



# Multipoint cure monitoring of CFRP laminates using a flexible matrix sensor

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## ABSTRACT

Since a carbon fiber reinforced plastic (CFRP) structure is complicated in the adoption of integral molding, local molding faults such as under curing and dry spots are liable to occur. To solve this problem, the distribution of the degree of curing for the entire composite structure must be measured. In the present study, we propose a patch-type flexible matrix sensor based on permittivity measurements. Multiple electrodes and wirings are readily fabricated simultaneously using a photolithographic process. Moreover, the sensor has only  $m + n$  wirings for  $m \times n$  sensors, and is thereby suitable for multipoint cure monitoring. We also constructed a method for estimating the degree of curing considering the effect of frequency dependence of the permittivity of resin and viscosity variation due to temperature change. Experiments of multipoint cure monitoring are carried out using a CFRP plate and an actual aircraft structure. As a result, we confirmed the effectiveness of this method by comparing with results using a conventional differential scanning calorimeter.

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## 1. Introduction

The integral molding technique has been developed in recent years to save weight and improve reliability by reducing the number of parts in a system. Since the structure molded by integral molding is complicated, local molding faults such as under curing and overheating are liable to occur. To overcome this problem, a measurement of the distribution of cure-degree throughout the entire structure is highly sought after, since it enables heat to be applied locally to an area with delayed curing. This leads to a reduction in curing faults and residual strain and a shorter molding time [1,2]. Even when the molding condition is proper, defective products may occur owing to deterioration of the resin itself [3]. Therefore it is necessary to measure the distribution of cure-degree in terms of not only determining the molding condition but also ensuring quality.

For measurements of the cure index, the use of dielectric sensors [4], optical fiber sensors [5–6] and piezoelectric sensors [7] have been proposed. These methods require the sensors to be embedded in the mold; this would possibly affect the mechanical properties of the structure [8,9]. Moreover, these sensors are point-sensors. Consequently, as a number of sensors are required, complicated wiring and an expensive measurement device to

check the distribution of cure-degree of the whole structure are also required. Therefore, these methods increase the molding cost and are not suitable for monitoring the distribution of the cure index.

Sensors embedded in a structure during molding are not used during service and increase the structure's weight. Thus, there are problems in applying these methods to practical sensing systems. Although a distributional measurement using a fiber Bragg grating (FBG) [10] has been proposed, the measurement of the degree of curing using an FBG is limited to along the fiber. Furthermore, the measurement method is based on a strain measurement and does not calculate the degree of curing directly. Thus, a new monitoring method that measures the distribution of cure-degree of a complicated shape at low cost and uses sensors that can be utilized again after measuring the degree of curing is required.

In the present study, we propose a flexible matrix sensor in which multiple sensors and wirings are readily fabricated simultaneously by photolithography for measurements of cure-degree at multiple points. Since the wirings form a grid of rows and columns, the sensor requires only  $m + n$  wirings for  $m \times n$  sensors and thereby is suitable for the multiple-point measurement. The thickness of the sensor is 85  $\mu\text{m}$  and is sufficiently flexible. Thus, the sensor can be applied to a complicated shape by attaching to the surface of a carbon fiber reinforced plastic (CFRP) molding and the mechanical properties of the molding are hardly affected.

In a previous study, the Todoroki et al. [11] proposed a system that detects internal damage from the change in electrical resis-

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tance of the CFRP itself. After cure monitoring, the flexible matrix sensor can be used as electrodes for an impressed voltage and applied to monitoring the health of the structure. Therefore, the sensor we proposed is not removed after curing. It remains attached to the structures during its service period. The proposed flexible matrix sensor can be used from the start of manufacturing to the end of the product's life and is called a "cradle-to-grave monitoring sensor", an novel sensor for the total life monitoring of a structure.

When the degree of curing of CFRP is measured using a conventional dielectric sensor, the carbon fibers in the CFRP short-circuit the sensor and become a resistor or capacitor. Therefore, the flow of carbon fibers leads to a change of capacitance during molding. Thus it is impossible to monitor the degree of curing of CFRP using a conventional dielectric sensor.

A conventional dielectric sensor measures capacitance or dissipation factor  $D$ . The capacitance is affected by changes of distance and the area of electrodes resulting from curing shrinkage. The dissipation factor  $D$  is also affected by the change of electrical resistance from the flow of carbon fibers during curing [12,13]. Moreover, as most cure estimation methods proposed before are based on empirical equations between a dielectric sensor and differential scanning calorimetry results, several experiments are required for estimating the degree of curing. Therefore conventional dielectric sensors cannot accurately measure the degree of curing of CFRP. But in the proposed method, capacitance is measured on several frequencies and uses the frequency dependence of capacitance. Because the principle is based not on the electrical effect of capacitance or electrical resistance, but on the relationship between permittivity and rheology, such as relaxation time, this method is not affected by sensor deformation or the flow of carbon fibers.

In the present study, we propose a new method for cure monitoring using the frequency dependence of permittivity in a dielectric resin such as epoxy and the viscosity variation due to temperature changes. Since the method is not based on empirical equations but on rheological equations representing the relationship between permittivity and viscosity, the viscosity can be obtained without pre-experiments. In addition, the method uses the frequency dependence of permittivity of the resin and thus monitoring is not affected by the absolute values of the electrical resistance (or carbon fiber flow during molding) and capacitance (or the configurations of electrodes). Since the viscosity changes depend only on the initial temperature before curing, the effects of the temperature on the viscosity can be obtained at this stage. As a consequence, the method estimates the degree of curing neglecting the influence of the temperature change during molding.

To examine the validity of the proposed method, measurements of the distribution of cure-degree are carried out using CFRP plates made from prepreg during molding with the flexible matrix sensor. As practical applications, we apply the proposed method to measuring the distribution of cure-degree of aircraft structures that have curved shapes and thus undergo uneven heating, and the applicability is experimentally investigated by comparing with differential scanning calorimetry results.

## 2. The principle of cure monitoring

The degree of curing is estimated using the relationship between viscosity and the frequency dependence of a polar polymer such as epoxy resin. Since viscosity estimation via dielectrometry is not based on empirical equations but on resin rheological equations, the viscosity of resin can be estimated without pre-experiments.

Since the rotation of the electric dipole during orientational polarization is delayed owing to viscous resistance, the polar polymer shows the frequency dependence of permittivity caused by dielectric relaxation [14–16]. In general, complex permittivity  $\varepsilon^*$  is expressed by Eq. (1) using the relaxation time  $\tau$  and alternating current frequency  $\omega$

$$\varepsilon^* = \varepsilon_\infty + \Delta\varepsilon/(1 + i\omega\tau) \quad (1)$$

where  $\varepsilon_\infty$  and  $\Delta\varepsilon$  are the permittivity at the ultimate high frequency and relaxation strength, respectively. The real part  $\varepsilon'$  of the complex permittivity  $\varepsilon^*$  is given by

$$\varepsilon' = \varepsilon_\infty + \Delta\varepsilon/(1 + \omega^2\tau^2) \quad (2)$$

At high frequency, Eq. (2) can be approximated as Eq. (3) for  $\omega^2\tau^2 \gg 1$

$$\varepsilon' \approx \varepsilon_\infty + \Delta\varepsilon/(\omega^2\tau^2) \quad (3)$$

By means of a Taylor series expansion around  $\omega_0$ , Eq. (3) can be rewritten as

$$\varepsilon' \approx \varepsilon_\infty + \frac{\Delta\varepsilon}{\omega_0^2\tau^2} - \frac{2\Delta\varepsilon}{\omega_0^3\tau^2}\Delta\omega = b_0 + b_1\Delta\omega \quad (4)$$

where  $\Delta\omega$  represents a minute change in frequency. In general, the alternating current frequency  $\omega$  is from several kHz to several GHz. If  $\Delta\omega$  is up to several kHz, the approximate accuracy of Eq. (4) is good. Using the constant  $m$  from Eq. (4)

$$\tau = m/\sqrt{-b_1} \quad (5)$$

where  $m$  is  $(E\sqrt{2\Delta\varepsilon})/\sqrt{\omega_0^3}$ . According to the Rouse principle [17], (a) the relaxation spectrum is proportional to  $\rho RT$  and (b) the relaxation time is approximately proportional to  $\xi/T$ ; where  $\rho$ ,  $R$  and  $\xi$  are the density at absolute temperature  $T$ , the general gas constant and the coefficient of friction per monomeric substance. When the polymer changes from a density  $\rho_0$  and temperature  $T_0$  to a density  $\rho$  and temperature  $T$ , the whole spectrum becomes  $(T\rho/T_0\rho_0)$  times that of the former condition according to principle (a). On the other hand, the relaxation time becomes  $a_T$  times that for  $T_0$  according to principle (b). Thus, the relaxation time  $\tau$  and viscosity  $\mu$  are respectively given by

$$\mu_T = \frac{T\rho \cdot a_T}{T_0\rho_0} \mu_{T_0} \quad (6)$$

$$\tau_T = a_T \tau_{T_0} \quad (7)$$

As the fractional temperature and density variations are infinitesimal compared with the fractional viscosity variation,  $(T\rho/T_0\rho_0)$  can be approximated as 1. Therefore, the following relation is constructed between relaxation time  $\tau$  and viscosity  $\mu$  using constant  $n$

$$\mu = n/\sqrt{-b_1} \quad (8)$$

Here, the viscosity  $\mu$  of the resin of the thermosetting polymer depends on the absolute temperature  $T$  and degree of curing  $\beta$  [18,19], and is represented as

$$\mu = \mu_\infty \exp\left(\frac{U}{RT} + K\beta\right) \quad (9)$$

where  $\mu_\infty$  and  $K$  are material constants. From Eqs. (8) and (9), the index of viscosity  $1/\sqrt{-b_1}$  is given by

$$\frac{1}{\sqrt{-b_1}} = \frac{\mu_\infty}{n} \exp\left(\frac{U}{RT} + K\beta\right) \quad (10)$$

The following equation is obtained to estimate the degree of curing  $\beta$ :

$$\beta = \frac{1}{K} \left[ \ln\left(\frac{1}{\sqrt{-b_1}}\right) - \frac{U}{RT} - \ln\left(\frac{\mu_\infty}{n}\right) \right] \quad (11)$$

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