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Theoretical Considerations for Viscoelastic Characterization of Biomaterials

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RINGKASAN

Untuk sesuatu rekaan, pembinaan serta ujian jentera-jentera memproses dan mesin-mesin pengangkutan bahan-bahan biological, sifat-sifat mekanikal bahan-bahan tersebut perlu diketahui terlebeh dahulu. Oleh kerana percubaan-percubaan yang lampau dapat mempastikan bahawa bahan-bahan biological bersifat viscoelastic, perlulah ianya dinyatakan supaya dapat dipastikan tindak balas sifat-sifat tersebut dalam kerja-kerja mengangkut dan memproses. Untuk bahan-bahan kejuruteraan, penggunaan teori viscoelasticity adalah terbukti dan nyata, tetapi bagi bahan-bahan biological perkara ini tidaklah sedemikian. Untuk bahan-bahan biological, teori asas viscoelasticity perlu diubah untuk penyesuaian dengan sifat-sifat biological. Oleh itu, teori-teori yang perlu adalah dibincangkan dalam kajian ini. Ianya juga memperkenalkan istilah-istilah bio-engineering dan menghuraikan gambaran viscoelastic terutamanya dibidang bio-creep dan relaxation.

Kajian ini berusaha untuk menunjukkan persamaan sifat tersebut dengan model-model mekanikal dan persamaan mereka secara am. Ringkasan barrel-effect untuk bahan viscoelastic yang berunsur biological juga diberikan. Peranan shift-factors sebagai sifat bahan juga diliputi dalam perbincangan ini.

SUMMARY

The mechanical properties of biomaterials need to be known and standardised before design, construction and testing of processing and handling machinery can be undertaken. Since experiments in the past have proved the viscoelastic nature of biomaterials, it is necessary to characterize them in order to ascertain their response to handling and processing. For engineering materials, the application of the theory of viscoelasticity is now quite well known; but this is not the case for biomaterials. In the characterization of biomaterials it will be necessary to modify the basic theory in the context of the bio-effects. The theoretical considerations are discussed in this study. The various bio-engineering terms are defined and viscoelastic representation with particular emphasis on bio-creep and relaxation is described.

An attempt is made to show analogy with the mechanical models and their generalization. Barreleffect for viscoelastic materials of bio-origin is outlined. The shift-factors and their significance as a material property are also included.

INTRODUCTION

One of the basic needs in the food industry is knowledge of mechanical properties of the products requiring processing and handling. The structural complexity of food products presents a challenge in measuring the rheological properties. In order to provide food of higher quality it is necessary to understand the physical laws governing the response of biological materials to handling and processing. Damage and spoilage have to be controlled to increase the efficiency of harvesting, handling and storage facilities. The importance of knowledge of these basic engineering parameters for food products and the application of this information to an engineering analysis is obvious. For example, the response of specific food products to load or deformation at various temperature conditions is required. Only recently, food and agricultural engineers have begun to apply the basic theories of engineering to the behaviour of food and agricultural products. The scope of Biomaterials Science coming under Biomechanics or Bioengineering seems unlimited.

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Biomaterials are, in fact, plant and animal materials. They are the principal raw materials for food and agricultural industries. They also include the final products after primary, secondary or tertiary processing; and the agricultural and food engineers and scientists are concerned with their physical behaviour. The nature of biomaterials, e.g. specimen for testing (size, shape, non-availability of prepared specimen, ripeness, type, moisture, temperature, variety of the same kind, harvesting time effect, soil and irrigation effect, fertilizer and pesticide effect) present numerous obstacles in actual experimentation. It is, therefore, necessary for these problems and conditions to be specified for each testing of biomaterials. Since the citing of examples of even a single biomaterial will require mention and discussion of the whole spectrum under which testing is done and results obtained, no attempt has been made to analyse any example in this study.

Based on experimental evidences attributed to Zoerb and Hall (1960), Mohsenin (1963), Timbers (1964) and Morrow (1965), agricultural products are viscoelastic. From the very limited data available in this area it would appear that viscoelastic behaviour is non-linear. In an attempt to explain the rheological behaviour of agricultural products, simplified assumptions have been made and the theories of linear viscoelasticity applied.

BIOENGINEERING TERMINOLOGY

The following terms and definitions are used in this study. These are illustrated in Fig. 1.

Bioyield point is that point on the forcedeformation curve at which an increase in deformation occurs with a decrease or no change of the applied force. In many agricultural products, the presence of this bioyield point is an indication of initial cell rupture in the cellular structure of the material.

Rupture point is that point on the forcedeformation curve at which the crack is visible to the unaided eye. It indicates a failure in the macrostructure while bioyield point indicates a failure in microstructure of the specimen.

The degree of elasticity, D, is defined as the ratio of elastic deformation to the sum of elastic and plastic deformation when a material is loaded and then unloaded to zero load.





The hysteresis is defined as the energy absorbed by the material in a cycle of loading and unloading and is evaluated as the area between loading and unloading curves.

Viscoelasticity, in general, is a combined solid-like and liquid-like behaviour in which the stress-strain relationship is time dependent.

Linear viscoelasticity is defined as a viscoelastic behaviour in which the ratio of stress to strain is a function of time alone and not of the stress magnitude.

Stress relaxation is the decay of stress with time after the material is suddenly deformed to a given deformation or constant strain.

Creep is the deformation as a function of time when the material is suddenly subjected to a constant load. Relaxation time is the time required for the stress in the Maxwell model, to decay to 1/e or approximately 37 per cent of its original value.

Retardation time is the time required for the Kelvin model to deform to 1-1/e or approximately 63 per cent of its total deformation.

VISCOELASTIC REPRESENTATION

The mechanism of response of an engineering material may be studied on any of three basic levels: the molecular, structural, and the phenomenological level. At the first level, the response of the material is inferred from the properties of its microscopic elementary particles; at the second, the material is considered as being made up of nonhomogeneous, visible units whose interaction produces the observed behaviour; at the third, the material is assumed to be macroscopically homogeneous and isotropic such that the behaviour of any part of it, in any direction, is the same as that of the whole.

The behaviour of all real materials will fall somewhere between the two extremes of the Euclid-solid and Pascalian-liquid. The Euclidsolid is a completely rigid body, which is incompressible, while the Pascalian-liquid is also incompressible but offers absolutely no resistance to deformation. Next to the Euclid-solid in hierarchy is the Hooke-solid, for which stress is directly proportional to strain, while next to the Pascalian-liquid is the Newtonian-liquid, for which stress is directly proportional to the timerate of strain. Another type of idealized behaviour is that of the St. Venant-solid, which, up to a certain stress, called the yield stress, acts like a Hooke-solid, but once that stress is reached, it deforms plastically at constant stress.

In general, the response of all real materials is made up of a complex combination of the many idealized responses mentioned above. In fact, any quantitative representation of this complex response will in itself be idealized, the degree of idealization depending on the accuracy desired or achieved.

The fundamental problem in a study of material response is the determination of the functional relationship between the strain, and the stress, and their time derivatives. This is the rheological equation of state of the material. One of the fundamental methods of representing the linear viscoelastic materials is that of the Linear operator equation,

$$P_{\delta ij} = Q \epsilon_{ij}$$

Here the linear operators P and Q are defined as,

$$P = \sum_{o}^{m} \chi_{m} \frac{\lambda^{m}}{\lambda^{tm}}$$
$$Q = \sum_{o}^{n} \beta_{n} \frac{\lambda^{n}}{\lambda^{tn}}$$

where α and β are non-zero coefficients.

From the superposition principle creep is expressed mathematically as,

$$\epsilon$$
 (t) = $\delta\left(\frac{1}{E} + \psi(t)\right)$

and relaxation as,

$$\delta(t) = \epsilon [E + \phi(t)]$$

where ψ and ϕ are creep and relaxation functions respectively.

The phenomena of creep and relaxation, according to Gross (1953), are represented by a finite series of exponential functions with time distributed over a finite interval. This leads to two distribution functions of time constants i.e. distribution functions of both retardation and relaxation times.

Despite their mathematical elegance the practical application of the distribution functions, is difficult to quantify. It is suggested, therefore, that experimental data should not be expressed in terms of distributions, but rather in experimental parameters which are read or calculated directly from the experimental data.

Another fundamental way and one of the most widely used and easily interpreted methods of representing the behaviour of a viscoelastic material is to seek an analogy with mechanical models made up of various combinations of Hookean springs and Newtonian dashpots. These are the two primary elements of a mechanical model.

MECHANICAL MODELS

The combination of elastic and vicouss elements in series forms a Maxwell model as shown in Fig. 2(a) and the relationship between stress and strain is,



(A) MAXWELL MODEL

(b) KELVIN-VOIGT MODEL



(C) GENERALIZED MAXWELL MODEL

Fig. 2. Mechanical Models.

$$\dot{\sigma} \quad \sigma = \dot{\epsilon}$$

 $\frac{1}{E} \quad \eta = \dot{\epsilon}$

In a Kelvin-Voigt model, the two primary elements are connected in parallel as shown in Fig. 2(b), and the stress-strain relationship is

$$\sigma = \mathbf{E} \boldsymbol{\epsilon} + \eta \, \dot{\boldsymbol{\epsilon}}$$

There are two ways of systematically building up more complicated models, the Kelvin chain (or generalized Kelvin-Voigt model) and the generalized Maxwell model. In the former an arbitrary number of Kelvin units are in series. In the generalized Maxwell model, Maxwell units are in parallel as shown in Fig. 2(c).

In the generalized Maxwell model, if it is given a sudden deformation,

 $\epsilon = k H(t)$

where Heaviside function H (t) is

$$H(t) = 0, t < 0 \text{ and } H(t) = 1, t \ge 0$$

The relaxation behaviour of a linear viscoelastic material can be represented as

$$\boldsymbol{\sigma}(t) = k E_{1} H(t) + k \eta_{2} \delta(t) + k \frac{\eta_{2}}{\Sigma} E_{1} \exp(-E_{1} \frac{t}{\eta_{1}}) H(t)$$
where
$$\delta(t) = \frac{d}{dt} H(t)$$

The force response to a unit extension is defined by Bland (1960) as the relaxation function. It is therefore,

$$\phi(t) = \sum_{i=3}^{n} E_i \exp(-E_i - \frac{t}{\eta_i}) H(t)$$

Stress relxation in materials can be represented by a generalized Maxwell model and creep can be represented by Kelvin chains. This is the limitation of the rheological models in that one model may adequately represent relaxation while creep may be extremely difficult to represent.

It can therefore be postulated that the relaxation behaviour of a biomaterial is represented by generalized Maxwell model and the number of Maxwell units can be determined from the experiment.

If the stress in a material falls to zero for large values of time, then there should be no spring in parallel with the other elements when a generalized Maxwell model is postulated to simulate the behaviour of the biomaterials. If on the other hand, the stress does not approach zero as time approaches infinity, then obviously this type of behaviour should be represented by an elastic element in parallel with the generalized model.

PRONY-DIRICHLET SERIES

In a generalized Maxwell model, the step input in strain gives

$$\sigma(t) = \left(\begin{array}{c} E_{o} + \sum_{i=1}^{n} E_{i} e^{-t} \\ \end{array} \right) \epsilon_{o}$$

where the constant term E_0 has been added to allow for an elastic response. This model gives relaxation function (stress divided by constant strain) as

$$\boldsymbol{\phi}(\mathbf{t}) = \mathbf{E}_{\mathbf{o}} + \sum_{i=1}^{n} \mathbf{E}_{i} \mathbf{e} \mathbf{u}_{i}$$

The exponential nature of these functions makes it convenient when using Laplace Transforms for analytical manipulations. The above exponential series is called Prony-Dirichlet series. Under certain conditions it is mathematically complete. To determine the coefficients of this series, Brisbane (1966) has outlined the following steps;

(a) Choose η decades of time over the time interval of interest from the relaxation function curve. For example 1, 10, 10³, 10² seconds.

(b) Choose values of $_{i}$ such that one falls within each decade on the curve.

(c) Make use of the fact that at a sufficient long time,

$$\phi$$
 (∞) = E₀

(d) Equate the series value of E (t) to the experimental values at (n-1) points.

(e) Get a set of algebraic equations for the coefficients Ei.

The resulting set of equations for the coefficients E_i can be solved on a computer.

BARREL EFFECT

Bartenev and Zuyev (1968) proposed that under compressive loads the viscoelastic material fails usually either by shear or by rupture and not by a combination of both shear and rupture provided the compression proceeds without slipping or sliding on bearing surfaces. They further suggested the existence of the 'barrel' effect as shown in Fig. 3. Under compression of the specimen the points A and B are in tension as the free side surfaces bulge in the form of a barrel. Along the line AB of the barrel a growth of small tears takes place from the surface into the depth of the material.

SHIFT FACTORS

The temperature and moisture shift factors are used to describe the effects of temperature and moisture on any viscoelastic function. The shift factor is an inherent property of the material.

The thermo-rheologically simple nature of a material indicates that an increase in temperature corresponds to an increase in time. The same reasoning is used in a hydro-rheologically simple material so that an increase in moisture content would correspond to an increase in either temperature or time.



Fig. 3. Barrel effect under compression.

Temperature shift factor $a_T(T)$ is defined as $a_T = e^{f(T)}$

f(T) is a shift function of temperature and shift is positive as temperature increases when the same viscoelastic function is plotted against logarithm of time, as shown in Fig. 4.



Fig. 4. Shift-Principle.

Let $\phi(t, T_0)$ be the relaxation function at reference temperature T_0 and $\phi(t, T)$ at any temperature T.

Let us change independent variable such that

$$\phi (t, T_0) = L_1 (log t)$$

In other words, relaxation function is plotted against logarithm of time.

For thermo-rheologically simple materials, viscoelastic functions when plotted against log of time exhibit a shift but no change in shape when temperature is changed. The relationship is, therefore,

$$\phi (t, T) = L_1 (log t + f(T))$$

where

$$f(T_0) = 0$$
 and $\frac{df(T)}{dt} > 0$

Substituting the log shift-factor in place of the shift-function, we obtain

$$\begin{aligned} \phi (t, T) &= L_1 \left(\log t + \log a_T \right) \\ &= L_1 \left(\log t \cdot a_T \right) \\ &= L_1 \left(\log \frac{2}{3} \right) \end{aligned}$$

where $\dot{a} = t \cdot a_T$

Thus the relaxation function at any temperature can be directly obtained from the relaxation function at base temperature T_0 by replacing t with 3.

Based on the temperature shift factor, an analogy is established for a moisture shift factor. It is also defined as

 $a_{M} = e^{f(M)}$

where f(M) = shift function of moisture.

Hydro-rheologically simple materials are defined as those materials which have all material functions shifting in the same way and in the same amount.

Developing the relaxation function at any moisture content in the same way it is given that

 φ (t, M₀) = L₂ (log t) and φ (t, M) = L₂ (logt. a_n)

Christensen (1971) described the method for determining the shift factors which are defined this study. The viscoelastic mechanical in property-relaxation function, creep function or complex moduli, when plotted against the logarithm of time can be superimposed to form a single curve merely by shifting the various curves at different temperatures along the logarithm of time axis. If the curves do coincide within experimental error the basic postulate of thermorheologically simple material is verified. further claimed that there are no general inclusive guide lines that can be given to answer the question whether a given material can be expected to exhibit the thermo-rheologically simple type of behaviour. The only safe and certain answer lies in experimentally verifying or invalidating the shifting procedure for every material studied.

It can, therefore, be postulated that a biomaterial is both thermo and hydro-rheologically simple. That is, the material has only one timetemperature shift factor and one time-moisture shift factor.

CONCLUSIONS

- (i) Biomaterials are viscoelastic and need viscoelastic characterization.
- (ii) The Theory of Viscoelasticity as applied to engineering materials is standardised, but it should be modified when biomaterials are to be characterised.
- (iii) Viscoelastic behaviour can be represented mathematically by the linear operator equation and/or by an analogy of mechanical models.
- (iv) Stress-relaxation of biomaterials can be represented by a generalized Maxwell model which is expressed as a PRONY-DIRICH-LET series. The coefficients of this series are found by BRISBANE's method using computer.
- (v) "Barrel-effect" is observed in viscoelastic materials.
- (vi) The biomaterials can be postulated to be both thermo and hydro-rheologically simple.

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