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Exploring the Nonlinear Viscoelasticity of a High Viscosity Silicone Oil with LAOS

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Abstract:
Measurements and modeling of the nonlinear viscoelastic properties of a high viscosity silicone oil (polydimethylsiloxane, PDMS) are reported. LAOS test were performed with a high precision rotational rheometer to probe the nonlinear response. The measurements show that the material can be safely considered linear below strain amplitude 1. The viscous Lissajous-Bodwitch curves indicate intracycle shear thinning, whereas the elastic Lissajous-Bodwitch curves indicate intracycle strain stiffening in the nonlinear regime. Secondary loops in some of the measured viscous stress curves are attributed to a non-sinusoidal shear rate signal. A multi-element White-Metzner model is used as a constitutive equation, which accurately describes the LAOS data in all measured cases. Based on the extension of the measured data by simulations, nonlinear properties are analyzed both for the elastic and for the viscous part. It is observed that the nonlinearity considerably increases the weight of the higher harmonics in the shear stress signal. It is predicted that the viscous nonlinearity has a maximum around 50 rad/s angular frequency, and that the elastic nonlinearity becomes nearly independent of the angular frequency above 30 rad/s.

Key words:
LAOS, PDMS, nonlinear viscoelasticity, White-Metzner model, intracycle shear thinning

1 INTRODUCTION

Silicone oils are important viscoelastic fluids, with a broad range of use from fundamental science to industrial applications [1 – 12]. Linear viscoelastic properties of silicone oils, usually with rather low zero-shear viscosity, have been studied in a number of papers, as well as their shear thinning behavior [2, 13]. High viscosity silicon oils were investigated only in a few cases [14 – 17], including high-frequency measurements [18, 19]. If the deformations of the silicone oil are large enough, nonlinear viscoelastic properties control its material response. This is of increasing importance, both in polymer research and in engineering applications. Amplitude sweep test were already published in our previous work [19], but a more extensive analysis has not been done yet on the nonlinear viscoelasticity of high viscosity silicone oils, to the best of our knowledge. Now we extend our former investigations and report on LAOS measurement with high viscosity PDMS to explore deeper the nonlinear viscoelastic properties. We use the silicone oil AK 1.000.000 from Wacker as the test material, since the viscoelastic properties of this silicone oil well represent that of a whole range of silicone oils with different zero-shear viscosities. Large amplitude oscillatory shear (LAOS) is a commonly used method to characterize the nonlinear viscoelastic properties of the materials [20].

The most common method to quantify LAOS tests is Fourier transform (FT) rheology [21]. For a sinusoidal strain input, the stress response could be described by a Fourier series [22]. Only odd harmonics are included in these series because the stress signal is assumed to be of odd symmetry with respect to directionality of shear strain or shear rate, i.e., the material response is unchanged if the coordinate system is reversed [23]. Another way to quantify nonlinear viscoelasticity in LAOS is the stress decomposition (SD) technique. Here the stress response is decomposed to elastic and viscous stresses. The elastic stress should exhibit odd sym-

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meter. Earlier results already have shown that this silicone oil is shear thinning, and both the storage and the loss moduli decrease with increasing oscillation amplitude in an amplitude sweep test. Based on our new LAOS tests, the total viscoelastic stress response can be safely considered linear below strain amplitude 1 and angular frequency 100 rad/s. In the nonlinear regime, the elastic Lissajous-Bodwitch curves show intracycle strain stiffening whereas the viscous Lissajous-Bodwitch curves show intracycle shear thinning. We carefully analyzed also the raw data of our new LAOS tests and discarded from the LAOS analysis those cases where the input shear-rate signal was not sinusoidal sufficiently. However, we successfully utilized these cases as a verification of our nonlinear viscoelastic model, a 6 element White-Metzner type constitutive equation (Equations 9 and 10). The analysis of S in comparison with v_3/v_1 and that of T in comparison with e_3/e_1 gives a more complete explanation of the nonlinear viscoelastic properties.

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