Prediction of the mechanical behaviour of heterogeneous polymer systems, based on their microstructure

Citation for published version (APA):
Smit, R. J. M., Brekelmans, W. A. M., Meijer, H. E. H., & Govaert, L. E. (1997). Prediction of the mechanical behaviour of heterogeneous polymer systems, based on their microstructure. In *Deformation, Yield and Fracture of Polymers : 10th International Conference, 7-10 April, 1997, Cambridge, UK* (pp. 494-497). The Institute of Materials.

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.
• 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: 26. Mar. 2025

PREDICTION OF THE MECHANICAL BEHAVIOUR OF HETEROGENEOUS POLYMER SYSTEMS, BASED ON THEIR MICROSTRUCTURE

R.J.M. Smit*, W.A.M. Brekelmans*, H.E.H. Meijer* and L.E. Govaert*

The overall mechanical behaviour of heterogeneous hour-glass-shaped polycarbonate tensile bars was predicted from the microstructural properties by using a multi-level finite element method (MLFEM). The multi-level method, providing an objective relationship between the deformation and stresses at macro level (FE model of tensile bar) and micro level (FE model of matrix material with inclusions), is introduced briefly. The simulations indicate that the addition of a fine dispersion of non-adhering low-modulus rubbery particles seems to result in toughening of the sample: the deformation spreads out over the whole specimen. The basic mechanism for this toughness enhancement is explained.

INTRODUCTION

Microstructural adaptations in polymer systems are often used for mechanical property enhancement. In particular the toughness and impact resistance of glassy and semicrystalline polymers are strongly improved by the addition of a fine dispersion of a low modulus phase (1,2). Accurate predictions of these property enhancements are of major importance for polymer blend development and applications. These property predictions are often inaccurate or impossible by (i) complicated constitutive behaviour of the microstructure (e.g. large deformations, visco-elastic behaviour, crazing) and (ii) the unclear relationship between the properties on user scale (typical length scale $10^{-3} - 10^{-1}$ m) and morphological scale (typical length scale $10^{-8} - 10^{-6}$ m).

This paper presents a novel approach to simulate the large strain time-dependent mechanical behaviour of heterogeneous polymeric solids based on the microstructure. A recently developed large-strain elasto-viscoplastic constitutive equation (3,4) for the modelling of pre-failure mechanical behaviour of polymer glasses, is combined with a novel accurate objective homogenisation method that has recently been developed (5). The method is used to study the influence of the microstructure on shear band formation in plane strain hour-glass-shaped polycarbonate specimen with a fine dispersion of non-adhering low-modulus rubbery particles.

HOMOGENISATION OF HETEROGENEOUS SYSTEMS

The homogenisation method originates from the classical assumption of local spatial periodicity of the morphology (6), allowing a macroscopic structure with different microstructures at different points. The microstructure is identified by a representative volume element (RVE, e.g. a cube with irregularly distributed spherical inclusions) with periodic boundary conditions. The macro-micro relationship is obtained by the assumption that

---

*Faculty of Mechanical Engineering, Eindhoven University of Technology,
P.O. Box 513, 5600 MB Eindhoven, The Netherlands

the local macroscopic deformation and stress tensors are equal to the RVE averaged deformation and stress tensors. Consequently, the macroscopic displacement field is decoupled from the microscopic displacement field: to each macroscopic point (in fact an integration point in the macroscopic mesh) a periodic RVE is assigned that relates the macroscopic deformation tensor to the macroscopic stress tensor. This decoupling has severe implications for the finite element formulation of this homogenisation method. The macroscopic structure is considered as a homogeneous solid and is discretised accordingly. In each macroscopic integration point the local deformation tensor is applied to the associated discretised RVE, such that the RVE averaged deformation tensor equals the local macroscopic deformation tensor. The resulting inhomogeneous RVE stress field is averaged and returned to the macroscopic integration point as the local macroscopic stress. This procedure is repeated for each macroscopic integration point in order to obtain an estimation of the macroscopic stress field. Multi-level (macro-micro) finite element procedures are applied to converge iteratively to a deformed macroscopic equilibrium state.

CONSTITUTIVE MODELLING OF GLASSY POLYMERS

For the constitutive modelling of the large-strain time-dependent mechanical behaviour of the individual polymeric components, a constitutive model has been adopted from Tervoort *et al.* (3) and Timmermans (4). 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.

RESULTS AND DISCUSSION

Numerical simulations of shear band formation in heterogeneous plane strain hour-glass-shaped tensile specimen have been performed, using the aforementioned homogenisation method and Leonov model. The systems considered are pure polycarbonate and polycarbonate with 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. 1a. 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 microstructures at the macroscopic integration points, are visualised in Fig. 1b,c. The rubbery inclusions 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 material parameters adopted from Timmermans (4).

The predicted averaged stress-strain responses of the RVEs under isothermal uniaxial plane strain extension (strain rate 0.01 s^{-1}) are depicted in Fig. 2. The homogeneous polycarbonate shows the typical mechanical behaviour that is representative for a range of glassy polymers: an initial elastic response, followed by intrinsic strain softening and

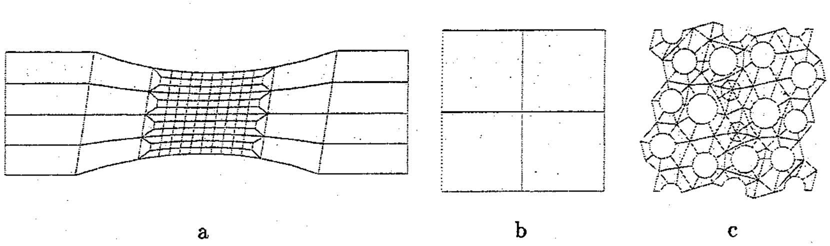

Figure 1: Geometry and mesh of (a) the undeformed plane strain hour-glass-shaped tensile specimen, and the spatially periodic RVEs that represent the local microstructure: (b) homogeneous polycarbonate, (c) polycarbonate with 30 vol.% voids.

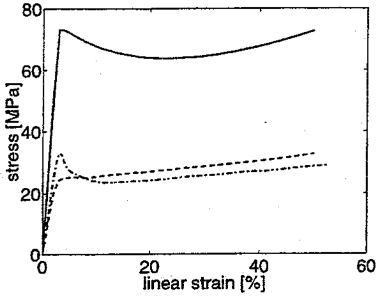

Figure 2: Volume averaged stress-strain responses for polycarbonate RVEs containing no voids (solid), and 30 vol.% voids in an irregular stack (dashed) and a regular cubic stack (dash-dot).

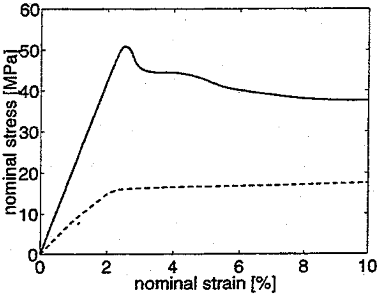

Figure 3: Predicted mechanical response of the macroscopic polycarbonate specimen with no voids (solid) and 30 vol.% voids (dashed).

subsequent strain hardening. The introduction of voids has a pronounced influence on the deformation behaviour: stiffness and yield stress are reduced considerably and the stress drop by intrinsic strain softening is diminished. Evidently the reduction of stiffness and yield stress is caused by the decrease of load-bearing material by the presence of 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 strain hardening, before they coalesce into large shear bands that dominate the RVE deformation behaviour. This is in contrast with the behaviour of RVEs with regularly stacked inclusions: a cubic stack causes a simultaneous formation of shear bands between all the holes, and the overall material behaviour is dominated by the shear band formation, which is accompanied by intrinsic softening (compare the stress-strain responses of the RVEs with regular and irregular distributed voids in Fig. 2). Since intrinsic strain softening is known to be the main cause of unstable material behaviour, often resulting in shear band formation, the decrease of softening will 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

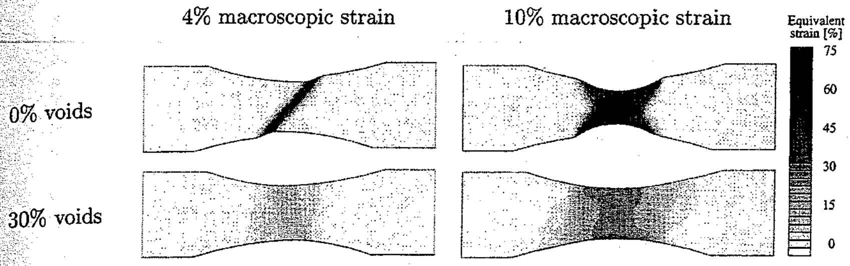

Figure 4: Contours of equivalent strain in the deformed polycarbonate sample with 0 and 30 vol.% voids at a nominal macroscopic strain of 4 and 10%.

total nominal strain of 10%. The predicted macroscopic nominal stress responses are shown in Fig. 3. The equivalent strain in the macroscopic samples at 4 and 10% strain is plotted in Fig. 4. Pure polycarbonate shows a characteristic deformation behaviour: at a nominal strain of 0-2%, initial stiff overall elastic response; 2-3%, load fall by shear band formation; 3-4.5%, some stabilisation and growth of the first shear band; 4.5-5.5%, load fall by a second shear band formation; 5.5-10%, formation and growth of the neck. Hence, the behaviour of the pure polycarbonate is dominated 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 occur. The addition of voids results in a broader shear band almost perpendicular to the load direction. Instead of a neck, a broad dilatation zone is formed and the deformation is distributed over a large part of the sample. As a result, the maximum equivalent strain at 10% nominal strain decreases with a factor 3. A decrease in stiffness and yield stress results, and the post-yield stress drop disappears.

Many experimental evidence is available indicating that toughness is enhanced by the addition of easily cavitating and/or non-adhering rubbery particles (1,2). However, the basic mechanism of this toughness enhancement was not really understood yet. The results of the multi-scale analyses 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.

REFERENCES

1. van der Sanden, M.C.M., De Kok, J.M.M., Meijer, H.E.H., *Polymer* **35** (1994) 2995.
2. Magelhães, A.M.L., Borggreve, R.J.M., *Macromolecules* **28** (1995) 5841.
3. Tervoort, T.A., Klompen, E.T.J., Govaert, L.E., *J. Rheol.* **40** (1996) 779.
4. Timmermans, P.H.M., 'Evaluation of a constitutive model for solid polymeric materials'. Ph.D. thesis, Eindhoven University of Technology, Eindhoven, The Netherlands (1997).
5. Smit, R.J.M., Brekelmans, W.A.M., Meijer, H.E.H., *Comp. Meth. Appl. Mech. Engng.*, submitted (1996).
6. Bensoussan, A., Lions, J.L., Papanicolaou, G., 'Asymptotic Analysis for Periodic Structures'. North-Holland, Amsterdam (1978).