Carbopol: From a simple to a thixotropic yield stress fluid?

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There is an ongoing discussion in the literature about the flow behaviour of the widely used model yield-stress fluid Carbopol. Some papers show that it is indeed the simple model yield-stress fluid that many people believe it to be. However, other authors report rheological hysteresis in the flow curve, transient shear banding that persists for a very long time, and the breaking of fore-aft symmetry in a falling ball experiment. Such behaviours have in the past been associated with thixotropic yield stress fluids, which are very different from simple ones. Recently, it was suggested that both types of behaviour may be found in the same type of Carbopol, depending on the preparation (J. Rheol. 62, 773-780, 2018): Strong stirring breaks the polymers into smaller fragments, some of which are so small that they exhibit Brownian motion. This generates a depletion interaction that leads to gel formation, which in turn leads to the thixotropy, and is usually interpreted in terms of a simple toy-model for the evolution of the microstructure and the viscosity (Soft Matter 2, 274–283, 2006). The basic assumptions of the model are that there exists a structural parameter, λ, that describes the local degree of interconnection of the microstructure, and that the viscosity increases with increasing λ. In addition, for an aging system at low or zero shear rate λ increases, while the flow at sufficiently high shear rates breaks down the structure and λ decreases to a low steady state value.

This work presents a systematic quantitative assessment of this model by means of a thorough theoretical and numerical analysis and by comparison with experimental results. The general features of the model dynamics are considered in the framework of the dynamical systems theory, and its dynamic response to selected forcing schemes typically used during rheological tests on yield stress fluids was obtained numerically. To better understand the physical significance of the outcomes of the theoretical and numerical analysis, identical forcing patterns were applied to Carbopol® gel samples prepared following different protocols.

Surprisingly, when a stepped ramp (either in controlled stress mode or in controlled shear rate mode) is applied, the model returns multiple-valued functions, irrespective of the magnitude of the characteristic recombination time. In addition, irrespective of the forcing scheme and of the parameters choice, during the transient the magnitude of the structural parameter, λ, can attain values significantly higher than one; the physical interpretation of this result is not possible based on the present knowledge. Finally, irrespective of the magnitudes of the applied stress or shear rate and of the parameters choice, when the fluid becomes fully de-structured (i.e., when $\lambda = 0$) the viscosity becomes constant, i.e., the fluid becomes Newtonian. It appears most of the observed behaviours are not intrinsic features of the model, but are rather introduced into the model through the (arbitrary) relationship between viscosity and the structural parameter.

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