Numerical Laplace inversion in rheological characterization

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Abstract

A new algorithm available for the numerical inversion of Laplace transforms using multi-precision computational environment provides controlled accuracy, that is the inversion can be carried out to yield any pre-specified number of significant digits. One important application is characterizing rheological phenomena, because the modeler is no longer restricted to a narrow class of “analytically treatable” constitutive equations, and the numerical inversion provides de facto “analytic” results for any reasonable model formulated in the Laplace domain. The advantages of Laplace domain models are discussed. In addition, idealized but important abstract flows (such as the Rayleigh or Stokes first problem) are calculated. As an illustration the fractional-power model of Low Density Polyethylene (LDPE) at 150 °C (for small shear strains) is considered and the arrival of a sinusoidal wave is calculated.

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1. Introduction

The goal of this paper is to draw attention to new algorithms for the numerical inversion of Laplace transforms using multi-precision computational environments. These algorithms provide controlled accuracy, that is the inversion can be carried out to yield any pre-specified number of significant digits. When applied to rheological phenomena, the modeler is no longer restricted to a narrow class of “analytically treatable” constitutive equations, because the numerical inversion provides de facto “analytic” results for any reasonable model formulated in the frequency (Laplace) domain. In addition, idealized but important abstract flows (such as the Rayleigh or Stokes first problem) can be calculated easily for real fluids.

The inversion algorithm with controlled accuracy has been described and extensively tested in our publication [1]. In this work we emphasize the application of the technique to real-fluid characterization,
involving multi-modal Maxwell, fractional and more complex rheological models. For illustrative purposes we use the well studied Low Density Polyethylene (LDPE) as a model fluid, see e.g., Tanner [2], Lodge and Meissner [3], Wagner and Laun [4], Debbaut et al. [5], Rubio and Wagner [6], Inkson et al. [7], Venerus et al. [8] and Chodenkar et al. [9]. As a by-product, we suggest a comprehensive description of the rheological behavior. Nevertheless, we are aware that the actual form of the model and the values of the parameters are subject to change and the final word (if there is one at all) will be said by the experimental investigators. Therefore, this paper is more concerned about the methodology of using numerical Laplace inversion than the actual form and parameter values of the constitutive equation.

2. Numerical Laplace transform inversion

There are over 100 algorithms available for the numerical inversion of Laplace transforms. Those that have passed the test of time fall into one of four categories. The four groupings of algorithms are according to the basic approach of the method as follows:

- Fourier-series expansion
- Laguerre-function expansion
- Bromwich contour deformation
- Combination of Gaver functionals

There have been about 40 algorithms developed which are based on the Fourier-series expansion of the time-domain function. For a discussion of these various methods see the survey paper of Abate and Whitt [10].

The next most popular approach is based on the Laguerre-function expansion of the original function. It seems to be the oldest approach to numerical inversion, and there have been about 15 such algorithms developed over the years, see the survey paper of Abate et al. [11].

One of the best approaches to numerical inversion is to deform the Bromwich contour of the inversion integral. The seminal paper on this approach is due to Talbot [12]. Another very good approach to numerical inversion is the method due to Gaver [13] and Stehfest [14], see Appendix.

There has been some use of numerical inversion algorithms to calculate transient responses for time-dependent problems related to viscoelasticity. Several authors have used algorithms based on the Fourier-series approach, see for example Warhola and Pipkin [15], Georgiadis and Rigatos [16], Gaul and Schanz [17] and Ezzat et al. [18]. In [17], the authors also recommend use of the Talbot method, but this seems to be the only paper to do so. Again, with respect to viscoelastic phenomena, a few authors have used the Gaver–Stehfest technique, see for example Chatterjee and Loring [19] and De Chant [20].

We would like to point out that there is a fundamental problem associated with almost all numerical inversion algorithms. Inherent in these procedures are a number of free parameters. Unless the user has some insight for setting these parameters, the algorithm is not reliable. In fact, the techniques presented for parameter settings involve intricate rules which are usually muddled. In this work we use the Gaver–Wynn–Rho algorithm which was studied in [1]. It is an improvement over the Gaver–Stehfest technique, see Appendix for details. It contains only one free parameter \( M \), the number of terms to be considered. That is, all other parameters are fixed internal to the algorithm. This is achieved by systematic multi-precision computing. In fact, the algorithm determines the required number of decimal digits for the inner calculation. As a result, our algorithm continues to provide more accuracy as \( M \) increases (for
Table 1
Numerical inversion of (6) at $t_1 = 1$ using the Gaver–Wynn–Rho algorithm

<table>
<thead>
<tr>
<th>$M$</th>
<th>Precision digits</th>
<th>$n/u_0$</th>
<th>Significant digits in result</th>
</tr>
</thead>
<tbody>
<tr>
<td>8</td>
<td>17</td>
<td>0.66849167...</td>
<td>8</td>
</tr>
<tr>
<td>16</td>
<td>34</td>
<td>0.66849167310784970...</td>
<td>16</td>
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<td>24</td>
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<tr>
<td>32</td>
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<tr>
<td>40</td>
<td>84</td>
<td>0.66849167310784970...</td>
<td>41</td>
</tr>
</tbody>
</table>

example see Table 1). It has long been recognized that this is only possible in a multi-precision computational environment. However, very little work has been done investigating numerical inversion using multi-precision computing.

A deficiency of all numerical inversion algorithms is the inability to handle transforms of non-differentiable functions. Fortunately, in many rheological applications we can avoid such problems, by a simple translation of the time-domain function. If the transform $\hat{f}(s)$ to be inverted has its inverse $f(t)$, such that $f(t) = 0$ in the interval $0 < t < a$ but $f(t)$ is smooth for $t > a$, then it is more efficient to consider the transform

$$\hat{f}^*(s) = \hat{f}(s) \exp(\lambda s),$$

(1)

for numerical inversion at the shifted time $t_1 = t - a$. That is, the operational formula (1) results in the shifting of the time-domain function.

To illustrate the above points, first we consider a simple problem of rheology that has a known analytic solution in the form of a definite integral. Tanner [21] obtained the solution to the Rayleigh problem for some simple viscoelastic constitutive equations. In this problem a semi-infinite body of fluid of density $\rho$ is in contact with a plane wall initially at rest. At time zero the wall starts to move in its own plane (in the $x$-direction) with constant velocity, $u_0$. If the rheological constitutive equation in Laplace space is given by

$$\hat{G}(s) = \frac{\eta_0}{1 + \lambda s},$$

(2)

where $\eta_0$ and $\lambda$ are parameters of the Maxwell model, then the velocity of the fluid at distance $y$ from the wall in time $t$ can be obtained by inverting the following Laplace transform:

$$\hat{u}/u_0 = \hat{f}(s) = \frac{1}{s} \exp\left[-\sqrt{\rho s/\hat{G}(s)}\right].$$

(3)

Combining (2) and (3), we have

$$\frac{\hat{u}}{u_0} = \hat{f}(s) = \frac{1}{s} \exp[-\sqrt{\rho/\eta_0}(1 + \lambda s)],$$

(4)

where $r = y \sqrt{\rho/(\eta_0 \lambda)}$. The analytical inverse of (4) was derived by Tanner [21]:

$$\frac{u}{u_0} = U(r^* - r) \left[ \exp\left(-\frac{r}{2}\right) + r \int_r^{r^*} \exp(\frac{-z}{2}) \sqrt{\frac{\rho}{\eta_0 \lambda}} \frac{1}{2} \left(1 + \frac{r^2 - z^2}{r^2}ight) dz \right],$$

(5)
where \( t^* = t/\lambda \), \( I_1 \) is the modified Bessel function of the first kind and of order one, and \( U \) the Heaviside step function.

We now consider the numerical inversion of (4). Our approach is to use the shifting rule (1), where in this case the time shift \( a = \lambda r \). Hence the transform to be inverted is given by

\[
\hat{f}(s) = \frac{1}{s} \exp\left[-r\sqrt{\lambda s(1 + \lambda s)}\right] \exp(r \lambda s). \tag{6}
\]

We can check the accuracy of our inversion by calculating (5). Selecting \( \rho = 1000 \text{ kg/m}^3 \), \( \eta_0 = 1000 \text{ Pa s} \), \( \lambda = 1 \text{ s} \) and \( y = 1 \text{ m} \) the inverse at \( t_1 = 1 \text{ s} \) can be calculated with the Gaver–Wynn–Rho algorithm as shown in Table 1.

Of course in most rheological calculations we do not need 41 significant digits. The real significance of Table 1 is that the number of accurate (or significant) digits is about equal to \( M \). This error estimate seems to work for a large class of transforms, see [1]. Also note, that to achieve this level of accuracy the computation must be performed with the number of decimal digits of precision equal to \( 2M \); for example, at \( M = 24 \), we used 50 precision digits.

In Table 1, we see that the accuracy of the result increases with \( M \). Therefore, we do not need the exact answer to determine the accuracy. The simple prescription is to run the procedure twice, once at \( M \) and then, e.g., at \( M + 10 \); the number of common digits determine the number of significant figures for the result.

### 3. Solving the Rayleigh and related shear flow problems for a real fluid

The shear relaxation modulus of LDPE is often described using the multi-modal Maxwell model. Recently, Chodenkar et al. [9] used 12 terms at temperature 150 °C

\[
\hat{G}(s) = \sum_{i=1}^{12} \frac{\eta_0 i}{1 + \lambda_i s}, \tag{7}
\]

where the coefficients are shown in Table 2.

<table>
<thead>
<tr>
<th>( i )</th>
<th>( \lambda_i ) (s)</th>
<th>( \eta_0 ) (Pa s)</th>
</tr>
</thead>
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<td>1</td>
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<td>220.942</td>
</tr>
<tr>
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<td>3,636.28</td>
</tr>
<tr>
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</tr>
<tr>
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<td>5.446</td>
<td>10,343.6</td>
</tr>
<tr>
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<td>16.86</td>
<td>13,923.7</td>
</tr>
<tr>
<td>10</td>
<td>52.18</td>
<td>15,676.4</td>
</tr>
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<td>11</td>
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</tr>
<tr>
<td>12</td>
<td>52.18</td>
<td>5,290.0</td>
</tr>
</tbody>
</table>
In this section, first we solve the Rayleigh problem for LPDE at at 150 °C where the density is \( \rho = 917 \text{ kg/m}^3 \) ([9]). We select the observation point at \( y = 1 \text{ m} \). Note that analytical solution is not available, so we must rely on numerical inversion. The time of arrival of the velocity wave can be obtained directly from the Laplace transform:

\[
\tau = \frac{y \sqrt{\rho}}{\lim_{s \to \infty} \sqrt{s \hat{G}(s)}},
\]

yielding \( \tau = 0.06898 \text{ s} \). The result of the numerical inversion of (3) with \( \hat{G}(s) \) given by (7) and shift \( a = \tau \) are shown in Fig. 1.

Though the Rayleigh problem has its own significance for boundary layer calculations, the arrival of the wave generated by a sinusoidal wall motion is of more practical interest. In this case, Eq. (3) changes to

\[
\frac{\hat{u}}{u_0} = \hat{f}(s) = \frac{1/\omega^2}{1 + s/\omega^2} \exp \left[ -\frac{y \sqrt{s \hat{G}(s)}}{\sqrt{\rho}} \right],
\]

where \( u_0 \) is now the maximum velocity of the wall and \( \omega \) the circular frequency. Again, we invert the shifted Laplace transform. As previously, the observations are made at \( y = 1 \text{ m} \). Results for 1 Hz (\( \omega = 2\pi \text{ Hz} \)) and 30 Hz are shown in Figs. 2 and 3. Comparing the two figures we see the well-known larger attenuation at the higher frequency, but also observe a large “overshoot”. This secondary overshoot does not contradict linear viscoelasticity, in fact at very large frequencies the shape of the arrival curve tends to the derivative curve of the solution of the Rayleigh problem.

So far we have used the Laplace inversion to solve simple flow problems with traditional fluid models. In the next section we take full advantage of the controlled accuracy inversion in the identification of the rheological model. At source, rheological characterization requires computerized inversion of experimental data and the problem of ill-posedness cannot be neglected.
At this point, we must make a careful distinction between numerical Laplace transform inversion and inversion of experimental data. In this work numerical Laplace transform inversion is used to solve the forward problem. Whether the forward model is available in an analytical form (such as in the case of multi-modal Maxwell model) or in the form of a Laplace transform that has to be numerically inverted to calculate model response (as will be the case in the next section), has little direct effect on the ill-posedness of the parameter identification process. What really matters is: how many parameters we need to incorporate into the model. A model may well successfully represent the data in the range of the independent variable being measured, but the asymptotics of the model outside that range might be very unrealistic. The more parameters we need to incorporate, the less relevant is the model for extrapolation.
As we will see, the real advantage of formulating the model in Laplace space (in contrast to using simple analytically solvable models, such as multi-modal Maxwell model) is that the number of parameters can be drastically reduced without losing the ability to represent the measured data accurately. Simply put, well selected Laplace space models with a few parameters are as flexible as the many parameter multi-modal Maxwell model. Laplace space models are basically generalized differential-integral equations and in general, they cover a much wider spectrum of behavior than models constructed with analytic solvability in mind. While this has been well known since Boltzman, so far the model forms practically applied have been very much limited by the unreliability of the numerical Laplace transform inversion. This difficulty has been finally overcome by multi-precision Laplace inversion. (Multi-precision Laplace inversion does not mean that we are interested in the 41st figure of a calculated response, rather we want a limited number of significant digits but we want to be sure that those figures are correct.)

4. Fractional-power rheological model

It is well known that the multi-modal Maxwell model has very limited extrapolation power. This is due to the large number of parameters and the associated non-uniqueness of the relaxation spectra. Fig. 4 shows the two components of the complex relaxation modulus calculated from 

\[ G'(\omega) = \text{Re}[i\omega \hat{G}(i\omega)] \]

and 

\[ G''(\omega) = \text{Im}[i\omega \hat{G}(i\omega)], \]

and also the dynamic viscosity function 

\[ \eta^* = \frac{1}{\omega} \sqrt{G'(\omega)^2 + G''(\omega)^2}. \]

The behavior at \( \omega > 100 \text{ rad/s} \) is obviously the consequence of the truncation of the spectrum. One may argue that in that region no experiments are available and hence the use of the model for large frequencies is not justified anyway. However, any time space use involves the use of the full spectrum and hence, if we use the model to predict flow behavior, the high frequency behavior can not be marginalized.

Many authors have suggested other dynamic shear modulus models. For instance, following the pioneering work of Smits and deVires [22] and Bagley and Torvik [23], fractional Maxwell models have gained popularity. Those models are often used in conjunction with analytical solutions for simple flows.
(mostly in the form of Mittag–Leffler functions), see for instance Enelund and Olsson [24], Mainardi and Goren [25], Hernández-Jiménez et al. [26] and Wenchanga et al. [27]. While the Mittag–Leffler functions are relatively easily calculated using Computer Algebra Systems, the investigator is again limited to models of simple forms that might not capture the complexity of the real behavior.

On the other hand, the numerical inversion of the Laplace (frequency) space model allows us to capture all the experimental details and yet to preserve some other desirable properties of the model, such as robustness and extrapolating power, all being related to the low number of parameters.

As an example we fit the following functional form of the Laplace transformed shear relaxation modulus to the data reconstructed from Table 2:

$$\hat{G}(s) = \eta_0 \left( 1 + (\lambda s)^m \right)^n.$$  

With \( n = 1 \) this model reduces to the fractional derivative Maxwell model studied in many recent publications. However, without allowing \( n \) to vary, we found the fit not satisfactory.

The least squares (in the logarithmic sense) fit of the functional form (10) was obtained by a standard search algorithm and yielded the parameter values \( \eta_0 = 74029.3 \) Pa s; \( \lambda = 24.9833 \) s; \( m = 0.572729 \) and \( n = 0.976889 \). We will call this the fractional-power model of LDPE (valid for small strain). The fit is shown in Fig. 5, where the points represent values calculated from the analytical inverse of Eq. (7). The four parameter fractional-power model reproduces the points as well as the 24 parameter multi-modal Maxwell model. Fig. 6 provides the same information for the fractional-power model as Fig. 4 does for the multi-modal Maxwell model. While the behavior in the middle range of frequencies is similar, the improved extrapolation power of the fractional-power model is obvious at large frequencies.

Other viscoelastic functions are easily generated from (10) since “within additive constants, each of the viscoelastic functions are mathematically equivalent to one other, . . . although the information they emphasize is different” [28]. For instance, the memory kernel shown in Fig. 7 can be calculated as the inverse of \(-s \hat{G}(s)\).
5. Arrival of a sinusoidal wave for the fractional-power model

As previously for the multi-modal Maxwell model, now we study the effect of starting a sinusoidal motion of the wall. The arrival of the sinusoidal wave with \( \omega = 2 \pi \) rad/s and \( \omega = 30 \times (2\pi) \) rad/s, as observed at \( y = 1 \) m, is shown on Figs. 2 and 3, respectively. Calculating the arrival time from Eq. (8) now yields \( \tau = 0 \) s. Contrary to the multi-modal Maxwell fluid, but similarly to the Newtonian fluid, the wall motion generates an instantaneous disturbance at \( y = 1 \) m. In other words, the differential equation is of parabolic type, similar to the one describing the dissipation of momentum in a Newtonian fluid. Nevertheless, there is an arrival time in the practical sense and it is basically the same as for the
multi-modal Maxwell model. At low frequency, the arrival of the sinusoidal wave is virtually the same for both models. A notable difference can be observed, however, if we compare the high frequency cases: the fractional-power model predicts more intense attenuation—obviously a result of the different way of extrapolating to high frequencies.

6. Conclusions

Using the recently available technique of numerical Laplace inversion with controlled accuracy opens up new possibilities in rheological characterization of polymeric (and other) materials. Since the numerical inversion of a Laplace transform can be easily done (for instance with the multi-precision Gaver–Wynn–Rho algorithm), the model can be conveniently formulated in Laplace space without worrying about “solvability” and concentrating more on the actual rheological behavior. The Laplace-space models are basically generalized differential-integral equation models with a very low number of parameters but high flexibility. Because of the low number of parameters to identify, the actual model fitting becomes less ill-conditioned and hence more robust and reliable. As an illustration we introduced the fractional-power model of LDPE and determined the constants (for low shear strains) at 150°C. The fractional-power model has only four parameters (\(\eta_0 = 74029.3\) Pa s, \(\lambda = 24.9833\) s, \(m = 0.572729\), and \(n = 0.976889\)) and it predicts various shear flows with considerable success. Even though taking into account more experimental information may necessitate further improvement of the suggested model, our point was to show that artificial restrictions on the form of the constitutive equation can be avoided by using numerical Laplace inversion with controlled accuracy.

Appendix A

The Post-Widder formula states that if we introduce

\[
\phi_k(t) = \left(\frac{-1}{k!}\right)^k \left(\frac{k}{7}\right)^{k+1} \left(\frac{k}{7}\right) \hat{f}(\frac{k}{7}),
\]

then as \(k \to \infty\), \(\phi_k(t) \to f(t)\).

In 1966, Gaver [13] presented the discrete analog of (A.1), namely

\[
f_k(t) = \left(\frac{-1}{k!}\right) \left(\frac{2\alpha}{(k-1)!}\right) \hat{f}\left(\frac{ka}{7}\right),
\]

where \(\alpha = \ln(2)\) and \(\Delta f(n\alpha) = \hat{f}(n+1) - \hat{f}(n\alpha)\), that is the difference operator.

Unfortunately, the Gaver functionals provide a very poor approximation because \(|f(t) - f_k(t)| \sim c/k\) as \(k \to \infty\). For example, \(f_{100}(t)\) may yield an estimate to \(f(t)\) with only two or three digits of accuracy.

To achieve a good approximation, a convergence acceleration algorithm is required for the sequence \(f_k(t)\). For this problem a very powerful acceleration scheme called Saffier summation (see pp. 35–38 of Wimp [29]) can be used as first suggested by Stehfest [14]. For the sequence of Gaver functionals \(f_1(t), f_2(t), \ldots, f_M(t)\), we write

\[
f(t, M) = \sum_{k=1}^{M} W_k f_k(t),
\]

(A.3)
where the weights are given by
\[ W_k = (-1)^k \frac{k^M}{M!} \frac{1}{k} \]
Formulas (A.2) and (A.3) are the components of the Gaver–Stehfest method. The Salzer summation may be considered as a Richardson extrapolation process (see Wimp [29]). In fact this was the approach used by Stehfest [14].

In a recent work [1], we showed that a more effective acceleration scheme for the Gaver functionals is the Wynn rho algorithm. It is given by
\[
\rho(n)_{-1} = 0, \quad \rho(n)_0 = f_n(t), \quad n \geq 0 \\
\rho(n)_{k} = \rho(n+1)_{k} - \frac{k}{\rho(n+1)_{k-1} - \rho(n)_{k-1}}, \quad k \geq 1 \quad (A.4)
\]
see [29] (p. 168). The approximant is then obtained as \( f(t, 2M) = \rho(0)_{2M} \). It was found that the number of precision decimals in the internal calculations must be at least \( 2.1M \), where \( M \) is the largest index used in the Gaver functionals.

A Mathematica realization of the algorithm, called GWR is available from the Mathematica Information Center, http://library.wolfram.com/database/MathSource/4738/. In general, it is enough to call the function with three arguments: GWR[\( F, t, M \)], where \( F \) is the name of a function computing the Laplace transform, \( t \) the time and \( M \) the number of terms. (For experienced users a fourth parameter: precin is available to depart from the default precision (2.1M), but it is hardly needed. We calculated all results in this paper with the default.)

As an example we consider the solution of the Rayleigh problem for a Maxwell fluid. First we define the Laplace transform function:
\[ \eta_0 = 1000; \lambda = 1; \rho = 1000; \]
\[ G[s_] = \frac{\eta_0}{1 + \lambda s} \]
\[ F[s_] = \frac{1}{s} \cdot \text{Exp}\left[-y \cdot \text{Sqrt}\left[\frac{\rho s}{G[s]}\right]\right] \]

The function should be prepared for arbitrary precision evaluation, that is it can not contain approximate numbers with decimal point. Instead, rational fractions (such as 996889/1000000) should be used.

The next step is to call the GWR routine and print the result:
\[ t = 2; \quad M = 256; \quad \text{uratio} = \text{GWR}[F, t, M]; \quad \text{Print}[M, \text{uratio}]; \]
To obtain a 17-digit accurate result we need \( M = 256 \) requiring considerable computational time (about a minute on a current PC). On the other hand, \( M = 32 \) gives already the accurate answer, if we use translation in time:
\[ r = y \cdot \text{Sqrt}[\frac{\rho}{\lambda \eta_0}] \]
\[ F1[s_] = F[s] \cdot \text{Exp}[\lambda r s]; \quad t1 = 1; \quad M = 32; \quad \text{uratiol} = \text{GWR}[F1, t1, M]; \quad \text{Print}[M, \text{uratiol}]; \]
and the computation is done in less than a second.
References


