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# CURE MONITORING OF THERMOSETTING RESIN COMPOSITES BY LACOMTECH DIELECTROMETRY

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**SUMMARY:** The properties of thermosetting resin composites are dependent on the degree of cure and consolidation quality. During the consolidation process of thermosetting resin composites, the viscosity of the resin of composite has a dominant role for uniform and quality products.
In this study, the dissipation factor that is a function of viscosity was measured during the cure process of thermosetting resin composites by the newly developed Lacomtech dielectrometry apparatus and sensors. Using the measured dissipation factor, the relationship between the dissipation factor and degree of cure with respect to environmental temperature was investigated.

**KEYWORDS:** Dielectrometry, Cure Monitoring, Dissipation Factor

## INTRODUCTION

The cure process of thermosetting resin matrix fiber composites accompanies not only chemical reaction in a multiphase but also physical movement of heat and mass [1-3], which is dependent on resin viscosity. The chemical reactions of thermosetting polymeric materials are exothermic by the cross link between monomers with complex tendencies. Since the conductivities of polymeric materials are usually lower than those of metallic materials, deformation and distortion in composite products after curing occur due to temperature gradient. Therefore, the on-line measurement and process control of the cure process are important for uniform and quality composite products: the control of ambient temperature and pressure through on-line cure monitoring yields reliable composite products efficiently in short time.
Much research has been performed in this area. Kim and Lee investigated the relationship between the dissipation factor and the viscosity of composites, and modeled the degree of cure as function of dissipation factor for on-line cure monitoring [4, 5]. Stephan et al. performed on-line real-time dielectric measurement during compression molding for composites and controlled the process temperature and pressure [6].
In this paper, a new sensor and on-line cure monitoring system using dielectrometry were developed and their performance were compared with the previous systems. Also, the dielectric constant and temperature during the cure of composites were measured.

## MEASUREMENT OF DISSIPATION FACTOR

The Lacomtech dielectrometry consists of an electric circuit and several sensors with the shape of two planar inter-digital type electrodes as shown in Fig. 1. When the sensors are embedded in composite materials and connected to an alternating electric field, the combination of the electrodes and the composites forms a capacitor because the thermosetting resins in composite materials are dielectric. The charge accumulated in the capacitor depends on the mobility of dipoles and ions present in the resin to follow the alternating electric field and varies with the stage of cure.

The degree of cure is related with dipoles and ions movements. They have high mobility when the resin is uncured, but they move little when the resin is fully cured. The degree of the movement can be expressed by the dissipation factor $D$ that represents the ratio of energy loss by movements of dipoles and ions to supplied energy. Using the dielectrometry sensor that is modeled as a parallel circuit of resistance $R_m$ and capacitance $C_m$ as shown in Fig. 2, the dissipation factor of equivalent circuit can be obtained as follows.

$$D = \left|\frac{I_R V_m}{I_C V_m}\right| = \left|\frac{I_R}{I_C}\right| = \left|\frac{Z_C}{Z_R}\right| = \frac{1}{wR_m C_m} \qquad (1)$$

where $w$ is the angular frequency of alternating current.

## EXPERIMENTAL SETUP

In order to increase the sensitivity of signals of dielectrometry sensors, the Lacomtech dielectrometry employs the Wheatstone bridge circuit as shown in Fig. 3. The accuracy of Wheatstone bridge circuit becomes higher as the amplitude of output signal approaches zero. The relationship between the alternating input signal $V_i$ and the output signal $V_o$ is expressed as follows.

$$\frac{V_o}{V_i} = \frac{Z_m}{Z_m + Z_1} - \frac{1}{2} = \frac{Z_m - Z_1}{2(Z_m + Z_1)} \qquad (2)$$

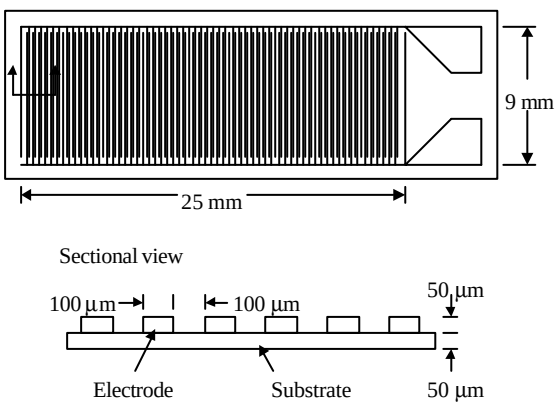



Fig. 1 Shape of the dielectrometric sensor

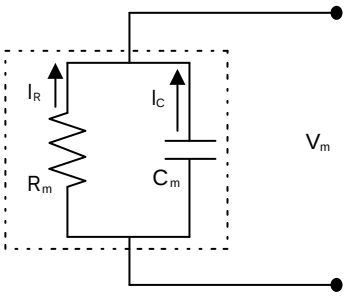



Fig. 2 Equivalent circuit of composite materials.

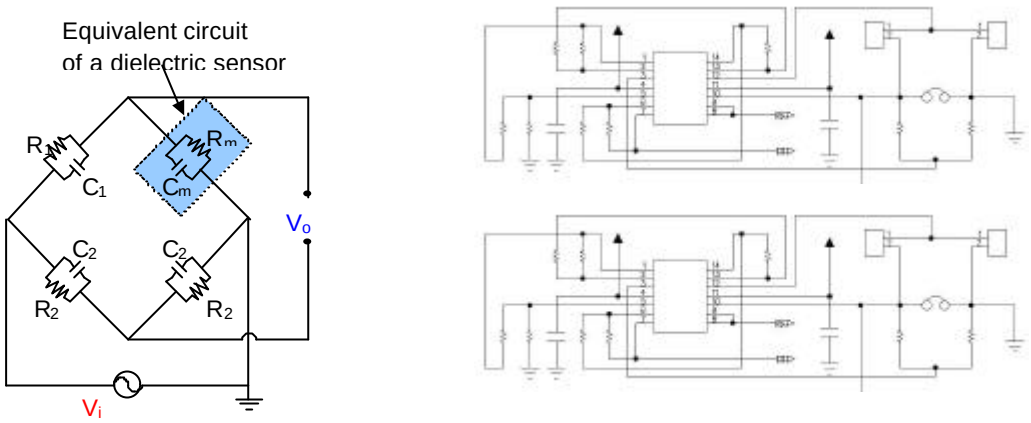



Fig. 3 Electric circuit employing Wheatstone bridge for measuring the dissipation factor.

where $Z_m$ and $Z_1$ represent the impedance of the sensor and the element of circuit, respectively.

$$Z_m = \frac{-Z_1\left(2\frac{V_o}{V_i}+1\right)}{2\frac{V_o}{V_i}-1} \qquad R_m = \frac{|Z_m|^2}{\mathrm{Re}[Z_m]} \qquad C_m = -\frac{\mathrm{Im}[Z_m]}{w|Z_m|^2} \qquad (3)$$

Using Equation (3), the dissipation factor can be obtained from Equation (1). Since $R_m$ and $C_m$ change according to the degree of cure, a variable resistance and a condenser were used for $R_1$ and $C_1$ to make the output signal of Wheatstone bridge circuit zero. In this manner, the dissipation factor was obtained with respect to ambient temperature. Also the process variables such as degree of cure and temperature were related statistically to the dissipation factor.

Since the distance between dielectrometry instrument and composite manufacturing apparatus such autoclave is usually not short, long lead wires for signal transfer are

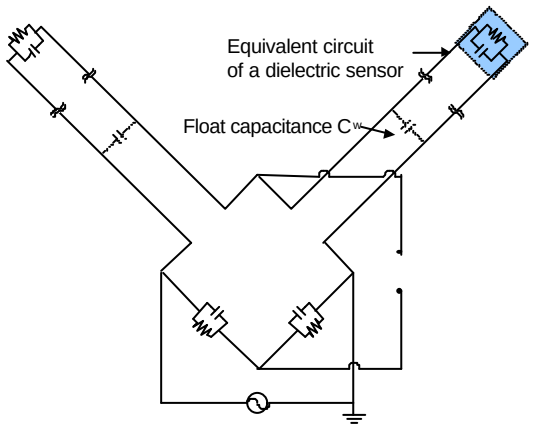



Fig. 4 Electric circuit employing Wheatstone bridge and long lead wire for measuring the dissipation factor.

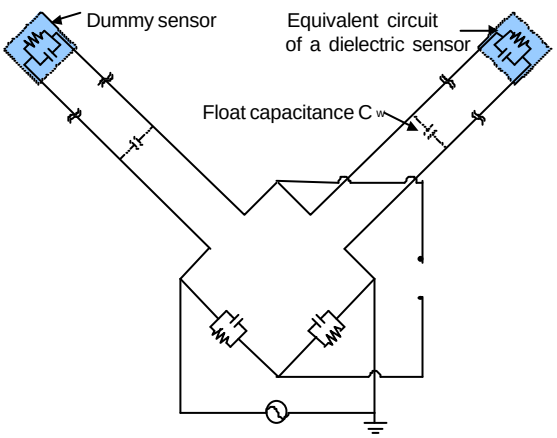



Fig. 5 Electric circuit employing Wheatstone bridge, long lead wire and dummy sensor for measuring the dissipation factor.

necessary as shown in Fig. 4, which induces float capacitance. Also, if the composite manufacturing apparatus uses electricity, the dielectrometry measurement may be affected by environmental electric field that causes error and nonlinearity. In this case, dielectrometry sensors should be shielded from environmental electrical field. If the shielding of lead wires is not easy, the effect of environmental electric field must be reduced. In order to reduce the float capacitance and environmental effect, a dummy sensor was substituted for an element of Wheatstone bridge whose wire length was equal to that of the dielectrometry sensor as shown in Fig. 5. The dummy sensor located close to the dielectrometry sensor will be exposed to the same environments. Then Equation (2) can be rewritten as follows.

$$\begin{aligned}\frac{V_o}{V_i} &= \frac{(Z_m + \Delta Z_m) + Z_w}{\{(Z_m + \Delta Z_m) + Z_w\} + \{(Z_1 + \Delta Z_1) + Z_w\}} - \frac{1}{2} \\ &= \frac{(Z_m - Z_1) + (\Delta Z_m - \Delta Z_1)}{2\{(Z_m + \Delta Z_m) + 2Z_w + (Z_1 + \Delta Z_1)\}}\end{aligned} \tag{4}$$

where $\Delta Z_m$ and $\Delta Z_1$ represent the change of $Z_m$ and $Z_1$ by the environment effect, respectively. Since the impedance $Z_1$ and its change $\Delta Z_1$ of the dummy sensor are almost equal to $Z_m$ and $\Delta Z_m$ of the dielectrometry sensor, respectively, the numerator of Equation (4) is very small, which makes the error and the nonlinearity very small.

In order to reduce the float capacitance of the lead wire, two wires were twisted each other 50 turns per meter. Float capacitance of the twisted wire and the normal wire were 40 and 170 pF/m, respectively, while the sensor capacitance itself was about 100 pF. Since the lead wire was usually 2 ~ 3 m long, the float capacitance of common wire were hundreds pF larger than that of the twisted wire. The float capacitance was reduced by employing the twisted wire. To measure the dielectrometry up to 200°C,

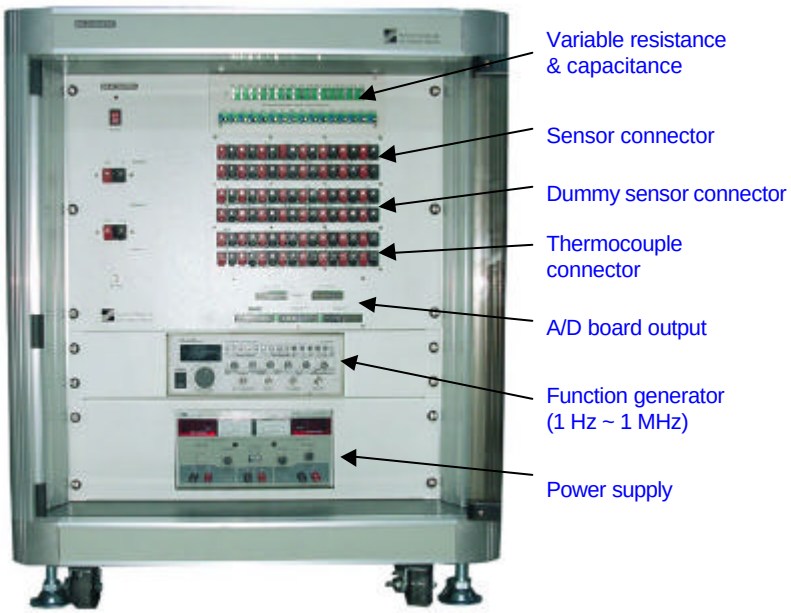



Fig. 6 Newly developed Lacomtech dielectrometry apparatus.

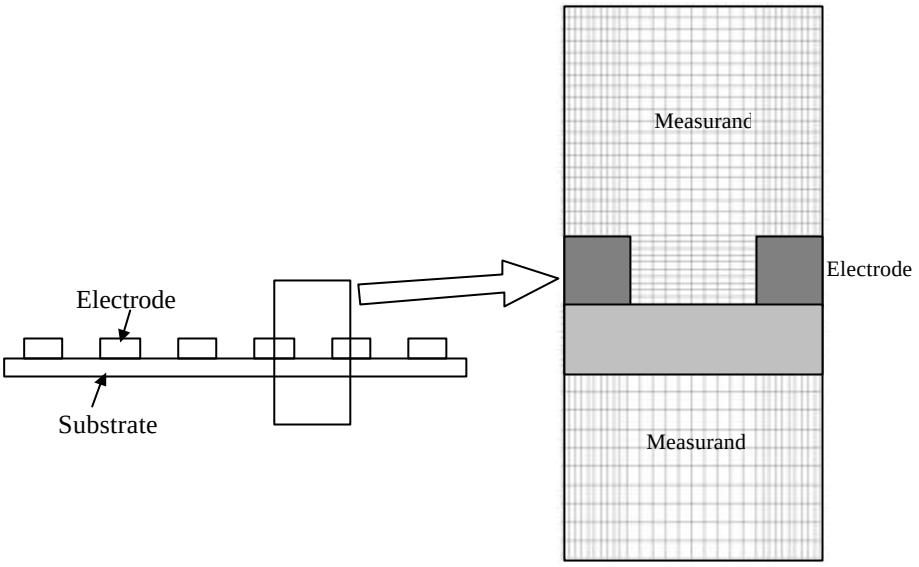



Fig. 7 Finite element model of the electric field for the dielectrometric sensor.

the heat resistant cable covered by Teflon was used. Also the cable was shielded to reduce environmental electric field. Fig. 6 shows the photograph of the developed cure monitoring apparatus that measures temperatures and dielectric constants simultaneously at 16 channels.

## DIELECTROMETRY SENSOR

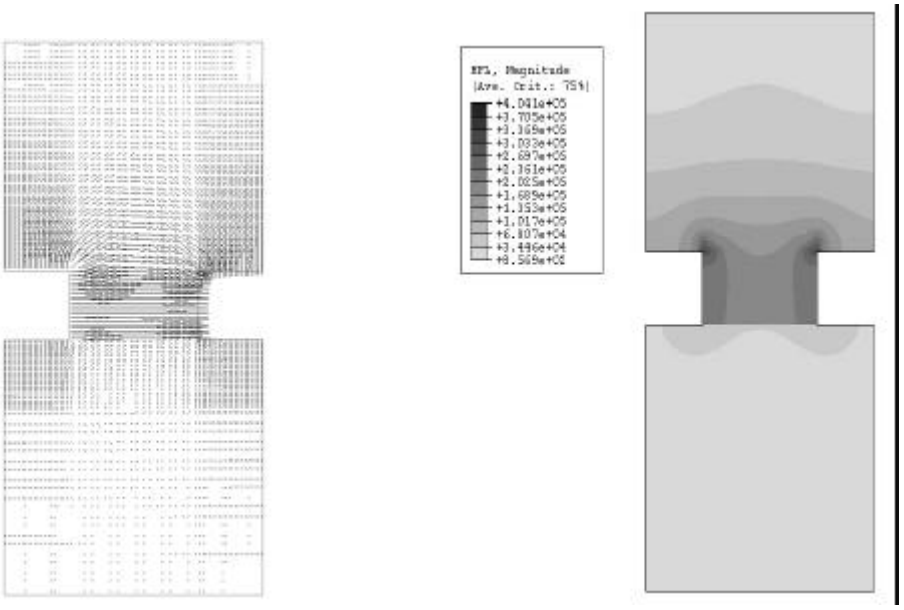
The planar inter-digital dielectrometry sensor used in the experiment was composed of a substrate and two electrodes with opposite polarity on the same plane. Since this sensor shape increases the adjacent area between the electrodes in the unit area of parallel plates, the capacitance of the dielectrometry sensor is increased [1, 7]. The electrodes were fabricated by photolithographic etching of copper film. The substrate was made of polyimide film for high temperature use. The thicknesses of the polyimide base and the copper film were equally 50 μm. As shown in Fig. 1, the sensor area, the width of the electrode and the distance between the electrodes were 25 mm × 9 mm, 100μm and 100μm, respectively.

The electric field governed by Poisson's equation as depicted in Equation (5), was analyzed to obtain the capacitance of the dielectrometry sensor.

$$\nabla \cdot (\varepsilon \nabla V) = -\rho \qquad (5)$$

where $e$, $V$ and $r$ represent the dielectric constant, electric potential and charge density, respectively.

In this study, a commercial finite element analysis package was used for calculation of the sensor capacitance. Due to the symmetry and repetition of electrodes, the sensor was modeled as shown in Fig. 7. The value of charge $Q$ was calculated by multiplying volumes of elements by their charge densities obtained from the finite element analysis. Then the sensor capacitance was obtained from the relation of capacitance $C$



(a) (b)

Fig. 8 Analysis of electric field for the dielectrometric sensor

(a) Electric field line (b) Charge density.

$= Q / V$. Fig. 8 shows the electric field line and the charge density. Fig. 9 shows the sensor capacitance w.r.t. the ratio of dielectric constant ε of the measurand to that of vacuum $\varepsilon_0$. When the measurand was air, the sensor capacitance was 25 pF. When the dielectric constant of measurand was $10\varepsilon_0$, the sensor capacitance was 52 pF.

In order to verify the accuracy of the calculated results, the capacitances of dielectrometry sensor were measured using the air ($1.0\boldsymbol{e}_0$) and the silicon oil of known dielectric constant ($3.15\boldsymbol{e}_0$) as the measurands. The sensor capacitances were 24 pF and 50 pF, respectively; whose errors were less than 4 %.

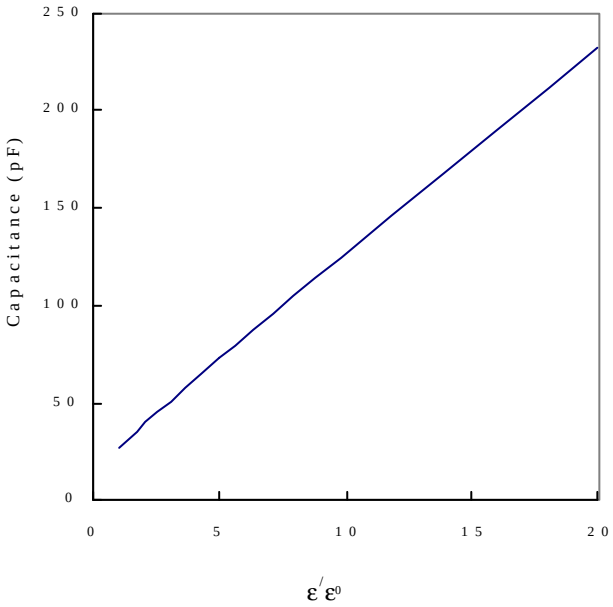



Fig. 9 Capacitance of dielectrometric sensor w.r.t. the dielectric constant of measurand.

## MEASUREMENT

Unidirectional glass fiber epoxy prepreg whose dimensions were 30 mm × 30 mm ×1.2 mm was used to measure the dissipation factor during autoclave vacuum bag degassing molding process. The dielectrometry sensor and thermocouple were inserted between the middle plies of the specimen. The specimen was cured at 125°C under 0.1 MPa. The dissipation factor and temperature of the specimen were measured using both the conventional and the newly

developed apparatuses.

Fig. 10 (a), (b) and (c) show the test results when the circuits of Fig. 3 (case a), Fig. 4 (case b) and Fig. 5 (case c) were used, respectively. The values of $R_2, C_2$ and frequency of sinusoidal wave for the stable set-up of circuit were 10 MΩ, 100 pF and 1 kHz, respectively. In Fig. 10, the dissipation factors of case (a) and case (b) were 0.25 and 0.2 at their start points, respectively. Since case (b) had long lead wires that countervailed the float capacitance of the wire, it had a lower dissipation factor than (a). Case (c) shows noise reduction compared with cases (a) and (b) because a dummy sensor was implemented to the case (c), which reduced the effects by the environmental temperature and electric field as well as the common mode noise. While, in case of (b), $DZ_m$ in equation (4) was not eliminated because $DZ_1$ was nonexistent.

From the experiment, it was found that the new Lacomtech dielectrometry apparatus successfully measured stable signals with less noise. Also it was found that the apparatus was easier to obtain the start and end points of cure reaction and to relate dissipation factors to the environmental temperature and degree of cure.

## CONCLUSION

In this study, the cure monitoring using during the cure of thermoset matrix fiber composites was investigated by the newly developed Lacomtech dielectrometry

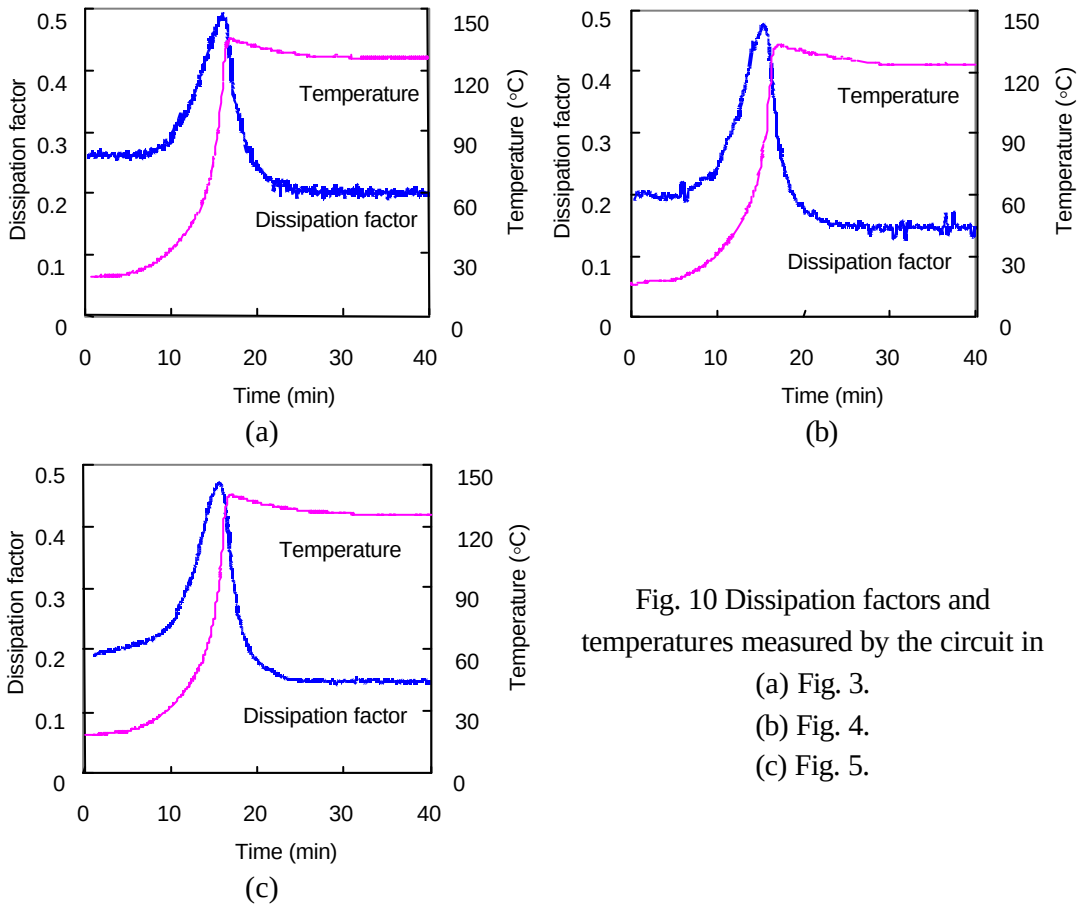



Fig. 10 Dissipation factors and temperatures measured by the circuit in
(a) Fig. 3.
(b) Fig. 4.
(c) Fig. 5.

apparatus employing Wheatstone bridge circuit. Also new dielectrometry sensors incorporating dummy sensor were used in order to improve the accuracy of measurement by reducing the float capacitance, effects of environmental temperature and electric field. From the test, it was found that the stable dissipation factor could be obtained by the newly developed system, which could be used to control process variables such as temperature, pressure and manufacturing time.

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