

New approach for understanding the different behavior of polymer melts and solutions under non-linear extensional flow

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While the non-universality of polymer melts and solutions under non-linear extensional flow is now well established experimentally, its molecular origin is not yet fully understood.

Indeed, if two linear polymers have the same number of entanglements, whatever their chemical structure or concentration, their normalized viscoelastic response are identical. However, this universality seems to break down for their normalized extensional viscosity under non-linear extensional flow [1]: while polymer melts exhibit an exclusive monotonic extension-thinning behavior, polymer solutions exhibit a strong extension-hardening after an initial extension-thinning behavior. In addition, recent results [2] showed that the extensional viscosity of well-entangled polymer blends actually exhibit a c dependence and have a similar behavior to polymer melts, while polymer solutions exhibit a \sqrt{c} dependence that is difficult to understand from the current molecular theories applied for linear flows.

Several approaches have been proposed to model the behavior of polymer solutions, such as the Extended Interchain Pressure concept [3], Monomeric Friction Reduction combined with nematic interactions [4] or an effective stretch relaxation time related to the dual tube picture [5]. However, while these pictures enable a quantitative description of particular polymer blends, they usually fail to describe a wide range of polymer blends within the same model.

In this approach, we propose to take into account the dual tube picture, already used to model binary blends in the linear regime of deformation, to bridge the gap between the extensional behavior of polymer solutions and well-entangled bidisperse blends in the non-linear regime of deformation (Fig.1).

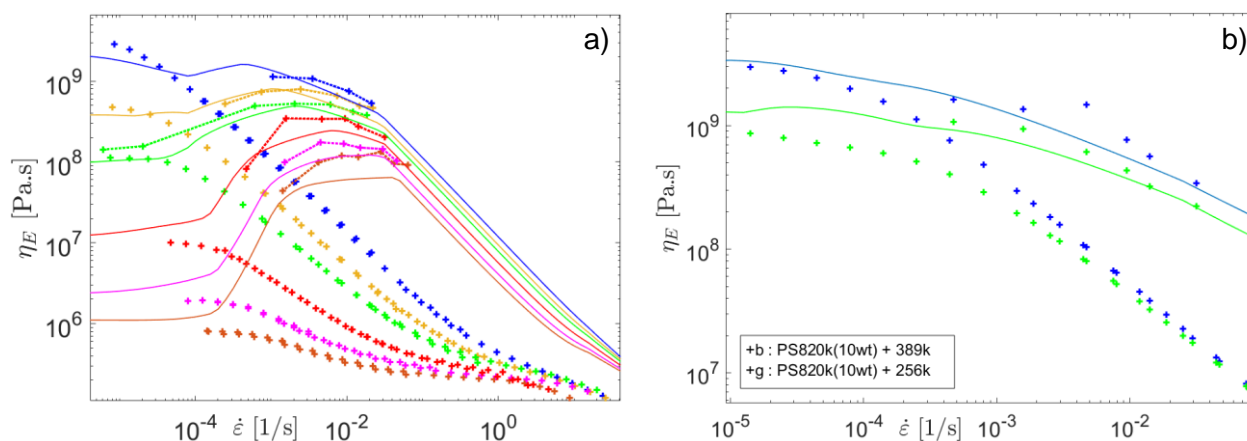


Fig.1. Measured extensional viscosity (symbols) of PS820k oligomeric solutions diluted from top to bottom to 50%, 30%, 20%, 10%, 5% and 3% (a) and PS820k well-entangled blends (b) compared with preliminary theoretical results

[1] Q. Huang, L. Hengeller, N. Alvarez, O. Hassager *Macromolecules*. **2015**, *48*, 4158-4163.

[2] A. André, T. Shahid, F. Oosterlinck, C. Clasen, E. van Ruymbeke *Macromolecules*. **2021**, *54*, 2797-2810.

[3] M. Wagner, E. Narimissa *J. Rheol.* **2021**, *65*, 1413.

[4] Q. Huang, N. Alvarez, Y. Matsumiya, H. K. Rasmussen, H. Watanabe, O. Hassager *ACS Macro Lett.* **2013**, *2*, 741-744.

[5] D. J. Read, K. Jagannathan, S. K. Sukuruman, D. Auhl *J. Rheol.* **2012**, *56*, 823.