Exploiting Large-Amplitude Oscillatory Shear Flow

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Large-amplitude oscillatory shear flow (LAOS) is the most popular laboratory experiment for investigating the nonlinear rheological behavior of polymeric liquids (see Fig. 1). This nonlinear experiment brings out distortion in the stress response. Attempts to analyze this distortion using continuum approaches have led us to quantitative responses but these teach us little about the molecular origins of the distortions, namely molecular structure and orientation. Here, we investigate stress responses from LAOS from continuum and molecular approaches. We also explore traverse the bridge between these two approaches, which we call macromolecular continua.

In the continuum framework, the Oldroyd 8-constant framework is a versatile set of popular fluid models. Moreover, when generalized to multimode fluid using the Spriggs relations, this fluid model can successfully predict nonlinear behaviour of polymeric liquids. Finally, the Oldroyd 8-constant framework yields exact solutions, for both shear stress and normal stress difference responses. Whereas these exact solutions give us general idea on how material reacts to nonlinear input, using them can be cumbersome. We can exploit these exact solutions to get simpler forms called Padé approximants. Under some circumstances, the best of these, for either the shear stress or the normal stress differences, gives the same level of accuracy as the exact expression. Lastly, we can also exploit these exact solutions for the thermodynamic analysis of flow stability to depict when LAOS is unstable (see Fig. 2).

Molecular structure and orientation are the primary causes of nonlinearity in viscoelastic behaviors of polymeric liquids. To explain the molecular origins of polymer fluid elasticity, we find the rigid dumbbell suspension to be the simplest relevant. By relevant we at least mean, predicting distortion in the stress response. The stress expressions for molecular models take the approximate form of truncated power series. We can thus exploit the best Padé approximants to perfect these truncated expansions. Since arriving at these truncated expansions involves difficulty, perfecting these so easily is useful.

References

Bienvenue à tous!