# AN ORTHOTROPIC CLOSURE APPROXIMATION FOR THE FOURTH ORDER MOMENT OF A STRAND VECTOR AND ITS CONSEQUENCES ON PARTIALLY EXTENDING STRAND CONVECTION MODELS FOR POLYMER MELTS

**Roney L. Thompson, rthompson@mec.uff.br**
LFTC-LMTA, Department of Mechanical Engineering (PGMEC), Universidade Federal Fluminense, Rua Passo da Patria 156, 24210-240, Niteroi, RJ, Brasil

***Abstract.*** *The partially extending strand convection model of Larson (1984) is analyzed from the framework perspective constructed by Pasquali and Scriven (2004) and it is shown that the terms related to stretching and rotation concerning a strand vector do not originate these same phenomena as far as the second moment of the probability density function, the conformation tensor, is considered. In order to produce each of these phenomena on the mesoscopic level from the correspondent phenomena at the microscopic level, it is necessary to change the classical closure approximation for the fourth moment of the probability density function of a representative microelement vector (the quadratic closure approximation) to a special one that belongs to the class of orthotropic ones presented by Cintra and Tucker (1995). A decomposition theorem presented by Thompson (2008) is used to provide such a closure approximation which are more physically consistent in the sense stated above. The new class of models for the evolution equation of the second order conformation tensor is presented with the same framework (Pasquali and Scriven, 2004). An interesting improvement is the fact that these models do not lead to 3-D instabilities in a 2-D flow. The concept of the natural convected time derivative (Thompson, 2008) is used in order to split the general evolution equation into two parts: a stretching-evolution equation and a rotation-evolution equation. This framework gives an alternative simulation algorithm that can be coupled with the matrix-logarithm approach presented by Fattal and Kupferman (2004).*

***Keywords:*** *Stretching and rotating decoupling; Partially extended convected models; Orthotropic closure approximations*

## 1. INTRODUCTION

A large variety of flowing systems of complex microstructured fluids is described by a probability density function, $\psi(\mathbf{R}, \mathbf{x}, t)$ where $\mathbf{R}$ is a representative vector that describes the main aspect of the microstructure, $\mathbf{x}$ is the position vector and $t$ is the current time. In suspensions, $\mathbf{R}$ is the vector of the major axis of a given particle. In immiscible blends, $\mathbf{R}$ is the outwardly directed unit vector normal to the droplet surface. In dilute solutions of polymeric liquids, $\mathbf{R}$ is the end-to-end distance of the polymer while in melts it is the difference between two entanglements.

The molecular theories developed to understand the behavior of such fluids are, therefore, based on an evolution equation for the probability density function. The probability density function obeys the probability continuity equation of the form

$$\frac{D\psi}{Dt} + \nabla_{\mathbf{R}} \cdot \mathbf{J}_\psi = \psi_p - \psi_d \tag{1}$$

where $\mathbf{J}_\psi = \psi\dot{\mathbf{R}}$ is the probability density flux, the operator $\nabla_{\mathbf{R}} \cdot ()$ is the divergence resolved in the configurational space $R^3$, and $\psi_p$ and $\psi_d$ are, respectively, the local (in the sense of the Euclidean and configurational spaces) rates of production and destruction of segments per unit mass. The calculation of the elastic stress tensor was identified as strongly related (in fact proportional in most cases) to the second moment of the probability density function. Besides that, the second moment can capture the main features of a microstructure, namely size and orientation. Hence, its evolution equation can model stretching and change on orientation. Therefore, in order to gain economy on computational calculations, a simpler approach considers the evolution equation of the second moment of the probability density function defined as

$$\langle \mathbf{RR} \rangle (\mathbf{x}, t) = \int_{R^3} \psi(\mathbf{R}, \mathbf{x}, t)\, \mathbf{RR}\; d\mathbf{R} \tag{2}$$

$\mathbf{\Gamma} \equiv \langle \mathbf{RR} \rangle$ represents a *conformation* tensor. The approach that considers its evolution (instead of considering the evolution of the probability density function) is a *mesoscopic* approach.

In order to compute the evolution equation for the conformation tensor $\langle \dot{\mathbf{RR}} \rangle$ (suppressing the dependence on $(\mathbf{x}, t)$) we have to use eqs.(1) and (2).

$$\langle \dot{\mathbf{RR}} \rangle = \langle \dot{\mathbf{R}}\mathbf{R} \rangle + \langle \mathbf{R}\dot{\mathbf{R}} \rangle = \int_{R^3} \left( \mathbf{J}_\psi \mathbf{R} + \mathbf{R}\mathbf{J}_\psi \right)\; d\mathbf{R} \tag{3}$$

Depending on the form of the flux $\mathbf{J}_\psi$ and the rate of production and destruction of segments (and the interaction with the medium), different evolution equations are obtained.

## 2. THE THERMODYNAMIC FRAMEWORK OF PASQUALI & SCRIVEN (2004)

Pasquali and Scriven (2004) developed an elegant thermodynamic framework that accounts for microstructural features of flowing complex fluids at the level of mesoscopic models. In addition to the classical (Newtonian) variables, the local thermodynamic state of the liquid is defined by the conformation tensor and entanglement concentration.

The general evolution equation for the conformation tensor is given by [1]

$$\dot{\mathbf{\Gamma}} = 2\xi\frac{\mathbf{D}:\mathbf{\Gamma}}{\mathbf{\Gamma}:\mathbf{\Gamma}}\mathbf{\Gamma} + \zeta\left(\mathbf{\Gamma}\cdot\mathbf{D} + \mathbf{D}\cdot\mathbf{\Gamma} - 2\frac{\mathbf{D}:\mathbf{\Gamma}}{\mathbf{\Gamma}:\mathbf{\Gamma}}\mathbf{\Gamma}\right) + \mathbf{W}\cdot\mathbf{\Gamma} - \mathbf{\Gamma}\cdot\mathbf{W} + \mathbf{G}\left(\mathbf{\Gamma}\right) \tag{4}$$

where $\xi$ and $\zeta$ are, respectively, the stretching and rotation resistant coefficients, and $\mathbf{G}\left(\mathbf{\Gamma}\right)$ is an isotropic tensor valued function of the conformation tensor that accounts for relaxation and other sources (such as entanglement production and destruction).

We can roughly divide the complexity of the flowing system in three categories. The first one is obtained by the class of affine models, models in which $\xi = \zeta = 1$. In this case Eq.(4) is simplified to

$$\overset{\triangledown}{\mathbf{\Gamma}} = \mathbf{G}\left(\mathbf{\Gamma}\right) \tag{5}$$

where $\overset{\triangledown}{\mathbf{\Gamma}}$ is the upper convected time derivative of $\mathbf{\Gamma}$. The FENE models, Giesekus, are examples which belong to this class. The second class is composed by non-affine deformation of the molecules, but the resistance to stretching is the same as the rotation one. In other words $\xi = \zeta \neq 1$. Equation (4), therefore, can be written as

$$\overset{\triangledown}{\mathbf{\Gamma}} = \mathbf{G}\left(\mathbf{\Gamma}\right) - \left(1-\xi\right)\left(\mathbf{\Gamma}\mathbf{D} + \mathbf{D}\mathbf{\Gamma}\right) \tag{6}$$

The main representative models from this class are the Jonhson-Segalman and the PTT. Finally, the third class is composed by those models in which $\xi \neq \zeta$. The two models which will be discussed here that belong to this third group are .

### 2.1 The third class

In the first two categories we are not able to discriminate the origins of stretching and rotation of the evolution equation of the conformation tensor from the microscopic model for the strand, since $\xi = \zeta$. Therefore, the main feature of the third class of models is that we really have to think separately about stretching and rotation. For this reason we will analyze Eq.(4) for the case where $\xi \neq \zeta$. It is worth noticing that the partially extending strand convection model of Larson (1984) is special cases of this third class.

Looking to Eq.(4) we are induced to identify the term $2\xi\frac{\mathbf{D}:\mathbf{\Gamma}}{\mathbf{\Gamma}:\mathbf{\Gamma}}\mathbf{\Gamma}$ as *the* stretching term and the term $\zeta\left(\mathbf{\Gamma}\mathbf{D} + \mathbf{D}\mathbf{\Gamma} - 2\frac{\mathbf{D}:\mathbf{\Gamma}}{\mathbf{\Gamma}:\mathbf{\Gamma}}\mathbf{\Gamma}\right)$ with a change on the conformation due to rotation. This induction comes from the fact that the former is multiplied by $\xi$, the stretching resistance, while the latter is multiplied by $\zeta$, the rotation resistance.

However, if we identify the term $2\xi\frac{\mathbf{D}:\mathbf{\Gamma}}{\mathbf{\Gamma}:\mathbf{\Gamma}}\mathbf{\Gamma}$ as *the* stretching term we will have to work with the fact that the rate of stretch in a certain eigendirection is proportional to the eigenvalue of that direction irrespective of the relative direction between $\mathbf{D}$ and $\mathbf{\Gamma}$. This happens because $2\xi\frac{\mathbf{D}:\mathbf{\Gamma}}{\mathbf{\Gamma}:\mathbf{\Gamma}}$ is a scalar, and, therefore, the stretches are proportional to $\mathbf{\Gamma}$. Hence, if we use the change of variable suggested by Fattal and Kupferman (2004), the group of molecules is obliged to have the same rate of log-stretching in every direction. The nonphysical consequences of this behavior are easy to point out. For example, let us consider that the deformation tensor $\mathbf{D}$ and the conformation tensor $\mathbf{\Gamma}$ have the same eigenvectors and that the vorticity tensor is null ($\mathbf{W} = \mathbf{0}$) since $\tau = -\infty$. If we consider incompressible materials then there is at least one eigenvalue of $\mathbf{D}$ positive ($\lambda_1^D$) and another negative ($\lambda_3^D$). The influence of $\mathbf{D} = \mathbf{L}$ on the flow should be to stretch the log-conformation tensor at direction 1 and to compress the conformation tensor in direction 3 even if $\lambda_3^\Gamma > \lambda_1^\Gamma$, for example. If $\lambda_3^D = -\lambda_1^D$, a two-dimensional flow, the term $2\xi\frac{\mathbf{D}:\mathbf{\Gamma}}{\mathbf{\Gamma}:\mathbf{\Gamma}}\mathbf{\Gamma}$ produces stretching on the neutral direction also.

There is also a problem on identifying the term $\zeta\left(\mathbf{\Gamma}\mathbf{D} + \mathbf{D}\mathbf{\Gamma} - 2\frac{\mathbf{D}:\mathbf{\Gamma}}{\mathbf{\Gamma}:\mathbf{\Gamma}}\mathbf{\Gamma}\right)$ with a change on the conformation due to rotation. The reason is because this term is not, in general, orthogonal to $\mathbf{\Gamma}$. This is easily verified if we take the double dot product (which for symmetric tensors is also the inner product) $\zeta\left(\mathbf{\Gamma}\mathbf{D} + \mathbf{D}\mathbf{\Gamma} - 2\frac{\mathbf{D}:\mathbf{\Gamma}}{\mathbf{\Gamma}:\mathbf{\Gamma}}\mathbf{\Gamma}\right) : \mathbf{\Gamma}$ and find that this is not necessarily a null tensor. Therefore, there is stretching in this term.

The considerations above indicate that, in fact, the term $2\xi\frac{\mathbf{D}:\mathbf{\Gamma}}{\mathbf{\Gamma}:\mathbf{\Gamma}}\mathbf{\Gamma}$ is not the stretching term (in general) of the conformation tensor, while the term $\zeta\left(\mathbf{\Gamma}\mathbf{D} + \mathbf{D}\mathbf{\Gamma} - 2\frac{\mathbf{D}:\mathbf{\Gamma}}{\mathbf{\Gamma}:\mathbf{\Gamma}}\mathbf{\Gamma}\right)$ does not represent a change on orientation due to purely rotation for materials which can be represented by Eq.(4) when $\xi \neq \zeta$, like the model developed by Larson (1984).

As pointed by Pasquali and Scriven (2004), another intrinsic problem of models of this third class is the fact that they can present 3-D instabilities of the 2-D flow.

[1] Note that here we adopt as the vorticity tensor the quantity $\mathbf{W} \equiv \frac{1}{2}\left(\mathbf{L} - \mathbf{L}^T\right)$ while Pasquali and Scriven (2004) use $\mathbf{W} \equiv \frac{1}{2}\left(\nabla\mathbf{v} - \nabla\mathbf{v}^T\right)$, where $\mathbf{L} \equiv \nabla\mathbf{v}^T$

Since the quantities $\xi$ and $\zeta$ are the resistant parameters associated, respectively, to stretching and rotation of a single microelement, the approximation introduced to filter the information from microstructure to the mesoscopic level is responsible for an influence on the other phenomenon. More specifically, we will show that in the backbone represented by Eq.(4) the rotation of a microelement is part of the stretching of the conformation tensor.

### 2.2 A decomposition of a tensor with respect to another

In Thompson (2008) a decomposition theorem was presented. Here we will synthesize considering two second order symmetric tensors $\boldsymbol{\mathcal{U}}$ and $\boldsymbol{\mathcal{V}}$. Let us call $\mathbf{e}_i^{\mathcal{U}}$ and $\lambda_i^{\mathcal{U}}$ the (real) unit eigenvectors and eigenvalues of tensor $\boldsymbol{\mathcal{U}}$, respectively. Let us define a fourth order tensor $\mathbf{1}^{\mathcal{U}\mathcal{U}}$ as

$$\mathbf{1}^{\mathcal{U}\mathcal{U}} = \sum_{k=1}^{3} \mathbf{e}_k^{\mathcal{U}}\mathbf{e}_k^{\mathcal{U}}\mathbf{e}_k^{\mathcal{U}}\mathbf{e}_k^{\mathcal{U}} \tag{7}$$

and a decomposition of tensor $\boldsymbol{\mathcal{V}}$, such that

$$\boldsymbol{\mathcal{V}} = \boldsymbol{\Phi}_{\mathcal{V}}^{\mathcal{U}} + \widetilde{\boldsymbol{\Phi}}_{\mathcal{V}}^{\mathcal{U}} \tag{8}$$

where

$$\boldsymbol{\Phi}_{\mathcal{V}}^{\mathcal{U}} = \mathbf{1}^{\mathcal{U}\mathcal{U}} : \boldsymbol{\mathcal{V}} \tag{9}$$

$$\widetilde{\boldsymbol{\Phi}}_{\mathcal{V}}^{\mathcal{U}} = \left(\mathbf{1}^{\delta\delta} - \mathbf{1}^{\mathcal{U}\mathcal{U}}\right) : \boldsymbol{\mathcal{V}} \tag{10}$$

where $\mathbf{1}^{\delta\delta}$ is the fourth order identity tensor that when applied to any second order tensor maps this tensor to itself, as follows

$$\mathbf{1}^{\delta\delta} : \boldsymbol{B} = \boldsymbol{B} \tag{11}$$

Then, the decomposition given by Eqs.(8), (9), and (10) have the following properties

1. $\boldsymbol{\Phi}_{\mathcal{V}}^{\mathcal{U}}$ and $\widetilde{\boldsymbol{\Phi}}_{\mathcal{V}}^{\mathcal{U}}$ are orthogonal.
2. $\boldsymbol{\mathcal{U}}$ and $\boldsymbol{\Phi}_{\mathcal{V}}^{\mathcal{U}}$ are coaxial (commute).
3. $\boldsymbol{\mathcal{U}}$ and $\widetilde{\boldsymbol{\Phi}}_{\mathcal{V}}^{\mathcal{U}}$ are orthogonal.
4. $\boldsymbol{\mathcal{U}} : \boldsymbol{\mathcal{V}} = \boldsymbol{\mathcal{U}} : \boldsymbol{\Phi}_{\mathcal{V}}^{\mathcal{U}}$
5. $\widetilde{\Xi}\left[\boldsymbol{\mathcal{U}}, \boldsymbol{\mathcal{V}}\right] = \widetilde{\Xi}\left[\boldsymbol{\mathcal{U}}, \widetilde{\boldsymbol{\Phi}}_{\mathcal{V}}^{\mathcal{U}}\right]$

where the second order tensor-valued function $\widetilde{\Xi}\left[\boldsymbol{\mathcal{U}}, \boldsymbol{\mathcal{V}}\right]$ denotes the Lie product between any two second order tensors, defined as

$$\widetilde{\Xi}\left[\boldsymbol{\mathcal{U}}, \boldsymbol{\mathcal{V}}\right] \equiv \boldsymbol{\mathcal{U}} \cdot \boldsymbol{\mathcal{V}} - \boldsymbol{\mathcal{V}} \cdot \boldsymbol{\mathcal{U}} \tag{12}$$

Because of the above properties, tensors $\boldsymbol{\Phi}_{\mathcal{V}}^{\mathcal{U}}$ and $\widetilde{\boldsymbol{\Phi}}_{\mathcal{V}}^{\mathcal{U}}$ were called (Thompson (2008)) the *in-phase* and *out-of-phase* parts of $\boldsymbol{\mathcal{V}}$ with respect to $\boldsymbol{\mathcal{U}}$. The following properties are important in the present context

1. When tensors $\boldsymbol{\mathcal{U}}$ and $\boldsymbol{\mathcal{V}}$ commute, $\boldsymbol{\Phi}_{\mathcal{V}}^{\mathcal{U}} = \boldsymbol{\mathcal{V}}$ and $\widetilde{\boldsymbol{\Phi}}_{\mathcal{V}}^{\mathcal{U}} = \mathbf{0}$.
2. When $\boldsymbol{\mathcal{V}} = \mathbf{A}^S\mathbf{B}^W - \mathbf{B}^W\mathbf{A}^S$, where $\mathbf{A}^S$ is symmetric and $\mathbf{B}^W$ is skew-symmetric, and $\boldsymbol{\mathcal{U}}$ commutes with $\mathbf{A}^S$, $\boldsymbol{\Phi}_{\mathcal{V}}^{\mathcal{U}} = \mathbf{0}$ and $\widetilde{\boldsymbol{\Phi}}_{\mathcal{V}}^{\mathcal{U}} = \boldsymbol{\mathcal{V}}$.

## 3. APPLICATION OF THE DECOMPOSITION THEOREM ON THE GENERAL EQUATION OF PASQUALI & SCRIVEN (2004)

As shown by Thompson (2008) the part of tensor $\mathbf{D}\cdot\boldsymbol{\Gamma} + \boldsymbol{\Gamma}\cdot\mathbf{D}$ which is coaxial to the conformation tensor is $2\boldsymbol{\Phi}_{\boldsymbol{D}}^{\boldsymbol{\Gamma}}\cdot\boldsymbol{\Gamma}$. Therefore, we can write Eq.(4) as

$$\dot{\boldsymbol{\Gamma}} = 2(\xi - \zeta)\frac{\mathbf{D} : \boldsymbol{\Gamma}}{\boldsymbol{\Gamma} : \boldsymbol{\Gamma}}\boldsymbol{\Gamma} + 2\zeta\boldsymbol{\Phi}_{\boldsymbol{D}}^{\boldsymbol{\Gamma}} \cdot \boldsymbol{\Gamma} +$$

$$\zeta\left(\mathbf{\Gamma}\cdot\mathbf{D}+\mathbf{D}\cdot\mathbf{\Gamma}-2\mathbf{\Phi}_{\boldsymbol{D}}^{\mathbf{\Gamma}}\cdot\mathbf{\Gamma}\right)+\mathbf{W}\cdot\mathbf{\Gamma}-\mathbf{\Gamma}\cdot\mathbf{W}+\mathbf{G}\left(\mathbf{\Gamma}\right) \tag{13}$$

The term $2(\xi-\zeta)\frac{\mathbf{D}:\mathbf{\Gamma}}{\mathbf{\Gamma}:\mathbf{\Gamma}}\mathbf{\Gamma}+2\zeta\mathbf{\Phi}_{\boldsymbol{D}}^{\mathbf{\Gamma}}\cdot\mathbf{\Gamma}$ is, therefore, the stretching term (of the conformation tensor) due to the flow. This shows that in the backbone constructed by Pasquali and Scriven (2004) predicts a *stretching* of the conformation tensor that is originated by *rotation* of the microelement. The term $\zeta\left(\mathbf{\Gamma}\cdot\mathbf{D}+\mathbf{D}\cdot\mathbf{\Gamma}-2\mathbf{\Phi}_{\boldsymbol{D}}^{\mathbf{\Gamma}}\cdot\mathbf{\Gamma}\right)+\mathbf{W}\cdot\mathbf{\Gamma}-\mathbf{\Gamma}\cdot\mathbf{W}$ is the rate of rotation (of the conformation tensor) due to the flow. This tensor is always orthogonal to $\mathbf{\Gamma}$.

## 4. APPLICATION OF THE DECOMPOSITION THEOREM ON THE MICRO-MACRO EQUATION USING A NEW ORTHOTROPIC CLOSURE APPROXIMATION

Following Pasquali and Scriven (2004) we can divide the contributions to the evolution of strand into three parts: a flow contribution, an entropic relaxation contribution, a brownian motion contribution. In other words

$$\langle\dot{\mathbf{R}\mathbf{R}}\rangle=\langle\dot{\mathbf{R}\mathbf{R}}\rangle_{flow}+\langle\dot{\mathbf{R}\mathbf{R}}\rangle_{ent-relax}+\langle\dot{\mathbf{R}\mathbf{R}}\rangle_{brown}+\langle\dot{\mathbf{R}\mathbf{R}}\rangle_{entang} \tag{14}$$

The contribution of the flow to the evolution of a strand, considering a resistance to stretching, $\xi$, and to rotation, $\zeta$, is given by

$$\dot{\mathbf{R}}_{flow}=\xi\mathbf{R}\mathbf{e}^R\mathbf{e}^R:\mathbf{D}+\zeta\left(\mathbf{D}\cdot\mathbf{R}-\mathbf{R}\mathbf{e}^R\mathbf{e}^R:\mathbf{D}\right)+\mathbf{W}\cdot\mathbf{R} \tag{15}$$

$$\langle\dot{\mathbf{R}\mathbf{R}}\rangle_{flow}=\int_{R^3}2\xi\psi\mathbf{R}\mathbf{R}\mathbf{e}^R\mathbf{e}^R:\mathbf{D}\;d\mathbf{R}+$$
$$\int_{R^3}\zeta\psi\left(\mathbf{R}\mathbf{R}\cdot\mathbf{D}+\mathbf{D}\cdot\mathbf{R}\mathbf{R}-2\mathbf{R}\mathbf{R}\mathbf{e}^R\mathbf{e}^R:\mathbf{D}\right)+$$
$$\int_{R^3}\psi\left(\mathbf{W}\cdot\mathbf{R}\mathbf{R}-\mathbf{R}\mathbf{R}\cdot\mathbf{W}\right)\;d\mathbf{R} \tag{16}$$

If we consider that the length scale where the velocity gradient does not change is larger when compared to the length scale of a representative configurational space, we can take the tensors which are related to the mean flow out of averaging. Therefore we have that

$$\dot{\mathbf{\Gamma}}=2\xi\langle\mathbf{R}\mathbf{R}\mathbf{e}^R\mathbf{e}^R\rangle:\mathbf{D}+$$
$$\zeta\left(\langle\mathbf{R}\mathbf{R}\rangle\cdot\mathbf{D}+\mathbf{D}\cdot\langle\mathbf{R}\mathbf{R}\rangle-2\langle\mathbf{R}\mathbf{R}\mathbf{e}^R\mathbf{e}^R\rangle:\mathbf{D}\right)+$$
$$\mathbf{W}\cdot\langle\mathbf{R}\mathbf{R}\rangle-\langle\mathbf{R}\mathbf{R}\rangle\cdot\mathbf{W} \tag{17}$$

where $\langle R^2\rangle\equiv\mathrm{tr}\langle\mathbf{R}\mathbf{R}\rangle$.

The hypothesis considered to make a closure approximation for the fourth order tensor as a function of the second order is to maintain the phenomena of stretching and rotation with the same compliance factors $\xi$ and $\zeta$. This result is achieved if eigenstructure of the second order moment is maintained for the fourth moment as

$$\langle\mathbf{R}\mathbf{R}\rangle.\mathbf{x}=\lambda\mathbf{x}\Rightarrow\langle\mathbf{R}\mathbf{R}\mathbf{R}\mathbf{R}\rangle:\mathbf{x}\mathbf{x}=\lambda^2\mathbf{x}\mathbf{x} \tag{18}$$

In other words if we considered that $\lambda_i^\Gamma$ and $\mathbf{e}_i^\Gamma$ are, respectively the eigenvalues and eigenvectors of $\mathbf{\Gamma}\equiv\langle\mathbf{R}\mathbf{R}\rangle$ the maintenance of the eigenstructure means that $\left(\lambda_i^\Gamma\right)^2$ and $\mathbf{e}_i^\Gamma\mathbf{e}_i^\Gamma$ are the eigenvalues and eigendyadics which are solution of the second equation of (18). Therefore, we can write tensor $\langle\mathbf{R}\mathbf{R}\mathbf{e}^R\mathbf{e}^R\rangle$ as

$$\langle\mathbf{R}\mathbf{R}\mathbf{e}^R\mathbf{e}^R\rangle=\sum_1^3\lambda_i^\Gamma\mathbf{e}_i^\Gamma\mathbf{e}_i^\Gamma\mathbf{e}_i^\Gamma\mathbf{e}_i^\Gamma \tag{19}$$

Therefore, if we apply three times Eq.(9) we have that

$$\langle\mathbf{R}\mathbf{R}\mathbf{e}^R\mathbf{e}^R\rangle:\mathbf{D}=\sum_1^3\lambda_i^\Gamma D_{ii}\mathbf{e}_i^\Gamma\mathbf{e}_i^\Gamma=\mathbf{\Gamma}\cdot\mathbf{\Phi}_D^\Gamma=\mathbf{\Phi}_{\boldsymbol{D}}^{\mathbf{\Gamma}}\cdot\mathbf{\Gamma} \tag{20}$$

where $\mathbf{\Phi}_{\boldsymbol{D}}^{\mathbf{\Gamma}}$ is the in-phase part of $\mathbf{D}$ related to $\mathbf{\Gamma}$

### 4.1 Modified evolution equation

The discussion above suggests the following modification on Eq.(4)

$$\dot{\mathbf{\Gamma}} = 2\xi\mathbf{\Phi}^{\mathbf{\Gamma}}_{D} \cdot \mathbf{\Gamma} + \zeta\left(\mathbf{\Gamma}\cdot\mathbf{D} + \mathbf{D}\cdot\mathbf{\Gamma} - 2\mathbf{\Phi}^{\mathbf{\Gamma}}_{D}\cdot\mathbf{\Gamma}\right) + \mathbf{W}\cdot\mathbf{\Gamma} - \mathbf{\Gamma}\cdot\mathbf{W} + \mathbf{G}\left(\mathbf{\Gamma}\right) \tag{21}$$

Here we can identify the term $2\xi\mathbf{\Phi}^{\mathbf{\Gamma}}_{D}\cdot\mathbf{\Gamma}$ with a real anisotropic log-stretching that, depending on the form of $\mathbf{D}$, gives different rates of log-stretching in different directions. The term $\zeta\left(\mathbf{\Gamma}\cdot\mathbf{D} + \mathbf{D}\cdot\mathbf{\Gamma} - 2\mathbf{\Phi}^{\mathbf{\Gamma}}_{D}\cdot\mathbf{\Gamma}\right)$ is orthogonal to $\mathbf{\Gamma}$. As was alluded previously, Eq.(21) is no different from the original Eq.(4) when $\xi = \zeta$, the first two categories discussed.

Concerning the three-dimensional stability of two-dimensional flows, as denounced by Pasquali and Scriven (2004), an interesting improvement of this backbone of models represented by Eq.(21) is that the admitting steady solution does not require restrictions on the values of $\xi$ and $\zeta$ to be stable, since now they do not appear in the equation of change of the eigenvalue of the eigenvector correspondent to the neutral direction, since $\mathbf{D}$ is orthogonal to this direction.

The modified Doi-equation for the evolution of the conformation of rodlike polymers would be

While the modified Larson-equation for the evolution of the conformation of partially strand retracted polymers would be

### 4.2 Decoupling the evolution equation

Analyzing Eq.(21), we can easily identify, looking to the righthand side of the equation, that the first and last terms are in-phase with the conformation tensor (commute) while the second and third terms are out-of-phase terms. Therefore, if we split the lefthanded side of Eq.(21) in the same fashion, i.e. in-phase and out-of-phase with respect to $\mathbf{\Gamma}$, we can decouple the evolution into the stretching-evolution and the rotation-evolution equations of the conformation tensor. Hence, for $\dot{\mathbf{\Gamma}} = \mathbf{\Phi}^{\mathbf{\Gamma}}_{\dot{\mathbf{\Gamma}}} + \widetilde{\mathbf{\Phi}}^{\mathbf{\Gamma}}_{\dot{\mathbf{\Gamma}}}$ we have that

$$\mathbf{\Phi}^{\mathbf{\Gamma}}_{\dot{\mathbf{\Gamma}}} = 2\xi\mathbf{\Phi}^{\mathbf{\Gamma}}_{D}\cdot\mathbf{\Gamma} + \mathbf{G}\left(\mathbf{\Gamma}\right) \tag{22}$$

$$\widetilde{\mathbf{\Phi}}^{\mathbf{\Gamma}}_{\dot{\mathbf{\Gamma}}} = \zeta\left(\mathbf{\Gamma}\cdot\mathbf{D} + \mathbf{D}\cdot\mathbf{\Gamma} - 2\mathbf{\Phi}^{\mathbf{\Gamma}}_{D}\cdot\mathbf{\Gamma}\right) + \mathbf{\Gamma}\cdot\mathbf{W} - \mathbf{W}\cdot\mathbf{\Gamma} \tag{23}$$

Thompson (2008) presented the definition of the so called $\Gamma$-natural convected time derivative. This is an objective time derivative (if tensor $\mathbf{\Gamma}$ is objective) as seen by an observer attached on the basis of the eigenvectors of $\mathbf{\Gamma}$. As shown by Thompson (2008), for any symmetric tensor $\mathbf{\Gamma}$, the in-phase part its material time derivative with respect to itself, $\mathbf{\Phi}^{\mathbf{\Gamma}}_{\dot{\mathbf{\Gamma}}}$, is the natural time derivative of tensor $\mathbf{\Gamma}$, represented by $\mathbf{\Gamma}'$. The out-of-phase part, $\widetilde{\mathbf{\Phi}}^{\mathbf{\Gamma}}_{\dot{\mathbf{\Gamma}}}$ can be associated to the rate rotation of the eigenvectors of $\mathbf{\Gamma}$ (see Thompson (2008)) as

$$\widetilde{\mathbf{\Phi}}^{\mathbf{\Gamma}}_{\dot{\mathbf{\Gamma}}} = \mathbf{\Omega}^{\Gamma}\cdot\mathbf{\Gamma} - \mathbf{\Gamma}\cdot\mathbf{\Omega}^{\Gamma} \tag{24}$$

where $\mathbf{\Omega}^{\Gamma}$ is the skew-symmetric tensor that represents the rate of rotation of the eigenvectors of $\mathbf{\Gamma}$ given by

$$\mathbf{\Omega}^{\Gamma} = \dot{\mathbf{e}}^{\Gamma}_{i}\mathbf{e}^{\Gamma}_{i} \tag{25}$$

In other words, Eqs.(22) and (23) can be rewritten as

$$\mathbf{\Gamma}' = 2\xi\mathbf{\Phi}^{\mathbf{\Gamma}}_{D}\cdot\mathbf{\Gamma} + \mathbf{G}\left(\mathbf{\Gamma}\right) \tag{26}$$

$$\mathbf{\Omega}^{\Gamma}\cdot\mathbf{\Gamma} - \mathbf{\Gamma}\cdot\mathbf{\Omega}^{\Gamma} = \zeta\left(\mathbf{\Gamma}\cdot\mathbf{D} + \mathbf{D}\cdot\mathbf{\Gamma} - 2\mathbf{\Phi}^{\mathbf{\Gamma}}_{D}\cdot\mathbf{\Gamma}\right) + \mathbf{W}\cdot\mathbf{\Gamma} - \mathbf{\Gamma}\cdot\mathbf{W} \tag{27}$$

## 5. THE NEW APPROACH

### 5.1 The analysis of Fattal and Kupferman (2004)

It is worth pointing that the decomposition theorem presented by Fattal and Kupferman (2004) shares some similarities with the one presented by Thompson (2008). This we will explore next.

These authors considered the particular case of Eq.(4) or Eq.(21) of an affine deformation as represented by Eq.(5). Interestingly, before they apply the transformation of variable $\mathcal{T} : \mathbf{\Gamma} \rightarrow \log\mathbf{\Gamma}$, they rewrote Eq.(5) by decomposing the velocity gradient into three parts as

$$\nabla\mathbf{u} = \mathbf{\Omega} + \mathbf{B} + \mathbf{N}\cdot\mathbf{\Gamma}^{-1} \tag{28}$$

where $\mathbf{\Omega}$ and $\mathbf{N}$ are skew-symmetric tensors and $\mathbf{B}$ is symmetric and commutes with $\mathbf{\Gamma}$. When applied to Eq.(5), this decomposition leads to

$$\dot{\mathbf{\Gamma}} = 2\mathbf{B}\cdot\mathbf{\Gamma} + \mathbf{\Omega}\cdot\mathbf{\Gamma} - \mathbf{\Gamma}\cdot\mathbf{\Omega} + \mathbf{G}(\mathbf{\Gamma}) \tag{29}$$

Examination of Eqs.(22) and (23) together with Eq.(29) reveals that

$$\mathbf{B} = \mathbf{\Phi}_{\boldsymbol{D}}^{\mathbf{\Gamma}} \tag{30}$$

$$\mathbf{\Omega} = \mathbf{\Omega}^{\mathbf{\Gamma}} \tag{31}$$

In other words, the tensor $\mathbf{B}$ of the decomposition given by Eq.(28) is the in-phase part of $\mathbf{D}$ related to $\mathbf{\Gamma}$ and the tensor $\mathbf{\Omega}$ of the same decomposition is the rate-of-rotation of the eigenvectors of $\mathbf{\Gamma}$, for the case of affine deformation.

Even for non-affine deformation, Eq.(30) holds, but, as it will be shown, Eq.(31) has to be slightly modified.

Using the decomposition theorem presented by Fattal and Kupferman (2004) we can show the following corollary. The out-of-phase part of $\mathbf{\Gamma}\cdot\mathbf{D}+\mathbf{D}\cdot\mathbf{\Gamma}$ related to $\mathbf{\Gamma}$, represented by the term $\mathbf{\Gamma}\cdot\mathbf{D}+\mathbf{D}\cdot\mathbf{\Gamma}-2\mathbf{\Phi}_{\boldsymbol{D}}^{\mathbf{\Gamma}}\cdot\mathbf{\Gamma}$ can be written as $\mathbf{\Gamma}\cdot\mathbf{\Omega}_D^{\Gamma}-\mathbf{\Omega}_D^{\Gamma}\cdot\mathbf{\Gamma}$ where $\mathbf{\Omega}_D^{\Gamma}$ is skew-symmetric.

**Proof.** From Eq.(28), $\mathbf{D}$ can be written as

$$\mathbf{D} = \mathbf{\Phi}_{\boldsymbol{D}}^{\mathbf{\Gamma}} + \frac{1}{2}\left(\mathbf{N}\cdot\mathbf{\Gamma}^{-1} - \mathbf{\Gamma}^{-1}\cdot\mathbf{N}\right) \tag{32}$$

Therefore, the term $\mathbf{\Gamma}\cdot\mathbf{D}+\mathbf{D}\cdot\mathbf{\Gamma}-2\mathbf{\Phi}_{\boldsymbol{D}}^{\mathbf{\Gamma}}\cdot\mathbf{\Gamma}$ can be rewritten as

$$\begin{aligned}
&\mathbf{\Gamma}\cdot\mathbf{D}+\mathbf{D}\cdot\mathbf{\Gamma}-2\mathbf{\Phi}_{\boldsymbol{D}}^{\mathbf{\Gamma}}\cdot\mathbf{\Gamma} = \mathbf{\Gamma}\cdot\mathbf{\Phi}_{\boldsymbol{D}}^{\mathbf{\Gamma}} + \frac{1}{2}\left(\mathbf{\Gamma}\cdot\mathbf{N}\cdot\mathbf{\Gamma}^{-1} - \mathbf{\Gamma}\cdot\mathbf{\Gamma}^{-1}\cdot\mathbf{N}\right) + \\
&\mathbf{\Phi}_{\boldsymbol{D}}^{\mathbf{\Gamma}}\cdot\mathbf{\Gamma} + \frac{1}{2}\left(\mathbf{N}\cdot\mathbf{\Gamma}^{-1}\cdot\mathbf{\Gamma} - \mathbf{\Gamma}^{-1}\cdot\mathbf{N}\cdot\mathbf{\Gamma}\right) - 2\mathbf{\Phi}_{\boldsymbol{D}}^{\mathbf{\Gamma}}\cdot\mathbf{\Gamma} = \\
&\frac{1}{2}\left(\mathbf{\Gamma}\cdot\mathbf{N}\cdot\mathbf{\Gamma}^{-1} - \mathbf{\Gamma}^{-1}\cdot\mathbf{N}\cdot\mathbf{\Gamma}\right) = \\
&\mathbf{\Gamma}\cdot\left[\frac{1}{2}\left(\mathbf{N}\cdot\mathbf{\Gamma}^{-1} + \mathbf{\Gamma}^{-1}\cdot\mathbf{N}\right)\right] - \left[\frac{1}{2}\left(\mathbf{N}\cdot\mathbf{\Gamma}^{-1} + \mathbf{\Gamma}^{-1}\cdot\mathbf{N}\right)\right]\cdot\mathbf{\Gamma}
\end{aligned} \tag{33}$$

And, therefore

$$\mathbf{\Omega}_D^{\Gamma} = \frac{1}{2}\left(\mathbf{N}\cdot\mathbf{\Gamma}^{-1} + \mathbf{\Gamma}^{-1}\cdot\mathbf{N}\right) \tag{34}$$

Therefore Eq.(27) can be rewritten as

$$\mathbf{\Omega}^{\Gamma}\cdot\mathbf{\Gamma} - \mathbf{\Gamma}\cdot\mathbf{\Omega}^{\Gamma} = \left(\mathbf{W} - \zeta\mathbf{\Omega}_D^{\Gamma}\right)\cdot\mathbf{\Gamma} - \mathbf{\Gamma}\cdot\left(\mathbf{W} - \zeta\mathbf{\Omega}_D^{\Gamma}\right) \tag{35}$$

The equation above gives an interpretation for $\mathbf{\Omega}_D^{\Gamma}$. When the retraction is not complete, $\zeta \neq 0$,

$$\mathbf{\Omega}_D^{\Gamma} = \frac{1}{\zeta}\left(\mathbf{W} - \mathbf{\Omega}^{\Gamma}\right) \tag{36}$$

$$\mathbf{\Omega} = \mathbf{\Omega}^{\Gamma} - (1-\zeta)\mathbf{\Omega}_D^{\Gamma} = \mathbf{\Omega}^{\Gamma} - \frac{1-\zeta}{\zeta}\left(\mathbf{W} - \mathbf{\Omega}^{\Gamma}\right) \tag{37}$$

Therefore, $\mathbf{\Omega}_D^{\Gamma}$ is an objective spin tensor. For affine deformations, in the sense of rotation, $\mathbf{\Omega}_D^{\Gamma}$ is the vorticity relative to the rate of rotation of the eigenvectors of $\mathbf{\Gamma}$. Equation (37) shows how tensor $\mathbf{\Omega}$, from the decomposition of Fattal and Kupferman (2004), detaches from $\mathbf{\Omega}^{\Gamma}$ for non-affine deformations.

The equations for stretching evolution and rotation evolution are given, respectively, by

$$\mathbf{\Phi}_{\dot{\mathbf{\Gamma}}}^{\mathbf{\Gamma}} = 2\xi\mathbf{\Phi}_{\boldsymbol{D}}^{\mathbf{\Gamma}}\cdot\mathbf{\Gamma} + \mathbf{G}\left(\mathbf{\Gamma}\right) \tag{38}$$

$$\widetilde{\mathbf{\Phi}}_{\dot{\mathbf{\Gamma}}}^{\mathbf{\Gamma}} = \left[\zeta\mathbf{\Omega} + (1-\zeta)\mathbf{W}\right]\cdot\mathbf{\Gamma} - \mathbf{\Gamma}\cdot\left[\zeta\mathbf{\Omega} + (1-\zeta)\mathbf{W}\right] \tag{39}$$

This result states that for affine deformation in the sense of rotation, where $\zeta = 1$, Eq.() is recovered. On the other hand, for complete retraction, where $\zeta = 0$, the rotation of the eigenvectors of the conformation tensor is the vorticity.

Although Eqs.(38) and (39) are tensorial, they can be easily seen as vectorial equations. Equation (38) is an equation for the evolution of the eigenvalues of the conformation tensor while Eq.(39) is an equation for change in orientation of the conformation tensor a change of its eigenvectors.

### 5.2 Partially extended convected derivative

Larson (1984) considered $\zeta = 1$ to avoid spurious oscillations and came to a model of the following form

$$\overset{\circ}{\boldsymbol{\sigma}}_{PE} = \mathbf{G}(\boldsymbol{\Gamma})$$

where the partially extended convected derivative $\overset{\circ}{\boldsymbol{\sigma}}_{PE}$ is giving by

$$\overset{\circ}{\boldsymbol{\sigma}}_{PE} = \overset{\triangledown}{\boldsymbol{\sigma}} + \frac{2(1-\xi)}{\mathrm{tr}\boldsymbol{\sigma}}\mathbf{D} : \boldsymbol{\sigma}\boldsymbol{\sigma}$$

The partially extended convected derivative using the orthotropic closure approximation considered here and $\zeta = 1$ is

$$\overset{\circ}{\boldsymbol{\sigma}}_{PE2} = \overset{\triangledown}{\boldsymbol{\sigma}} + 2(1-\xi)\boldsymbol{\Phi}^{\sigma}_{D} \cdot \boldsymbol{\sigma}$$

In a step strain, Larson's approximation leads to

$$\overset{\circ}{\boldsymbol{\sigma}}_{PE} = \overset{\triangledown}{\boldsymbol{\sigma}} + \frac{2(1-\xi)}{\mathrm{tr}\boldsymbol{\sigma}}\mathbf{D} : \boldsymbol{\sigma}\boldsymbol{\sigma} = 0$$

$$\Rightarrow \mathrm{tr}\dot{\boldsymbol{\sigma}} = 2\xi\mathbf{D} : \boldsymbol{\sigma}$$

which is the same result of the present approximation as shown below

$$\overset{\circ}{\boldsymbol{\sigma}}_{PE2} = \overset{\triangledown}{\boldsymbol{\sigma}} + 2(1-\xi)\boldsymbol{\Phi}^{\boldsymbol{\Gamma}}_{\boldsymbol{D}} \cdot \boldsymbol{\sigma} = 0$$

$$\Rightarrow \mathrm{tr}\dot{\boldsymbol{\sigma}} = 2\mathbf{D} : \boldsymbol{\sigma} - 2(1-\xi)\boldsymbol{\Phi}^{\sigma}_{\mathrm{D}} : \boldsymbol{\sigma}$$

$$\Rightarrow \mathrm{tr}\dot{\boldsymbol{\sigma}} = 2\xi\mathbf{D} : \boldsymbol{\sigma}$$

## 6. FINAL REMARKS

The closure approximation for the fourth moment of the probability density function can be seen as a mapping from stretching and rotation of the microelement to stretching and rotation of the microstructure as represented by a filtered (average) quantity. A new orthotropic (in the sense discussed by Cintra and Tucker (1995)) closure approximation is proposed for partially extended convection models as proposed by Larson (1984). The new formulation does not have the 3D instabilities in 2D flows presented by Larson's model. Concerning the general thermodynamic framework presented by Pasquali and Scriven (2004) the backbone presented, given by

$$\dot{\boldsymbol{\Gamma}} = \frac{2\xi}{\mathrm{tr}\boldsymbol{\Gamma}}\mathbf{D} : \boldsymbol{\Gamma}\boldsymbol{\Gamma} + \zeta\left(\boldsymbol{\Gamma}\cdot\mathbf{D} + \mathbf{D}\cdot\boldsymbol{\Gamma} - \frac{2}{\mathrm{tr}\boldsymbol{\Gamma}}\mathbf{D} : \boldsymbol{\Gamma}\boldsymbol{\Gamma}\right) + \mathbf{W}\cdot\boldsymbol{\Gamma} - \boldsymbol{\Gamma}\cdot\mathbf{W} +$$

$$G(\boldsymbol{\Gamma}, e) + H(\mathbf{L}, \boldsymbol{\Gamma}, e)$$

is changed to

$$\dot{\boldsymbol{\Gamma}} = 2\xi\boldsymbol{\Phi}^{\boldsymbol{\Gamma}}_{\boldsymbol{D}} \cdot \boldsymbol{\Gamma} + \zeta\left(\boldsymbol{\Gamma}\cdot\mathbf{D} + \mathbf{D}\cdot\boldsymbol{\Gamma} - 2\boldsymbol{\Phi}^{\boldsymbol{\Gamma}}_{\boldsymbol{D}}\cdot\boldsymbol{\Gamma}\right) + \mathbf{W}\cdot\boldsymbol{\Gamma} - \boldsymbol{\Gamma}\cdot\mathbf{W}$$

$$G(\boldsymbol{\Gamma}, e) + H(\mathbf{L}, \boldsymbol{\Gamma}, e)$$

The new formulation, based on the maintenance of the eigenstructure when passing from the second moment to the fourth moment, keeps the compliance coefficients for stretching and rotation of the strand to the change in conformation. It is interesting to observe that another closure approximation needed to construct models usually called, by the literature of liquid crystals as the "nematic term", $\langle \mathbf{e}^R\mathbf{e}^R\mathbf{e}^R\mathbf{e}^R \rangle : \boldsymbol{\Gamma}$, with the present formulation would lead to the same result as the quadratic closure since $\boldsymbol{\Phi}^{\Gamma}_{\Gamma} = \boldsymbol{\Gamma}$. It is known that the quadratic closure give better results for the nematic term then the flow term $\langle \mathbf{e}^R\mathbf{e}^R\mathbf{e}^R\mathbf{e}^R \rangle : \boldsymbol{D}$. In fact, we can consider a maintenance of the eigenvector structure and change the eigenvalues by a factor. This factor would be a ratio between stretching the microstructure and stretching the microelement.

For general cases it would be interesting to combine this approximation with other orthotropic ones, but this time with a better interpretation for the mapping microelement-microstructure.

## 7. ACKNOWLEDGEMENTS

This research was funded by grants from PETROBRAS, ANP (Petroleum National Agency), and CNPq (the Brazilian Research Council).

## 8. REFERENCES

Cintra, J., Tucker, C., 1995. Microstructural evolution in polymer blends. J. Rheology 34, 177–210.

Fattal, R., Kupferman, R., 2004. Constitutive laws for matrix-logarithm of the conformation tensor. J. Non-Newt. Fluid Mech. 123, 281–285.

Larson, R. G., 1984. A constitutive equation for polymer melts based on partially extending strand convection. J. Rheology 28, 545–571.

Pasquali, M., Scriven, L. E., 2004. Theoretical modeling of microstructured liquids: a simple thermodynamic approach. J. Non-Newt. Fluid Mech. 120, 101–135.

Thompson, R. L., 2008. Some perspectives on the dynamic history of a material element. Int. J. Engng. Sci 46, 524–549.

## 9. Responsibility notice