Network density : a key factor in macroscopic toughness?

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H.G.H. van Melick, L.E. Govaert and H.E.H. Meijer

Eindhoven University of Technology,
Faculty of Mechanical Engineering,
Section Materials Technology,
P.O. Box 513, 5600 MB Eindhoven, the Netherlands

/Introduction

Although amorphous polymers like Polystyrene (PS, cd-boxes) and Polycarbonate (PC, cd's) are similar materials, their macroscopic deformation behaviour in tension is quite different (**fig.1a**).

- PS: crazing → brittle, small macroscopic strains
- PC: necking → ductile, large macroscopic strains

fig.1a *Brittle and tough*

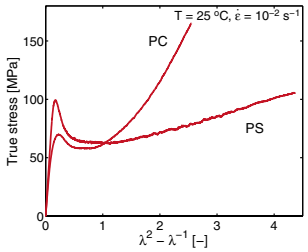

fig.1b *Compression tests*

PS suffers from extreme localisation due to (**fig.1b**):

- much strain softening. Elimination of strain softening inhibits localisation and brittle fracture [1, 2].
- weak strain hardening. Strain hardening: entropic contribution of entangled polymer network [3]

/Objective

Determine relation between molecular structure and post-yield behaviour. Alter molecular structure of PS:

- blending with Polyphenylene oxide (PS/PPO)
- cross-linking with DEGDMA (x-PS)

/Results and conclusions

Materials are characterised by DMTA.

- Dynamic modulus in rubbery region → molecular weight between entanglements → network density

With increasing %PPO in PS/PPO and cross-linker in x-PS, the rubber modulus (G_{N^0}) and thus the network density (ν_e) increase (**fig.2**).

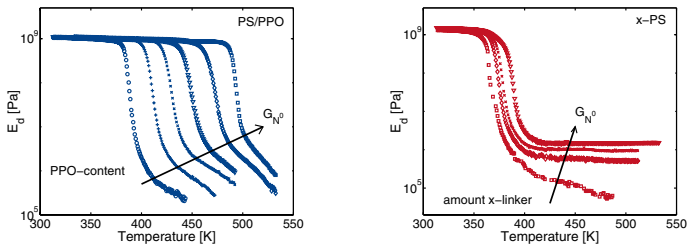

fig.2 *DMTA PS/PPO (left) and x-PS*

Compression tests are performed to determine the intrinsic properties (strain softening and hardening).

- In Gaussian theory → true stress proportional to $\lambda^2 - \lambda^{-1}$ → strain hardening modulus (G_p)

With increasing network density in PS/PPO and x-PS the strain hardening modulus increases (**fig.3**).

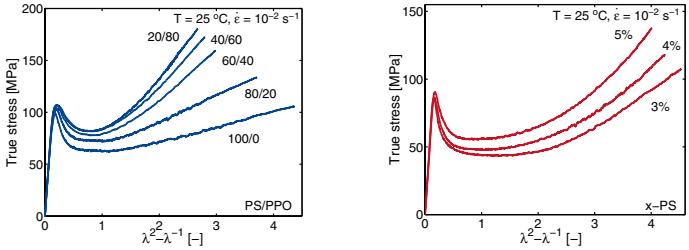

fig.3 *Compression tests PS/PPO (left) and x-PS*

Strain hardening modulus vs. network density gives a linear relationship for PS/PPO and x-PS (**fig.4a**).

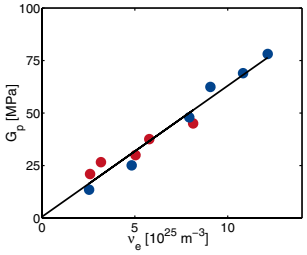

fig.4a G_p *vs.* ν_e

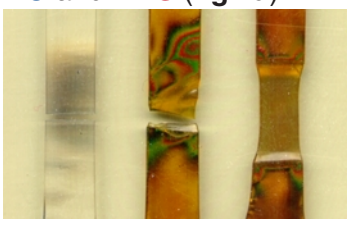

fig.4b *PS/PPO 100/0 to 20/80: crazing to shear yielding*

For high PPO content in the PS/PPO blends ductile macroscopic deformation behaviour is observed in tension (**fig.4b**), whereas x-PS remains brittle. The amount of softening (yield-drop) of PS/PPO in compression reduces strongly with increasing %PPO and temperature (**fig.3** & **5a**, encircled: ductile in tension).

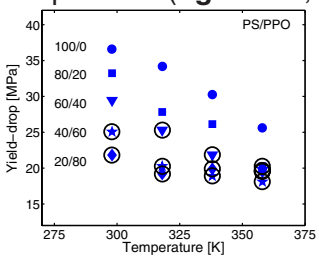

fig.5a *Yield-drop in PS/PPO*

fig.5b *Embrittled PC*

As under a certain threshold of yield-drop ductile behaviour is observed, strain softening appears to be the key factor in macroscopic toughness. This is confirmed by embrittlement of strongly annealed PC in which yield-drop has increased considerably (**fig. 5b**).

/References:

[1] TERVOORT, T.A. *Constitutive modelling of polymer glasses: finite nonlinear viscoelastic behaviour of polycarbonate, PhD thesis, Eindhoven University of Technology, Eindhoven, The Netherlands, 1996*

[2] SMIT, R.J.M *Toughness of heterogeneous polymeric systems: a modelling approach, PhD thesis, Eindhoven University of Technology, Eindhoven, The Netherlands, 1998*

[3] HAWARD, R.N.: *Strain hardening of thermoplastics, Macromolecules, v26, n22, p5860-5869, 1993*