Abstract

In product design it is important to choose the correct material for a specific application. Viscoelastic behavior let us know how much energy the material can dissipate on its internal structure or either return it to the surroundings, and the property that describe this is the Complex Modulus $G^*$, it is a complex quantity that can be separated in a real and an imaginary part called $G'$ storage modulus and $iG''$ loss modulus respectively. These properties can be measured experimentally from a small material sample easily by performing Dynamical Mechanical Analysis (DMA). In Product Design process there are both, computational and physical validations and there is the need of improving computational studies by understanding the physics of each component. Viscoelastic characteristics of materials can be represented by Prony series, also known as relaxation modulus in function of time. Relaxation modulus can be defined in most of Computer Aided Engineering (CAE) Software. In this article the procedure for calculating Prony Series from DMA data will be explained.

Keywords: Prony series, viscoelasticity, complex modulus, storage modulus, loss modulus, DMA.

Resumen

En el diseño de un producto es importante elegir el material correcto para una aplicación específica. El comportamiento viscoelástico nos muestra qué tanta energía puede disipar un material en su estructura interna, o bien, devolver a sus alrededores; la propiedad que describe esto es el módulo complejo $G^*$, que es una cantidad compleja que se puede separar en una parte real y una imaginaria llamada $G'$ módulo de almacenamiento e $iG''$ módulo de pérdida, respectivamente. Estas propiedades se pueden medir experimentalmente de una pequeña muestra de material, aplicando Análisis Mecánico Dinámico (DMA). En el proceso de diseño de un producto existen las validaciones computacionales y las validaciones físicas, por lo que existe la necesidad de mejorar los estudios computacionales mediante el entendimiento de la física de cada componente. Las características viscoelásticas de los materiales se pueden representar por Series de Prony, también conocidas como el módulo de relajación en función del tiempo. El módulo de relajación se puede definir en la mayoría de los paquetes de Software para Ingeniería Asistida por Computadora (CAE). En este artículo se explicará el procedimiento para calcular las Series de Prony de datos obtenidos con DMA.

Descriptores: Serie de Prony, viscoelasticidad, módulo complejo, módulo de almacenamiento, módulo de pérdida, DMA.
**INTRODUCTION**

In this article the method to calculate Prony series from DMA data will be explained with a real example. The Prony series of a piece of structural adhesive used in automotive industry will be obtained showing in detail the considerations and background of each decision taken during the analysis with the purpose of being a good reference for the reader. The motivation for writing this article is the lack of literature available on this subject.

Background on this article analysis is as follows: Automotive body is manufactured from steel or aluminum plates (Davies, 2003). Structural adhesives are used to fill the cavities between these plates and therefore increasing the strength of the structure. Normally they are installed before paint process as it is shown in Figure 1, where they will be cured with the furnace temperature. The installation process consists on place them on the surface of a metal plate in order they adhere by themselves to it. This process is most of the times done manually by an operator. Some of these adhesives when heated liberate gases that will create small bubbles on them, and they will increase their volumes, this is the expansion mechanism that promotes the filling of the closed section in the body structure as shown in Figure 2.

Energy during the impact will be dissipated by the structural adhesives. The properties that describe the energy dissipation capacity of materials are known as dynamic properties. The complex modulus \( G^* \) is a complex quantity that can be divided in a real part and an imaginary part, called storage modulus \( G' \), and the loss modulus \( G'' \), respectively. Their relationship is indicated in equation 1.

\[
G^* = G' + iG''
\]

(1)

The storage modulus \( G' \) describes the capacity of a material of storing and returning energy to the surroundings, and the loss modulus \( G'' \) describes the capacity of a material of dissipating energy on its internal structure. A graphical explanation of these concepts are often explained with the Figure 3.

**DINAMICAL MECHANICAL ANALYSIS**

The dynamical properties are obtained by an experimental method called dynamical mechanical analysis, DMA from now on. DMA is performed in a laboratory using a Dynamic Mechanical Analyzer, a device commonly used in the study of polymers rheology. Dynamical properties are studied by applying a cyclic stress into a material sample (Sepe, 1998), it can be applied in different modes: tension, bending, cantilever, dual cantilever, torsional, etc. Often the results are shown as frequency response diagrams, also named frequency sweep, where in the vertical axis are shown the values of Storage Modulus \( G' \) and loss Modulus \( G'' \), and in the horizontal axis are shown the frequencies where the analysis was performed. Most of the times horizontal axis is presented in a logarithmical scale since it is important to show high and low frequency values. An example of a frequency scan can be observed in Figure 4.
El Analizador Dinámico Mecánico es un dispositivo que tiene abrazaderas fijas y móviles. Las abrazaderas fijas se mantienen estáticas mientras que la abrazadera móvil se desplaza hacia arriba y abajo por un eje de transmisión, deformando el material en un patrón de oscilación, como se ilustra en la Figura 5. La magnitud y la frecuencia de la oscilación se progranan basadas en las características de cada material. El fondo matemático para el cálculo de $G'$ y $G''$ se puede encontrar en (Ferry, 1980).

En este trabajo se evaluó una muestra de vinilo estructural con baja capacidad de expansión (alrededor del 15 % de aumento de volumen) y se realizó una prueba de rep全家从0.1 a 100 Hz con 10 mediciones en cada década de frecuencia. Un década debe entenderse como un orden de magnitud, por ejemplo, desde 0.1 hasta 1 tenemos una década y desde 1 hasta 10 tenemos otra. Esta gama de frecuencias fue seleccionada basándose en las vibraciones comúnmente detectadas en vehículos por las irregularidades del pavimento por el sistema de suspensiones. La entrada típica va desde 1 a 2 Hz, como se mencionó en (Spinola, 2012), sin embargo, se decidió medir en un rango más amplio para tener los datos necesarios para hacer la aproximación de la curva $G'$ o $G''$ en función de la frecuencia $\omega$ como se expone en este trabajo.

Es deseable tener una gama de mediciones amplia de 3 ó más décadas para cualquier material.
der study to have a good material characterization. Also it is recommended that the common usage input frequency of the product that is under study is included in this range (in this case the structural adhesive used in automotive body). The advantage of perform testing in the frequencies domain is that it requires less time than carry out them on time domain, since the frequency is an inverse quantity with regards of time. This characterization in the frequencies domain will allow a good curve fitting that will let us know the numerical values that will be finally substituted in the Prony series that are functions in the time domain.

The deformation applied to the sample depends on the nature of the material. One method is to impose a small deformation (less than 1 % of the sample size) and keep increasing it until a constant value of the moduli is obtained.

The measured values obtained by performing DMA to this sample are shown in the Table I.

**MODELS OF VISCOELASTIC BEHAVIOR**

Most of all materials have a viscoelastic behavior and in the case of adhesives it is remarkable due to their polymeric nature. Elastic behavior is modeled by Hooke law that can be represented by the following equations:

$$\sigma = E\varepsilon$$  \(3\)

In the case of tensile stress, and:

$$\tau = G\dot{\gamma}$$  \(4\)

In the case of shearing stress; where:

- \(\sigma\) = tensile stress
- \(E\) = Young’s modulus
- \(\varepsilon\) = tensile strain
- \(\tau\) = shear stress
- \(G\) = shear modulus
- \(\gamma\) = strain by shear

Viscous behavior is modeled by the Newton’s law:

$$\tau = \eta\dot{\gamma}$$  \(5\)

Where:

- \(\tau\) = shear stress
- \(\eta\) = viscosity
- \(\dot{\gamma}\) = strain rate

In the case of viscoelasticity of polymers, a more complex model is needed because it depends on several facts: the size of the molecules, if they are crosslinked or not (Ferry, 1980), and also stress relaxation due to polymeric chains re-ordering with time. By these reasons, the equation that models viscoelasticity uses a modulus that is a function of time, as follows:

$$\tau = G(t)\dot{\gamma}$$  \(6\)

Where \(G(t)\) is the stress relaxation modulus of the material.

Ferry (1980) shows the same constitutive equation with the following notation:

$$\sigma_i(t) = \int G(t - t')\dot{\gamma}_i(t')dt'$$  \(7\)

Where:

- \(\sigma_i\) = stress tensor
- \(\dot{\gamma}_i\) = strain rate tensor
- \(t'\) = all the past times were the integration must be carried out over all of them, this must be considered due to the memory of the material during deformation.

Two basic mechanical models that predict viscoelastic properties of materials are Voigt and Maxwell elements, that are represented by springs and dashpots connected between them, as illustrated in Figure 7. The materials behavior can be described as combinations of Maxwell and Voigt elements connected between them. One simple model is the generalized Maxwell model that is just a set of Maxwell elements connected in parallel.

In generalized Maxwell model, the contribution in relaxation modulus of each Maxwell element is given by the equation 8:

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 depends on the number of terms desired for the sum. One theoretical method is exposed by Baumgaertel & Winter in (1989). For practical uses, this can be done using software to approximate the curve to these custom equations. The first step is to decide the number of terms. For this work, we will approximate a curve to the first two terms of the sum shown in equation 12.

\[ G'(\omega) = \left[ G_0 \right] + \sum_{i} \frac{G_i \omega^2 \tau_i^2}{1 + \omega^2 \tau_i^2} \]

In equation 14 there are five unknown values: \( G_0, G_i, \tau_1, \tau_2 \), these values will be approximated by fitting the curve with the form of equation 14 to the data obtained by DMA on the “Storage Modulus” column in Table 1. Software such as Matlab® can be used for a fast procedure capable to meet the Automotive Industry demands on quick analysis. The command used in Matlab® to make the curve approximation is “cftool” (curve fitting tool). This command will display an interactive fitting tool where the column vectors previously created with the information of the columns “Storage Modulus” and “Frequency” in the Table 1 can be loaded as vertical and horizontal axis respectively. In the case of solid-like materials is convenient to use the storage modulus instead of the loss modulus, for example, in this material it was possible to make the approximation of the curve with 30 points over 3 decades, but in the case of loss modulus the experiment should be made in a wider range of frequencies to get a better approximation of loss modulus curve. The loss modulus values are small in comparison to storage modulus values (by a factor of 30 times approximately). This also can be noticed in \( \tan \delta \) values that approach zero. By this reason it is very difficult to get a good approximation with loss modulus curve, since dominant behavior is described by storage modulus. Only storage modulus curve approximation will be discussed in this work.
The obtained values from storage modulus $G'(\omega)$ curve approximation are the following: $G'_c = 1947.00$, $G'_1 = 85.79$, $G'_2 = 97.63$, $\tau_1 = 0.0659$, $\tau_2 = 1.6950$ replacing these values in equation 14 it is obtained a function of the frequency $\omega$ only, as is shown in equation 15:

$$G'(\omega) = 1947.00 + \frac{85.79\omega^2(0.0659)^2}{1 + \omega^2(0.0659)^2} + \frac{97.63\omega^2(1.695)^2}{1 + \omega^2(1.695)^2}$$

(15)

By plotting the equation 15 as a function of the frequency $\omega$ is obtained the chart in Figure 8.

The values obtained in the curve approximation can also be replaced in the equation 11, in this case for the two first terms of the series, obtaining:

$$G(t) = 1947.00 + 85.79e^{-0.0659\cdot t} + 97.63e^{-1.695\cdot t}$$

(16)

The terms of equation 16 are the Prony series of the structural adhesive, also known as the relaxation modulus of the material in function of time. In Figure 9 is showed a plot of equation 16 as a function of time $t$, it is possible to observe the behavior of the relaxation modulus, where can be noticed that it’s value when a load
have been applied for a long time tends to the value of $G_0 = 1947.00 \text{ MPa}$, as mentioned previously.

It can be confirmed the accuracy of the model since the approximated values of $G'(\omega)$ calculated with equation 15 have less than 1 % of error with regards of the data obtained by experimental measurement through DMA experiment. Table 2 shows the error calculation for each DMA data point vs. the calculated value with the approximated function.

These results allow us to confirm that model showed in equation 15 is a good prediction for $G'(\omega)$ since the error values are below 1%, and this assure that $G(t)$ values will have good correlation with real behavior.

**CAE modeling using Prony series**

As mentioned previously, the Prony Series of equation 16, are also known as the discrete relaxation modulus in function of time. When equation 16 is substituted in equation 7, the constitutive equation is completely defined, the resulting is in equation 17.

$$\sigma_{ij} = \int_0^t \left[ 1947.00 + 85.79 e^{-(t-t^*)/0.0899} + 97.63 e^{-(t-t^*)/1.695} \right] G_{ij}(t^*) dt^*$$

(17)

CAE makes possible to incorporate the geometry of the product under analysis by dividing a complex geometry in small elements through the creation of a mesh. Equation 17 will be discretized and solved for each one of the elements of this mesh, since $\sigma_{ij}$ is the stress tensor of an infinitesimal element as illustrated in Figure 10.

The definition of Prony Series may be different on each commercial CAE software package. In the case of Abaqus® when the model is viscoelastic and defined in the time domain, the software displays a table similar to Table 3, where the requested parameters are: $g_i$, $k_i$, and $t_i$. The $g_i$ terms are the normalized Prony coefficients for shear (deviatoric) behavior, the $k_i$ terms are the normalized Prony coefficients for volumetric behavior, and the $t_i$ values are the relaxation times of Prony series. Abaqus® assumes that the frequency dependence of $g_i$ and $k_i$ is independent. In this work the volume changes in the material will not be considered, therefore the column for $k_i$ values will be left blank. The changes in shape are described by $g_i$, the deviatoric behavior, described by $g_i$ terms, they are defined as follows (Chen, 2000):

$$g_i = \frac{G_i}{G_0}$$

(18)

Where the $G_i$ values are the coefficients of the Prony series, and $G_0$ is the relaxation modulus $G(t)$ evaluated in $t = 0$. 

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Figure 8. Curve approximation of storage modulus to data obtained by DMA

Figure 9. Relaxation modulus $G(t)$ in function of time

Figure 10. Stress tensor components
### Table 2. Error calculation of values measured with DMA vs. values calculated with the fitted function $G'(\omega)$ (equation 15)

| Frequency | Calculated storage modulus with equation 15 | Measured storage modulus | Error $= \frac{|measured\ value - calculated\ value|}{measured\ value}$ |
|-----------|---------------------------------------------|---------------------------|-----------------------------------------------------------------|
| Hz        | MPA                                        | MPA                       | %                                                               |
| 0.1       | 1949.7303                                  | 1941                      | 0.4498                                                          |
| 0.13      | 1951.5271                                  | 1944                      | 0.3872                                                          |
| 0.16      | 1953.6981                                  | 1953                      | 0.0357                                                          |
| 0.2       | 1957.0780                                  | 1961                      | 0.2000                                                          |
| 0.25      | 1961.882                                   | 1968                      | 0.3107                                                          |
| 0.32      | 1969.2311                                  | 1976                      | 0.3426                                                          |
| 0.4       | 1977.8047                                  | 1983                      | 0.2620                                                          |
| 0.5       | 1987.0930                                  | 1990                      | 0.1054                                                          |
| 0.63      | 1999.1612                                  | 1998                      | 0.0581                                                          |
| 0.79      | 2009.9052                                  | 2007                      | 0.1448                                                          |
| 1         | 2019.7899                                  | 2014                      | 0.2875                                                          |
| 1.3       | 2028.5758                                  | 2021                      | 0.3749                                                          |
| 1.6       | 2033.8793                                  | 2027                      | 0.3394                                                          |
| 2         | 2038.2660                                  | 2033                      | 0.2590                                                          |
| 2.5       | 2041.7259                                  | 2039                      | 0.1337                                                          |
| 3.2       | 2045.0396                                  | 2046                      | 0.0469                                                          |
| 5         | 2051.6141                                  | 2058                      | 0.3103                                                          |
| 6.3       | 2056.2783                                  | 2064                      | 0.3741                                                          |
| 7.9       | 2062.1219                                  | 2069                      | 0.3280                                                          |
| 10        | 2070.0282                                  | 2076                      | 0.2877                                                          |
| 12.6      | 2079.1046                                  | 2082                      | 0.1391                                                          |
| 15.8      | 2088.7098                                  | 2087                      | 0.0819                                                          |
| 19        | 2096.4331                                  | 2093                      | 0.1640                                                          |
| 25        | 2106.6909                                  | 2100                      | 0.3186                                                          |
| 31.6      | 2113.6681                                  | 2106                      | 0.3641                                                          |
| 39.8      | 2118.8206                                  | 2112                      | 0.3229                                                          |
| 50        | 2122.4477                                  | 2119                      | 0.1627                                                          |
| 63        | 2124.9605                                  | 2121                      | 0.1867                                                          |
| 79.5      | 2126.6367                                  | 2137                      | 0.4849                                                          |
| 100       | 2127.7134                                  | 2141                      | 0.6206                                                          |

### Table 3. Dimensionless Prony series coefficients input in Abaqus®

| $\alpha_i$ | $k_i$ | $\tau_i$ |
|------------|-------|---------|
| 0.0403     | -     | 0.0659  |
| 0.0458     | -     | 1.695   |

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Calculating the values of \( G_0 \), \( g_1 \) and \( g_2 \):

\[
G_0 = G(0) = 2130.42 \text{ MPa}
\]

\[
g_1 = \frac{G_1}{G_0} = \frac{85.79 \text{ [MPa]}}{2130.42 \text{ [MPa]}} = 0.0403
\]

\[
g_2 = \frac{G_2}{G_0} = \frac{97.63 \text{ [MPa]}}{2130.42 \text{ [MPa]}} = 0.0458
\]

\( g_1 \) and \( g_2 \) are also known as the dimensionless Prony series coefficients.

Table 3 shows the input of values in Abaqus®. This table can have as many rows as known terms of the Prony series are available. By filling this table, the viscoelastic characteristics of the material under study are completely defined.

The method to calculate the relaxation modulus in function of time presented in this work has many application possibilities. Examples of other applications in automotive industry that used Prony series to describe viscoelastic behavior are: Prediction of damping of railway sandwich type dampers (Merideno et al., 2014), Modeling of viscoelastic parameters for automotive seating applications (Deng et al., 2003), Stress concentration analysis near holes in viscoelastic bodies (Levin et al., 2013), Analysis of adhesion of thermoplastics to steel (Golaz et al., 2011), Finite element analysis of seat cushion and soft-tissue materials focused on passenger-vehicle occupants comfort (Grujicic et al., 2009), Modeling of composites viscoelastic behavior (Machado et al., 2016; Ishikawa et al., 2018).

**Conclusions**

Viscoelastic properties of materials applied on CAE studies can provide more accurate results since it is a better approximation to real behavior of the material. The relaxation modulus of the analyzed structural adhesive showed a significant dependence with time since modulus values change from the maximum value in \( G_0 = 2130.42 \text{ MPa} \) to the value of \( G = 1949.00 \text{ MPa} \) due to stress relaxation. This shows that for vehicle CAE impact evaluation that occurs at a high speed the modulus will have also a high value since the stress relaxation will not occur at this speed. In the case of durability CAE evaluation, where the load is applied in long periods of time or in a time dependant load variation the stress relaxation will occur, therefore the modulus value will achieve the \( G_0 \) value. Viscoelastic properties of structural adhesives can be incorporated to CAE by estimating Relaxation modulus in function of time through Prony series by fitting a model to DMA experimental data in function of frequency.

Model fitting for \( G'(\omega) \) with respect to experimental DMA data showed an error smaller than 1 % using only two terms of the Prony series.

Through this methodology, parameters required to model viscoelasticity behavior in function of time in CAE software like Abaqus® can be obtained from the information of a single DMA experiment performed to a small material sample.

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