



Loss modulus measurement of a viscoelastic polymer at acoustic and ultrasonic frequencies using vibrothermography

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ABSTRACT

This paper describes a method to calculate the loss modulus of a viscoelastic polymer material at high frequencies based on vibrothermography. The polymer material is coated on the surface of a metallic substrate and resonant mechanical vibrations are generated in the substrate. The vibrating polymer coating undergoes hysteretic heating because of its viscoelastic nature. By measuring the surface temperature of the polymer coating and by solving the heat transfer equation, the heat intensity dissipated at the polymer is estimated. The loss modulus is then calculated from the estimated heat intensity and the measured dynamic vibrational strain on the coating surface. The measurement method is demonstrated with two different polymer materials at several frequencies in the range of 12–30 kHz and these values are compared with results from dynamic mechanical analysis experiments.

1. Introduction

Polymers are a class of materials that contain repeating chains of organic or inorganic molecules. These materials are used in various forms such as composites, adhesives, and coatings. Composites are increasingly used in several industries because of their lighter weight and equivalent mechanical properties compared to conventional metal alloys [1]. Unlike metallic materials where presence of defects such as fatigue cracks result in component damage, polymer composites have the ability to repair themselves when infused with repair agents during the cure process [2,3]. These are known as self-healing composites and have the potential to be used as structural components in aerospace and automotive industries [4]. The performance of polymer composites can be further enhanced by adding appropriate nano-sized particles to the matrix. Nanocomposites exhibit superior material properties compared to that of polymer composites and development of these materials is an active area of research [5–7]. Polymer adhesives can be used in composite-composite as well as metal-composite joining [8]. The advantages of using polymer adhesives compared to conventional metallic fasteners are that adhesive joints are less prone to fatigue damage and corrosion, and add lesser weight to the components. Polymers are also used in the development of functional materials such as corrosion resistant coatings, coatings to improve wettability, and electroactive actuators [9–11].

In all the above applications, mechanical characterization of polymers is an important step to understand the suitability and limitations of the material for the intended application. In this paper, we introduce a novel method to measure the loss modulus of a polymer adhesive coating using vibrothermography. Loss modulus of a polymer determines the amount of energy lost by the material when subjected to cyclical external loading. The proposed method uses vibration induced surface heating of the polymer along with the dynamic strain to measure the loss modulus. This method can be applied to any polymeric system applied as a surface coating on a metal or non-metallic substrate.

Vibrothermography is a nondestructive evaluation method to detect defects in metals and composites based on the heat generated between contacting asperities when subjected to vibrational excitation [12–14]. When a discontinuity such as crack or delamination is present in a specimen, the energy dissipated at this discontinuity manifests as heat which flows towards the surface and can be detected using an infrared camera. In vibrothermography, the specimen vibrates at its natural resonance frequencies and the component deformation profile takes shape of the corresponding resonant mode. For efficient defect detection, the vibration amplitude near the defect region should be non zero. Hence for a given resonant mode shape, any defects that are present at the nodal positions cannot be detected. It is possible to measure the resonant mode shape and in turn the vibration coverage

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by applying a layer of viscoelastic polymer adhesive to the specimen surface prior to excitation [15]. Due to the dynamic strain continuity between substrate and adhesive, the polymer material dissipates heat according to the resonant mode shape of the underlying substrate. The heat intensity dissipated by the adhesive depends on the loss modulus of the material, along with the dynamic strain and vibration frequency. Therefore, by calculating the heat intensity and dynamic strain from the measured surface temperature and vibration respectively, the loss modulus of polymer adhesive can be estimated.

Polymers are viscoelastic in nature. These materials exhibit characteristics of both elastic and viscous materials and therefore are absorptive. When subjected to mechanical loading, relaxation effects in the material cause a lag between deformation and load. The Young's modulus of viscoelastic materials is a complex number with real and imaginary components of the modulus called *storage* and *loss* modulus respectively. This complex modulus is often measured as a function of frequency or temperature using dynamic mechanical analysis (DMA) [16]. In DMA, oscillatory loads are applied to the specimen at a predefined set of frequencies at various temperatures and the deformation is measured from which the modulus is calculated. The principle of Time temperature superposition (TTSP) can be used to extend the results of the experiment to higher or lower frequencies than those measured [17]. When a viscoelastic material is subjected to cyclical load, its absorptive nature causes hysteresis because of which energy is lost during the deformation process. This results in net heat energy dissipation at the end of each loading cycle. If the loading is uniaxial, the heat intensity dissipated (g) is

$$g = \frac{1}{\tau} \int_0^\tau \text{Re} \left(\frac{1}{2} \sigma \dot{\epsilon}^* dt \right) \quad (1)$$

where, σ and ϵ are the stress and strain components along the loading direction and τ is the time period of cyclic loading. The time derivative of strain can be written as $\dot{\epsilon} = j2\pi f \epsilon$ where f is the frequency of loading in Hz. Writing the stress in terms of strain, we can rewrite this equation as

$$g = \frac{1}{\tau} \int_0^\tau \text{Re} \left(\frac{1}{2} (E' + jE'') \epsilon (j2\pi f \epsilon)^* dt \right) \quad (2)$$

where $E = E' + jE''$ is the complex Young's modulus. This integral evaluates to

$$g = \pi f E'' \epsilon_0^2 \quad (3)$$

where ϵ_0 is the strain amplitude [18].

In order to measure the loss modulus using vibrothermography, a metallic substrate is coated with a layer of the viscoelastic polymer and subjected to mechanical vibrations using piezoelectric actuator. If there is no crack present in the substrate, it can be assumed that any rise in the surface temperature of the coating is generated by the absorption in the polymer alone and not by the substrate. Because of displacement continuity between substrate and the polymer, the dynamic vibrational strain ϵ on the entire polymer surface can be calculated based on the displacement measurement at a single point and the known resonant mode shape. Since the vibration frequency f is also a known quantity, the loss modulus E'' of the polymer can be calculated from Eq. (3) if the thermal power g is estimated from the measured surface temperature rise in the polymer.

2. Thermal power estimation

The temperature rise in the polymer adhesive coating can be defined as a boundary value heat transfer problem. Mathematically, this problem can be approximated as a case of one dimensional heat conduction with plane heat sources uniformly distributed across the coating thickness. The heat intensity of these plane heat sources is the average thermal power dissipated by the coating. Our objective is to solve this boundary value problem to calculate the thermal power

required to generate the measured temperature rise in the coating surface. We first solve the boundary value problem for unit source heat intensities and fit this analytical solution to the measured coating surface temperature rise to determine the exact value of thermal power. The interface between the adhesive and the metal substrate is modeled as a constant temperature heat sink (Dirichlet boundary condition, $T = 0$) and the other surface of the polymer adhesive is modeled as an insulating surface (Neumann boundary condition, $\frac{\partial T}{\partial x} = 0$). We also assume zero initial conditions since the coating starts to heat only after the vibration begins. The boundary value problem to solve is therefore:

$$\alpha \frac{\partial^2 T(x, t)}{\partial x^2} - \frac{1}{\rho c} g = \frac{\partial T(x, t)}{\partial t} \quad (4)$$

$$\frac{\partial T}{\partial x} = 0 \text{ at } x = 0 \quad (5)$$

$$T = 0 \text{ at } x = L \quad (6)$$

$$T = 0 \text{ at } t = 0 \quad (7)$$

where, T is the temperature rise in the coating ($^{\circ}\text{C}$), g is the thermal power (heat source intensity per unit volume, $\frac{\text{W}}{\text{m}^3}$), L is the coating thickness (m), x is the coordinate along the direction of heat propagation ($x = 0$ at the coating substrate interface, and $x = L$ at the coating surface), α is the thermal diffusivity ($\frac{\text{m}^2}{\text{s}}$), the product term ρc is the volumetric heat capacity of the coating, ($\frac{\text{J}}{\text{m}^3 \text{K}}$) and t is time (s).

The Green's function to calculate temperature field generated by a thin planar volumetric heat source at $x = x'$ which delivers an energy pulse at $t = \tau$ for the above boundary conditions is given as [19]:

$$G_{X21}(x, t|x', \tau) \approx \begin{cases} (4\pi\alpha(t-\tau))^{-\frac{1}{2}} \left[e^{\left[\frac{-(x-x')^2}{4\alpha(t-\tau)} \right]} + e^{\left[\frac{-(x+x')^2}{4\alpha(t-\tau)} \right]} - e^{\left[\frac{-(2L-x-x')^2}{4\alpha(t-\tau)} \right]} \right] & \text{if } \frac{\alpha(t-\tau)}{L^2} < 0.022 \\ \frac{2}{L} \sum_{m=1}^{\infty} e^{\left[\frac{\beta_m^2 \alpha(t-\tau)}{L^2} \right]} \cos\left(\beta_m \frac{x}{L}\right) \cos\left(\beta_m \frac{x'}{L}\right) & \text{if } \frac{\alpha(t-\tau)}{L^2} > 0.022 \end{cases} \quad (8)$$

where $\beta_m = \pi(m - 0.5)$. The solution to the boundary value problem in Eqs. (4) through (7) is given by the convolution of the Green's function with the temporal and spatial distribution of the heat source. The convolution is calculated for unit intensity heat sources uniformly distributed across the entire coating thickness.

In vibrothermography, the heat dissipation in the coating begins at the onset of vibration ($t = t_0$) and ends at the end of the vibration ($t = t_1$). The heat source distribution is a rectangular function in both time and space whose limits are $\tau = [t_0, t_1]$ and $x' = [0, L]$ respectively:

$$g(x', \tau) = \begin{cases} 1.0 & t_0 \leq \tau \leq t_1, 0 \leq x' \leq L \\ 0 & \text{otherwise} \end{cases} \quad (9)$$

The convolution between Green's function and the above unit intensity source distribution is:

$$T(x, t) = \begin{cases} \int_{t_0}^t \int_0^L G_{X21}(x, t|x', \tau) dx' d\tau & t_0 < t \leq t_1 \\ \int_{t_0}^{t_1} \int_0^L G_{X21}(x, t|x', \tau) dx' d\tau & t > t_1 \end{cases} \quad (10)$$

If we evaluate the spatial integral in Eq. (10), it reduces to:

$$T(x, t) = \begin{cases} \int_{t_0}^t G_0(x, t, \tau) d\tau & t_0 < t \leq t_1 \\ \int_{t_0}^{t_1} G_0(x, t, \tau) d\tau & t > t_1 \end{cases} \quad (11)$$

Table 1
Manufacturer specifications of vibration damping adhesives.

Sl.No	Adhesive	Thickness (mm)	Density (kg/m ³)	Color
1	VHB4914	0.25	800	White
2	VHB4905	0.5	960	Clear

where G_0 is defined as:

$$G_0(x, t, \tau) = \frac{2}{15\pi\rho c} \left[5 \exp\left(\frac{4(t-\tau)\pi^2\alpha}{L^2}\right) \sin\left(\frac{3(L-x)\pi}{2L}\right) + 5 \exp\left(\frac{4(t-\tau)\pi^2\alpha}{L^2}\right) \sin\left(\frac{3(L+x)\pi}{2L}\right) + 15 \exp\left(\frac{6(t-\tau)\pi^2\alpha}{L^2}\right) \sin\left(\frac{(L-x)\pi}{2L}\right) + 15 \exp\left(\frac{6(t-\tau)\pi^2\alpha}{L^2}\right) \sin\left(\frac{(L+x)\pi}{2L}\right) + 3 \sin\left(\frac{5(L-x)\pi}{2L}\right) + 3 \sin\left(\frac{5(L+x)\pi}{2L}\right) \right] \exp\left(-\frac{25(t-\tau)\pi^2\alpha}{4L^2}\right) \quad (12)$$

Since a closed form solution does not exist for the above temporal integral, it has to be evaluated numerically to obtain the coating temperature as a function of x and t . Computing this numerical integral for $x = L$ gives the coating surface temperature for unit heat intensity. The unit intensity temperature rise is fitted to the experimentally measured surface temperature and the resulting fit coefficient gives the actual dissipated heat intensity g of the coating.

The next step in estimating the loss modulus is calculation of dynamic strain. Vibration amplitude (displacement) is measured at a single point on the surface of the coating using a laser vibrometer. This vibration measurement is used to calculate the dynamic strain on the entire surface using the known resonant mode shape of the substrate [20]. The resonant mode shapes can be computed numerically using a finite element analysis software such as CalculiX [21]. Using the measured thermal power g , measured vibrational strain ϵ_0 , and known excitation frequency f , loss modulus of the coating can be calculated from Eq. (3) as:

$$E'' = \frac{g}{\pi\epsilon_0^2 f} \quad (13)$$

3. Experiment

In order to measure the loss modulus, vibrothermography experiments were performed on two different polymer adhesives VHB3914 and VHB3095 (manufactured by 3M Company) at several frequencies in the range of 12 kHz and 30 kHz. A 228 mm × 25 mm × 6 mm rectangular Ti-6Al-4V alloy bar without any cracks was used as the substrate. Table 1 lists the nominal properties of these adhesives as specified by the manufacturer.

The first set of experiments were performed to validate the numerically evaluated resonance frequencies of the substrate and their corresponding mode shapes. To measure the mode shapes experimentally, the substrate was subjected to repeated swept frequency excitations while a laser vibrometer scanned across the vibrating substrate measuring the displacement on a finely spaced grid of points along the entire coating surface. Resonance frequencies were identified from the Fourier Transform of the vibration spectrum.

A second set of vibrothermography experiments were performed during which the coated substrate was subjected to tone burst excitations at several transverse flexural resonance frequencies of the substrate. For each excitation, the surface temperature rise on the entire coating surface was recorded using an infrared camera, along with the vibration amplitude at a single point on the coating.

Fig. 1 shows the set up used for performing vibrothermography experiments. A broadband piezoelectric transducer was used to generate vibrations in the substrate for a duration of 1 second [22].

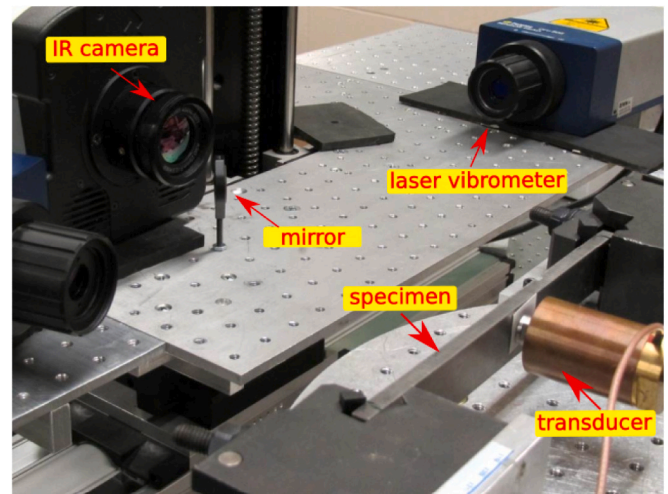


Fig. 1. Experimental setup for measuring loss modulus of a viscoelastic adhesive using vibrothermography.

A calibrated FLIR SC6000 infrared camera was used to record the thermal images of the coating during the tone burst excitations. The camera measured 150 thermal images at a frame rate of 90 fps. The background thermal image was subtracted from each thermal image obtained during and after the excitation so that each processed image contains the temperature rise relative to the ambient environment.

In order to measure the baseline loss modulus of the polymer adhesive, dynamic mechanical analysis measurements were performed using a TA Q800 DMA instrument. To measure the modulus at the same frequency range as that of vibrothermography, measurements were performed at frequency range of 1 Hz to 100 Hz at temperatures of -30°C to 30°C with 10°C increments. Then the principle of time temperature superposition (TTSP) was applied at a reference temperature of 20°C to generate master curves for loss modulus as a function of frequency.

4. Results and discussion

Fig. 2(a) shows the measured surface heating in the polymer adhesive when excited at a resonance frequency. The bright red regions indicate high temperature rise and the dark black regions indicate negligible temperature rise. Fig. 2(b) shows the vibration amplitude across the coating at the same resonance frequency experimentally evaluated using scanning laser vibrometer measurements. The bright red bands in this image indicate regions with high vibrational amplitude and the dark blue bands indicate regions with almost zero vibrational amplitude. Comparing these two images, it can be observed that the coating generated high heat in the regions where vibration amplitude was high and almost no heat in the regions where the vibration amplitude was low. This is consistent with our assumption that the heat generation in the adhesive depends on the vibrational strain in the substrate.

The thermal images were used to estimate the thermal power according to the process developed in section “Thermal power estimation”. Fig. 3 shows the observed temperature rise as a function of time at a single point on the coating surface and the analytical solution fitted to measured coating temperature. The shape of the measured temperature rise and the cooling curves correlates well with that of analytical solution.

Finally the loss modulus was calculated from the estimated thermal power and the measured dynamic strain using Eq. (13) for all the resonance frequencies. Table 2 compares the loss modulus estimated from vibrothermography against DMA measurements. The average error between the DMA and the vibrothermography measurements was 8% and 7% for VHB4914 and VHB4905 coatings respectively. Fig. 4

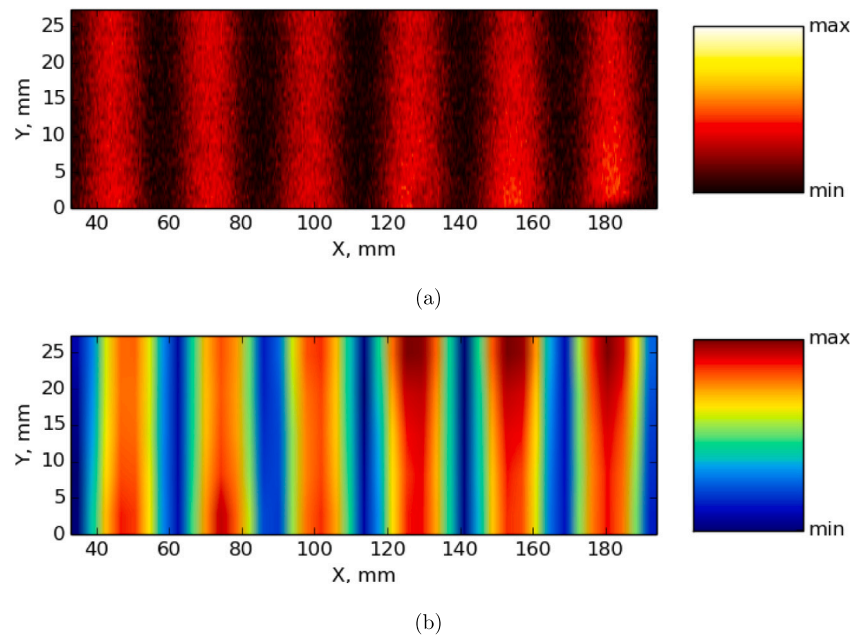


Fig. 2. Comparison of (a) measured surface temperature rise and (b) resonant mode shape.

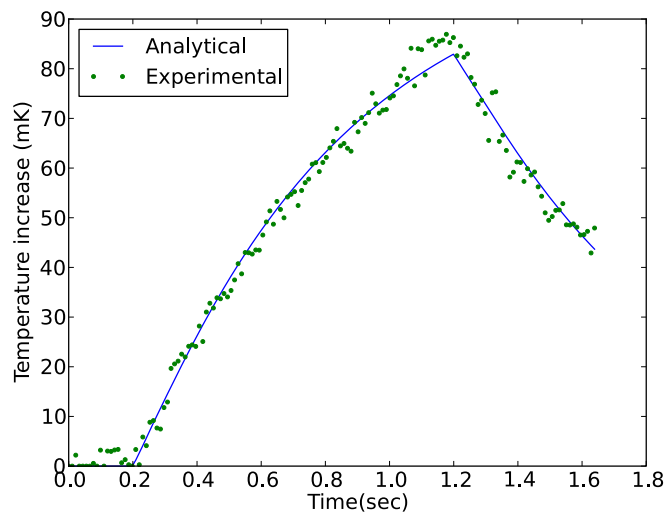


Fig. 3. Measured surface temperature (green) and analytical solution (blue) at a point on the coating surface.

Table 2

Loss moduli measurement comparison of VHB4914 and VHB4905.

Adhesive	Frequency (Hz)	Loss modulus, thermal (MPa)	Loss modulus, DMA (MPa)
VHB4914	12380	81.5 \pm 0.82	92.2
	16590	103.8 \pm 3.26	98.6
	18350	94.2 \pm 1.81	100.9
	20880	100.6 \pm 1.67	104.1
	30490	101.4 \pm 1.13	113.4
VHB4905	12340	60.7 \pm 1.14	52.3
	18140	57.1 \pm 0.74	57.5
	20710	62.3 \pm 0.48	59.2

shows a plot of DMA measurement of loss modulus after applying TTSP along with the results from thermal method. It should be noted that since the relation between loss modulus and the vibrational strain is quadratic, any error in strain measurement affects the loss modulus data significantly. Also, when the coating is not an ideal black body,

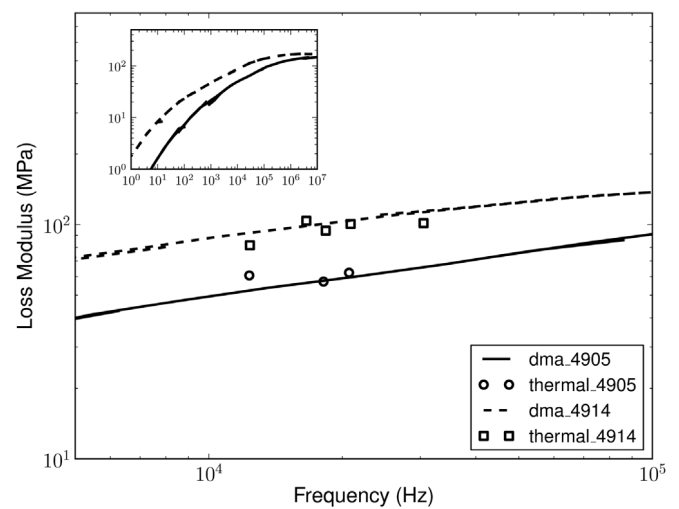


Fig. 4. Comparison of loss modulus measured from vibrothermography and DMA measurement.

the calculated thermal power may not match its true value. The DMA measurement depends on several external factors such as pre-load, oscillation amplitude etc. and is predicated on the assumptions behind TTSP. Despite these possible sources of measurement and calculation errors, the estimated values of loss moduli correlate well to the conventional loss modulus measurement process.

5. Conclusions

A method was introduced to measure the loss modulus of a polymer adhesive in the range of 12–30 kHz based on vibrothermography nondestructive evaluation. The measured loss modulus values were comparable with that of the conventional DMA based measurement. The proposed method is quicker than conventional DMA and still gives comparable loss modulus values. One disadvantage of this method is that for the coating to generate detectable amount of heat, sufficiently high vibrational amplitude has to be applied to the substrate. This restricts the measurement frequencies only to the resonant frequencies of

the substrate where it is possible to achieve high amplitude vibrations. Also, polymers with very low loss modulus may not generate detectable heat even at substantially high vibration amplitudes.

CRedit authorship contribution statement

Jyani S. Vaddi: Investigation, Formal analysis, Visualization, Writing - original draft. **Stephen D. Holland:** Funding acquisition, Project administration, Conceptualization, Supervision, Writing - review & editing. **Michael R. Kessler:** Resources, Supervision, Validation, Writing - review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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