ON THE INTERCONVERSION BETWEEN VISCOELASTIC MATERIAL FUNCTIONS

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ABSTRACT

The problem of numerical conversion of different viscoelastic material functions is reviewed. A number of approximation formulae are given, together with bounds for their errors.

1. STATEMENT OF THE PROBLEM

It is well known[1,3] that the linear viscoelastic behaviour of materials obeying the superposition principle may be characterized by various material functions, for instance:

(*a*) *Creep compliance*, $J(t)$, defined as the strain as a function of time, t, produced by a unit step in stress at time zero;

(*b*) *Relaxation modulus*, $G(t)$, defined as the stress effected by a unit step in strain at time zero;

(*c*) *Storage compliance*, $J'(\omega)$, and *loss compliance*, $J''(\omega)$, defined as functions of angular frequency ω; these are the amplitudes of the in-phase component and the out-of-phase component of strain under conditions of steady state response to a harmonic stress of angular frequency ω and unit amplitude;

(*d*) *Storage modulus*, $G'(\omega)$, and *loss modulus*, $G''(\omega)$, defined as the amplitudes of the in-phase component and the out-of-phase component of stress under conditions of steady state response to a harmonic strain of angular frequency ω and unit amplitude;

(*e*) *Retardation spectrum*, $f(\tau)$, as a function of retardation time, τ; it is defined by the equation:

$$J(t) = J_0 + \int_0^\infty f(\tau)\{1 - \exp(-t/\tau)\}\,d\tau + t/\eta \qquad (1)$$

where J_0 is the limit of the creep compliance for $t \to 0$ and $1/\eta$ is the limit of the rate of the creep compliance for $t \to \infty$; $f(\tau)$ is assumed to be a non-negative function of retardation time. This assumption is supported by overwhelming experimental evidence; it manifests itself experimentally by the fact that the rate of creep is a completely monotonic function of time $[\dot{J}(t) \geqslant 0; \ddot{J}(t) \leqslant 0; \dddot{J}(t) \geqslant 0;$ etc.].

(*f*) *Relaxation spectrum*, $g(\tau)$, as a function of relaxation time, τ; it is defined by the equation:

$$G(t) = G_\infty + \int_0^\infty g(\tau) \exp(-t/\tau)\, d\tau \tag{2}$$

where G_∞ is the limit of the relaxation modulus for $t \to \infty$. If the retardation spectrum is a non-negative function of the retardation time, the relaxation spectrum will be a non-negative function of the relaxation time, and vice versa[2].

Now, one of the problems encountered frequently in the investigation of relaxation behaviour is that of converting these characteristic material functions from one into the other. Formally, these problems have been solved by the theory of linear viscoelastic behaviour. According to this theory, the interconversion of various characteristic functions may be performed by application of (linear) integral transformations, or their inversions. However, as has been shown elsewhere[3], the actual application of those integral transformations to experimental data gives rise to basic difficulties, and to tedious calculations besides.

Therefore, we have recently studied the following questions:—

Assume that one of the characteristic material functions defined above† has been measured; that a finite number of measuring points is available at discrete times (frequencies) extending over a finite range in the time axis (frequency axis). Neither the behaviour at very short times (high frequencies) nor that at very long times (low frequencies) will be known. Each measurement has a finite experimental error.

1. Is it, under these assumptions, still possible to perform the conversion into other material functions?
2. How much experimental information is really needed for this conversion?
3. What is the most simple numerical procedure for this purpose?
4. What are the truncation errors, i.e. the errors due to the fact that information is only available over a finite range in time (frequency) scale?
5. What are the approximation errors, i.e. the errors due to the fact that we use simple approximation formulae, instead of the integral transforms, even within the limited range where the behaviour has been measured?
6. How, during the conversion process, is the experimental error transmitted to the new function?

Answers to these questions have been obtained for some of the conversion problems. Although the investigations are still in progress, it seems worthwhile to report the results obtained so far.

2. CHOICE OF TIMES FOR MEASUREMENT OF TRANSIENT BEHAVIOUR, AND CHOICE OF FREQUENCIES FOR MEASUREMENT OF DYNAMIC BEHAVIOUR

In the early investigations of relaxation behaviour, the choice of measuring times or frequencies was made more or less by chance within the time or

† We exclude the spectra for the moment.

frequency region where information was wanted. With modern techniques of automation available, use of preselected sampling times for the creep or relaxation experiment and use of preselected sampling frequencies for the vibration experiment have gradually become the usual practice. The question arises, therefore, which times or frequencies should be selected?

It is well known that relaxation behaviour is best represented by using a logarithmic time scale for plotting results of stress relaxation or creep experiments, and by using a logarithmic frequency scale for plotting results of dynamic measurements. Therefore, in order to avoid lack of information on the one hand and an excess on the other, the sampling times (frequencies) should be chosen in such a manner that they cover the logarithmic time scale (frequency scale) as regularly as possible. Furthermore, considerable gain in convenience for performing all calculations will be obtained by choosing sampling times (frequencies) that are exactly equally spaced in logarithmic time (frequency) scale. We have chosen the ratio of succeeding times (frequencies) to correspond to a factor of two.

This binary type of sampling was selected with regard to the technique of creep measurement employed at our research institute[4]. Using a logarithmic clock[5], the digital registration unit of the creep apparatus is activated at the above mentioned logarithmic sequence of times: 2, 4, 8, 16 sec etc., after the start of the creep experiment. Therefore, the item of information needed for the conversion is just the one obtained by the digital creep technique.

The system of approximations may also be used, if one starts with measurements which are distributed in a different way. One should then first deduce the results corresponding with binary sampling times (or frequencies) by interpolation. Subsequently the entire system of approximative equations can be applied without any further interpolation.

3. DERIVATION OF APPROXIMATION FORMULAE AND ERROR LIMITS

The method that we used to derive all approximation formulae was that of Ninomiya and Ferry[6], who proposed the following formula for calculation of $J(t)$:

$$J(t) \sim A(t) = J'(1/t) + 0{\cdot}40J''(0{\cdot}40/t) - 0{\cdot}014J''(10/t) \tag{3}$$

To prove this formula, $J(t)$ is written as an integral transform by using equation 1. The integrand consists of the retardation spectrum times a function of $x = t/\tau$, which is called the intensity function of $J(t)$ and which is equal to:

$$\chi(x) = 1 - \exp(-x) \tag{4}$$

The intensity function $\chi(x)$ is approximated by a linear combination of the intensity functions of the expressions $J'(1/t)$, $J''(0{\cdot}40/t)$ and $J''(10/t)$, which are respectively:

$$\chi'(x) = x^2/(1 + x^2)$$

$$\chi''(x/0{\cdot}40) = (x/0{\cdot}40)/\{1 + (x/0{\cdot}40)^2\}$$

$$\chi''(x/10) = (x/10)/\{1 + (x/10)^2\}$$

in the following way:

$$\chi(x) \sim \chi'(x) + 0{\cdot}40\chi''(x/0{\cdot}40) - 0{\cdot}014\chi''(x/10) \tag{5}$$

Multiplication of this equation by $f(\tau)$ and integration with respect to τ from 0 to ∞ yields equation 3. If the problem of the approximation of the intensity function has been solved appropriately, it will be found that also the terms outside the integrals in equation 3 are automatically accounted for. In the resulting expressions for the approximation formulae, neither the instantaneous compliance, J_0, nor the viscosity, η, occur explicitly. This is important, as those quantities are not accessible within an experiment of finite duration.

This method of deriving approximations was supplemented[3] by a consideration of the error of the approximation, which is defined as the difference between the approximation and the exact value of the desired quantity. So, in the case described above:

$$E(t) = A(t) - J(t) \tag{6}$$

We write the error as an integral transform of the retardation spectrum and find for the intensity function:

$$\psi(x) = \chi'(x) + 0{\cdot}40\chi''(x/0{\cdot}40) - 0{\cdot}014\chi''(x/10) - \chi(x) \tag{7}$$

We compare the intensity function of the error, $\psi(x)$, with the intensity function of the quantity which is to be calculated, i.e. $\chi(x)$. The quotient, $\chi(x)/\chi(x)$, is bounded for all positive values of x between a small negative lower limit and a small positive upper limit:

$$-0{\cdot}007 \leqslant \psi(x)/\chi(x) \leqslant 0{\cdot}101 \tag{8}$$

This inequality yields the following bounds for the relative error of the approximation:

$$-0{\cdot}007 \leqslant E(t)/J(t) \leqslant 0{\cdot}101 \tag{9}$$

It is also useful to compare the intensity function of the error, $\psi(x)$, with the intensity function, $\chi''(x)$, of $J''(\omega)$. We thus find the inequality:

$$-0{\cdot}028 \leqslant \psi(x)/\chi''(x) \leqslant 0{\cdot}096 \tag{10}$$

which yields limits for the absolute error in terms of $J''(\omega)$ with $\omega = 1/t$:

$$-0{\cdot}028J''(\omega) \leqslant E(t) \leqslant 0{\cdot}096J''(\omega) \tag{11}$$

If we use the well-known inequality:

$$J''(\omega) = J'(\omega)(\tan\delta) \leqslant J(t)(\tan\delta) \tag{12}$$

we find limits for the relative error expressed as functions of the value of $\tan\delta$ at the angular frequency $\omega = 1/t$:

$$-0{\cdot}028(\tan\delta) \leqslant E(t)/J(t) \leqslant 0{\cdot}096(\tan\delta) \tag{13}$$

A number of useful approximations has been derived in this way; they are listed in *Tables 1, 2, 3* and *4*. In the last columns of those tables, limits for the relative errors are expressed in percentages.

4. DISCUSSION

Figure 1 gives an illustration of the interrelationships treated so far. The quantities that are experimentally accessible have been placed in large blocks; furthermore the relaxation spectrum and the retardation spectrum, which could be known from the prediction of a molecular theory, are shown. Arrows between accessible functions indicate the existence of numerical methods for

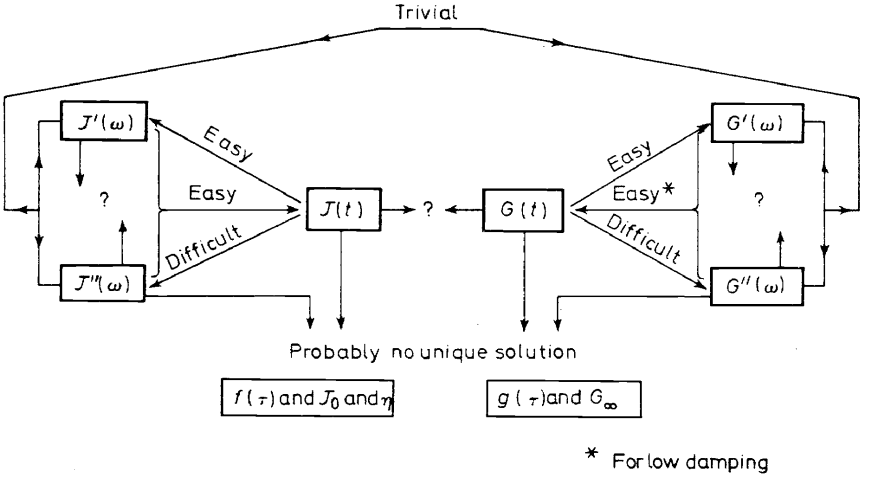

Figure 1. Experimentally accessible response functions, spectra, and numerical relationships between them.

conversion in the corresponding directions, which can be error bounded. The adjectives easy or difficult are used in connection with the amount of information needed for the conversion concerned. Arrows which are interrupted by interrogation marks indicate that the corresponding problem has not yet been treated in detail.

1. Calculation of storage compliance from the time dependence of the creep compliance

Approximation formulae for calculation of the storage compliance from the time dependence of the creep compliance have been treated elsewhere in full detail[7]. A list of these formulae, together with their error limits, is reproduced in *Table 1*. Whenever the damping, tan δ (or the double logarithmic slope of the creep compliance with respect to time), is small, a very simple formula will be sufficient:

$$J'(\omega) \sim J(t) - 0{\cdot}86\{J(2t) - J(t)\} \tag{14}$$

Bounds for the relative error of this equation are: $-15(\tan\delta)$ per cent and $+15(\tan\delta)$ per cent. Therefore, formula 14 will have a relative error smaller than 1 per cent in all cases where tan δ is smaller than 0·07.

Knowledge of two measuring points of the creep compliance suffices to

Table 1. Numerical formulae for calculation of storage compliance from creep compliance ($t = 1/\omega$)

$$J'(\omega) \sim J(t) - a[J(32t) - J(16t)] - b[J(16t) - J(8t)] - c[J(8t) - J(4t)] - d[J(4t) - J(2t)] - e[J(2t) - J(t)] - f[J(t) - J(t/2)] - g[J(t/2) - J(t/4)] - h[J(t/4) - J(t/8)]$$

a	b	c	d	e	f	g	h	*Limits for relative error*; %
				0·855				14·6 tan δ; – −14·6 tan δ; –
			0·445	—	0·376			7·8 tan δ; – 7·7 tan δ; –
		−0·0990	0·608	—	0·358			7·5 tan δ; 5·2 −7·5 tan δ; −9·1
		−0·119	0·680	—	0·225	—	0·0429	2·1 tan δ; 5·9 −2·1 tan δ; −20
	0·0108	−0·168	0·734	—	0·235			8·8 tan δ; 1·5 −1·9 tan δ; −1·5
	0·0109	−0·169	0·739	—	0·214	—	0·0451	2·3 tan δ; 1·6 −2·3 tan δ; −1·6
−0·000715	0·0185	−0·197	0·778	—	0·181	—	0·0494	3·1 tan δ; 0·8 −3·1 tan δ; −0·8

calculate one point of the storage compliance. If the creep compliance is known at n successive times of the binary series, formula 14 will allow calculation of $(n-1)$ values of the storage compliance at successive frequencies. These values may be used for further calculations within the same frame of approximations. This will demonstrate the advantage of the *a priori* choice of logarithmically equidistant measuring points.

For larger values of the damping, more involved formulae, also given in *Table 1*, should be used. It will always be possible to find formulae with a relative error smaller than 1 per cent, even at extremely high values of the damping. In the worst case, the last formula of *Table 1*, which is always within 1 per cent, would involve knowledge of the creep behaviour between $t/8$ and $32t$ for calculation of $J'(1/t)$.

To summarise, we can state that calculation of the storage compliance from creep is extremely simple for low values of the damping; it might be somewhat more involved for higher damping values, but the adjective easy is then still justified. No serious truncation problems will occur in connection with this conversion.

2. Calculation of loss compliance from the time dependence of the creep compliance

Approximation formulae for calculation of the loss compliance from the time dependence of the creep compliance were discussed in detail elsewhere[7] and are reproduced in *Table 2*. The calculation of the loss compliance from creep is much more difficult than the calculation of the storage compliance. Moreover, the error limits of the formulae of *Table 2* are decreasing functions of the damping. Consequently, the calculation of the loss compliance from creep is the more difficult, the lower the value of $(\tan \delta)$.

There is a formula that only involves knowledge of the creep compliance at two successive times, *viz.*:

$$J''(\omega) \sim 2{\cdot}12\{J(t) - J(t/2)\} \tag{15}$$

But this formula will be a very rough approximation. Even at high values of the damping, viz. for $(\tan \delta) = 1$, the error limits of this formula are 16 per cent and -16 per cent.

To reach higher accuracies, more complicated formulae have to be applied. If we want a formula with a relative error smaller than 3 per cent, the second formula of *Table 2* may be used for values of $(\tan \delta)$ between 1·5 and ∞; the third between 0·45 and ∞; the fifth between 0·2 and ∞ and the sixth between 0·075 and ∞. There is a formula which is within 3 per cent for all values of $(\tan \delta)$, namely:

$$\begin{aligned} J''(\omega) \sim & -0{\cdot}470\{J(4t) - J(2t)\} + 1{\cdot}674\{J(2t) - J(t)\} \\ & + 0{\cdot}198\{J(t) - J(t/2)\} + 0{\cdot}620\{J(t/2) - J(t/4)\} \\ & + 0{\cdot}012\{J(t/4) - J(t/8)\} + 0{\cdot}172\{J(t/8) - J(t/16)\} \\ & + 0{\cdot}043\{J(t/32) - J(t/64)\} + \cdots \end{aligned} \tag{16}$$

This formula is assumed to consist of an infinite number of terms. Each term following the one with coefficient 0·043 will be shifted a factor of 4 in time

Table 2. Numerical formulae for calculation of loss compliance from creep compliance: ($t = 1/\omega$)

$$J''(\omega) \sim d[J(4t) - J(2t)] + e[J(2t) - J(t)] + f[J(t) - J(t/2)] + g[J(t/2) - J(t/4)] + h[J(t/4) - J(t/8)] + j[J(t/8) - J(t/16)] + l[J(t/32) - J(t/64)] + n[J(t/128) - J(t/256)]$$

d	e	f	g	h	j	l	n	*Limits for relative error*; %
		2·12						$8[1 + 1/(\tan\delta)]$; 26 $-8[1 + 1/(\tan\delta)]$; $-26/(\tan\delta)$
−0·470	1·715	—	0·902					$0{\cdot}7[1 + 1/(\tan\delta)]$; 2·3 $-4{\cdot}6/(\tan\delta)$
−0·505	1·807	—	0·745	—	0·158			$1{\cdot}1[1 + 1/(\tan\delta)]$; 3·5 $-1{\cdot}3/(\tan\delta)$
−0·470	1·674	0·196	0·627	—	0·194			$0{\cdot}7[1 + 1/(\tan\delta)]$; 1·3 $-2{\cdot}5[1 + 0{\cdot}5/(\tan\delta)]$; $-2{\cdot}7/(\tan\delta)$
−0·470	1·674	0·197	0·621	0·011	0·172	0·0475		$0{\cdot}7[1 + 1/(\tan\delta)]$; 2·3 $-2{\cdot}5[1 + 0{\cdot}12/(\tan\delta)]$; $-2{\cdot}7/(\tan\delta)$
−0·470	1·674	0·198	0·620	0·012	0·172	0·0430	0·0122	$0{\cdot}7[1 + 1/(\tan\delta)]$; 2·7 $-2{\cdot}5[1 + 0{\cdot}03/(\tan\delta)]$; $-2{\cdot}7/(\tan\delta)$
−0·470	1·674	0·198	0·620	0·012	0·172	0·0433	0·0108	$0{\cdot}7[1 + 1/(\tan\delta)]$; 2·7 $-2{\cdot}7$; $-2{\cdot}7/(\tan\delta)$

scale into the direction of shorter times relative to its predecessor, and will have a coefficient which is exactly 1/4 of the coefficient of its predecessor.

Formula 16 constitutes the first example of a short time truncation problem. Calculation of $J''(\omega)$ at $\omega = 1/t$ is strongly influenced by the behaviour of the logarithmic derivative of the creep compliance at short times. The corresponding coefficients decrease only weakly, namely inversely proportional with the distance. Therefore, experimental evidence on the magnitude of the short time creep behaviour is needed before these terms can be rejected on the argument of their smallness.

In summary, we state that calculation of the loss compliance from creep is always difficult but will be especially cumbersome at small damping values.

3. Calculation of the creep compliance from the frequency dependence of storage and loss compliance

This problem has been treated in detail elsewhere[8]. We first remark that the knowledge of the frequency dependence of the loss compliance only is insufficient to calculate the creep compliance, because the value of the instantaneous compliance, J_0, is not contained in $J''(\omega)$. For this calculation one should in addition know at least the value of the storage compliance at one frequency. Formulae of this type are listed in *Table 3*.

In cases where the damping is small, a very simple (2-point) formula will be sufficient, *viz.*:

$$J(t) \sim J'(\omega) + 0{\cdot}566J''(\omega/2) - 0{\cdot}203J''(\omega) \qquad (17)$$

Limits for the relative error of this formula are: -8 (tan δ) per cent and $+8$ (tan δ) per cent. Therefore, this formula will have a relative error smaller than 1 per cent in all cases that (tan δ) is smaller than 0·125.

For large values of tan δ, more involved formulae are available. It will always be possible to find a formula with a relative error smaller than 1 per cent. In the worst case, the last formula of *Table 3*, which is always within 1 per cent, would involve knowledge of the loss compliance between $\omega/16$ and 8ω for calculation of $J(1/\omega)$.

In summary, we state that calculation of the creep compliance from the value of the storage compliance at one frequency and the frequency dependence of the loss compliance is an easy problem. It will be extremely simple for low values of the damping. No truncation problems will occur.

It is also possible to calculate the creep compliance from the value of the loss compliance at one frequency and the frequency dependence of the storage compliance. Formulae of this type are given in *Table 4*. These formulae all have a very low bound for the relative error at large damping values. They will, therefore, be especially appropriate for use in the region of higher damping. However, when applying these formulae in the case of low damping[8], some caution should be taken. Under these circumstances it will be safe to use the last formula of *Table 4*:

$$\begin{aligned} J(t) \sim\ & J'(\omega) + 0{\cdot}496J''(\omega/2) - 0{\cdot}065\{J'(\omega/4) - J'(\omega/2)\} \\ & - 0{\cdot}073\{J'(\omega/2) - J'(\omega)\} - 0{\cdot}111\{J'(\omega) - J'(2\omega)\} \\ & - 0{\cdot}030\{J'(8\omega)\} - J'(16\omega)\} - 0{\cdot}007\{J'(32\omega) - J'(64\omega)\} \cdot \cdot \cdot \cdot \end{aligned} \qquad (18)$$

Table 3. Numerical formulae for calculation of creep compliance from storage compliance and loss compliance ($t = 1/\omega$)

$$J(t) \sim J'(\omega) + aJ''(\omega/16) + bJ''(\omega/8) + cJ''(\omega/4) + dJ''(\omega/2) + eJ''(\omega) + fJ''(2\omega) + gJ''(4\omega) + hJ''(8\omega)$$

a	b	c	d	e	f	g	h	*Limits for relative error*; %
			0·446					22·3 tan δ; 7·9 −10·8 tan δ; −10·8
			0·566	−0·203				8·0 tan δ; 7·6 −7·9 tan δ; −7·0
			0·482	—	−0·0920			8·2 tan δ; 8·7 −8·2 tan δ; −8·2
			0·534	−0·129	−0·0312			7·6 tan δ; 7·8 −7·6 tan δ; −7·6
		0·0743	0·388	−0·112				10·0 tan δ; −6·2 tan δ; −4·2
		0·0872	0·319	—	−0·0532			7·5 tan δ; 3·9 −7·5 tan δ; −3·9
		0·103	0·278	—	—	—	−0·0166	3·5 tan δ; 3·6 −3·5 tan δ; −3·6
	0·0198	0·0375	0·339	—	−0·0122	—	−0·0152	3·6 tan δ; 2·1 −3·6 tan δ; −2·1
	0·0509	−0·116	0·635	−0·254	0·0383	0·0547	−0·0404	1·3 tan δ; 1·3 −1·3 tan δ; −1·3
0·00807	0·00719	−0·00616	0·467	−0·0918	−0·0534	0·0800	−0·0428	1·3 tan δ; 0·8 −1·3 tan δ; −0·8

Table 4. Numerical formulae for calculation of creep compliance from storage compliance and loss compliance: ($t = 1/\omega$)

$$J(t) \sim J'(\omega) + DJ''(\omega/2) - a[J'(\omega/4) - J'(\omega/2)] - b[J'(\omega/2) - J'(\omega)] - c[J'(\omega) - J'(2\omega)] - f[J'(8\omega) - J'(16\omega)] - h[J'(32\omega) - J'(64\omega)]$$

D	a	b	c	f	h	*Limits for relative error*; %
0·485		0·200				24 (tan δ); 3·0 −3·0 (tan δ); −3·0
0·495	0·0930	—	0·163			25 (tan δ); 1·0 −2·0 (tan δ); −1·0
0·496	0·0651	0·0729	0·111	0·0340		0·19[1 + 6(tan δ)]; 0·8 −1·7 (tan δ); −0·8
0·496	0·0652	0·0723	0·112	0·0293	0·00809	0·046[1 + 30(tan δ)]; 0·8 −1·4 (tan δ); −0·8
0·496	0·0651	0·0731	0·111	0·0300	0·00683	1·4 (tan δ); 0·8 −1·4 (tan δ); −0·8

This is assumed to consist of an infinite number of terms. Each term following the one with coefficient 0·007 will be shifted a factor of 4 in frequency scale into the direction of higher frequencies relative to its predecessor, and will have a coefficient which is exactly $\frac{1}{4}$ of the coefficient of its predecessor.

In general, the high frequency truncation problem in formula 18 will not cause serious trouble, as the corresponding coefficients are very low.

Finally, it is possible to give formulae which make use of the frequency dependence of storage and loss compliance. A useful example will be:

$$J(t) \sim J'(\omega) + 0{\cdot}496J''(\omega/2) - 0{\cdot}027J''(8\omega) - 0{\cdot}0583\{J'(\omega/4) - J'(\omega/2)\} - 0{\cdot}0906\{J'(\omega/2) - J'(\omega)\} - 0{\cdot}0854\{J'(\omega) - J'(2\omega)\} \quad (19)$$

It has these error limits: $-3{\cdot}2$ (tan δ) per cent; $-1{\cdot}0$ per cent and $+3{\cdot}2$ (tan δ) per cent; 1·0 per cent.

4. Calculation of storage modulus from the time dependence of the relaxation modulus

There exists a well-known parallelism between formulae which connect compliances and the corresponding formulae which connect moduli with each other. The reason is the far reaching similarity of the intensity functions of the compliances when written as integral transforms of the retardation spectrum, on the one hand, and those of the moduli when written as integral transforms of the relaxation spectrum on the other.

Consequently, each approximation formula between compliances may be transformed into one between moduli by performance of the simultaneous substitutions:

$$J(t) \to -G(t); \quad J'(\omega) \to -G'(\omega); \quad J''(\omega) \to G''(\omega) \quad (20)$$

However, the error limits of the new approximation for the moduli, which is obtained in this way, will generally differ from the error limits, that were valid for the approximation for the compliances. It is therefore not possible to translate the optimized systems as given in *Tables 1, 2, 3* and *4* by means of this substitution into optimized systems for the conversion of moduli. The problem of the conversion of moduli has to be considered separately.

It is only for small damping values that, by means of equation 20, a useful approximation for compliances can be translated into a useful approximation for moduli.

We derive from equation 14 a formula for the calculation of the storage modulus for small values of the damping (or the negative double logarithmic slope of the relaxation modulus with respect to time):

$$G'(\omega) \sim G(t) + 0{\cdot}86\{G(t) - G(2t)\} \quad (21)$$

Limits for the relative error of this equation will be -15 (tan δ) per cent and $+15$ (tan δ) per cent. It is also possible to translate the second formula or the fourth formula of *Table 1*; they will have these error limits: -8 (tan δ) per cent, $+8$ (tan δ) per cent and $-2{\cdot}1$ (tan δ) per cent, 2·1 (tan δ) per cent respectively, and will constitute still better approximations.

Finally, we can construct approximations for $G'(\omega)$ which will be accurate for arbitrary high damping values. An example is the following equation:

$$G'(\omega) \sim -0{\cdot}145\{G(4t) - G(8t)\} + 0{\cdot}729\{G(2t) - G(4t)\} + 0{\cdot}00288\{G(t) - G(2t)\} + 0{\cdot}111\{G(t/2) - G(t)\} + 0{\cdot}102\{G(t/4) - G(t/2)\} + 0{\cdot}0074\{G(t/16) - G(t/8)\} + \cdots \quad (22)$$

The limits for the relative error in this approximation are: $-1{\cdot}1$ (tan δ) per cent; $-8{\cdot}2$ per cent and $1{\cdot}1$ (tan δ) per cent; $8{\cdot}2$ per cent. Equation 22 is assumed to represent an infinite series. Each term after the one with coefficient 0·0074 will be shifted a factor of 4 in time scale into the direction of shorter times relative to its predecessor and will have a coefficient which is exactly $\frac{1}{16}$ of the coefficient of its predecessor.

Though equation 22 presents a short time truncation problem, this will not be serious in practice. The coefficients of the short time tail decrease strongly, namely inversely proportional to the square of their distance.

We may conclude, therefore, that calculation of the storage modulus from the relaxation modulus will be easy for all damping values; it is extremely simple for low damping.

5. Calculation of loss modulus from the time dependence of the relaxation modulus

Calculations of the loss modulus from stress relaxation will be even more difficult than that of loss compliance from creep. The only formula for this purpose that can be error bound is the transposed form of equation 16:

$$G''(\omega) \sim -0{\cdot}470\{G(2t) - G(4t)\} + 1{\cdot}674\{G(t) - G(2t)\} + 0{\cdot}198\{G(t/2) - G(t)\} + 0{\cdot}620\{G(t/4) - G(t/2)\} + 0{\cdot}012\{G(t/8) - G(t/4)\} + 0{\cdot}172\{G(t/16) - G(t/8)\} + 0{\cdot}043\{G(t/64) - G(t/32)\} + \cdots \quad (23)$$

The limits for the relative error of equation 23 will be: $-2{\cdot}7$ per cent and $+2{\cdot}7$ per cent, irrespective of the value of (tan δ).

For creep it was possible to truncate equation 16, if the value of tan δ at angular frequency $\omega = 1/t$ was sufficiently high. For stress relaxation, a truncation of equation 23 is not possible. Every truncated formula will have a lower limit for the relative error equal to -100 per cent for all tan δ values.

The only way to reject terms of the short time tail of equation 23 is by use of experimental knowledge about the order of magnitude of those terms. For this purpose it is necessary to know upper bounds of the negative value of the slope of the relaxation modulus with respect to the logarithm of time in the entire time region left to point $t = 1/\omega$, where the calculation is performed. A measure for this slope may be found in the value of the loss modulus at angular frequencies higher than ω, because of the inequality[3]:

$$-\mathrm{d}G(t)/\mathrm{d}\ln t \leqslant G''(\omega) \quad (24)$$

We conclude that calculation of $G''(\omega)$ from $G(t)$ will always present a cumbersome truncation problem.

6. Calculation of relaxation modulus from storage and loss moduli

Three types of formulae may be considered:

(a) calculation from the frequency dependence of the loss modulus and the value of the storage modulus at one frequency;

(b) calculation from the frequency dependence of the storage modulus and the value of the loss modulus at one frequency;

(c) calculation from the frequency depedence of the storage modulus.

The optimized systems have not yet been developed. However, the following remarks may be important:

For small values of the damping we may transpose equation 17 to:

$$G(t) \sim G'(\omega) - 0{\cdot}566G''(\omega/2) + 0{\cdot}203G''(\omega) \tag{25}$$

The limits for the *relative* error of equation 17 had been: -8 (tan δ) per cent and $+8$ (tan δ) per cent; they had been derived from limits for the *absolute* error of equation 17, which had been equal to $-0{\cdot}08J''(\omega)$ and $+0{\cdot}08J''(\omega)$. Consequently, we find as limits for the absolute error of equation 25: $-0{\cdot}08G''(\omega)$ and $+0{\cdot}08G''(\omega)$. These may be transposed again into limits for the relative error of equation 25, which become complicated functions of tan δ; for small values of tan δ these are again: -8 (tan δ) per cent and $+8$ (tan δ) per cent; for higher values of (tan δ), however, the limits will tend to $-\infty$ and $+\infty$ and increase much faster than in proportion to (tan δ).

As an example of type (b), we may transpose equation 18 into:

$$\begin{aligned} G(t) \sim\ & G'(\omega) - 0{\cdot}496G''(\omega/2) + 0{\cdot}065\{G'(\omega/2) - G'(\omega/4)\} \\ & + 0{\cdot}073\{G'(\omega) - G'(\omega/2)\} + 0{\cdot}111\{G'(2\omega) - G'(\omega)\} \\ & + 0{\cdot}030\{G'(16\omega) - G'(8\omega)\} + 0{\cdot}007\{G'(64\omega) - G'(32\omega)\} + \cdots \end{aligned} \tag{26}$$

Limits for the absolute error of this approximation will be: $-0{\cdot}014G''(\omega)$ and $+0{\cdot}014G''(\omega)$.

In contrast with the problems which have been considered so far, it is not possible to derive formulae for calculation of $G(t)$, which can be error bound for all (tan δ) values. Every formula will have limits for the relative error which tend to $-\infty$ and $+\infty$ for large (tan δ); and, therefore, every formula must finally fail in the high (tan δ) region.

We conclude that calculation of $G(t)$ from $G'(\omega)$ and $G''(\omega)$ will be easy for low damping, cumbersome for intermediate value of the damping, and impossible for very high values of the damping.

7. Calculation of loss modulus from frequency dependence of storage modulus and vice versa

In view of the existence of the Kramers[9]–Kronig[10] relations, one would expect that the loss modulus could be calculated from the frequency dependence of the storage modulus, and that the storage modulus could be calculated, apart from an arbitrary constant, from the frequency dependence of the loss modulus. The corresponding numerical relations, however, have not yet been investigated.

8. Calculation of relaxation and retardation spectrum from experimentally accessible functions

The famous problem of calculating the relaxation and retardation spectrum may be reconsidered at this stage. If we try to apply the same methods that have been used to derive the numerical relations between experimentally accessible functions, we immediately discover that no approximations for the spectra can be found which could be error bound.

The intensity functions of the spectra are delta functions that are infinite at one value of τ and zero elsewhere. Such a 'function' can never be approximated with *finite* accuracy by means of the intensity functions of measurable quantities, which are all finite and differentiable functions of time. Therefore, each approximation formula for the spectrum will have limits for the relative error, which are $-\infty$ and $+\infty$.

We believe that the spectra as defined in equations 1 and 2 are not experimentally accessible functions in a unique sense: Each finite set of experimental data can be described by a large number of spectra that are very different.

It is possible to redefine the spectra by an additional smoothing condition in such a way that the redefined spectra become experimentally accessible. The redefined spectrum $f_{red}(\tau)$ can be written by a formula:

$$f_{red}(t) = \int s(t/\tau)\, f(\tau)\, d\tau \tag{27}$$

where $s(x)$ is a function which approximates the delta function, i.e. it has a maximum at $x = 1$, tends to zero for $x \to 0$ and $x \to \infty$, and is normalized. The 'value' of the redfined spectrum, however, will then depend on the choice of the smoothing function $s(x)$.

The reader will appreciate that about 16 years ago functions of type 27 were developed as 'approximations to the spectra'[11].

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