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<td>Lim, Chun Ping; Han, J.; Lam, Yee Cheong</td>
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Chaos analysis of viscoelastic chaotic flows of polymeric fluids in a micro-channel
C. P. Lim, J. Han, and Y. C. Lam

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Chaos analysis of viscoelastic chaotic flows of polymeric fluids in a micro-channel

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Many fluids, including biological fluids such as mucus and blood, are viscoelastic. Through the introduction of chaotic flows in a micro-channel and the construction of maps of characteristic chaos parameters, differences in viscoelastic properties of these fluids can be measured. This is demonstrated by creating viscoelastic chaotic flows induced in an H-shaped micro-channel through the steady infusion of a polymeric fluid of polyethylene oxide (PEO) and another immiscible fluid (silicone oil). A protocol for chaos analysis was established and demonstrated for the analysis of the chaotic flows generated by two polymeric fluids of different molecular weight but with similar relaxation times. The flows were shown to be chaotic through the computation of their correlation dimension ($D_2$) and the largest Lyapunov exponent ($\lambda_1$), with $D_2$ being fractional and $\lambda_1$ being positive. Contour maps of $D_2$ and $\lambda_1$ of the respective fluids in the operating space, which is defined by the combination of polymeric fluids and silicone oil flow rates, were constructed to represent the characteristic of the chaotic flows generated. It was observed that, albeit being similar, the fluids have generally distinct characteristic maps with some similar trends. The differences in the $D_2$ and $\lambda_1$ maps are indicative of the difference in the molecular weight of the polymers in the fluids because the driving force of the viscoelastic chaotic flows is of molecular origin. This approach in constructing the characteristic maps of chaos parameters can be employed as a diagnostic tool for biological fluids and, more generally, chaotic signals.

Fluid flows in microfluidic channels are often laminar and not chaotic due to the low Reynolds' number (Re) for flows in micro-scale dimensions, i.e. negligible inertial forces. Nonetheless, non-inertia driven chaotic flows can be induced in micro-channels by other forces such as acoustic forces,\textsuperscript{1} interfacial tension\textsuperscript{2,3} and viscoelastic forces.\textsuperscript{4–6} With the addition of a trace amount of polymers, a fluid becomes viscoelastic, exhibiting both viscous and elastic characteristics. Chaotic flows of viscoelastic, polymeric fluids were demonstrated in a micro-channel by exploiting their non-linear behavior through the introduction of curved streamlines\textsuperscript{4} and sudden changes in geometry.\textsuperscript{6} Viscoelastic chaotic flows are deterministic but not easily predictable; this is the hallmark of chaos, in the sense that minute differences in initial conditions would lead to significantly different flow states. Characterization of these non-linear complex flows requires chaos analysis.

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Through chaos analysis, chaotic flows can be characterized by their chaos parameters such as the largest Lyapunov exponent ($\lambda_1$), the correlation dimension ($D_2$), the correlation entropy ($K_2$), etc.\textsuperscript{7} Chaos analysis was previously employed in flow regime identification of two phase flows in horizontal pipe,\textsuperscript{8} in T-junction\textsuperscript{9} and in bubble column.\textsuperscript{10,11} The onset of chaos in a mixing process\textsuperscript{12} and chaotic advection of particles\textsuperscript{13} could also be determined by such analysis. In this investigation, viscoelastic chaotic flows of polymeric fluids were generated in a micro-channel. The chaotic flows were characterized by $D_2$ (fractional $D_2$ indicates chaos) and $\lambda_1$ (positive $\lambda_1$ indicates chaos). $D_2$ represents the complexity of a chaotic flow while $\lambda_1$ its unpredictability. These two parameters can be described by maps that portray the differences in the chaos generated by systems with close similarity.

By their very nature, many fluids, including biological fluids such as mucus and blood, are viscoelastic. Conventional rheological measurements measure the bulk rheological properties of different fluids but are not adequate in capturing all their (non-linear) viscoelastic properties, often originating from molecular level differences. In this paper, we propose a novel characterization method of viscoelastic fluids by inducing chaotic flows in a micro-channel and the construction of characteristic chaos parameter maps. This method can potentially be employed as a diagnostic tool for biological fluids such as mucus and blood, potentially revealing subtle differences that are not clearly discernible by bulk rheological parameters (e.g. viscosity). In addition, this analysis can bring new paradigm to the analysis of chaotic signals such as electrocardiograms\textsuperscript{14} and signals from fetal heart rate monitoring techniques.\textsuperscript{15}

Viscoelastic chaotic flows of two polymeric fluids were generated in an H-shaped micro-channel (H-micro-channel) as shown in Fig. 1(a). It has a Center Channel (CC) with the width of 50\textmu m, and two main channels, \textit{i.e.} Main Channel 1 (MC 1) and Main Channel 2 (MC 2) with the

![Figure 1](image_url)

**FIG. 1.** (a) Viscoelastic chaotic flow generation by steady infusion of polymeric fluid and silicone oil. (b) Measurement of $s_{avg}$ as the chaotic flow signal.
width of 100μm. All channel depths are 50 μm. The junctions between the two main channels and CC will be abbreviated as MC1-CC and CC-MC2 respectively. The micro-channel was fabricated using NOA 81 (Norland Products Inc.) ultraviolet-curable adhesive based on the method developed by Hung et al. \(^\text{16}\) Briefly, a poly(dimethylsiloxane) (PDMS) mold of the micro-channel was casted from a SU-8 (MicroChem Corp.) negative mold fabricated by photolithography. \(^\text{17}\) NOA 81 micro-channel was casted from the PDMS mold and semi-cured by ultraviolet light. Subsequently, the micro-channel was sealed by a flat semi-cured NOA 81 (with inlet ports) that was backed by a block of PDMS. The assembly was fully cured under ultraviolet light and baked in a convection oven at 80 °C for 2 hours.

Viscoelastic chaotic flow was generated by infusing a viscoelastic polymeric fluid into MC 1 at a steady flow rate, \(Q_{\text{fluid}}\), and a Newtonian fluid (silicone oil) immiscible with the polymer solution into MC 2 at a steady flow rate \(Q_{\text{oil}}\). Fluids were infused by syringe pumps (Legato 210p from KD Scientific Inc.). \(Q_{\text{oil}}\) was adjusted to allow the polymer solution to displace the immiscible fluid in CC and to flow into MC 2 as shown in Fig. 1(a). \(Q_{\text{fluid}}\) was increased systematically until flow instability occurred, which manifested as the fluctuation of the interface between the two fluids that were infused at steady flow rates. We measured the average position of the interface, \(s_{\text{avg}}\), between the polymeric fluids and the immiscible fluid at the junction CC-MC2, as shown in Fig. 1(b). The measured \(s_{\text{avg}}\) were subjected to chaos analysis, in which \(D_2\) and \(\lambda_1\) were computed, for the identification of chaos and characterization of the chaotic flow.

Two polyethylene oxide (PEO) solutions that have similar elasticity (shear and extensional relaxation times) were employed. Fluid 1 (1.0 % wt/wt PEO of molecular weight \(M_w = 5 \times 10^6\) g/mol, namely PEO1) and Fluid 2 (0.65 % wt/wt PEO of molecular weight \(M_w = 8 \times 10^6\) g/mol, namely PEO2) were prepared to have a polymer concentration that is 15.33 times of their respective critical overlap concentrations (\(c^\ast\)). \(c^\ast\) is the concentration beyond which polymer molecules in a polymer solution begin to overlap. Thus, the polymer molecules in both Fluid 1 and Fluid 2 have the same degree of overlap. For ease of preparation, Fluid 1 was chosen to be 1.0 % wt/wt PEO which is equivalent to 15.33 times its \(c^\ast\). Hence, Fluid 2 was prepared to 0.65 % wt/wt PEO2 to arrive at 15.33 times its \(c^\ast\). This ensures that both fluids would exhibit non-linear viscoelastic properties. The shear rheological properties of the fluids were measured using Malvern Gemini HR-Nano rotational rheometer (Malvern Instruments Ltd.), with a 4 mm cone-plate (1° cone angle) measuring system. The Maxwell relaxation time, \(\lambda_M\), was computed using Equation (1). \(^\text{18}\)

\[
\lambda_M = \lim_{\omega \to 0} \frac{G'}{\omega^2 \eta_0}
\]

where \(G'\) is storage modulus, \(\omega\) angular frequency and \(\eta_0\), zero-shear viscosity.

The extensional relaxation time, \(\lambda_e\), was measured using Haake CaBER extensional rheometer (Thermo Scientific Inc.). Fluid 1 (\(\lambda_M = 1.97 \pm 0.18\) s, \(\lambda_e = 121.8 \pm 8.5\) ms) and Fluid 2 (\(\lambda_M = 2.14 \pm 0.11\) s, \(\lambda_e = 116.2 \pm 5.1\) ms) were determined to have similar relaxation times. Both Fluid 1 and Fluid 2 contain 1 mM disodium fluorescein for visualization. The immiscible fluid employed was silicone oil with a viscosity of 50 mPa·s (Sigma-Aldrich Company). Images of the interface position between the polymeric fluids and silicone oil were captured on an epi-fluorescence inverted microscope (Nikon Ti-eclipse) by a high speed camera (Photron Fastcam SA5) at 60 frames per second. \(s_{\text{avg}}\) was measured from the captured images through image processing using MATLAB.

Fig. 2(a) shows a typical \(s_{\text{avg}}\) time evolution signal obtained from the fluctuating interface between Fluid 1 and silicone oil at \(Q_{\text{fluid}} = 2.00\) ml/hr and \(Q_{\text{oil}} = 0.50\) ml/hr. Fig. 3(a) shows the \(s_{\text{avg}}\) signal for Fluid 2 at \(Q_{\text{fluid}} = 2.00\) ml/hr and \(Q_{\text{oil}} = 0.50\) ml/hr. Both signals show that the interface fluctuated strongly over the long term, even though the flow rates remained steady. The signals were measured after 240s of setting the required \(Q_{\text{fluid}}\) and \(Q_{\text{oil}}\) to ensure that the fluctuations are not due to the transient effect of changing the flow rates. The power spectral densities (PSD) of the signals, which were computed by Fast Fourier Transform (FFT), are plotted in Fig. 2(b) and Fig. 3(b) for Fluid 1 and Fluid 2 respectively. The PSD of both Fluid 1 and Fluid 2 have a monotonous low frequency region (\(< 1\) Hz) and decays at high frequency (\(> 1\) Hz) which is a characteristic of a chaotic flow. \(^\text{19}\) Nevertheless, FFT is a linear analysis; it alone is not adequate to quantify the
non-linear dynamics of the chaotic flows. Therefore, chaos analysis, which is a non-linear analysis, had to be employed for further analysis and quantification of the chaotic flows.

The signals obtained were further subjected to chaos analysis to compute $D_2$ and $\lambda_1$ through state space reconstruction by time-delay embedding. Let a signal measured at frequency $f_s$ be a time series \( \{ x_1, x_2, \ldots, x_N \} \). With a time delay of $\tau$, an $m$-dimensional ($m$ is the dimension of the state

FIG. 2. (a) $s_{avg}$ and (b) the corresponding PSD of Fluid 1 at $Q_{\text{fluid}} = 2.00$ ml/hr and $Q_{\text{oil}} = 0.50$ ml/hr.

FIG. 3. (a) $s_{avg}$ and (b) the corresponding PSD of Fluid 2 at $Q_{\text{fluid}} = 2.00$ ml/hr and $Q_{\text{oil}} = 0.50$ ml/hr.
FIG. 4. Protocol for consistent selection of \( m \) and \( \tau \).

space) state space \( \mathbf{X} = (\mathbf{X}_1 \mathbf{X}_2 \ldots \mathbf{X}_M)^T \) can be constructed by mapping the time series against time-lagged quantities of itself, where \( \mathbf{X}_i \) is the state in the reconstructed space at time step \( i \) with \( \mathbf{X}_i = (x_i \ x_{i+\tau} \ldots \ x_{i+(m-1)\tau}) \). Using \( \mathbf{X} \), \( D_2 \) was computed using the Grassberger-Procaccia algorithm\(^ {20,21} \) and \( \lambda_1 \) was computed using the algorithm proposed by Rosenstein \textit{et al.}\(^ {22} \). The computation of \( D_2 \) and \( \lambda_1 \) requires the selection of a suitable \( m \) and \( \tau \). Based on the false nearest neighbor (FNN) method\(^ {23} \) and the autocorrelation time \( \tau_c \),\(^ {24} \) a protocol (as shown in Fig. 4) was established to enable consistent selection of these parameters across different runs of chaos analysis. \( m \) was chosen to be the smallest \( m \) that gives the fraction of FNN to be less than 0.05. The minimum permissible \( \tau \) is chosen from the range of permissible \( \tau \) given by Equation (2):

\[
\frac{\tau_c}{m-1} \leq \tau \leq \tau_c
\]

The fluctuating flows of Fluid 1 and Fluid 2 at \( Q_{\text{fluid}} = 2.00 \text{ ml/hr} \) and \( Q_{\text{oil}} = 0.50 \text{ ml/hr} \) were determined to be chaotic with their \( D_2 \) being fractional and \( \lambda_1 \) being positive as summarized in Table I.

TABLE I. \( D_2 \) and \( \lambda_1 \) of viscoelastic chaotic flows at \( Q_{\text{fluid}} = 2.00 \text{ ml/hr} \) and \( Q_{\text{oil}} = 0.50 \text{ ml/hr} \) in H-micro-channel.

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<tr>
<th>Viscoelastic Fluid</th>
<th>Fluid 1</th>
<th>Fluid 2</th>
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</thead>
<tbody>
<tr>
<td>( m )</td>
<td>5</td>
<td>5</td>
</tr>
<tr>
<td>( \tau )</td>
<td>2</td>
<td>3</td>
</tr>
<tr>
<td>( D_2 )</td>
<td>4.33</td>
<td>5.5</td>
</tr>
<tr>
<td>( \lambda_1 )</td>
<td>6.25</td>
<td>5.16</td>
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FIG. 5. $D_2$ characteristic map in the operating space of (a) Fluid 1 and (b) Fluid 2.

The chaotic flows of Fluid 1 and Fluid 2 for other $Q_{\text{fluid}}$ and $Q_{\text{oil}}$ combinations in the operating space, which are limited to $0.50 \text{ml/hr} \leq Q_{\text{fluid}} \leq 4.00 \text{ml/hr}$ and $0.125 \text{ml/hr} \leq Q_{\text{oil}} \leq 2.00 \text{ml/hr}$, were analyzed similarly based on the protocol described in Fig. 4. Characteristic maps, which are two dimensional contour maps of $D_2$ and $\lambda_1$ plotted with $Q_{\text{fluid}}$ and $Q_{\text{oil}}$ being the axes, were constructed to characterize the chaotic flows generated by Fluid 1 and Fluid 2 in the operating space. The $D_2$ maps and $\lambda_1$ maps are plotted in Fig. 5 and Fig. 6 respectively. It was observed that the two fluids, which have similar relaxation times, have characteristic $D_2$ and $\lambda_1$ maps that bear similar trends but are generally distinct. From the $D_2$ maps, both Fluid 1 and Fluid 2 have peaks at high $Q_{\text{fluid}}$ of beyond 3.0 ml/hr; however, the $D_2$ peak of Fluid 2 occurs at low $Q_{\text{oil}}$ of 0.25 ml/hr compared to that of Fluid 1 which occur at $Q_{\text{oil}}$ of 0.50 ml/hr and 1.50 ml/hr. In the $\lambda_1$ maps, high $\lambda_1$ occurs at high $Q_{\text{fluid}}$ of beyond 2.5 ml/hr for both fluids; however, absent in Fluid 2, Fluid 1 exhibited a low peak at $Q_{\text{fluid}} = 1.5 \text{ ml/hr}$ and $Q_{\text{oil}} = 0.75 \text{ ml/hr}$. The bulk viscoelastic relaxation properties ($\lambda_M$ and $\lambda_v$) of Fluid 1 and Fluid 2 are similar. However, the driving force of the chaotic flows is of molecular origin and thus the differences in terms of the constituent molecules give rise to the distinct characteristic maps of the two fluids. This distinction offers a mean to differentiate between the two fluids in addition to characterizing the individual system. Furthermore, interface instability of viscoelastic fluid is the topic of relevance in many practical applications, such as mucus-air interface or biofilm stability; this analysis could potentially identify conditions for maximum instability and their correlation with molecular composition of biopolymers in each viscoelastic fluid.

In summary, chaotic flows of polymeric fluids were generated, at low Re, in a simple geometry with steady infusion of fluids driven by viscoelastic forces. The viscoelastic chaotic flows generated by two fluids, which are similar in terms of their relaxation times, were analyzed systematically with a protocol for consistent selection of $m$ and $\tau$ across different analyses. $D_2$ and $\lambda_1$ of the flows showed that they are indeed chaotic with $D_2$ being fractional and $\lambda_1$ being positive. In addition, the $D_2$ and $\lambda_1$ characteristic maps of the fluids were constructed, showing the differences in the
chaotic flows generated by two similar fluids over the operating space defined by $Q_{\text{fluid}}$ and $Q_{\text{oil}}$. Furthermore, the representation of the chaos parameters as characteristic maps could function as a tool to select the appropriate operating parameters to generate a desired chaotic state (instability), which is relevant in many situations involving viscoelastic fluids. This approach can be extended to the analysis of other chaotic signals such as electrocardiograms to identify abnormality through the characteristic chaos parameter maps.

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