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Supplementary information for "Sub-Newtonian coalescence in polymeric fluids"

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Theory

Giesekus and linear PTT equation under high Weissenberg creeping flow limit

To show that the upper convected derivative is the predominant term in high Wi low Re flows as proposed by Renardy[1] we decompose the stress tensor under quasi-steady assumption such that we are able to look at the magnitude of individual component in Eq (4) of manuscript in elasticity dominated regime.

Eq (4) under high Wi creep flow and quasi-steady assumption is simplified to S-1 Giesekus:

$$\overset{\triangledown}{\boldsymbol{\tau}} * + \kappa tr(\boldsymbol{\tau})^* \boldsymbol{\tau}^* = 0 \tag{S-1 PTT}$$

$$\overset{\triangledown}{\boldsymbol{\tau}} * + A \boldsymbol{\tau}^* \boldsymbol{\tau}^* = 0 \tag{S-1 Giesekus}$$

The analysis presented is for a two-dimensional plane. The velocity vector in 2D can be defined as $\mathbf{v} := (u, v)$. We use this to create a symmetric tensor $T^1 := \mathbf{v} \otimes \mathbf{v}$. Next we define a vector $\mathbf{w}$ such that $\mathbf{v} \times \mathbf{w} = \mathbf{1}$ and $\mathbf{v} \cdot \mathbf{w} = 0$. Using this we find $\mathbf{w} = (\frac{-v}{u^2+v^2}, \frac{u}{u^2+v^2})$. Similar to $\mathbf{v}$ we use $\mathbf{w}$ to define another symmetric tensor $T^2 := \mathbf{w} \otimes \mathbf{w}$. Using $\mathbf{v}$ and $\mathbf{w}$ we can define a third symmetric tensor as $T^3 := \mathbf{v} \otimes \mathbf{w} + \mathbf{w} \otimes \mathbf{v}$.

Stress tensor τ is symmetric, second-order tensor with rank= 3 and it can be written in the basis spanned by the three tensors T^1, T^2, T^3 as:

$$\tau = \alpha T^1 + \beta T^2 + \gamma T^3; \text{ where } \alpha, \beta, \text{and} \gamma \text{ are functions of (x,y)} \tag{S-2}$$

On substituting this decomposition of stress tensor in Eq (S-1), individual terms can be simplified to obtain three new equations owing to the linear independence of basis tensors. Few of the key results that will be important to proceed ahead:

$$\mathbf{v}.\mathbf{w} = 0 \Leftrightarrow v_i w_i = 0,\ \mathbf{w}.\mathbf{w} = \frac{1}{||\mathbf{v}||^2} \Leftrightarrow w_i w_i = \frac{1}{||\mathbf{v}||^2},\ \mathbf{v}.\mathbf{v} = ||\mathbf{v}||^2 \Leftrightarrow v_i v_i = ||\mathbf{v}||^2$$

The first term of eq (1) $\overset{\triangledown}{\boldsymbol{\tau}}*$ has three sub-terms. Note we will drop stars from now on for ease of notations.

- $\mathbf{v}.\nabla\boldsymbol{\tau} = \frac{\partial \tau_{ij}}{\partial x_k} v_k$ when double contracted with $\mathbf{v} \otimes \mathbf{v}$ we obtain: $\frac{\partial \tau_{ij}}{\partial x_k} v_k v_i v_j$

$$v_k v_i v_j \frac{\partial \tau_{ij}}{\partial x_k} = v_k v_i v_j \frac{\partial(\alpha v_i v_j + \beta w_i w_j + \gamma(v_i w_j + w_i v_j))}{\partial x_k}$$

$$= v_k v_i v_j \left(v_i v_j \frac{\partial \alpha}{\partial x_k} + \alpha v_i \frac{\partial v_j}{\partial x_k} + \alpha v_j \frac{\partial v_i}{\partial x_k} + \gamma v_i \frac{\partial w_j}{\partial x_k} + \gamma v_j \frac{\partial w_i}{\partial x_k} \right)$$

While expanding the derivative, we have dropped the terms corresponding to orthogonality of $\mathbf{v},\mathbf{w}$

$$= ||\mathbf{v}||^4 \nabla\alpha.\mathbf{v} + 2||\mathbf{v}||^2 \alpha((\nabla\mathbf{v})\mathbf{v}).\mathbf{v} + 2\gamma||\mathbf{v}||^2((\nabla\mathbf{w})\mathbf{v}).\mathbf{v}$$

- $(\nabla\mathbf{v})\boldsymbol{\tau}$ when double contracted with $\mathbf{v}\otimes\mathbf{v}$ we obtain: $\frac{\partial v_i}{\partial x_k}\tau_{kj}v_iv_j$

$$\frac{\partial v_i}{\partial x_k}\tau_{kj}v_iv_j = v_iv_j\frac{\partial v_i}{\partial x_k}(\alpha v_kv_j + \beta w_kw_j + \gamma(v_kw_j + w_kv_j))$$

$$= \|\mathbf{v}\|^2\Bigg(\alpha((\nabla\mathbf{v})\mathbf{v}).\mathbf{v} + \gamma((\nabla\mathbf{v})\mathbf{w}).\mathbf{v}\Bigg)$$

- $\boldsymbol{\tau}(\nabla\mathbf{v})^T$ when double contracted with $\mathbf{v}\otimes\mathbf{v}$ we obtain: $\tau_{ik}\frac{\partial v_j}{\partial x_k}v_iv_j$

$$\tau_{ik}\frac{\partial v_j}{\partial x_k}v_iv_j = (\alpha v_iv_k + \beta w_iw_k + \gamma(v_iw_k + w_iv_k))v_iv_j\frac{\partial v_j}{\partial x_k}$$

$$= \|\mathbf{v}\|^2\Bigg(\alpha((\nabla\mathbf{v})\mathbf{v}).\mathbf{v} + \gamma((\nabla\mathbf{v})\mathbf{w}).\mathbf{v}\Bigg)$$

- $\kappa tr(\boldsymbol{\tau})\boldsymbol{\tau}$ when double contracted with $\mathbf{v}\otimes\mathbf{v}$ we obtain: $\kappa\tau_{kk}\tau_{ij}v_iv_j$

$$\kappa\tau_{kk}\tau_{ij}v_iv_j = \kappa(\alpha v_kv_k + \beta w_kw_k + \gamma(v_kw_k + w_kv_k))(\alpha v_iv_j + \beta w_iw_j + \gamma(v_iw_j + w_iv_j))v_iv_j$$

$$= \kappa\|\mathbf{v}\|^4\alpha(\alpha\|\mathbf{v}\|^2 + \beta\|\mathbf{w}\|^2)$$

- $A\boldsymbol{\tau}\boldsymbol{\tau}$ when double contracted with $\mathbf{v}\otimes\mathbf{v}$ we obtain: $A\tau_{ik}\tau_{kj}v_iv_j = A(\alpha^2\|\mathbf{v}\|^2+\gamma^2\|\mathbf{w}\|^2)\|\mathbf{v}\|^4$

On putting all terms corresponding to Linear PTT together:

$$\|\mathbf{v}\|^4\nabla\alpha.\mathbf{v} + 2\gamma\|\mathbf{v}\|^2(((\nabla\mathbf{w})\mathbf{v}).\mathbf{v} - ((\nabla\mathbf{v})\mathbf{w}).\mathbf{v}) + \|\mathbf{v}\|^4\alpha\kappa(\alpha\|\mathbf{v}\|^2 + \beta\|\mathbf{w}\|^2) = 0 \tag{S-3}$$

$$(\mathbf{v}.\nabla)\alpha + 2\gamma\frac{(((\nabla\mathbf{w})\mathbf{v}).\mathbf{v} - ((\nabla\mathbf{v})\mathbf{w}).\mathbf{v})}{||\mathbf{v}||^2} + \alpha\kappa(\alpha||\mathbf{v}||^2 + \beta||\mathbf{w}||^2) = 0 \tag{S-4}$$

Similarly we will repeat the exercise but this time take a double contraction with $\mathbf{w}\otimes\mathbf{w}$

- $\mathbf{v}.\nabla\boldsymbol{\tau} = \frac{\partial\tau_{ij}}{\partial x_k}v_k$ when double contracted with $\mathbf{w}\otimes\mathbf{w}$ we obtain: $\frac{\partial\tau_{ij}}{\partial x_k}v_k w_i w_j$

$$v_k w_i w_j\frac{\partial\tau_{ij}}{\partial x_k} = v_k w_i w_j\frac{\partial(\alpha v_i v_j + \beta w_i w_j + \gamma(v_i w_j + w_i v_j))}{\partial x_k}$$

$$= v_k w_i w_j\left(w_i w_j\frac{\partial\beta}{\partial x_k} + \beta w_i\frac{\partial w_j}{\partial x_k} + \beta w_j\frac{\partial w_i}{\partial x_k} + \gamma w_j\frac{\partial v_i}{\partial x_k} + \gamma w_i\frac{\partial v_j}{\partial x_k}\right)$$

While expanding the derivative, we again drop the terms corresponding to orthogonality of $\mathbf{v},\mathbf{w}$

$$= ||\mathbf{w}||^4\nabla\beta.\mathbf{v} + 2||\mathbf{w}||^2\beta((\nabla\mathbf{w})\mathbf{v}).\mathbf{w} + 2\gamma||\mathbf{v}||^2((\nabla\mathbf{v})\mathbf{v}).\mathbf{w}$$

- $(\nabla\mathbf{v})\boldsymbol{\tau}$ when double contracted with $\mathbf{w}\otimes\mathbf{w}$ we obtain: $\frac{\partial v_i}{\partial x_k}\tau_{kj}w_i w_j$

$$\frac{\partial v_i}{\partial x_k}\tau_{kj}w_i w_j = w_i w_j\frac{\partial v_i}{\partial x_k}(\alpha v_k v_j + \beta w_k w_j + \gamma(v_k w_j + w_k v_j))$$

$$= ||\mathbf{w}||^2\left(\beta((\nabla\mathbf{v})\mathbf{w}).\mathbf{w} + \gamma((\nabla\mathbf{v})\mathbf{v}).\mathbf{w}\right)$$

- $\boldsymbol{\tau}(\nabla\mathbf{v})^T$ when double contracted with $\mathbf{w}\otimes\mathbf{w}$ we obtain: $\tau_{ik}\frac{\partial v_j}{\partial x_k}w_i w_j$

$$\tau_{ik}\frac{\partial v_j}{\partial x_k}w_i w_j = (\alpha v_i v_k + \beta w_i w_k + \gamma(v_i w_k + w_i v_k))w_i w_j\frac{\partial v_j}{\partial x_k}$$

$$= ||\mathbf{w}||^2\left(\beta((\nabla\mathbf{v})\mathbf{w}).\mathbf{w} + \gamma((\nabla\mathbf{v})\mathbf{v}).\mathbf{w}\right)$$

- $\kappa tr(\boldsymbol{\tau})\boldsymbol{\tau}$ when double contracted with $\mathbf{w} \otimes \mathbf{w}$ we obtain: $\kappa \tau_{kk} \tau_{ij} w_i w_j$

$$\kappa \tau_{kk} \tau_{ij} w_i w_j = \kappa(\alpha v_k v_k + \beta w_k w_k + \gamma(v_k w_k + w_k v_k))(\alpha v_i v_j + \beta w_i w_j + \gamma(v_i w_j + w_i v_j)) w_i w_j$$

$$= \kappa ||\mathbf{w}||^4 \beta(\alpha ||\mathbf{v}||^2 + \beta ||\mathbf{w}||^2)$$

- $A\boldsymbol{\tau}\boldsymbol{\tau}$ when double contracted with $\mathbf{w} \otimes \mathbf{w}$ we obtain: $A\tau_{ik}\tau_{kj} w_i w_j = A(\beta^2 ||\mathbf{w}||^2 + \gamma^2 ||\mathbf{v}||^2)||\mathbf{w}||^4$

On putting all terms corresponding to Linear PTT together:

$$||\mathbf{w}||^4 \nabla\beta.\mathbf{v} + 2\beta||\mathbf{w}||^2(((\nabla\mathbf{w})\mathbf{v}).\mathbf{w} - ((\nabla\mathbf{v})\mathbf{w}).\mathbf{w}) + ||\mathbf{w}||^4 \beta\kappa(\alpha||\mathbf{v}||^2 + \beta||\mathbf{w}||^2) = 0 \tag{S-5}$$

$$(\mathbf{v}.\nabla)\beta + 2\beta\frac{(((\nabla\mathbf{w})\mathbf{v}).\mathbf{w} - ((\nabla\mathbf{v})\mathbf{w}).\mathbf{w})}{||\mathbf{w}||^2} + \beta\kappa(\alpha||\mathbf{v}||^2 + \beta||\mathbf{w}||^2) = 0 \tag{S-6}$$

For the final equation we will double contract with: $\mathbf{v} \otimes \mathbf{w} + \mathbf{w} \otimes \mathbf{v}$

- $\mathbf{v}.\nabla\boldsymbol{\tau} = \frac{\partial \tau_{ij}}{\partial x_k} v_k$ when double contracted with $\mathbf{v} \otimes \mathbf{w} + \mathbf{w} \otimes \mathbf{v}$ we obtain: $\frac{\partial \tau_{ij}}{\partial x_k} v_k v_i w_j + \frac{\partial \tau_{ij}}{\partial x_k} v_k w_i v_j$

$$\frac{\partial \tau_{ij}}{\partial x_k} v_k v_i w_j + \frac{\partial \tau_{ij}}{\partial x_k} v_k w_i v_j = v_k v_i w_j \frac{\partial(\alpha v_i v_j + \beta w_i w_j + \gamma(v_i w_j + w_i v_j))}{\partial x_k} +$$

$$v_k w_i v_j \frac{\partial(\alpha v_i v_j + \beta w_i w_j + \gamma(v_i w_j + w_i v_j))}{\partial x_k}$$

$$= v_k v_i w_j \left(\alpha v_i \frac{\partial v_j}{x_k} + \beta w_j \frac{\partial w_i}{\partial x_k} + v_i w_j \frac{\partial \gamma}{\partial x_k} + \gamma w_j \frac{\partial v_i}{\partial x_k} + \gamma v_i \frac{\partial w_j}{\partial x_k} \right) +$$

$$v_k w_i v_j \left(\alpha v_j \frac{\partial v_i}{x_k} + \beta w_i \frac{\partial w_j}{\partial x_k} + w_i v_j \frac{\partial \gamma}{\partial x_k} + \gamma w_i \frac{\partial v_j}{\partial x_k} + \gamma v_j \frac{\partial w_i}{\partial x_k} \right)$$

While expanding the derivative, we again drop the terms corresponding to orthogonality of $\mathbf{v},\mathbf{w}$

$$= 2\|\mathbf{v}\|^2 \alpha (\nabla \mathbf{v})\mathbf{v}.\mathbf{w} + 2\|\mathbf{w}\|^2 \beta ((\nabla \mathbf{w})\mathbf{v}).\mathbf{v} + 2\|\mathbf{v}\|^2 \|\mathbf{w}\|^2 ((\nabla \gamma).\mathbf{v}) +$$

$$2\gamma \|\mathbf{w}\|^2 ((\nabla \mathbf{v})\mathbf{v}).\mathbf{v} + 2\gamma \|\mathbf{v}\|^2 ((\nabla \mathbf{w})\mathbf{v}).\mathbf{w}$$

- $(\nabla \mathbf{v})\boldsymbol{\tau}$ when double contracted with $\mathbf{v} \otimes \mathbf{w} + \mathbf{w} \otimes \mathbf{v}$ we obtain: $\frac{\partial v_i}{\partial x_k} \tau_{kj} (v_i w_j + w_i v_j)$

$$\frac{\partial v_i}{\partial x_k} \tau_{kj} (v_i w_j + w_i v_j) = (v_i w_j + w_i v_j) \frac{\partial v_i}{\partial x_k} (\alpha v_k v_j + \beta w_k w_j + \gamma (v_k w_j + w_k v_j))$$

$$= \|\mathbf{w}\|^2 (\beta ((\nabla \mathbf{v})\mathbf{w}).\mathbf{v} + \gamma ((\nabla \mathbf{v})\mathbf{v}).\mathbf{v}) + \|\mathbf{v}\|^2 (\alpha ((\nabla \mathbf{v})\mathbf{v}).\mathbf{w} + \gamma ((\nabla \mathbf{v})\mathbf{w}).\mathbf{w})$$

- $\boldsymbol{\tau}(\nabla \mathbf{v})^T$ when double contracted with $\mathbf{v} \otimes \mathbf{w} + \mathbf{w} \otimes \mathbf{v}$ we obtain: $\tau_{ik} \frac{\partial v_j}{\partial x_k} (v_i w_j + w_i v_j)$

$$\tau_{ik} \frac{\partial v_j}{\partial x_k} (v_i w_j + w_i v_j) = (v_i w_j + w_i v_j)(\alpha v_i v_k + \beta w_i w_k + \gamma (v_i w_k + w_i v_k)) \frac{\partial v_j}{\partial x_k}$$

$$= \|\mathbf{v}\|^2 (\alpha ((\nabla \mathbf{v})\mathbf{v}).\mathbf{w} + \gamma ((\nabla \mathbf{v})\mathbf{w}).\mathbf{w}) + \|\mathbf{w}\|^2 (\beta ((\nabla \mathbf{v})\mathbf{w}).\mathbf{v} + \gamma ((\nabla \mathbf{v})\mathbf{v}).\mathbf{v})$$

- $\kappa tr(\boldsymbol{\tau})\boldsymbol{\tau}$ when double contracted with $\mathbf{v} \otimes \mathbf{w} + \mathbf{w} \otimes \mathbf{v}$ we obtain: $\kappa \tau_{kk} \tau_{ij} (v_i w_j + w_i v_j)$

$$\kappa \tau_{kk} \tau_{ij} (v_i w_j + w_i v_j) = \kappa (\alpha v_k v_k + \beta w_k w_k + \gamma (v_k w_k + w_k v_k))(\alpha v_i v_j + \beta w_i w_j + \gamma (v_i w_j + w_i v_j))(v_i w_j + w_i v_j)$$

$$= 2\kappa\gamma(\alpha||\mathbf{v}||^2 + \beta||\mathbf{w}||^2)||\mathbf{v}||^2||\mathbf{w}||^2$$

- $A\boldsymbol{\tau\tau}$ when double contracted with $\mathbf{v}\otimes\mathbf{w}+\mathbf{w}\otimes\mathbf{v}$ we obtain: $A\tau_{ik}\tau_{kj}(v_iw_j+w_iv_j)=A(2\alpha\gamma||\mathbf{v}||^2+ 2\beta\gamma||\mathbf{w}||^2)||\mathbf{v}||^2||\mathbf{w}||^2$

On putting all terms corresponding to Linear PTT together:

$$||\mathbf{v}||^2||\mathbf{w}||^2\nabla\gamma.\mathbf{v} + \beta||\mathbf{w}||^2(((\nabla\mathbf{w})\mathbf{v}).\mathbf{v} - ((\nabla\mathbf{v})\mathbf{w}).\mathbf{v}) + \gamma||\mathbf{v}||^2(((\nabla\mathbf{w})\mathbf{v}).\mathbf{w} - ((\nabla\mathbf{v})\mathbf{w}).\mathbf{w})$$

$$+\kappa\gamma(\alpha||\mathbf{v}||^2 + \beta||\mathbf{w}||^2)||\mathbf{v}||^2||\mathbf{w}||^2 = 0 \tag{S-7}$$

$$(\mathbf{v}.\nabla)\gamma + \beta\frac{(((\nabla\mathbf{w})\mathbf{v}).\mathbf{v} - ((\nabla\mathbf{v})\mathbf{w}).\mathbf{v})}{||\mathbf{v}||^2} + \gamma\frac{(((\nabla\mathbf{w})\mathbf{v}).\mathbf{w} - ((\nabla\mathbf{v})\mathbf{w}).\mathbf{w})}{||\mathbf{w}||^2} +$$

$$\kappa\gamma(\alpha||\mathbf{v}||^2 + \beta||\mathbf{w}||^2) = 0 \tag{S-8}$$

Before proceeding ahead we will drive some sub-parts for final simplification. We know that $||\mathbf{v}||^2||\mathbf{w}||^2 = 1$ this in tensorial notation is $v_iv_iw_jw_j = 1$. Taking gradient of this expression gives

$$2v_i\frac{\partial v_i}{\partial x_k}w_jw_j + 2w_j\frac{\partial w_i}{\partial x_k}v_iv_i = 0 \text{ Taking dot with } \mathbf{v}$$

$$((\nabla\mathbf{w})\mathbf{v}).\mathbf{w}||\mathbf{v}||^2 + ((\nabla\mathbf{v})\mathbf{v}).\mathbf{v}||\mathbf{w}||^2 = 0 \tag{S-9}$$

eq (23) can be used to rewrite $\frac{((\nabla\mathbf{w})\mathbf{v}).\mathbf{w}-((\nabla\mathbf{v})\mathbf{w}).\mathbf{w}}{||\mathbf{w}||^2}$ as:

$$-(((\nabla\mathbf{v})\mathbf{v}).\mathbf{v}||\mathbf{w}||^2) + ((\nabla\mathbf{v})\mathbf{w}).\mathbf{w}||\mathbf{v}||^2)$$

At this point we can invoke definitions of individual terms and the continuity equation to simplify the result as.

$$-(((\nabla\mathbf{v})\mathbf{v}).\mathbf{v}||\mathbf{w}||^2)+((\nabla\mathbf{v})\mathbf{w}).\mathbf{w}||\mathbf{v}||^2) = -(u^2(\frac{\partial u}{\partial x}+\frac{\partial v}{\partial y})+v^2(\frac{\partial u}{\partial x}+\frac{\partial v}{\partial y})+uv(\frac{\partial v}{\partial x}+\frac{\partial u}{\partial y}-\frac{\partial v}{\partial x}-\frac{\partial u}{\partial y})$$

Therefore we can conclude: $$\frac{((\nabla\mathbf{w})\mathbf{v}).\mathbf{w}-((\nabla\mathbf{v})\mathbf{w}).\mathbf{w}}{||\mathbf{w}||^2} = 0 \qquad \text{(S-10)}$$

eq (S-10) is an important result as it simplies few of the terms in eqs (S-8) and (S-6) to 0. Next we look at $v_i w_i = 0$. Taking gradient of the expression, followed by a dot product with $\mathbf{v}$ gives:

$$(\nabla\mathbf{w})\mathbf{v}).\mathbf{v}+(\nabla\mathbf{v})\mathbf{v}).\mathbf{w} = 0$$

This result can be used to rewrite the expression:

$$\frac{((\nabla\mathbf{w})\mathbf{v}).\mathbf{v}-((\nabla\mathbf{v})\mathbf{w}).\mathbf{v}}{||\mathbf{v}||^2} = -\left(\frac{((\nabla\mathbf{v})\mathbf{v}).\mathbf{w}+((\nabla\mathbf{v})\mathbf{w}).\mathbf{v}}{||\mathbf{v}||^2}\right)$$

Similar to previous case we substitute the values of individual terms to obtain:

$$= \frac{1}{||\mathbf{v}||^2}\left[2uv\frac{\partial u}{\partial x}-2uv\frac{\partial v}{\partial y}-(u^2-v^2)\frac{\partial v}{\partial x}-(u^2-v^2)\frac{\partial u}{\partial y}\right]$$

The above expression is : $$\frac{1}{||\mathbf{v}||^2}\left[2uv\frac{\partial u}{\partial x}-2uv\frac{\partial v}{\partial y}-(u^2-v^2)\frac{\partial v}{\partial x}-(u^2-v^2)\frac{\partial u}{\partial y}\right] = \nabla.\mathbf{w} \quad \text{(S-11)}$$

This result can be obtained by finding divergence of $\mathbf{w}$ using its definition. Finally we use eq (S-10) and eq (S-11) to simplify eq (S-8), (S-6), and (S-4) to get:

$$(\mathbf{v}.\nabla)\alpha + 2\gamma\nabla.\mathbf{w} + \alpha\kappa(\alpha||\mathbf{v}||^2 + \beta||\mathbf{w}||^2) = 0 \tag{S-12}$$

$$(\mathbf{v}.\nabla)\beta + \beta\kappa(\alpha||\mathbf{v}||^2 + \beta||\mathbf{w}||^2) = 0 \tag{S-13}$$

$$(\mathbf{v}.\nabla)\gamma + \beta\nabla.\mathbf{w} + \kappa\gamma(\alpha||\mathbf{v}||^2 + \beta||\mathbf{w}||^2) = 0 \tag{S-14}$$

Similarly terms in Geisekus are combined to give:

$$(\mathbf{v}.\nabla)\alpha + 2\gamma\nabla.\mathbf{w} + A(\alpha^2||\mathbf{v}||^2 + \gamma^2||\mathbf{w}||^2) = 0 \tag{S-15}$$

$$(\mathbf{v}.\nabla)\beta + A(\beta^2||\mathbf{w}||^2 + \gamma^2||\mathbf{v}||^2) = 0 \tag{S-16}$$

$$(\mathbf{v}.\nabla)\gamma + \beta\nabla.\mathbf{w} + A(\alpha\gamma||\mathbf{v}||^2 + \beta\gamma||\mathbf{w}||^2) = 0 \tag{S-17}$$

Combined eq (S-12), (S-13), (S-14) is the final form of governing equation for the stress tensor $\boldsymbol{\tau}$ following linear PTT constitutive law under high Wi flow.

The term $\mathbf{v}.\nabla$ is a scalar operator that returns the rate of change of the corresponding input variable along the streamline. In eq (S-13) the term $\kappa(\alpha||\mathbf{v}||^2 + \beta||\mathbf{w}||^2)$ corresponds to the trace of stress tensor $\boldsymbol{\tau}$ and is always positive. Therefore, in eq (S-13) β has to monotonically decrease along a streamline. This is not always physical for instance, in closed streamline flows or even for open streamline flows it requires very large value of β upstream to ensure non-negative values of β. Using this argument we observe that $\beta = 0$ is the only value that always satisfies eq (S-13) without any loss of physical sense. Now on substituting $\beta = 0$ in eq (S-14) we again refer to our previous argument to conclude $\gamma = 0$ which then can be substituted in eq (S-12) to conclude $\alpha = 0$. Similarly we can use the above arguments in eq (S-15), (S-16) and, (S-17) to conclude $\alpha = \beta = \gamma = 0$. Having functions α, β, γ all equal to zero implies that the stess tensor $\boldsymbol{\tau} = 0$. This result is physically unacceptable as it implies 0 stress for non stationary flow.

Therefore, we can conclude that the terms corresponding to the $tr(\boldsymbol{\tau})\boldsymbol{\tau}$ and $\boldsymbol{\tau}\boldsymbol{\tau}$ are giving physically inconsistent results suggesting to drop them entirely from the equations. Physically it means that the order of the term $\kappa tr(\boldsymbol{\tau})\boldsymbol{\tau}$ and $A\boldsymbol{\tau}\boldsymbol{\tau}$ are lesser than the upper convected term. Retaining the term by assuming it to have the same order as the upper convected term is physically not possible as it means 0 stresses. In conclusion, at High Wi limit $\overset{\triangledown}{\boldsymbol{\tau}}* = 0$ is the final dimensionless form of the constitutive law.

Derivation of Eq (10)

The mass and momentum equation for the quasi-radial, quasi-steady, and axis symmetric flow conditions are given below with $u_r := u$ and $u_z := v$.

$$\frac{\partial u}{\partial r} + \frac{\partial v}{\partial z} + \frac{u}{r} = 0 \tag{1}$$

$$\rho\Big(u\frac{\partial u}{\partial r}\Big) = -\frac{\partial p}{\partial r} + \frac{\partial \tau_{rr}}{\partial r} + \frac{\tau_{rr}}{r} + \frac{\partial \tau_{rz}}{\partial z} \tag{2a}$$

$$0 = -\frac{\partial p}{\partial z} + \frac{\partial \tau_{zz}}{\partial z} + \frac{\tau_{rz}}{r} + \frac{\partial \tau_{rz}}{\partial r} \tag{2b}$$

Following are the components of Lower convected derivative in low Re and high Wi flow under axis symmetric assumption:

$$u\frac{\partial \tau_{rr}}{\partial r} + 2\tau_{rr}\frac{\partial u}{\partial r} = 2\frac{\eta}{\lambda}\frac{\partial u}{\partial r} \tag{3a}$$
$$u\frac{\partial \tau_{zz}}{\partial r} - 2\tau_{zz}\frac{\partial u}{\partial r} = -2\frac{\eta}{\lambda}\frac{\partial u}{\partial r} \tag{3b}$$
$$u\frac{\partial \tau_{rz}}{\partial r} - u\frac{\tau_{rz}}{r} = 0 \tag{3c}$$

To non dimensionalize the above equations we use the following scheme: $u^* = u/U_c$, $r^* = r/R_c$, $z^* = z/Z_c$, $t^* = t/T_c$, $\tau^*_{rr} = \tau_{rr}/\tau_{rc}$ and $\tau^*_{rz} = \tau_{rz}/\tau_{zc}$, where $T_c := R_c/U_c$, U_c, $R_c = R_o$, $Z_c = R_c^2/R_o$, τ_{rc} and τ_{zc} are the characteristic time, velocity, lengths and stresses respectively. Here $U_c := \sqrt{\eta/\rho\lambda}$ is the shear wave velocity, $\tau_{rc} = \eta/\lambda$ is shear modulus, $Z_c = R_o/2$, $\tau_{zc} := \tau_{rc}/Wi_c$ with $Wi_c = \lambda U_c/R_o$, and $P_c = \sigma/R_o$ is the characteristic pressure. On substituting these in Eq set 3a and 3c, we obtain:

$$u^*\frac{\partial \tau^*_{rr}}{\partial r^*} + 2\tau^*_{rr}\frac{\partial u^*}{\partial r^*} = 2\frac{\partial u^*}{\partial r^*} \tag{4a}$$
$$u^*\frac{\partial \tau^*_{rz}}{\partial r^*} - u^*\frac{\tau^*_{rz}}{r^*} = 0 \tag{4b}$$

On integrating along r direction in $z = 0$ plane, the dimensionless rr and rz components of stress tensor are:

$$\tau^*_{rr} = 1 + K_1/(u^*)^2 \tag{5a}$$

$$\tau^*_{rz} = K_2 r^* \tag{5b}$$

Here, K_1 and K_2 are dimensionless integrating constants which in general are functions of z locally. Further the radial momentum equation is simplified to obtain:

$$\left(\frac{\rho U_c^2}{R_o}\right)u^*\frac{\partial u^*}{\partial r^*} = -\frac{P_c}{R_o}\frac{\partial p^*}{\partial r^*} + \frac{\tau_{rc}}{R_o}\left(\frac{\partial \tau^*_{rr}}{\partial r^*} + \frac{\tau^*_{rr}}{r^*}\right) + \frac{\tau_{zc}}{Z_c}\frac{\partial \tau^*_{rz}}{\partial z^*}$$

First term on RHS $\frac{P_c}{R_o}\frac{\partial p^*}{\partial r^*}$ can be rewritten as $m_1 \frac{\sigma}{R_o^2}\frac{p^*}{R*}$ with $p^* = \left(\frac{1}{R^*} + \frac{2}{(R^*)^2}\right)$.[2]Here m_1 is the scaling coefficient.

$$\left(\frac{\rho U_c^2}{R_o}\right) u^* \frac{\partial u^*}{\partial r^*} = m_1 \frac{\sigma}{R_o^2}\frac{1}{R^*}\left(\frac{1}{R^*} + \frac{2R_o}{(R^*)^2 R_o}\right) + \frac{\tau_{rc}}{R_o}\left(\frac{\partial \tau_{rr}^*}{\partial r^*} + \frac{\tau_{rr}^*}{r^*}\right) + \frac{\tau_{zc}}{Z_c}\frac{\partial \tau_{rz}^*}{\partial z^*} \tag{6}$$

$$u^* \frac{\partial u^*}{\partial r^*} = m_1 \frac{\sigma\lambda}{\eta R_o}\frac{1}{R^*}\left(\frac{1}{R^*} + \frac{2}{(R^*)^2}\right) + \left(\frac{\partial \tau_{rr}^*}{\partial r^*} + \frac{\tau_{rr}^*}{r^*}\right) + \frac{2}{Wi_c}\frac{\partial \tau_{rz}^*}{\partial z^*}$$

Further m_2 and m_3 defined here as the scaling coefficients of elastic and viscous terms respectively along with $r^* \sim R^* = R/R_o$, $z^* \sim Z^* = (R/R_o)^2$ $u^* \sim U^* = U/U_c$ are introduced to obtain the scaled equation for neck evolution.

$$\frac{(U^*)^2}{R^*} = m_1 \frac{\sigma\lambda}{\eta R_o}\frac{1}{R^*}\left(\frac{1}{R^*} + \frac{2}{(R^*)^2}\right) + m_2 \frac{\tau_{rr}^*}{R^*} + \frac{2m_3}{Wi_c}\frac{\tau_{rz}^*}{Z^*} \tag{7}$$

From Eq 8a $\tau_{rr}^* \sim 1 + K_1/(U^*)^2 \sim K_1'\big(1 + 1/(U^*)^2\big)$ as $1/U^*$ is the dominant term and the new scaling coefficient K_1' will adjust the minor contributions by $1/K_1$ term similarly Eq 5b gives $\tau_{rz}^* \sim K_2 R^*$. These when substituted in Eq 7 gives:

$$\frac{(U^*)^2}{R^*} = M_1 \frac{1}{R^*}\left(\frac{1}{R^*} + \frac{2}{(R^*)^2}\right) + \frac{M_2}{R^*}\big(1 + \frac{1}{(U^*)^2}\big) + \frac{2M_3}{Wi_c}\frac{R^*}{Z^*} \tag{8}$$

Here $M_1 := m_1 \frac{\sigma\lambda}{\eta R_o} = \frac{m_1}{Ec_{-1}}$, $M_2 := m_2 K_1'$, and $M_3 := m_3 K_2$ are the final scaling coefficients that have absorbed all the previous parameters. Eq 8 is simplified further to obtain the dimensionless neck evolution equation as :

$$(U^*)^2 = M_1\left(\frac{1}{R^*} + \frac{2}{(R^*)^2}\right) + M_2\big(1 + \frac{1}{(U^*)^2}\big) + \frac{2M_3}{Wi_c}$$

$$(U^*)^4 - \left(M_1\left(\frac{1}{R^*} + \frac{2}{(R^*)^2}\right) + M_2 + \frac{2M_3}{Wi_c}\right)(U^*)^2 - M_2 = 0 \tag{9}$$

Experimental setup validation

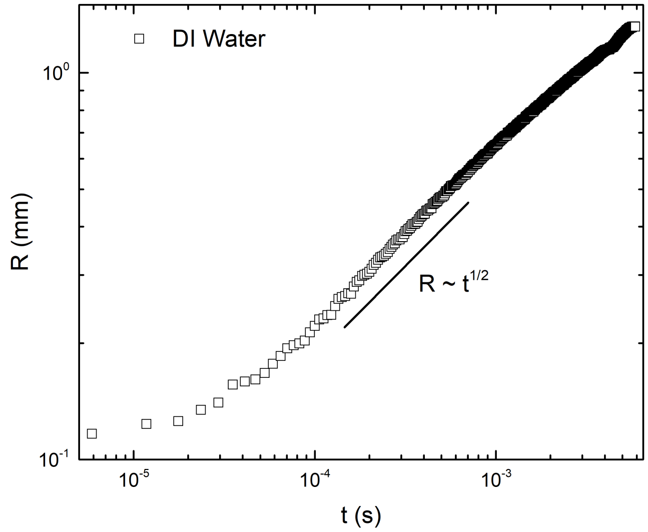

Figure S1: Neck radius evolution of DI water.

Power law fit

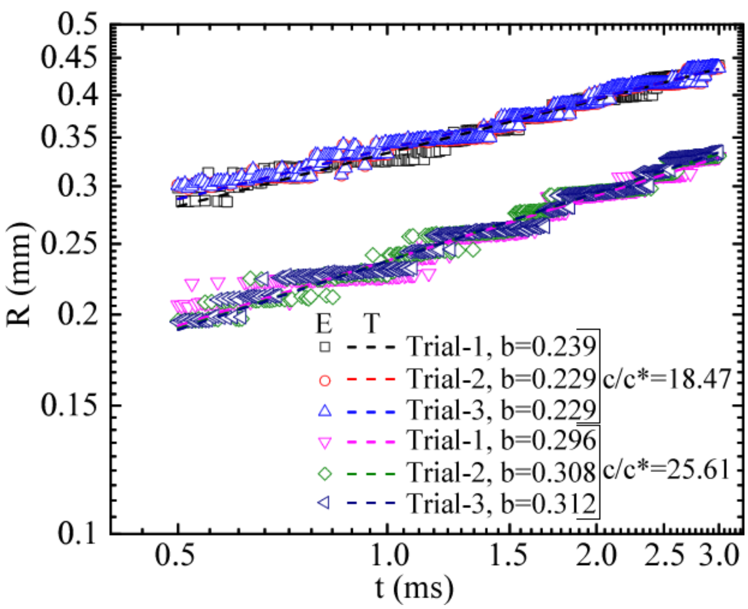

Figure S2: Temporal evolution of neck radius in the region of interest for different experimental trials of $c/c^* = 18.47, 25.61$.

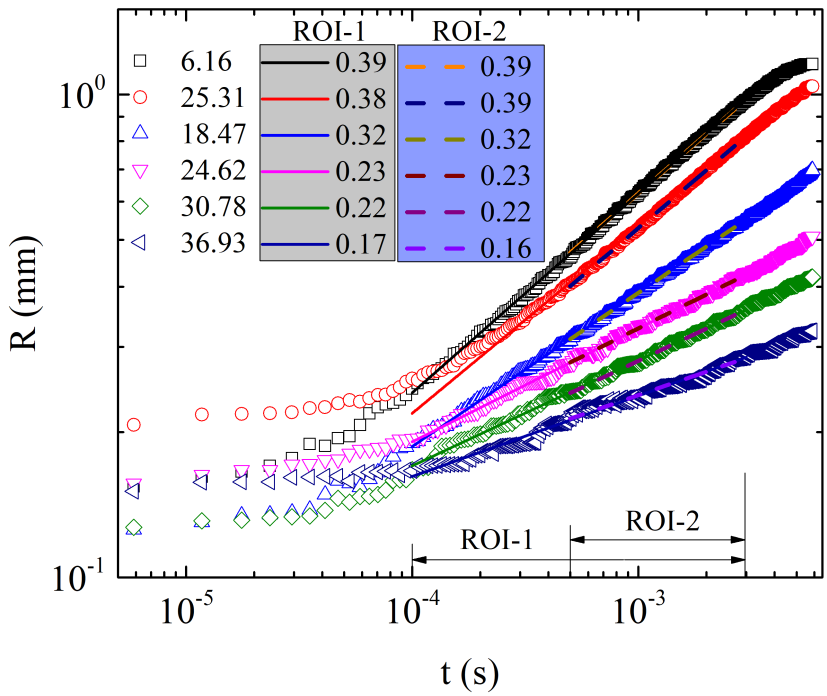

Figure S3: Power law fitting for different ROIs.

Constants and Dimensionless numbers

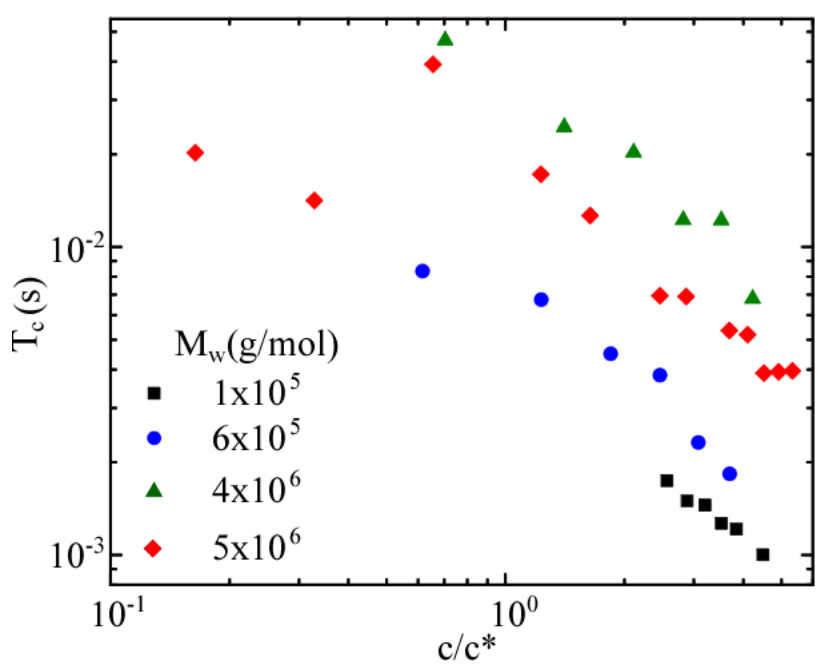

Figure S4: Variation of characteristic time T_c with c/c^* across molecular weights.

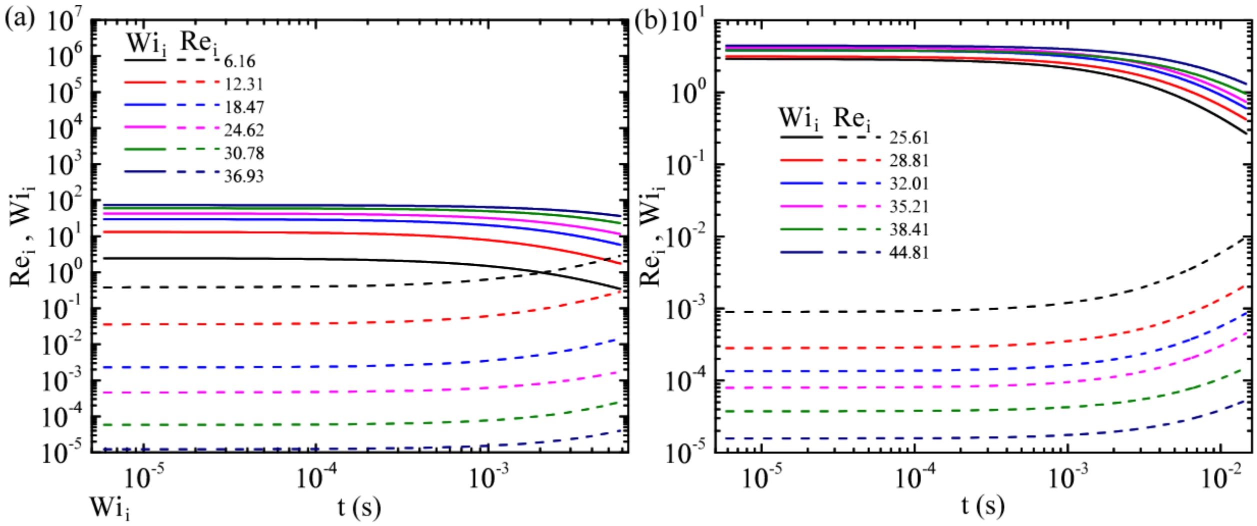

Figure S5: Variation of non-dimensional numbers Re and Wi with time for PEO of (a) $M_w = 6 \times 10^5$ and (b) $M_w = 1 \times 10^5$ g/mol.

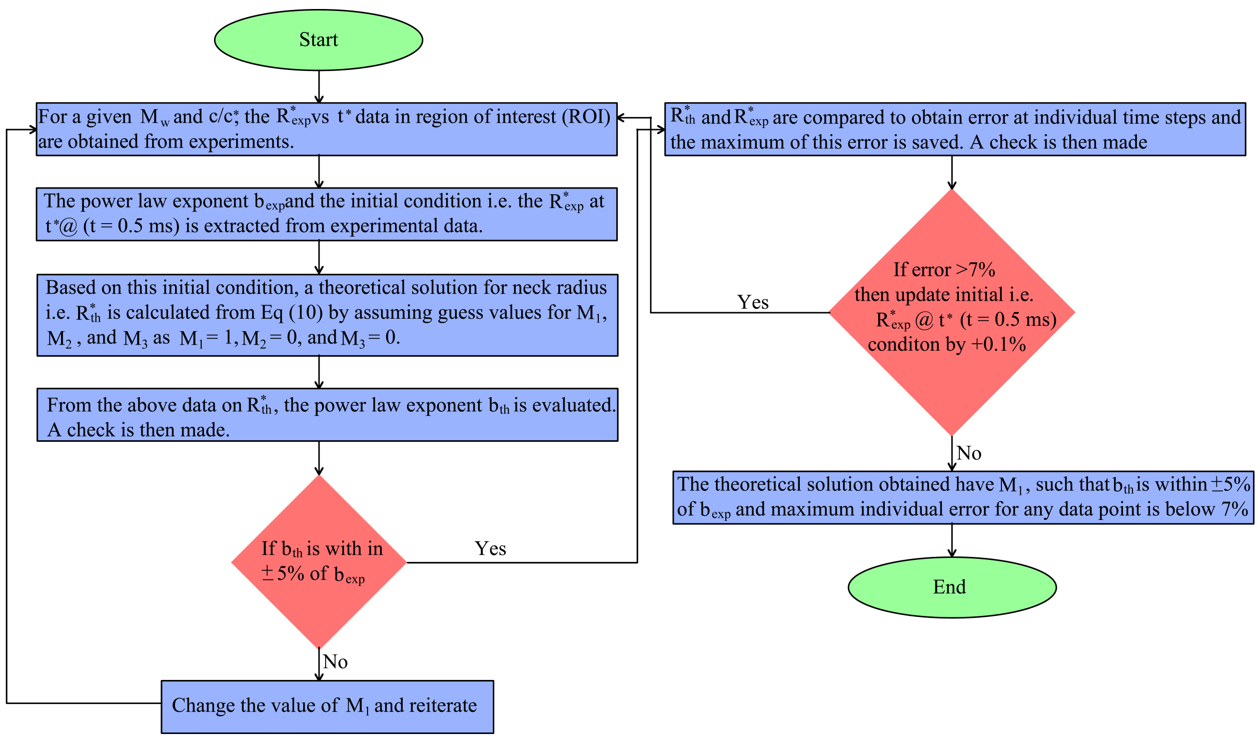

Figure S6: Flow chart represent the algorithm to obtain M_1 values.

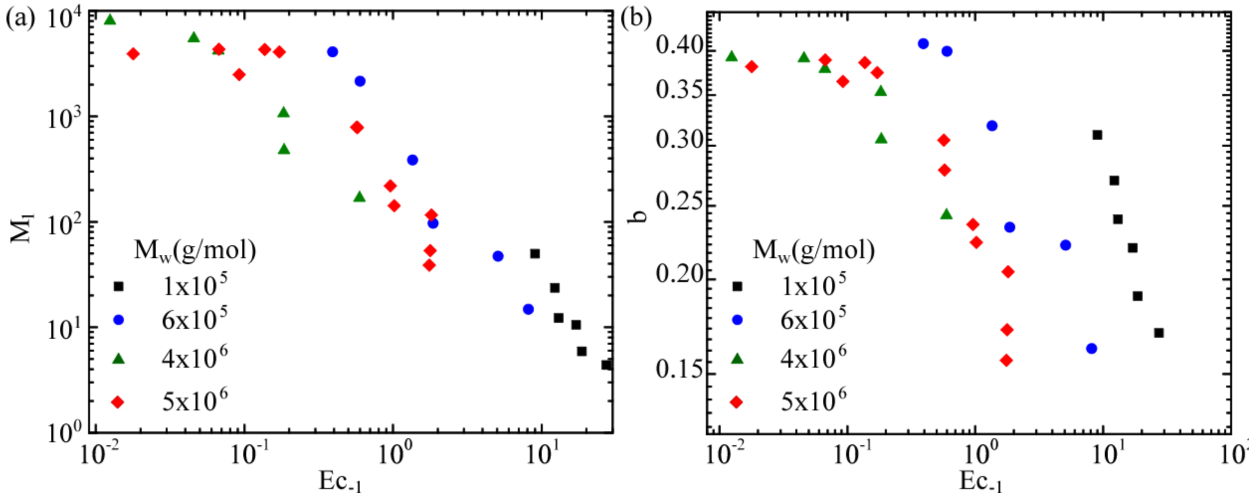

Figure S7: (a) Variation of M_1 with Ec_{-1} and (b) Variation of b with Ec_{-1} across molecular weights.

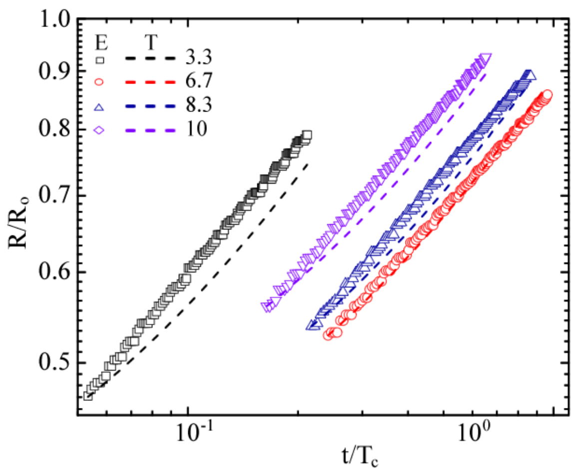

Figure S8: Comparison of dimensionless neck radius evolution obtained from experiments with solution of Eq (10) for various c/c^* of PAM $M_w = 5 \times 10^6$ g/mol. (obtained from Varma et al.[3]).

Arrested Coalescence

Analysis

$$-M_1 = \frac{M_2(1 + \frac{2}{Wi_c})}{\frac{1}{R^*} + \frac{2}{(R^*)^2}} \tag{10}$$

$$-\frac{m_1}{Ec_{-1}} = \frac{M_2(1 + \frac{2}{Wi_c})}{\frac{1}{R^*} + \frac{2}{(R^*)^2}}$$

The term in denominator $\mathcal{O}(\frac{2}{(R^*)^2}) > \mathcal{O}(\frac{1}{(R^*)})$ therefore we can neglect the lower order term to get an approximate value for θ_{arrest}. Similarly in numerator $1 > 2/Wi_c$ for high elasticity droplets(that will show arrest)

$$-\frac{m_1}{Ec_{-1}M_2} = \frac{(R^*)^2}{2} = \frac{\theta^2_{arrest}}{2}$$

$$\frac{20}{Ec_{-1}} = \theta^2_{arrest} \tag{11}$$

DI Water

$R(t = 0.5 \text{ ms}) = 0.41$ mm and $R(t = 3 \text{ ms}) = 0.89$ mm

$\dot{\gamma}_{DI} = \frac{R(t=3 \text{ ms}) - R(t=0.5 \text{ ms})}{R(t=0.5 \text{ ms})(3-0.5)}$

$\dot{\gamma}_{DI} = 0.47 \text{ ms}^{-1}$

PEO $M_w = 1 \times 10^5$ g/mol, $c/c* = 70$

$R(t = 0.5 \text{ ms}) = 0.24$ mm and $R(t = 1000 \text{ ms}) = 0.74$ mm

$\dot{\gamma}_{arrest} = \frac{R(t=1000 \text{ ms}) - R(t=0.5 \text{ ms})}{R(t=0.5 \text{ ms})(1000-0.5)}$

$\dot{\gamma}_{arrest} = 0.002 \text{ ms}^{-1}$

$\dot{\gamma}_{arrest}/\dot{\gamma}_{DI} * 100 = 0.43\%$

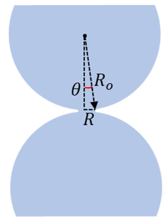

Figure S9: Schematic of the angle θ subtended at the neck.

Rheology, Non-Dimensional numbers, Non-Dimensionalising Variables

The relaxation time λ for all the solutions are obtained using the Zimm model[4] .

$$\lambda_z = \frac{1}{\zeta(3\nu)} \frac{[\eta] M_w \eta_s}{\mathrm{N_A k_B} T} \tag{12}$$

Here, η_s, $\mathrm{k_B}$, λ_z, T, ν are solvent viscosity, Boltzmann constant, Zimm relaxation time, absolute temperature and fractal polymer dimension determined using the relation $A = 3\nu - 1$, (where A is the exponent of Mark-Houwink-Sakurada correlation) respectively. The relaxation times of the solutions in semi-dilute unentangled λ_{SUE} and semi-dilute entangled λ_{SE} regimes, are obtained using these correlations : $\lambda_{\mathrm{SUE}} = \lambda_z \left(\frac{c}{c^*}\right)^{\frac{2-3\nu}{3\nu-1}}$ and $\lambda_{\mathrm{SE}} = \lambda_z \left(\frac{c}{c^*}\right)^{\frac{3-3\nu}{3\nu-1}}$ [5–7] respectively. The values of viscosity, relaxation time and concentration ratios for molecular weights $M_w = 5 \times 10^6$ and 4×10^6 g/mol are obtained from our previous study.[2]

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