Complex plane analysis of fractional derivative model and its use for parameter determination of viscoelastic material

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Abstract. The mechanical properties of viscoelastic materials are usually described by a rheological model composed of spring and dashpot in series or in parallel or their combination. The complex modulus as a function of angular frequency was firstly deduced, when fractional derivative rheological model is subjected to a sinusoidal perturbation in dynamic mechanical analysis (DMA) measurements. Then, the algebraic equations between the storage and loss moduli of the fractional Zener model in complex plane have been developed. The curve on the complex plane, a plot of loss modulus against storage modulus, is a repressed or distorted semicircle with its center below the real axis. The parameters of mechanical elements can be graphically estimated from its complex plane plot. The dynamic mechanical test of bituminous mixtures was carried out by frequency sweep over a wide range of 200 Hz to 0.02 Hz and its mechanical response can be described adequately by the fractional Zener model, whose element parameters are determined via geometric method.

1. Introduction

It is a common practice that creep and stress relaxation tests are used to measure the mechanical properties of viscoelastic material in time domain. In order to describe and imitate the viscoelastic behavior, rheological models have been proposed by an arrangement of basic elastic elements (springs) and viscous elements (dashpots) in series or parallel or their combination [1, 2]. The parameters are usually acquired through the time-dependent experimental data by fitting with a previously given rheological model. For the sake of simplicity and rapidity, measurement in frequency domain is developed on the basis of Laplace or Fourier integral transform. An oscillatory test has become very easy to implement with the advent of modern electronics. A sinusoidal stress or strain in a wide range of frequency is applied to the viscoelastic material, and the resultant mechanical response is monitored by using the controlled-stress mode or controlled-strain mode. The ratio of response and excitation (modulus or compliance) can be expressed as a complex quantity, which is function of loading frequency rather than time.

This paper aims to present a graphical method for the determination of element parameters on the basis of complex plane analysis from dynamic mechanical analysis (DMA) measurements. The Zener model and its fractional derivative model are simple rheological models of being able to describe the mechanical behavior of viscoelastic solids [3, 4]. In dynamic mechanical experiments, the complex modulus functions of fractional Zener model were derived and the relations between the storage and
loss moduli were generated. A complex plane of different frequency points is plotted for determination of the parameters. The dynamic mechanical behavior of the typical viscoelastic material bituminous mixtures was investigated by a dynamic mechanical analyzer (DMA) through frequency sweep in a wide frequency range between 200 Hz and 0.02 Hz. These rheological output in complex plane format were examined to explore the element parameters.

2. Theoretical background

2.1. Dynamic mechanical analysis (DMA)

For the sake of investigation of the viscoelastic characteristics of material, one may put a small sinusoidal signal excitation on it, and measure its response. In these oscillatory tests, samples are subjected to a periodically varying stress or strain. If the perturbation is strain, the corresponding linear response, stress, oscillates at the same angular frequency as the applied strain, but leads the strain by a phase shift [1, 2]. They can be given in complex notation by

\[ \varepsilon^* = \varepsilon_0 \exp(i\omega t) \quad (1) \]
\[ \sigma^* = \sigma_0 \exp[i(\omega t + \delta)] \quad (2) \]

where \( \varepsilon_0 \) and \( \sigma_0 \) are the strain amplitude and stress amplitude, respectively, \( \delta \) the phase shift, \( t \) the time in second (s), \( \omega \) the angular frequency in radians per second (rad./sec), \( \omega = 2\pi f \), \( f \) is the frequency in cycles per second (Hz), and \( i \) is the imaginary unit \( (i^2 = 1) \).

The ratio of the stress to the strain is the complex modulus, which can be expressed as

\[ E^* = \sigma^*/\varepsilon^* = (\sigma_0/\varepsilon_0)(\cos\delta + i\sin\delta) = E' + iE'' \quad (3) \]
\[ E' = (\sigma_0/\varepsilon_0)\cos\delta \quad (4) \]
\[ E'' = (\sigma_0/\varepsilon_0)\sin\delta \quad (5) \]
\[ \delta = \tan^{-1}(E''/E') \quad (6) \]

where \( E' \) and \( E'' \) are the storage and loss moduli, the real and imaginary parts of \( E^* \), respectively.

2.2. The response of fractional Zener model

The Zener model is known as a simple viscoelastic solid model, which is a parallel arrangement of a spring with an elastic modulus \( E_p \) and a Maxwell element (a spring of elastic modulus \( E \) in series with a dashpot of viscosity \( \eta \)), as shown in Figure 1.

![Figure 1. Zener model.](image)

Its differential constitutive equation is given by

\[ E\sigma(t) + \eta \frac{d\sigma(t)}{dt} = EE_p\varepsilon(t) + \eta(E + E_p) \frac{d\varepsilon(t)}{dt} \quad (7) \]

In the Maxwell unit, the ratio of a coefficient of viscosity \( \eta \) of a dashpot to an elastic modulus \( E \) of a spring is of time dimension and called relaxation time \( \tau \) which describe stress relaxation.

\[ \tau = \eta/E \quad (8) \]

By introduction of the term of relaxation time, equation (7) can be rewritten as

\[ \sigma(t) + \tau \frac{d\sigma(t)}{dt} = E_p\varepsilon(t) + (E + E_p)\tau \frac{d\varepsilon(t)}{dt} \quad (9) \]

It is known that the ideal Zener model may be too simplified to correlate with the dynamic behavior of
practical cases. The inadequacy of the Zener model in the frequency domain is that the variations of theoretical dynamic mechanical responses with frequency are always larger than those of the experimental observations [5]. The behavior of this simple model can be improved by introducing the so-called fractional derivatives in the differential constitutive equation in which the customary time derivatives of integer order are replaced by derivatives of fractional order [3-5]. Therefore, the differential constitutive equation of the fractional Zener model may be given by [4, 5]

$$\sigma(t) + \tau^a \frac{d^a \sigma(t)}{dt^a} = E_p \sigma(t) + (E + E_p) \tau^a \frac{d^a \tau(t)}{dt^a}$$  \hspace{1cm} (10)

The symbol $\alpha$ denotes the order of the differential equation and takes on a value between 0 and 1. If the order $\alpha$ equals 1, equations (9) and (10) become identical. The $\alpha$th order time derivative of a function, say $\varphi(t)$ is defined as [6]

$$\frac{d^\alpha \varphi(t)}{dt^\alpha} = \frac{1}{\Gamma(1-\alpha)} \frac{d}{dt} \int_0^t (t-u)^{\alpha-1} \varphi(u) \, du$$  \hspace{1cm} (11)

where $u$ is a dummy variable, and $\Gamma$ is the gamma function.

The fractional time derivative of equations (1) and (2) are given by

$$\frac{d^\alpha \varphi(t)}{dt^\alpha} = i(\omega)^\alpha \varphi(t)$$  \hspace{1cm} (12)

$$\frac{d^\alpha \sigma(t)}{dt^\alpha} = (i\omega)^\alpha \sigma(t)$$  \hspace{1cm} (13)

The complex modulus of the fractional Zener model can be derived by substitution of equations (12) and (13) into equation (10)

$$E^* = \frac{\sigma(t)}{\varphi(t)} = \frac{E_p + (E + E_p)(i\omega)^\alpha}{1 + (i\omega)^\alpha} = E_p + \frac{E(i\omega)^\alpha}{E + (i\omega)^\alpha}$$  \hspace{1cm} (14)

Since

$$i^\alpha = \cos(\alpha \pi/2) + i \sin(\alpha \pi/2)$$  \hspace{1cm} (15)

Equation (14) can be rearranged in the form

$$E^* = E_p + E \omega^\alpha \eta^\alpha \frac{\cos(\alpha \pi/2) + i \sin(\alpha \pi/2)}{[E^a + \omega^a \eta^a \cos(\alpha \pi/2)] + i \omega^\alpha \eta^\alpha \sin(\alpha \pi/2)}$$  \hspace{1cm} (16)

Separating the real and imaginary parts, complex modulus and its components can be expressed as

$$E^* = E_p + E \omega^\alpha \eta^\alpha \frac{[E^a \cos(\alpha \pi/2) + \omega^a \eta^a] + i E^a \sin(\alpha \pi/2)}{[E^a + \omega^a \eta^a \cos(\alpha \pi/2)]^2 + [\omega^a \eta^a \sin(\alpha \pi/2)]^2}$$  \hspace{1cm} (17)

or

$$E^* = E_p + E \omega^\alpha \eta^\alpha \frac{E^a \cos(\alpha \pi/2) + \omega^a \eta^a}{[E^a \cos(\alpha \pi/2) + \omega^a \eta^a]^2 + [E^a \sin(\alpha \pi/2)]^2}$$  \hspace{1cm} (18)

$$E' = E_p + E \omega^\alpha \eta^\alpha \frac{E^a \cos(\alpha \pi/2) + \omega^a \eta^a}{[E^a \cos(\alpha \pi/2) + \omega^a \eta^a]^2 + [E^a \sin(\alpha \pi/2)]^2}$$  \hspace{1cm} (19)

$$E'' = E \omega^\alpha \eta^\alpha \frac{E^a \sin(\alpha \pi/2)}{[E^a \cos(\alpha \pi/2) + \omega^a \eta^a]^2 + [E^a \sin(\alpha \pi/2)]^2}$$  \hspace{1cm} (20)

It can be found that

$$(E'-E_p)^2 + E''^2 = (E \omega^\alpha \eta^\alpha)^2 \frac{1}{[E^a \cos(\alpha \pi/2) + \omega^a \eta^a]^2 + [E^a \sin(\alpha \pi/2)]^2}$$  \hspace{1cm} (21)
Equation (19) can be expressed as
\[
E' - E_p = \frac{1}{E} \left[ (E' - E_p)^2 + E''^2 \right] + E\omega^2 \eta^2 + \frac{E\alpha^2}{[E\alpha^2 + \omega^2 \eta^2 + \frac{E\alpha^2}{2}]^2} \] (22)

Multiplying equation (20) by the trigonometric term \(\cot(\alpha\pi/2)\), equation (22) can be rewritten as
\[
E' - E_p = \frac{1}{E} \left[ (E' - E_p)^2 + E''^2 \right] + \cot(\alpha\pi/2) E'' \] (23)

\[
\left( E' - E_p - \frac{E}{2} \right)^2 + \left[ E'' + \frac{E}{2} \cot(\alpha\pi/2) \right] = \left( \frac{E}{2} \right)^2 \left[ 1 + \cot^2(\alpha\pi/2) \right] \] (24)

As the trigonometric relations
\[
1 + \cot^2 \beta = \frac{1}{\sin^2 \beta} \] (25)

Then equation (24) becomes
\[
\left( E' - E_p - \frac{E}{2} \right)^2 + \left[ E'' + \frac{E}{2} \cot(\alpha\pi/2) \right]^2 = \left( \frac{E}{2} \right)^2 \left[ \frac{1}{\sin(\alpha\pi/2)} \right]^2 \] (26)

For simplicity, we define
\[
\varphi = \pi/2 - \alpha\pi/2 \] (27)

Finally, for the fractional Zener model, the relations between \(E'\) and \(E''\) can be described of the form
\[
\left( E' - E_p - \frac{E}{2} \right)^2 + \left( E'' + \frac{E}{2} \tan \varphi \right)^2 = \left( \frac{E}{2} \right)^2 \left[ \frac{1}{\cos \varphi} \right]^2 \] (28)

This is the equation of a circle of radius \(\frac{E}{2\cos \varphi}\), and its center on the negative side of the imaginary axis. The loss modulus versus storage modulus over a sufficiently wide range of angular frequency can be plotted in a complex plane as illustrated in Figure 2 in which \(A (E_p, 0), B (E_p + E, 0), \) center \(C \left( E_p + \frac{E}{2}, -\frac{E}{2} \tan \varphi \right), \) \(D \left( E_p + \frac{E}{2}, 0 \right), \) \(E \left( E_p + E, -E \tan \varphi \right)\).

![Figure 2. Complex modulus plane plot for fractional Zener model.](image)

When \(\omega \to 0, E' \to E_p, \) and \(E'' \to 0.\) When \(\omega \to \infty, E' \to (E_p + E), \) and \(E'' \to 0.\) We can see that the storage modulus increases from \(E_p\) up to \((E_p + E).\) It is a right shift of \(E_p\) which is the elastic modulus of the parallel spring along the real axis from the origin. A semicircle is rotated clockwise at an angle of \(\varphi\) around the point \(A \left( E_p, 0 \right).\) The angle \(\varphi\) by which such a semicircle is rotated depends on the fractional order \(\alpha\) described in equation (27). It can also be regarded as a repressed semicircle, since the arc in the first quadrant is smaller than a full semicircle.
3. Determination of parameters
As for the non-ideal solid model, 4 parameters $E$, $E_p$, $\eta$, and $\alpha$ need to be determined and their solution procedure consists of the following steps.

(a) Determination of the spring constants.
It is shown that the spring parameters are obtained from storage modulus asymptotes at low and high frequencies. From Figure 2, the parameters $E_p$ and $E$ can be immediately obtained by the intersections $A$ and $B$.

(b) Determination of the fractional order $\alpha$.
The angle of deflection $\phi$ shown in Figure 2 is determined by the fractional order $\alpha$, described in equation (27). The value of $\phi$ may be precisely determined by the radius length $\left(\frac{E}{2 \cos \phi}\right)$ or the ordinate of the circle center $\left(-\frac{E}{2 \tan \phi}\right)$. Then, one may obtain an estimate of order $\alpha$ from the $\phi$ value using equation (29), the rewritten form of equation (27).

$$\alpha = 1 - 2\phi/\pi \quad (29)$$

According to the above analysis, the parameters, $E$, $E_p$, and $\alpha$ can be determined adequately from geometric characteristics of the complex plane plot.

(c) Approximate determination of the viscosity coefficient of the dashpot.
It may be approximately evaluated from the experimental modulus data collected at a variety of frequencies. Equation (20) can be also rearranged to be in the form of a quadratic equation of $\eta^a$.

$$\omega^{2a}E^a(\eta^a)^2 + \omega^a E^a [2E^* \cos(\alpha \pi/2) - E \sin(\alpha \pi/2)] \eta^a + E^{2a}E^* = 0 \quad (30)$$

Let

$$a = \omega^{2a}E^* \quad (31)$$
$$b = \omega^a E^a [2E^* \cos(\alpha \pi/2) - E \sin(\alpha \pi/2)] \quad (32)$$
$$c = E^{2a}E^* \quad (33)$$

Thus

$$a(\eta^a)^2 + b\eta^a + c = 0 \quad (34)$$

The solution for the quadratic equation is therefore

$$\eta^a = \frac{-b \pm \sqrt{b^2 - 4ac}}{2a} \quad (35)$$

For the determination of $\eta$, it is necessary to solve an algebraic equation. Since the loss modulus $E^*$ value at each experimental frequency can be obtained from the test, and the parameters $E$, $E_p$, and $\alpha$ have already been determined, equation (30) will be completely solved. For each angular frequency $\omega$, two roots $\eta_1$ and $\eta_2$ will be provided as shown in equation (35). Only if it is real and possesses practical physical meaning, the viscosity coefficient value can be chosen as an effective result. There are minute variations in different roots determined by different frequency in the light of equation (30), in that experimental data are not completely on the semicircular arc. The mean value for viscosity coefficient of each angular frequency is applied as the final approximate result.

4. Example

4.1. Experimental program

4.1.1. Specimen preparation. Neat asphalt 60/80 penetration grade, and limestone aggregates of 2.36mm as maximum size was employed for preparing asphalt mixtures. The asphalt mixtures were rolled in a mold of 300 mm×300 mm×50 mm with a wheel compactor. Thereafter, the sample was cut
to some small cuboid blocks, and then were cut to final prescribed dimensions, 60 mm in length, 15 mm in width and 5.5 mm in height approximately.

4.1.2. Dynamic mechanical analysis (DMA) test. The Dynamic Mechanical Analyzer Q 800, produced by TA Instruments, was employed in this work. The DMA Multi-Frequency-Strain mode was used, through varying the frequency from the high frequency (200 Hz) to the low frequency (0.02 Hz), covering 4 decades, with 12 points per decade. The three-point bending beam test in which a vertical deflection amplitude of 2 μm is applied to the specimen to remain in the linear viscoelastic (LEV) domain was selected. These tests were carried out on a constant temperature of 45°C.

4.1.3. Experimental results. The experimental data of storage and loss moduli at different frequency are shown in Figure 3. The detailed results can be found in Table A.1 in Appendix A.

The frequency dependence of the loss modulus versus storage modulus are plotted in the form of complex plane diagram (see Figure 4), as a locus of points, where each data point corresponds to the storage and loss moduli at a different measurement frequency shown in Figure 3. Fitting the experimental data with a circular equation, one obtains

\[(E' - 8930)^2 + (E'^* + 10730)^2 = 13930^2\]  (36)

with radius 13930, center (8930, −10730), and the intersection points (47,0) and (17813,0) on real axis, depicted in Figure 5. The experimental data shown in Figure 4 is found to be a part arc of a semicircle which is rotated at an angle of 50.4 degrees. It can be calculated by trigonometry.

4.2. Parameters determination
From the two intersections A and B, described in Figure 2 and Figure 5, one finds \(E_p=47\) MPa and
\[ E = 17766 \text{ MPa}. \]

According to the length of the radius, we have
\[ \left( -\frac{E}{2} \tan \phi \right) = -10730 \text{ MPa} \] (37)

in which, \( \tan \phi = 1.208 \), \( \phi = 0.879 \) in rad. Therefore, according to equation (29), \( \alpha = 0.440 \).

The viscosity coefficient is calculated by solving the equation (30). The viscosity coefficient for each experimental frequency is shown in Table A.1 in Appendix A. The values of \( \eta_1 \) for all the experimental frequencies are reasonable whereas those of \( \eta_2 \), unreasonable, so one takes the average value of the former as the final approximate result, which is 0.22 MPa\( \cdot \text{s}^{-1} \).

All the parameter values are listed in Table 1.

| \( E \) (MPa) | \( E_\rho \) (MPa) | \( \eta \) (MPa\( \cdot \text{s} \)) | \( \varphi \) (rad.) | \( \alpha \) |
|----------------|-----------------|----------------------|----------------------|--------|
| 17766          | 47              | 0.22                  | 0.879                | 50.4   |

5. Conclusions
An algebraic connection between storage and loss moduli of the dynamic mechanical response for the fractional Zener model is proposed. Analysis of experimental data, loss modulus as a function of storage modulus, which yield a skewed semicircular arc with its center below the real axis in the complex plane, can provide estimates of the element parameters. The spring constants can be easily obtained by the two intersections on the real axis. The order of fractional differential operator can be determined through the angle by which such a semicircle is rotated via trigonometry method.

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Appendix A.

Table A.1. Experimental results and viscosity coefficient results for different frequency.

| \( f \) (Hz) | \( E' \) (MPa) | \( E'' \) (MPa) | \( \eta_1 \) (MPa\( \cdot \text{s} \)) | \( \eta_2 \) (MPa\( \cdot \text{s} \)) | \( \varphi_1 \) (Hz) | \( \varphi_2 \) (Hz) | \( \eta_1 \) (MPa\( \cdot \text{s} \)) | \( \eta_2 \) (MPa\( \cdot \text{s} \)) |
|--------------|----------------|-----------------|----------------------|----------------------|-----------------|-----------------|----------------------|----------------------|
| 200          | 2286           | 1522            | 0.27                  | 745                  | 2               | 314             | 218                  | 0.19                  |
| 165          | 2158           | 1447            | 0.28                  | 1052                 | 1.7             | 293             | 201                  | 0.19                  | 14796052 |
| 136          | 1986           | 1344            | 0.27                  | 1588                 | 1.4             | 269             | 182                  | 0.18                  | 22804093 |
| 112          | 1838           | 1250            | 0.27                  | 2371                 | 1.1             | 249             | 164                  | 0.18                  | 36558330 |
| 92           | 1705           | 1164            | 0.27                  | 3530                 | 0.93            | 240             | 153                  | 0.18                  | 50938542 |
| 76.5         | 1580           | 1084            | 0.26                  | 5158                 | 0.77            | 220             | 138                  | 0.17                  | 78399336 |
| 63           | 1459           | 1023            | 0.27                  | 7342                 | 0.63            | 206             | 126                  | 0.17                  | 119102674 |
| 52           | 1328           | 937             | 0.26                  | 11246                | 0.52            | 198             | 118                  | 0.18                  | 165932898 |
| 43           | 1225           | 870             | 0.26                  | 16511                | 0.43            | 181             | 105                  | 0.16                  | 262386858 |
| 35.6         | 1119           | 805             | 0.26                  | 24465                | 0.36            | 175             | 99                   | 0.17                  | 358579664 |
| 29.4         | 1020           | 739             | 0.25                  | 36832                | 0.29            | 168             | 93                   | 0.18                  | 520711863 |
| 24.2         | 920            | 676             | 0.24                  | 56195                | 0.24            | 159             | 85                   | 0.18                  | 763446218 |
| 20           | 859            | 632             | 0.25                  | 80569                | 0.2             | 152             | 80                   | 0.19                  | 1073630514 |
| 16.5         | 765            | 562             | 0.23                  | 130472               | 0.17            | 145             | 75                   | 0.19                  | 1448878894 |
| 13.6         | 707            | 521             | 0.23                  | 191173               | 0.14            | 138             | 70                   | 0.20                  | 2061328002 |
| 11.2         | 654            | 483             | 0.23                  | 278565               | 0.11            | 130             | 64                   | 0.21                  | 3222370492 |
| 9.3          | 599            | 446             | 0.23                  | 407188               | 0.09            | 125             | 60                   | 0.22                  | 4579308127 |
| 7.7          | 545            | 404             | 0.22                  | 624198               | 0.08            | 121             | 57                   | 0.22                  | 5659637667 |
| 6.3          | 499            | 369             | 0.21                  | 953078               | 0.06            | 113             | 52                   | 0.23                  | 9698864053 |
| 5.2          | 465            | 341             | 0.21                  | 1392114              | 0.05            | 110             | 48                   | 0.24                  | 13419146828 |
| 4.3          | 418            | 303             | 0.19                  | 2223611              | 0.04            | 105             | 45                   | 0.25                  | 19813789520 |
| 3.6          | 395            | 283             | 0.20                  | 3122613              | 0.03            | 104             | 40                   | 0.26                  | 33647920330 |
| 2.9          | 366            | 259             | 0.20                  | 4781854              | 0.02            | 98              | 36                   | 0.30                  | 65568094411 |
| 2.4          | 336            | 235             | 0.19                  | 7257789              | 0.01            | 92              | 30                   | 0.30                  | 65568094411 |
References
[1] Ferry J D 1980 Viscoelasticity Properties of Polymers, 3rd ed. (New York: John Wiley & Sons, Inc.) pp 8-59
[2] Tschoegl N W 1989 The Phenomenological Theory of Linear Viscoelastic Behavior: An Introduction (Berlin: Springer-Verlag Berlin Heidelberg) pp 55-66
[3] Bagley R L and Torvik P J 1984 On the appearance of the fractional derivate in the behavior of real materials. J. Appl. Mech. 51 294-298
[4] Pritz T 1996 Analysis of four-parameter fractional derivate model of real solid materials. J. Sound Vib. 195 103-115
[5] Pritz T 1999 Verification of local Kramers-Kronig relations for complex modulus by means of fractional derivative model. J. Sound Vib. 228 1145-65
[6] Bagley R L and Torvik P J 1983 A theoretical basis for the application of fractional calculus to viscoelasticity. J. Rheol. 27 201-210