NUMERICAL ANALYSIS IN NONLINEAR VISCOELASTICITY

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Abstract—The well-known Volterra integral equation in linear viscoelasticity is treated to characterize the response of nonlinear viscoelastic materials. In this context, a procedure is presented which includes a differential approximation of the time-dependent kernel and an optimization procedure and leads to an optimum selection of certain parameters such that the deviation of the theoretical results from the experimental data is minimal. The formulations are then applied numerically to the case of natural cellulose fibres by using available experimental data on the relaxation of such fibres. The results of the investigation prove the close accuracy of the predictive ability of the model within the range of strain levels considered.

1. INTRODUCTION

From a phenomenological point of view, the concept of the relaxation function in linear viscoelasticity can be expressed in terms of Volterra[1] response equation as follows:

\[ \xi'(t) = E^e \left [ e'(t) - \int_0^t R'(t - \tau) e' (\tau) d\tau \right ] \]  

(1)

where \( \xi'(t) \), \( e'(t) \) are the stress and strain tensors respectively at time \( t \), \( E^e \) is the elastic tensor modulus and \( R'(t - \tau) \) is the relaxation function which is usually a positive decreasing function of time.

2. NONLINEAR RHEOLOGICAL RESPONSE

While equation (1) represents the linear viscoelastic response, the nonlinear response behaviour of real material is significant[2, 3]. Thus, for the formulation of the response in this case, the method first outlined by Distefano and Todeschini[4] is applied such that the Volterra equation read now for the uniaxial case as follows:

\[ \xi(t) = g(\epsilon(t), k_1, k_2, \ldots) + \int_0^t h(\epsilon(\tau), b_1 b_2, \ldots) R(t - \tau) d\tau \]  

(2)

in which \( \xi(t) \), \( \epsilon(t) \) are the scalar components of stress and strain respectively at time \( t \) in the sample subjected to uniaxial loading. In this equation, the function \( g(.) \) corresponds to the elastic response in a parametric form, whereas the function \( h(.) \) accounts for the nonlinear hereditary effects and is also given in a parametric form. In these two functions, unknown constants \( k_1, k_2, \ldots \) and \( b_1, b_2, \ldots \) are respectively involved. The choice of the functions \( g(.) \) and \( h(.) \) is guided by a qualitative knowledge of the viscoelastic behaviour of the material. The function \( R(.) \) is the relaxation function for the uniaxial case. In a strict sense, this function is assumed to satisfy an \( N \)th order differential equation with constant coefficients of the following form:

\[ a_0 R + a_1 R^{(1)} + a_2 R^{(2)} + \cdots + a_{N-1} R^{(N-1)} + R^{(N)} = 0 \]  

(3)

in which \( a_0, a_1, \ldots, a_{N-1} \) are unknown coefficients.

In order to simplify the analysis, one can introduce for the instantaneous response a single
elastic constant, \( E \), only. Let, also, the constant strain being applied to a specimen during a relaxation experiment be denoted by \( \varepsilon_i \), where \( i = 1, 2, \ldots, n \) (\( n \) corresponding to the number of experiments). Then, equation (2) can be rewritten in a reduced form for a fixed time \( t \) within the range of the whole relaxation experiment in the following manner:

\[
\dot{\varepsilon}_i(t) = E\dot{\varepsilon}_i + h(\varepsilon_i, b_1, b_2, \ldots) \int_0^t R(\tau) \, d\tau
\]  

(4)

where the oversign \( \dot{\varepsilon} \) is used to indicate the theoretical stress.

In order, however, to find the unknown constants \( b_1, b_2, \ldots \) and to solve for the relaxation kernel \( R(\tau) \), it is convenient to adopt a minimization procedure as outlined below. In this context, the adopted procedure will be presented whilst an actual numerical evaluation for the nonlinear rheological response of natural cellulose fibres is presented in Section 4.

3. APPROXIMATE SOLUTION OF THE RELAXATION KERNEL

Whilst equation (4) has been shown in a general form for the relaxation of a nonlinear viscoelastic material, a solution of the kernel \( R(\tau) \) can only be given in an approximate form. For this purpose equation (5), below, will be used which is based on experimental observations of the relaxation behaviour of the material. Thus, denoting the experimental stress by \( \varepsilon_i \), one can express analytically the experimental relaxation curves as a function of the applied stress and time for a number of experiments \( i = 1, 2, \ldots, n \) in the following manner:

\[
\varepsilon_i(t) = \varepsilon_i + G_i \theta_i(t, m_{i1}, m_{i2}, \ldots)
\]  

(5)

in which \( \varepsilon_i = E\dot{\varepsilon}_i \) is the applied stress at \( t = 0 \), \( G_i \) are constants and \( \theta_i(t, m_{i1}, m_{i2}, \ldots) \) are functions of time. The shape of the experimental relaxation curve suggests the form of the function \( \theta_i(.) \) for the type of material under consideration. In equation (5) the constants \( G_i \) and \( m_{i1}, m_{i2}, \ldots \) are required to be determined for each experiment \( i \) by a standard fitting procedure.

In order to use the experimentally available data of the relaxation of the material by means of equation (5), it is evident that the coefficients in (3) have to be found by an optimization procedure such that the kernel \( \int_0^t R(\tau) \, d\tau \) in equation (4) is approximated by functions \( \theta_i(t, m_{i1}, m_{i2}, \ldots) \) of equation (5). Thus, using this approximation, it may be convenient to identify:

\[
R(\tau) = \frac{d}{dt} \theta_i(t, m_{i1}, m_{i2}, \ldots) = \Gamma_i \int_0^T
\]  

(6)

where the time \( T \) represents the total time for each relaxation experiment.

By substituting the values of \( \Gamma_i \) corresponding to equation (6) into (3) and using the method of differential approximation[5], we require that the functional:

\[
\sum_{j=1}^{N} \left\{ a_0 \Gamma_i^{(1)} + a_1 \Gamma_i^{(2)} + \cdots + a_{N-1} \Gamma_i^{(N-1)} + \Gamma_i^{(N)} \right\}^2 \, dt
\]  

(7)

be a minimum with respect to all possible choices of the coefficients \( a_0, a_1, \ldots, a_{N-1} \). The limits of the integral in the above equation and the sequel are taken from 0 to \( T \).

The minimization of expression (7) with respect to the \( a_j, j = 0, 1, \ldots, N - 1 \) leads to a system of \( N\)-simultaneous linear algebraic equations which can be written in the following form:

\[
\sum_{j=1}^{N} \int \Gamma_i^{(j)} (a_0 \Gamma_i^{(1)} + a_1 \Gamma_i^{(2)} + \cdots + a_{N-1} \Gamma_i^{(N-1)}) \, dt = -\sum_{j=1}^{N} \int \Gamma_i^{(j)} \Gamma_i^{(N)} \, dt
\]  

(8)

where \( i = 1, 2, \ldots, n \), and \( j = 1, 2, \ldots, N - 1 \).

This system of \( N\)-simultaneous algebraic equations is to be solved for the coefficients \( a_j, j = 0, 1, \ldots, N - 1 \) of equation (3).
Having determined the coefficients of the linear differential equation (3), one solution of this equation may be assumed to be of the form \( R(\tau) = \exp(F\tau) \). In order that this solution satisfies the linear differential equation (3) identically, one must have the characteristic equation:

\[
a_0 + a_1 F + a_2 F^2 + \cdots + a_{N-1} F^{N-1} + F^N = 0. \tag{9}
\]

This expression yields \( N \)-roots \( F_I (I = 1, 2, \ldots, N) \) which correspond to the \( N \)-solutions of the original linear differential equation. Hence, the general solution can be written as a linear combination of the \( N \)-solutions of the form:

\[
R(\tau) = \sum_{I=1}^{N} D_I \exp(F_I \tau) \tag{10}
\]

where \( D_I (I = 1, 2, \ldots, N) \) are constants to be determined.

Now, by combining equations (4) and (10) it follows that:

\[
\dot{\xi}_I(t) = E^* e_i + h(*e_n b_1, b_2, \ldots) \sum_{I=1}^{N} D_I (\exp(F_I \tau) - 1)
\]

\[
\left( D_I = \frac{D_I}{F_I}, \quad I = 1, 2, \ldots, N \text{ and } i = 1, 2, \ldots n \right). \tag{11}
\]

The identification problem of the nonlinear viscoelastic response of the material can now be formalized in the following manner:

Given \( n \) independent experimental functions \( *\xi_I (i = 1, 2, \ldots, n) \), equation (5), corresponding to applied strains \( *e_n \), we wish to find the constants \( b_1, b_2, \ldots \) and \( D_I (I = 1, 2, \ldots, N) \) in the constitutive equation (11) such that the functional:

\[
II(b_1, b_2, \ldots, D_I) = \sum_{I=1}^{N} \gamma_I \int (\dot{\xi}_I(t) - *\xi(t))^2 dt \tag{12}
\]

is minimized. For the purpose of minimizing this functional, a quadratic expression of the difference between the theoretical stress \( \dot{\xi}(t), (i = 1, 2, \ldots, n) \) which is given by equation (11) and the experimental stress \( *\xi(t) \) as given by equation (5) is used and where \( \gamma_I (i = 1, 2, \ldots, n) \) represent suitable positive weighting factors.

Once the values of the unknown constants of equation (11) have been determined by the above procedure, this equation may be used as a “model equation” for the rheological behaviour of nonlinear viscoelastic material.

4. NUMERICAL EVALUATION

The foregoing procedure is applied for the case of natural cellulose fibres. The relaxation data as derived from the experimental relaxation curves of these fibres are first fitted to a convenient form of the analytical expression (5). Secondly, numerical schemes are developed for the differential approximation of the relaxation kernel as discussed above and, hence, for the minimization of the objective function (12).

4.1 Fitting the experimental data of relaxation

The experimental curves, Meredith[2], showing the relaxation stress against the logarithm of time are represented in Fig. 1 for natural cellulose fibres (cotton) in tension.

Referring to expression (5) and the shape of the relaxation curves of Fig. 1, one may express the relaxation stress of natural cellulose fibres as a function of time in the following manner:

\[
*\xi_I(t) = *\xi_i - G_i \tau^m_i \quad (i = 1, 2, \ldots, 5) \tag{13}
\]

where \( *\xi_I \) is the instantaneous stress at time \( t = 0 \) and \( G_i \) and \( m_i \) are constants.

It is thus required to fit for each experiment the parameters \( G_i \) and \( m_i \) in equation (13) such
that the relaxation stress $\xi(t)$ gives a reasonable representation of the outcome of the relaxation experiment. The values of these constants, Haddad [3], are given in Table 1.

4.2 Differential approximation of the relaxation kernel

With the foregoing obtained information from the experimental relaxation data of natural cellulose by means of equation (13), one can proceed using the method of differential approximation indicated in Section 3 to determine the coefficients of the linear differential equation (3).

Comparing equations (5) and (13) it is evident that for the case of natural cellulose the function $\theta(t)$ takes the following form:

$$\theta(t) = -t^m_i.$$

Consequently, one can write in view of equations (6) and (14) that:

$$\Gamma_i = \frac{d}{dt}(-t^{m_i})$$

from which the $j$th derivative required for equation (8) can be written as:

$$\Gamma^{(j)} = \frac{d^{j+1}}{dt^{j+1}}(-t^{m_i}) = -m_i(m_i-1)(m_i-2) \ldots (m_i-j)t^{m_i-j-1} \quad (j = 0, 1, 2, \ldots, N-1).$$

A straightforward matrix inversion method has been used to solve the system of the simultaneous algebraic equations (8) for the coefficients $a_i(j = 0, 1, \ldots, N-1)$. However, it should be mentioned that with reference to equation (16) and in view of the values of $m_i (i = 1, 2, \ldots, 5)$ indicated in Table 1, the singularity of the terms of (8) prevents the use of the lower limit of the integration of this equation to be zero. This problem has been solved here by taking the lower limit of the integral 0.09 hr. The results of the computations are presented in Table 2 for $N = 3, 4, \ldots, 8$.

Having determined the coefficients $a_i(j = 0, 1, \ldots, N-1)$ it is possible to continue to find the roots of the characteristic equation (9). In this context, the Secant method has been employed and the numerical values of the roots $F_i (i = 1, 2, \ldots, N)$ of equation (9) are given in Table 3.

4.3 Minimization of the objective function (12)

We continue to determine the values of the unknown constants of the model equation (11)

![Fig. 1. Experimental relaxation curves of natural cellulose fibres (cotton), from Meredith[2].](image-url)
Numerical analysis in nonlinear viscoelasticity

Table 1. Material parameters characteristic equation (13)—cotton fibres

<table>
<thead>
<tr>
<th>i</th>
<th>(e_i)</th>
<th>(\zeta_i)</th>
<th>(G_i)</th>
<th>(m_i)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.01</td>
<td>4.5</td>
<td>1.91883</td>
<td>0.06405</td>
</tr>
<tr>
<td>2</td>
<td>0.02</td>
<td>9.7</td>
<td>4.28641</td>
<td>0.06351</td>
</tr>
<tr>
<td>3</td>
<td>0.03</td>
<td>13.6</td>
<td>5.68172</td>
<td>0.06612</td>
</tr>
<tr>
<td>4</td>
<td>0.04</td>
<td>17.9</td>
<td>6.90827</td>
<td>0.05839</td>
</tr>
<tr>
<td>5</td>
<td>0.05</td>
<td>22.05</td>
<td>6.90621</td>
<td>0.06527</td>
</tr>
</tbody>
</table>

Table 2. Coefficients of the linear differential equation (3)—cotton fibres

<table>
<thead>
<tr>
<th>(a_j)</th>
<th>(N)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>4</td>
</tr>
<tr>
<td>(a_0)</td>
<td>(0.426757 \times 10^3)</td>
</tr>
<tr>
<td>(a_1)</td>
<td>(0.441519 \times 10^3)</td>
</tr>
<tr>
<td>(a_2)</td>
<td>(0.496572 \times 10^2)</td>
</tr>
<tr>
<td>(a_3)</td>
<td>(0.891147 \times 10^2)</td>
</tr>
<tr>
<td>(a_4)</td>
<td>(0.140015 \times 10^3)</td>
</tr>
<tr>
<td>(a_5)</td>
<td>(0.202358 \times 10^3)</td>
</tr>
<tr>
<td>(a_6)</td>
<td>(0.276144 \times 10^3)</td>
</tr>
<tr>
<td>(a_7)</td>
<td>(0.361374 \times 10^3)</td>
</tr>
</tbody>
</table>

Table 3. Roots of the characteristic equation (9)—cotton fibres

<table>
<thead>
<tr>
<th>(F_i)</th>
<th>(1/\text{hr})</th>
<th>(N)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>4</td>
<td>5</td>
</tr>
<tr>
<td>(F_1)</td>
<td>(-1.099525)</td>
<td>(-0.740515)</td>
</tr>
<tr>
<td>(F_2)</td>
<td>(-10.089640)</td>
<td>(-6.308098)</td>
</tr>
<tr>
<td>(F_3)</td>
<td>(-38.468035)</td>
<td>(-22.076275)</td>
</tr>
<tr>
<td>(F_4)</td>
<td>(-59.350363)</td>
<td>(-38.149942)</td>
</tr>
<tr>
<td>(F_5)</td>
<td>(-81.550000)</td>
<td>(-55.507862)</td>
</tr>
<tr>
<td>(F_6)</td>
<td>(-104.633550)</td>
<td>(-74.250533)</td>
</tr>
<tr>
<td>(F_7)</td>
<td>(-128.372337)</td>
<td>(-94.051185)</td>
</tr>
<tr>
<td>(F_8)</td>
<td>(-152.616160)</td>
<td></td>
</tr>
</tbody>
</table>

where the function \(h(.)\) appearing in this equation is assumed to take, for the present case of natural cellulose, the following form:

\[
h(.) = \exp(be). \quad (17)
\]

Substituting for \(h(.)\) and \(\theta(.)\), as given by (17) and (14) respectively into (11) and (5), one can write the functional (12) in the following manner:

\[
II\{b, D_i (I = 1, 2, \ldots, N)\} = \sum_{i=1}^{N} \gamma_i \int \left[G_t m_i - (\exp(b*e_i)) \sum_{j=1}^{N} D_t (\exp(F_i) - 1) \right]^2 dt. \quad (18)
\]
The computational analysis for minimizing the objective function (18) has been carried out by using the steepest descent method whereby the lower limit of the integral of this equation has been taken as 0.09 hr as previously mentioned. The values of the parameters $b$ and $D_i (i = 1, 2, \ldots, N)$ which minimize equation (18) are shown in Table 4 for $N = 3, 4, \ldots, 8$.

4.4 Testing the predictive ability of the model

To test the predictive ability of the model, the predicted values of stress have been computed for each experiment $i$, at different values of $t$ between 0.09 and 2 hr and for $N = 3, 4, \ldots, 8$. This has been carried out by using equation (11) whereby the function $h(.)$ appearing in this equation is given by equation (17). The predicted values of stress have been then compared with the experimental ones of the same time. The mean square error, $[\text{M.S.E.}]$, has been computed for each experiment at each value of $N$ through the relation:

$$[\text{M.S.E.}] = \frac{\sum_X \left( \hat{\xi}(t_i) - \xi(t_i) \right)^2}{X}$$

($i = 1, 2, \ldots, X, t_{X}$ within the domain of $T$).

Table 5 shows the mean square error for each experiment $i(i = 1, 2, \ldots, 5)$ as associated with an order of optimization $N$. The total mean square error reaches a minimum value of $N = 4$. The predictive ability of the model is also demonstrated in Fig. 2 for strain levels 0.01 and 0.02.

### Table 4. Values of the parameters characteristic equation (11)—cotton fibres

<table>
<thead>
<tr>
<th>$D_i$</th>
<th>$N$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>3</td>
</tr>
<tr>
<td>$D_1$</td>
<td>64.848773</td>
</tr>
<tr>
<td>$D_2$</td>
<td>61.622924</td>
</tr>
<tr>
<td>$D_3$</td>
<td>85.071193</td>
</tr>
<tr>
<td>$D_4$</td>
<td>34.74769</td>
</tr>
<tr>
<td>$D_5$</td>
<td>17.350358</td>
</tr>
<tr>
<td>$D_6$</td>
<td>20.684165</td>
</tr>
<tr>
<td>$D_7$</td>
<td>24.108952</td>
</tr>
<tr>
<td>$D_8$</td>
<td>27.570524</td>
</tr>
</tbody>
</table>

### Table 5. Mean square error

<table>
<thead>
<tr>
<th>$i$</th>
<th>$\epsilon_1$</th>
<th>$N$</th>
<th>Mean Square Error, [$10^8$ dyn/cm$^2$]$^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>3</td>
<td>4</td>
</tr>
<tr>
<td>1</td>
<td>0.01</td>
<td>0.088020</td>
<td>0.026856</td>
</tr>
<tr>
<td>2</td>
<td>0.02</td>
<td>0.506500</td>
<td>0.061094</td>
</tr>
<tr>
<td>3</td>
<td>0.03</td>
<td>3.302700</td>
<td>1.177800</td>
</tr>
<tr>
<td>4</td>
<td>0.04</td>
<td>5.690000</td>
<td>2.243800</td>
</tr>
<tr>
<td>5</td>
<td>0.05</td>
<td>3.598600</td>
<td>0.46755</td>
</tr>
<tr>
<td></td>
<td>Total mean square error</td>
<td>2.637164</td>
<td>0.79542</td>
</tr>
</tbody>
</table>
5. CONCLUDING REMARKS

The nonlinear viscoelastic response of real material has been considered in a generalized manner by using the hereditary relaxation relation due to Volterra. The analytical approach due to Distefano and Todeschini has been adopted. In this context, a procedure has been presented that leads to an optimum selection of certain parameters that characterize the relaxation response of the material so that the deviation of the theoretical results form the experimental data would be minimal. The theoretical model has been applied to the relaxation of natural cellulose fibres in tension where the experimental data are available due to Meredith. The results of the investigation prove the close accuracy of the predictive ability of the model within the range of strain levels considered, i.e. $*e = 0.01, 0.02, \ldots, 0.05$. In this range of strain, the total mean square error reaches a minimum value at $N = 4$ for the time range between 0.09 and 2 hr.

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REFERENCES