CONSTITUTIVE ANALYSIS OF RUBBER COMPOUNDS WITH VARYING MOLECULAR STRUCTURE

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1. Introduction

The numerical dimensioning of injection heads and tools for the extrusion of rubber profiles requires both, the experimental investigation and numerical consideration of the viscoelastic material behavior. Usage of finite element software allows drastic reduction of a time-consuming empiric adaptation process for the design of extrusion tools. In this context, viscoelasticity covers three different aspects:

- *shear-thinning:* The dynamic viscosity decreases with increasing shear strain rate, thus the material is called non-Newtonian. Experimental characterization of this behavior is commonly based on capillary viscometry (see Figure 1(a)). In such tests, a piston presses (at controlled velocity v) the heated material out of a reservoir into and through a capillary die (with a specific ratio of length to diameter L/D). After reaching a steady flow state within the die, the pressure p of the melted material is measured in the reservoir, in the immediate vicinity of the capillary entry. One capillary experiment allows various piston velocities, depending on available melt in the reservoir.
  Evaluation of experiments consists of transforming measured pressure-velocity curves into a relationship between viscosity η and shear strain rate $\dot{\gamma}$ [1].
- *die swell phenomenon:* Heated rubber melts expand upon exiting from capillary tubes. This swelling of the extrudate on emerging from a die is typical for non-Newtonian viscoelastic liquids. Investigation of this behavior is done during a capillary experiment under usage of a swell value measuring unit (see Figure 1(b)), giving access to a cross-sectional area ratio χ of strand diameter d and diameter of employed die D. After measurement of the melt pressure p, the piston stops. Then, for each piston velocity v, the strand diameter is measured under consideration of the relaxation of the material [2].
- *elasticity:* Performance of tests with a rubber process analyzer allows the separation of the stored energy in the melt, representing the elastic portion, and the energy dissipated as heat, representing the viscous portion. This type of torsional rheometer strains a sample in shear by oscillating the lower die sinusoidally (see Figure 1(c)). The upper die is fixed and connected with a measuring device, which records the applied bending moment S^*.
  The latter is split into an elastic component S' and a viscous component S'' by means of a Fourier transformation. Afterwards, by applying a form factor the storage modulus G' and the loss modulus G'' for all investigated angular frequencies ω are provided [3].

2. Materials

The tested materials are non-vulcanized rubber compounds used in industry, containing mainly EPDM (ethylene-propylene-diene-monomer) and carbon black and chalk as filler material. In total six compounds were investigated, two different chemical conditions of EPDM, amorphous and crystalline, with three different filler degrees each.

Such rubber compounds are used for window sealings, pipeline constructions, bridge dilatations and various parts of cars. Thus, the goal of this contribution is identification of crystallization and filler degree on the viscoelastic properties of EPDM rubber compounds.

Fig. 1: Devices for experimental investigation of viscoelastic properties of rubber blends: (a) capillary viscometer for viscosity, (b) swell value measuring unit for die swell phenomenon, (c) rubber process analyzer for elastic moduli.

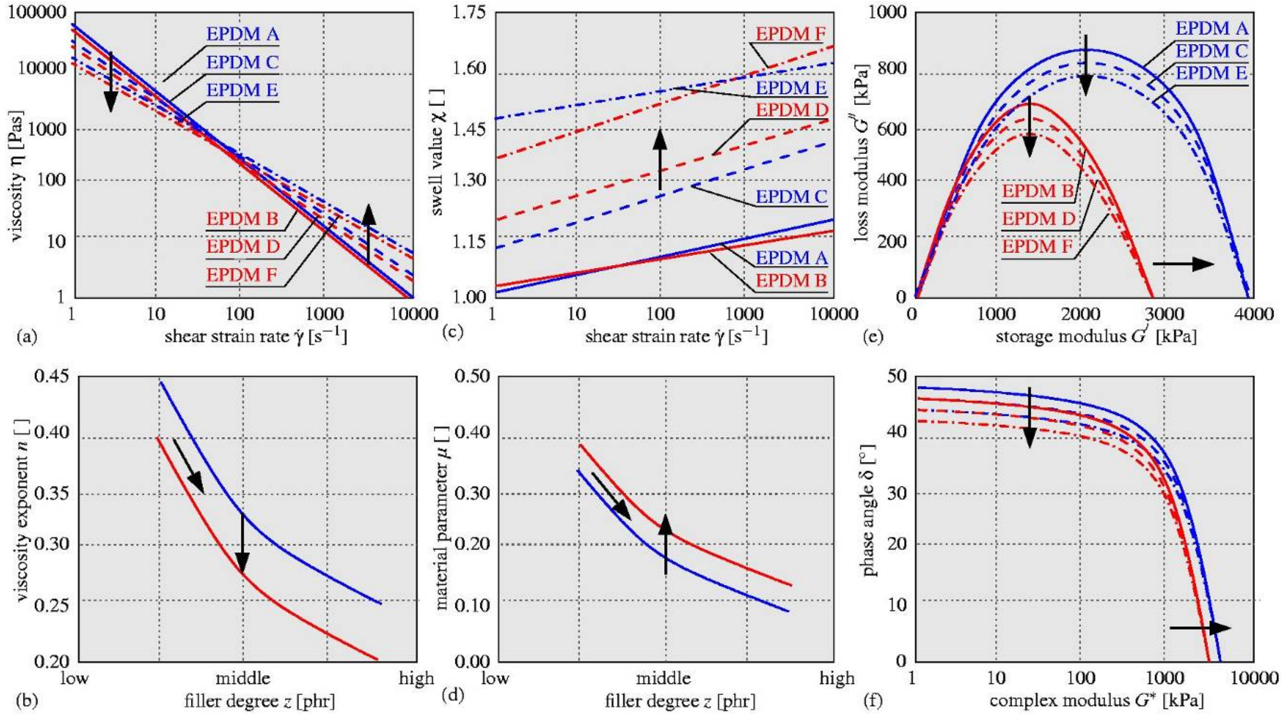

Fig. 2: Characteristics of viscoelasticity of investigated rubber compounds: (a) viscosity curves, (b) influence of filler degree on shear-thinning behavior, (c) relationship of swell value and shear strain rate, (d) influence of filler degree on material parameter describing die swell phenomenon, (e) COLE-COLE diagrams, (f) BLACK diagrams.

3. Dynamic viscosity – capillary viscometry

For the description of the nonlinear correlation between dynamic viscosity and shear strain rate (see Figure 2(a)) the power law after OSTWALD and DE WAELE is used, introducing two constants, the consistency factor k and the viscosity exponent n.

The application of the power law allows identification of the influence of both, the crystallization degree of the basic polymer and the amount of used filler material, on the shear-thinning properties of the investigated rubber compounds. Using a double-logarithmic plot for the viscosity function, a clear trend for both properties is detected. Using EPDM with amorphous chemical structure, i.e. a low crystallization degree, leads to an increase of the consistency factor k, i.e. the absolute value of the viscosity curve. Furthermore, an increase of the filler content of the rubber compound yields a decrease of the viscosity exponent. Thus, shear-thinning increases with increasing filler degree.

4. Die swell – swell value measuring unit

The test parameter of a capillary experiment, length L and diameter D of the employed capillary die, the melt temperature T, and the corresponding shear strain rate $\dot{\gamma}$, have a certain influence on the die swell of non-Newtonian fluids. With an empiric relationship [2] these influences on the die swell phenomenon are investigated. The equation allows characterization of the die swell phenomenon by means of only one material parameter μ. Using this equation, the influence of crystallization and filler degree on the die swell is covered by only one parameter, too. This material parameter is always higher for rubber compounds consisting of polymers with crystalline structure. For different filler degrees, a decrease of die swell with increasing filler content is detected.

5. Dynamic moduli – Huet model

For experimental investigation of rubber compounds by torsional rheometry, the simplified HUET model reproduces the viscoelastic behavior satisfactory [3]. This nonlinear spring-dashpot model requires only two material parameters, the creep compliance J_0 and the exponent m_1 of the nonlinear dashpot. Its application allows identification of the influence of both, the crystallization degree of the basic polymer and of the amount of used filler material, on the dynamic moduli of rubber compounds.

Under consideration of two material parameters, a clear trend for both properties is detected. Using EPDM with an amorphous chemical structure leads to an increase of the viscous properties of the corresponding rubber compound, i.e. to an increase of the phase angle. Furthermore, an increase of the filler content of the rubber compound yields an increase of the viscous properties, i.e. an increase of both the loss modulus and the phase angle, respectively.

6. References

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