Discussion of "Linear and Nonlinear Rheological Properties of Bituminous Mastics under Large Amplitude Oscillatory Shear Testing" by Aboelkasim Diab and Zhanping You

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The nonlinearity in an oscillatory shear test for bituminous materials is not well understood and the authors indeed touch upon a pertinent topic related to the same in the original paper. The discussers would like to complement the authors on this aspect and offer additional insight on nonlinearity in oscillatory shear in this short discussion.

The oscillatory shear test is being widely used in the characterization of bituminous binder. In this test, the bituminous material is subjected to load (stress)–controlled or deflection (strain)–controlled sinusoidal loading, with the deflection-controlled test being more common. The test is generally conducted in the linear regime so that the material functions such as storage modulus, loss modulus, dynamic modulus, and phase angle can be meaningfully interpreted. The strain amplitude for testing is carefully selected to maintain the response of the material in the linear regime.

In the traditional method of identifying the linear regime, the peak stress and peak strain from the oscillatory shear test is used and this test is widely termed as a small amplitude oscillatory shear (SAOS) test. In the SAOS test, the strain amplitude corresponding to linear behavior of the material is identified using the storage and loss moduli or dynamic modulus. When the response of the material is linear, the storage, loss, and dynamic moduli are independent of strain amplitude used for shearing. For ensuring the linearity using the modulus value, ASTM D7175 (ASTM 2008) recommends conducting the test with increasing strain amplitude and the linear regime of loading is defined as the range of strain where the dynamic modulus is 90% or more of the initial value. ASTM D7175 (ASTM 2008) recommends conducting this test at a frequency of 10 rad/s and at a strain amplitude in the range of 2%–12%. The linearity check based on ASTM D7175 (ASTM 2008) is more specific to a frequency and strain amplitude used in the performance grading of binder. Also, dynamic modulus is a linear measure and ensuring linearity and nonlinearity based on the linear parameter may not be appropriate.

Linearity can also be defined based on the geometry of the stress and strain waveform. Here if the response of the material is linear for the applied sinusoidal strain waveform, the stress response is expected to be sinusoidal in shape. The deviation of the response stress waveform from sinusoidal shape indicates the nonlinear behavior of the material. For the sinusoidal strain and sinusoidal stress waveform, the stress-strain plot, also termed as Lissajous plot, is a perfect ellipse. Hence, the elliptical-shaped Lissajous plot indicates the linear response of the material, and the magnitude of deviation of the Lissajous plot from the ellipse indicates the extent of nonlinearity. To quantify linearity based on the shape of the stress-strain plot, one needs complete stress and strain waveform data. The oscillatory shear testing with the complete stress and strain waveform data, termed as large amplitude oscillatory shear (LAOS) testing, is used for identifying linear and nonlinear behavior of the material. The linear and nonlinear behavior of the material from the stress-strain waveform can be identified based on the shape of the Lissajous plot, harmonics of the resultant stress waveform, and strain stiffening ratio (Ewoldt et al. 2008).

The discussers differ from the authors' idea in the method of defining nonlinearity of the material and relating the performance to the nonlinear measure. Following Ewoldt et al. (2008), the authors used Lissajous plots to delineate linear and nonlinear response of the material. For the perfect ellipse, the slope of the major axis (G'_L) and the tangent at zero instantaneous strain (G'_M) are the same. The authors calculated G'_M, G'_L, and S (strain stiffening ratio) at different strain levels for binder and mastics and used them as a measure of nonlinearity. The concern here is the sensitivity of the parameter to delineate linear and nonlinear response of the material.

Sensitivity of *S*

If G'_L and G'_M are identical and equal to storage modulus G', the value of S is zero, and this indicates the material response is linear. Now, let us consider the case of ±10% deviation of G'_L and G'_M from G'. Up to ±10% variation, G'_L and G'_M will be in the range of $0.90G'$ and $1.10G'$. The value of S for the ±10% difference in G'_L and G'_M from G' is

$$S = \pm\frac{1.1G' - 0.90G'}{1.1G'} = \pm 0.182 \tag{1}$$

Hence, if one allows up to ±10% of value difference in the experimental results (ASTM 2008), the response of the material can be considered linear if the value of S is within the range of -0.182 and $+0.182$. The value of S for some of the mastics reported by the authors at 10% and 30% strain amplitude was as low as -0.004 and 0.0072. Padmarekha et al. (2013) reported the value of S for different unmodified and modified asphalt for 1% and 5% strain amplitude and at the temperature of 30°C, 40°C and 50°C. The value of S at 5% strain amplitude was found to be as high as 0.546 for the modified binder.

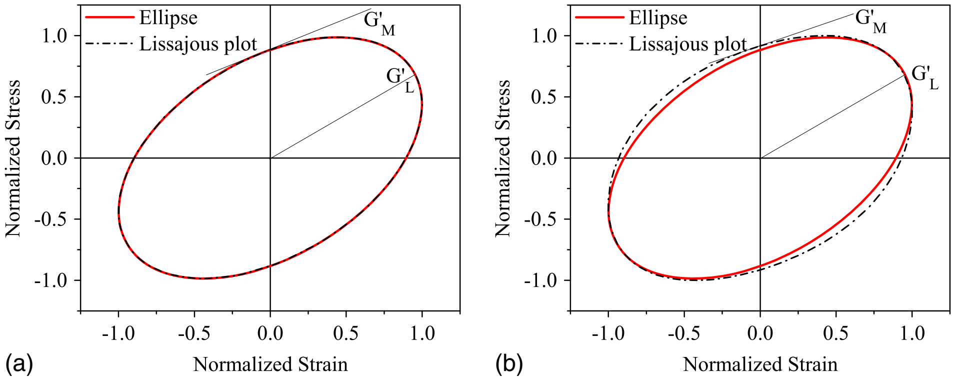

Fig. 1. Comparison of normalized Lissajous plot with ellipse shape: (a) 0.5% strain amplitude; and (b) 40% strain amplitude.

In this short discussion, we report the value of S for the sample binder tested at two different strain levels. For this purpose, the binder was sheared using large amplitude oscillatory shear protocol in a dynamic shear rheometer (MCR 302, Anton-Paar, Austria). Long-term-aged polymer-modified binder was used here for testing. The stress and strain (MCR 302, Anton-Paar) waveform data were collected for two different strain amplitude of 0.5% and 40%. These are the two extreme strain levels used by the authors. Both tests were conducted at 40°C and at a frequency of 1 Hz. The stress and strain waveform was normalized with respect to the maximum value, and the Lissajous plot was constructed and its shape was compared with the ideal ellipse shape. Fig. 1(a) shows the comparison of the Lissajous plot for 0.5% strain amplitude with the ideal ellipse. The Lissajous plot was observed to coincide with the ellipse. The value of S calculated using G'_L and G'_M was 0.067. Fig. 1(b) shows the comparison of the Lissajous plot for 40% strain amplitude with the ideal ellipse. One can observe the deviation of the Lissajous plot from the ellipse. The value of S in this case was calculated to be 0.101.

Sign of *S*

Another concern is the sign of S. The value of S is positive if $G'_L > G'_M$, and as per Ewoldt et al. (2008) this condition indicates strain stiffening behavior. The value of S is negative if $G'_L < G'_M$, and this indicates strain softening behavior. The value of S as per Fig. 13 in the original paper, especially for controlled binder and the binder and fly ash mastic, increases on the negative side with increase in strain amplitude. As per Ewoldt et al. (2008), this indicates the strain softening nature of the material, and not the decrease in the extent of nonlinearity as stated by the authors.

All the preceding discussion also holds for the thickening ratio (T) measured from η'_L and η'_M. Here, η'_L and η'_M indicate the instantaneous viscosity calculated at the smallest and largest shear rates using a stress-shear rate plot, also termed as a Bowditch plot (Ewoldt et al. 2008).

Harmonics of the Waveform

In the polymer literature, harmonic analysis is used to a large extent to verify the shape of the response stress waveform (Hyun et al. 2011). The response stress waveform can be mathematically represented using the Fourier series function as given in Eq. (2)

$$\sigma = \gamma_o \sum_{n=\text{odd}} [G'_n \sin(n\omega t) + G''_n \cos(n\omega t)] \qquad (2)$$

where σ = stress; γ_o = strain amplitude; ω = frequency; t = time; G'_n and G''_n are material functions; and n is the harmonic of waveform function, and with the stress being odd function (Bird et al. 1987), n takes only the odd numbers $(1, 3, 5, 7, \ldots)$. The presence of higher harmonics $(n = 3, 5, \ldots)$ in the stress waveform indicates the nonlinear response of the material. As of today, no studies have reported the intensity of third harmonics for bituminous binder. For this short discussion, the intensity of third harmonics of the stress waveform data collected using the LAOS protocol are reported. The intensity of the third harmonic (I_3) of the stress waveform of the bituminous binder when tested using 0.5% and 40% at 1 Hz frequency and 40°C was found to be 2,750 and 1.59×10^5 Pa. Polymer studies use relative intensity (the ratio of third harmonic intensity I_3 to first harmonic intensity I_1) to delineate linear and nonlinear response (Hyun et al. 2011). For polymeric materials, the value of I_3/I_1 is as low as 10^{-2} for linear response and as high as 10 for nonlinear response. Such a range is not established for bituminous binder and mastic.

To conclude, the response of the material can be linear at lower strain level, and as the strain in the material increases, the same material can exhibit nonlinear response. It is necessary to delineate the strain level corresponding to linear and nonlinear behavior of the material. Within the context of the LAOS data presented by the authors and supplemented by the discussers, it is not very clear how to delineate the linear and nonlinear behavior of bituminous material. Hence, at best, it is too early for the authors to make the following observations: "These local nonlinear measures could differentiate between the type and concentration of mineral fillers at different strain amplitudes; therefore they can be used to rank the properties of bituminous materials in the nonlinear regime" and "Overall, the nonlinear measures showed the ability of the mastics to withstand high strain amplitudes, which can be a good indicator of their ability to resist pavement distresses, especially high loading–associated distresses (e.g., rutting)."

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