ MPa, $\kappa = 1916$ MPa, $A_0 = 8.172 \cdot 10^{25} \text{ s}^{-1}$, $\tau_0 = 0.707$ MPa, $\mu = 0.098$, $h = 120 \text{ s}^{-2}$, $D_{inf} = 43$ and $H = 27$ MPa.

The averaged stress-strain and volume-strain responses of the RVEs under isothermal uniaxial plane strain extension (strain rate 0.01 s^{-1}) are depicted in Fig. 4. The homogeneous

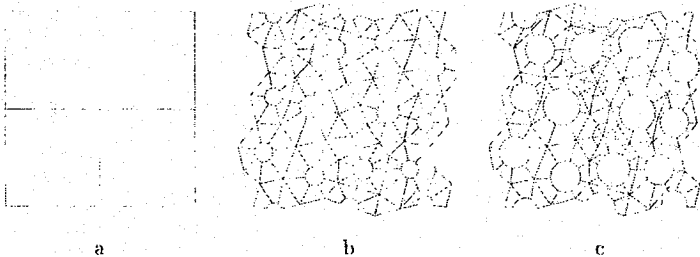


Figure 3: Geometry and mesh of the spatially periodic RVEs that represent the local microstructure in the macroscopic hour-glass-shaped sample: (a) solid polycarbonate; (b) polycarbonate with 10 vol.% voids; (c) polycarbonate with 30 vol.% voids.

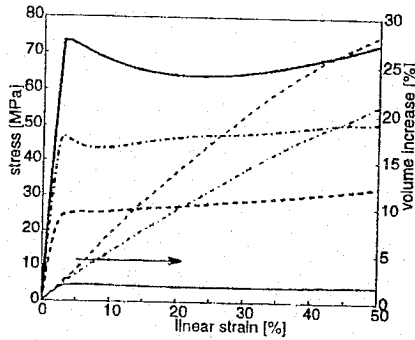


Figure 4: RVE averaged tensile stress (black) (defined with respect to the deformed cross sectional area of the RVE) and volume increase (grey) vs. natural strain for isothermal plane strain uniaxial tensile tests on the RVEs shown in Fig. 3: (solid) no voids; (dash-dot) 10 vol.% voids; (dashed) 30 vol.% voids.

polycarbonate shows the typical mechanical behaviour that is representative for a range of glassy polymers: a small-strain elastic response, followed by intrinsic strain softening and subsequent strain hardening. The maximum volume increase is approximately 3% and thus almost negligible.

The introduction of voids has a pronounced influence on the deformation behaviour: stiffness and yield stress are reduced considerably, the stress drop by intrinsic strain softening is diminished and the volume increase becomes relatively large. The reduction of stiffness and yield stress is evidently caused by the decrease of load-bearing material by the presence of voids. The strong volume response is also caused by the presence of (zero-bulk-modulus) voids. The reduction of strain softening originates from the irregular distribution of the voids. Due to this irregular distribution, the shear bands are formed between the holes in an arbitrary order and subsequently stabilised by the strain hardening, before they coalesce into large shear bands that dominate the RVE deformation behaviour. This is in contrast with the predictions of the heterogeneous material behaviour by computations on RVEs with regularly stacked inclusions. Then the overall material behaviour shows a considerable softening behaviour because all the shear bands in the material are formed at the same moment and join each other simultaneously. This is illustrated in Fig. 5 where the overall stress-strain response of a voided RVE with an irregular stack is compared with the response of an RVE with a cubic stack. Since intrinsic strain softening is known to be the main cause of unstable material behaviour, often resulting in a concentration of deformation in a relatively small volume of the specimen (e.g. shear bands), the decrease of softening will probably evolve in a more stable overall material behaviour.

The macroscopic specimen is stretched with a constant strain rate of 0.01 s^{-1} to a total nominal strain of 10%. The predicted macroscopic nominal stress responses (defined with respect to the undeformed cross sectional area of the specimen) are shown in Fig. 6. The equivalent strain and volume increase in the macroscopic samples at 4 and 10% strain are plotted in Fig. 7 and 8, respectively.

Pure polycarbonate shows a characteristic deformation behaviour: at a nominal strain

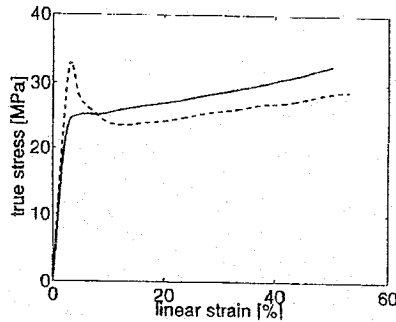


Figure 5: True Stress vs. natural strain for RVEs with voids in an irregular stack (solid) and in a cubic stack (dashed). The void volume fraction of both RVEs is 30%.

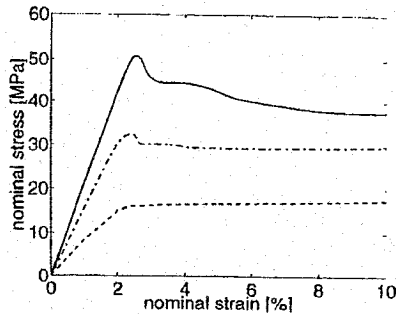


Figure 6: Nominal stress vs. nominal strain of the macroscopic polycarbonate specimen, predicted with no voids (solid), 10 vol.% voids (dash-dot) and 30 vol.% voids (dashed).

of 0-2%, initial stiff overall elastic response; 2-3%, load fall by the shear band formation (45° with load direction); 3-4.5%, some stabilisation and growth of the first shear band; 4.5-5.5%, load fall by the second shear band formation (-45° with the load direction); 5.5-10%, formation and growth of the neck. Hence, the behaviour of the pure polycarbonate is characterised by a strong concentration of deformation in the shear bands (up to 75% local strain), neck formation and a large unstable post-yield stress drop (much apparent softening). The volume plots show a small, almost negligible volume increase of approximately 3%.

The addition of voids results in a broader shear band under an angle of 22° (10% voids) or -5° (30% voids). As a result, the maximum equivalent strain at 10% nominal strain decreases with a factor 2-4. A decrease in stiffness and yield stress results, and the post-yield stress drop diminishes. The contour plot of the volume strain shows that the addition of 10% voids leads to concentration of volume deformation at small macroscopic strains. A higher volume fraction results to a broader dilatation zone. This is probably caused by decreasing intrinsic strain softening in the voided specimen, as shown in Fig.

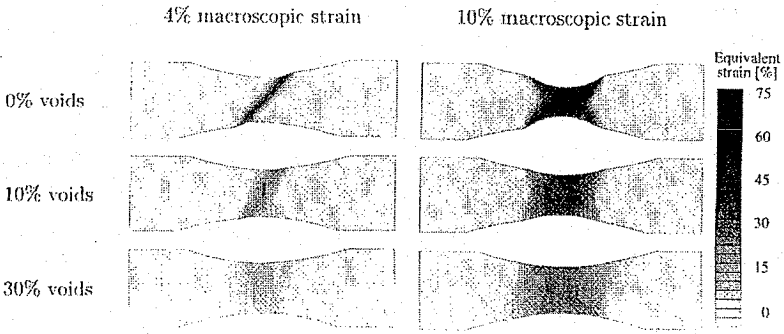


Figure 7: Contours of equivalent strain of the deformed polycarbonate sample with 0, 5 and 10 vol.% voids, at a nominal macroscopic strain of 4 and 10%.

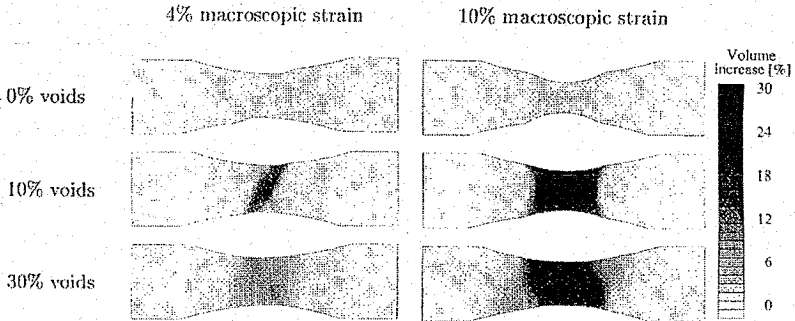


Figure 8: Contours of the volume increase of the deformed polycarbonate samples with 0, 5 and 10 vol.% voids, at a nominal macroscopic strain of 4 and 10%.

4: at 10% voids the decreased but still significant intrinsic softening results in relatively unstable post-yield deformation behaviour and thus into the formation of narrow shear bands; at 30% voids the heterogeneous specimen shows no intrinsic softening at all and the deformation spreads out over the whole specimen. It should be noted that the inhomogeneous microstructure implies that a low macroscopic strain could correspond to high local microscopic strains (e.g. stretching of the ligaments between the inclusions).

Many experimental evidence is available indicating that toughness is enhanced by the addition of easily cavitating and/or non-adhering rubbery particles [10, 11]. However, the basic mechanism of this toughness enhancement was not really understood yet. The results of these multi-scale analyses are believed to indicate that the random stack of easily cavitating rubbery particles has a pronounced influence on the toughness enhancement of heterogeneous polymeric systems by the removal of the intrinsic softening behaviour.

CONCLUSIONS AND RECOMMENDATIONS

A new homogenisation method has been used to predict the shear band formation in plane strain hour-glass-shaped heterogeneous polycarbonate specimen. The homogenisation method results in an unambiguous objective relationship between micro- and macroscopic properties. It has been demonstrated that the deformation behaviour changes dramatically by the addition of a high volume fraction of a fine dispersion of low-modulus non-adhering particles: the irregularly distributed heterogeneities result in a decreased intrinsic softening, a more stable deformation and a widespread of strains. This is thought to be a strong indication that the interaction between randomly distributed cavitating or non-adhering particles increases the toughness of polymeric systems.

Future research will focus on the influence of well-adhering cavitating rubbery particles on the overall deformation behaviour of heterogeneous polymers. Furthermore, the effect of local failure on the macrostructural behaviour will be investigated.

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