J. Non-Newtonian Fluid Mech. 165 (2010) 687–697

Contents lists available at ScienceDirect

Journal of Non-Newtonian Fluid Mechanics

journal homepage: www.elsevier.com/locate/jnnfm

Dynamic texture scaling of sheared nematic polymers in the large Ericksen number limit

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ARTICLE INFO

Article history:
Received 16 November 2009
Received in revised form 11 March 2010
Accepted 16 March 2010

PACS:
61.30.v
61.30Vx
67.40.Fd
67.55.Fa

Keywords:
Nematic polymers
Rod dispersions
High Ericksen number
Length scale
Heterogeneous properties
Defect core

ABSTRACT

In steady shear experiments, nematic polymers and rigid Brownian rod dispersions develop unsteady fine structure (Larson and Mead, 1992 [33]; Larson and Mead, 1993 [34]; Tan and Berry, 2003 [42]). These features are representative of the dynamic morphologies generated during film and mold processes, which translate to the two banes of materials engineering: heterogeneous properties on unknown length scales, and dynamic fluctuations in processing outcomes. Our goal here is to quantify the length scales of these dynamic textures in the realistic parameter regime of extremely high Ericksen number, *Er*, the ratio of viscous to elastic stress. To avoid model-specific or numerical anomalies, we simulate three distinct models of flowing nematic polymers employing three different algorithms run on different computer systems, following our earlier studies at moderate Ericksen numbers (Forest et al., 2008 [11,12]). Our strategy recognizes the rich history on this problem as well as practical prohibitive constraints. First, the relevant experimental and processing conditions dictate $Er \approx O(10^5)$ and order unity Deborah number (*De*, the bulk shear rate normalized by the rotational relaxation rate of the nematic phase). Second, theoretical estimates of the smallest length scales of morphology and their sources exist only for steady-state structures, originally from liquid crystal models (cf. Carlsson, 1984 [2]; Carlsson, 1976 [3]; Cladis and Torza, 1976 [4]; de Gennes, 1974 [21]; Manneville, 1981 [35]) and subsequently from nematic polymer orientation tensor and full kinetic models (cf. Cui, 2006 [5]; Forest and Wang, 2003 [13]; Forest et al., 2004 [15]; Forest et al., 2007 [19]; Marrucci, 1985 [36]; Marrucci, 1990 [37]; Marrucci, 1991 [38]; Marrucci and Greco, 1993 [39]; Zhou and Forest, 2006 [50]; Zhou et al., 2007 [52]). Third, numerical experiments for $Er \approx O(10^{2,3})$ and $De \approx O(1)$ confirm experimental observations of unsteady long-time attractors (Denn and Rey, 2002 [6]; Feng et al., 2001 [10]; Forest et al., 2008 [11,12]; Forest et al., 2005 [17]; Han and Rey, 1995 [23]; Klein et al., 2005 [30]; Kupferman et al., 2000 [32]; Sgalari et al., 2002 [40]; Tsuji and Rey, 1997 [43]; Yang et al., 2008 [46]; Yang et al., in press [47]). Fourth, the unsteady regime persists to $Er \approx O(10^5)$ (Sgalari et al., 2002 [40]), where single runs, much less two or three decades of Er, are prohibitively expensive in two or three space dimensions due to severe spatial and temporal resolution constraints. Here we propose and implement a new strategy to finesse these limitations. The key observation arises from comprehensive 2D simulations (Klein et al., 2005 [30]; Yang et al., 2008 [46]; Yang et al., in press [47]) which implicate transient defect cores (local isotropic or oblate disordered phases) as the source of the finest texture, or equivalently, the strongest gradients. In light of these results, we simulate 1D flow-orientational heterogeneity spanning the plate gap, which preserves the fine-scale transient defect cores and associated shear flow and stress features, while screening higher-dimensional, nonlocal defect topology and cellular flow patterns. We simulate spatio-temporal attractors for *Er* out to 10^5 for all three models. Each model reveals a transient staircase of tumbling-wagging layers spanning the shear gap, where the smallest length scales of morphology are associated with intermittent oblate defects between adjacent tumbling and wagging layers, which form precisely while neighboring layers become out of phase. With increasing *Er*, the number of tumbling, wagging and oblate defect layers grows, meanwhile layers propagate, collide, merge and reform *ad infinitum*.

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0377-0257/$ – see front matter
doi:10.1016/j.jnnfm.2010.03.003

To quantify these dynamic textures, we calculate a *time-averaged morphology length scale distribution function*, defined by the reciprocal of the gradient of the oblate defect metric. The peak of this PDF characterizes a dominant morphology length scale which, for all three models, remarkably adheres to the Marrucci scaling law $Er^{-(1/2)}$, consistent with earlier numerical studies (Sgalari et al., 2002 [40]) on yet another nematic flow-tensor model. This result yields an *a priori* estimate for the finest length scales in shear-dominated nematic polymer films and molds based on processing conditions and Frank elasticity constants, which confirms a prediction of Marrucci, 25 years ago (Marrucci, 1985 [36]), based on dimensional analysis in the dynamic tumbling regime.

1. Introduction

One of the challenging problems in film and mold processing of nematic polymers and nano-rod dispersions is to predict, *a priori*, the dominant length scales of shear-induced morphology [9,13,20,21,35]. We refer to the review by Tan and Berry [42] and the subsequent review by Rey and Denn [6]. These texture features determine the length scales of variability in material performance, for optical and electromagnetic transmission properties as well as conductive and mechanical behavior. By contrast, fiber (extension dominated) processing achieves highly uniform orientational alignment along the fiber axis. Another challenge is to understand the sources of small scale heterogeneity arising from shear-dominated hydrodynamics and physical confinement associated with film and mold processing; such information reveals whether or not the heterogeneity is inevitable in shear processing. There is evidence that disclination line and loop defects are responsible for the fine scales of structure [10,23,28,31-34,37-39]. If topological defects are indeed associated with the smallest structure length scales, then the finest texture should reside in their cores, where local disordered phases form to obviate an actual singularity in the principal axis of nematic orientation. Two-dimensional simulations of the Leal group [30] and the authors [19,46–49] confirm that local defect cores are always present in half-integer degree topological defects, and that the largest gradients of the orientational distribution arise in the cores. These studies for $Er \approx 10^2$, 10^3 are prohibitively expensive and generate quite large data sets in the realistic regime of Ericksen numbers in two space dimensions due precisely to the proliferation of small scales, requiring much higher spatial resolution, and the unsteady attractors which require extremely small time steps at such high Er. Data analysis for the purpose of extracting scaling behavior of the morphology therefore seems impractical.

The above evidence, however, suggests a numerical strategy which we implement in this paper to estimate the dominant length scales of texture in the dynamic regime of very high Ericksen numbers. Namely, we restrict to one-dimensional heterogeneity in the flow-gradient direction spanning the gap in a parallel-plate shear cell. This dimensional reduction suppresses topological defects, but allows defect cores in the form of locally disordered oblate or isotropic phases. These phases arise naturally in sheared nematic polymers as the minimum energy orientational configuration that can smoothly accommodate nearby strongly ordered nematic phases whose principal axes are out of phase. Instead of creating a discontinuity in the director (principal axis of orientation), the oblate phase corresponds to a multiplicity two eigenvalue degeneracy of the second moment of the orientational distribution, in which the director is no longer identifiable and spreads to a circle. The isotropic phase is a fully degenerate phase, where the director lies anywhere on the sphere, and the eigenvalue degeneracy is multiplicity 3. Whereas oblate and isotropic phases are always present in the cores of two and three dimensional topological defects, it is important to recognize that disordered phases exist in lower dimensional (zero and one) structures where topological defects are not possible. Both of these disordered phases play a prominent role in the Onsager hysteresis diagram for the isotropic-nematic phase transition, and in 1D shear flow studies of the authors at moderate Ericksen numbers [5,11,12,18,50,51].

Information on the length and time scales and sources of fine scale structure is critical for all applications of nano-rod and nano-platelet materials. If heterogeneity is unavoidable in shear-dominated flows, which is supported by experimental and numerical evidence, then quantitative estimates of the structure are needed. We present results from numerical simulations of three different flow-orientation models, together with post-processing diagnostics, which reveal both morphology scaling behavior and the sources of the strongest gradients, or equivalently, the smallest length scales. Our results are for one-dimensional heterogeneous structures arising from parallel plate driving conditions in the realistic regime of moderate Deborah numbers and very high Ericksen numbers, where the responses are not only unsteady but strongly intermittent.

The theoretical formulation of the morphology length scale problem begins with a choice of flow-nematic model and boundary conditions [14,16,22,37–39]. Key non-dimensional parameters are identified, notably the Deborah number (ratio of bulk flow rate to microstructure relaxation rate) and Ericksen number (the ratio of viscous to elastic stresses). From a given model formulation, one can attempt to ascertain how the length scales of morphology scale with the fundamental dimensionless parameters. Rarely can one infer this information from the equations and boundary conditions, so the primary approach has been to study special solutions to these equations. The main results of this nature have focused on 1D heterogeneous *steady state* solutions in parallel plate shear cells, with steady plate driving conditions and strong anchoring on the microstructure (nematic director, or rod orientational distribution). We refer to [15,17] where the literature is reviewed, and to the reviews of Tan and Berry and Rey and Denn cited above. The upshot regarding *steady-state structure scaling behavior* is that all evidence points to a modified Marrucci scaling of the smallest length scale, $Er^{-\alpha}$, where α is between 1/4 and 1. Since the Ericksen number is typically quite large, nailing down the exponent and confirming a power law behavior would be valuable. In [15], explicit steady solutions are derived in the dual limit of low Deborah number (equivalent to slow plates) and low Ericksen number (equivalent to very strong distortional elasticity which arrests tumbling). These asymptotic solutions reveal two distinct scaling properties of orientational morphology: Er^{-1} average scaling of director dominated structure that spans the shear gap, and an $Er^{-1/2}$ scaling of order parameter dominated boundary layers. In [17], these scaling properties are upheld with full kinetic-flow simulations for $Er \approx O(10^3)$. Sgalari et al. [40] used a different nematic tensor model and carried 1D flow-nematic simulations out to $Er \approx 10^6$. They used Fourier transforms of flow and morphology snapshots to identify dominant length scales, supporting the Marrucci $Er^{-1/2}$ scaling. To our knowledge, these results have not been improved upon nor extended.

The limitations of the available analytical results with respect to materials applications are fairly severe. First and foremost, they break down when either the Deborah or Ericksen number exceeds order unity, whereas realistic processing or rheological conditions