Cure Monitoring of Composite Carbon/Epoxy through Electrical Impedance Analysis

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ABSTRACT

Composite materials are increasingly used in aeronautic; they offer many benefits such as mechanical strength, mass and consumption reduction. However, their process development needs to be known and controlled, in order to adjust the process parameters and optimize the characteristics of structures made from these materials. This paper is focused on impedance spectroscopy measurement and analysis technique to characterize material's properties. In fact, the composites based on carbon fiber have electrical proprieties; therefore a three-dimensional modeling of the electrical conduction in the material is established by using a distributed allocation of an electrical resistance (R_P) in parallel with a capacitance (C_P). Then, thin electrodes (40 µm thick) are inserted inside the material and a specific impedance measurement bench is developed to perform real-time measurements of R_P and C_P on unidirectional (UD) mono-ply and multi-plies samples. During curing (in an oven) the change in values of both R_P and C_P in different stages of the curing cycle is showed. Then, problems that occur during the curing cycle (layup defect, loss of vacuum) were detected by a large gap of the measured electrical parameters in comparison with the ordinary case. Therefore, by this electrical measurement, we present a way to ensure an automated real-time monitoring of the composite curing process.

1. INTRODUCTION

Carbon Fiber Reinforced Polymers (CFRP) are highly used for the high mechanical performances as regards with their low density. An answer in how optimize their properties can be found in the knowledge and the control of parameters linked to the material itself (voids, percolation network, fiber and resin ratios, etc.) and to the cycle (temperature, pressure, vacuum) during curing.

In order to provide more elements to monitor composite cure process, various works have been undertaken by means of dielectric sensors, optical fiber sensors and piezoelectric sensors, etc. These techniques often required sensors to be embedded, thus could affect the mechanical properties of the structure or their use could be tricky. However, another interesting approach is to investigate the behavior of the material by considering the material itself as a sensor and measuring its electrical properties. Some studies are focused on the measurement of the resistance (R) or the capacitance (C) or better still on the measurement of the impedance (Z).

Ryan, Carolyn and Karim (2002) propose to use the measurement of the capacitance as an indicator to reduce the curing time; but notify the need of further studies to determine a relationship between the change of capacitance and temperature or the degree of cure. Inada and Todoroki (2005) use two electrodes placed on the surface of the material. They consider the material as a parallel RC circuit and perform a frequency analysis of the dielectric permittivity to study the change of the capacitance. They established an estimate of the degree of cure, but they talk about errors that are caused by the decision of the end point of perfect cure.

Shoukai and Chung (1999) burn out the ends of the material to expose the carbon fibers for the purpose of making electrical contacts. The exposed fibers are wrapped by pieces of copper foil, with silver paint between the copper and the fibers. They show that the resistance of the material depends on the direction of currant flow, temperature and pressure. Joung-Man, Sang-Il and Jin-Ho (2005) for their part, perform resistive measurements on a single carbon fiber embedded in an epoxy resin, they were able to assess the residual stresses and temperature during curing.

The previous works of Marguerès, Camps, Viargues and Olivier (2013) have been show that it is possible to monitor the evolution of the behaviour of the material during its cure

cycle by using flexible printed electrodes (inserted inside the material) and an associated acquisition system. They also consider the material as a parallel RC circuit and perform the impedance analysis to provide the online change of the resistance and the capacitance during curing. The evolution over time of the measured electrical parameters (R and C) matches the evolution of the rheological parameters studied by standard methods (obtained on a parallel plate rheometer); more than ten points of agreement were established.

Indeed, the purpose of this paper is to continue previous studies of Marguerès et all (2013). The composite material studied is made from T700/M21 prepregs (pre-impregnated plies) used in aeronautic and space industry. This material has conductive and insulating parts (long carbon fiber and epoxy resin) and its electrical conduction properties depend on the fibers orientation. Firstly, a three-dimensional (3D) model of the electrical conduction is made to describe the material's anisotropy. This model consists of a resistance and a capacitance connected in parallel (R_P and C_P). Thin copper electrodes (40 µm thick) inserted in the material, depending on the fiber orientation, serve as measuring elements. A specific impedance measurement bench has been developed to achieve real-time measurements of R_P and C_P. The sensitivity of this R_P and C_P depending on the different stages of the curing cycle to detect a defect is studied here. All this aims to provide a monitoring and possibly to real-time control the curing cycle to obtain the desired properties of the produced CFRP structures.

2. MODELING OF ELECTRICAL CONDUCTION

The used prepregs are unidirectional and 250 µm thick. The matrix is a M21 epoxy resin. The reinforcement is made of high strength carbon fibers (7 µm diameter). The studied materials are mono-ply samples but also unidirectional laminates (multi-plies with fibers oriented in the same direction). This unidirectional (UD) orientation confers anisotropic electrical properties.

Thus, this material contains a conductor part (fibers) and an insulator part (resin). So it is suitable to perform impedance analysis using a frequency sweep (here from 10 Hz to 1 MHz). The resistive conduction is linked to the conduction through the fibers and the percolation points, and it is predominant at low frequency. The capacitive conduction (through resin and voids) is predominant at high-frequency. To establish a three dimensional model of the electrical conduction inside the composite material, the axes of the electrical conduction are defined as follows:

1. In the fiber plane (intra-ply): Two types of conduction are possible. A longitudinal conduction in the fibers (intra-fibers intra-ply conduction), and a transverse conduction which is perpendicular to the fibers orientation (inter-fibers intra-ply conduction). The longitudinal impedance measurements are delicate, because the low value of the corresponding resistance induces distortions on capacity measurement. That is why, in our model, only the resistive conduction along the fibers is considered (measured at 10 Hz). The intra-fibers conduction corresponds to the current flow in the fibers and through the percolation points. But the inter-fibers conduction is mainly due to the current flow through the percolation points. At high frequency only the inter-fibers capacitive conduction in the resin is considered and measured at 100 kHz. Finally, the longitudinal conduction is modeled as distributed resistances, while transverse conduction as resistances and capacitances in parallel (figure 1).
2. In the thickness plane (inter-plies): The electrical conduction is considered as the same as the inter-fibers conduction (in red in figure 1).

Finally, the complete electrical model equivalent to a unidirectional multi-plies composite material can be considered as a cascaded structure of hexapole nodes.

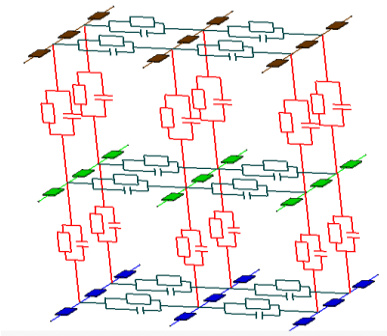


Figure 1. 3D electrical conduction model of the multi-plies composite material.

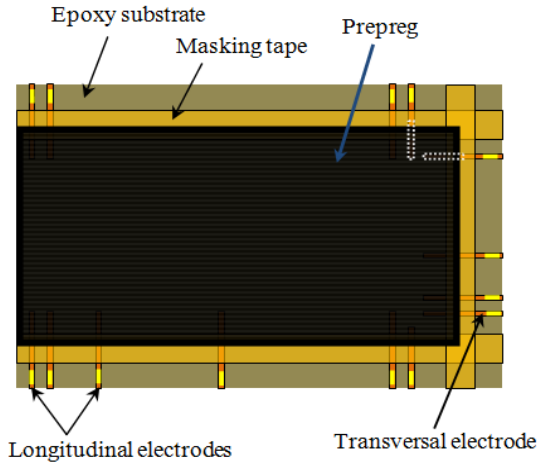
3. EXPERIMENTAL SET-UP

The mono-ply sample is a prepreg placed on an epoxy substrate. The measuring electrodes are inserted between prepreg and substrate (see figure 2.a). The multi-plies samples contain up to 24 plies (10 x 10 cm^2). Before curing, thin flexible electrodes (flexe) are inserted between two consecutive plies in order to reduce interfaces resistances (see figure 2.b). A flexe is a copper tape which is 40 µm-thick, 6mm-wide and 20 cm-long. It is covered with polyimide film (kapton, 35 µm thick) at masking areas (outside the material). A frequency sweep of the sinusoidal current at constant amplitude, combined with the alternating voltage measurement (amplitude and phase), allows establishing frequency evolution of complex electrical impedance (Z) for our electrical model, as shown in the following equation:

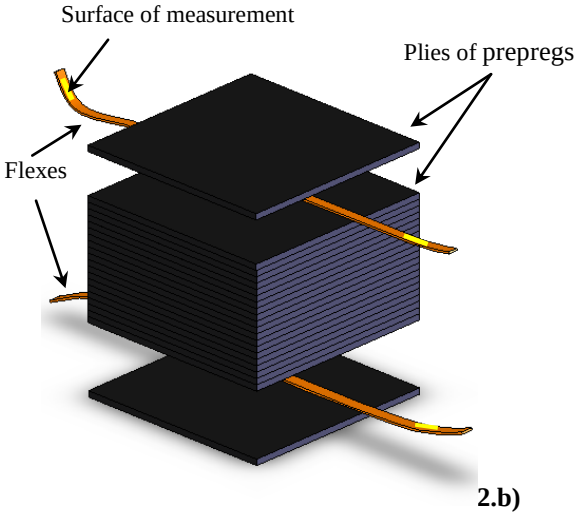
$$Z = \frac{R \times \frac{1}{jC\omega}}{R + \frac{1}{jC\omega}} = \frac{R}{1 + jRC\omega} \quad (1)$$

Where : R is the resistance ; C the capacitance ; f the frequency ; ω the angular velocity ($\omega = 2\pi f$).

From this expression, measurement at low frequency allows determination of resistance, while the capacitance measurement is optimal at high frequency. The developed acquisition bench allows real time measurement of the overall electrical impedance parameters R_P and C_P on samples during their curing process.



2.a)



2.b)

Figure 2. Mono-ply (2.a) and multi-plies (2.b) samples.

4. PRELIMINARY STUDIES

The post-curing measurements on the composite mono-ply and multi-plies samples were used to validate the model and to bring out the values levels of the measured parameters. The fibers resistivity in the longitudinal intra-ply measurement is 15.10^{-5} Ω.m; its determination is difficult because of the presence of significant contacts resistances R_C (1 to 4 Ω) which disrupts measurement. These contacts resistances have random values with a large dispersion (400%) and impose 4-points measurement method. The transverse inter-fibers measurement shows a resistivity about 1.5 Ω.m and the transverse inter-plies measurement shows the higher values of resistances with a resistivity equal to 4.5 Ω.m. In both previous transverse inter-fibers and inter-plies cases, 2 points measurement method is used because contacts resistances effects are negligible.

5. CURE MONITORING

The real-time measurements were achieved during curing, in an oven, using mono-ply samples (for longitudinal intra-fibers and transverse inter-fibers conductions) and multi-plies UD samples (inter-plies conduction).

The longitudinal intra-fibers measurements show sporadic variations of R_P and C_P; this is due to the low resistances values and also mainly the preponderance of the contact resistances between fibers and electrodes (as described above). Both transverse measurements show variations of R_P and C_P correlated to the changes in the material state.

The figure 3 shows the results of transverse inter-plies R_P and C_P measurements during curing. The electrodes are inserted between plies 1 and 2, and plies 23 and 24 (figure 2.b). As expected, the resistance R_P decreases over time from 30 kΩ to few hundred ohms (220 Ω). This is due to the contacts improvements between fibers and electrodes and also to the increasing of the percolation network. The changes in C_P during curing show two peaks; the first corresponds to the point of polymer liquefaction and the second to the gel point.

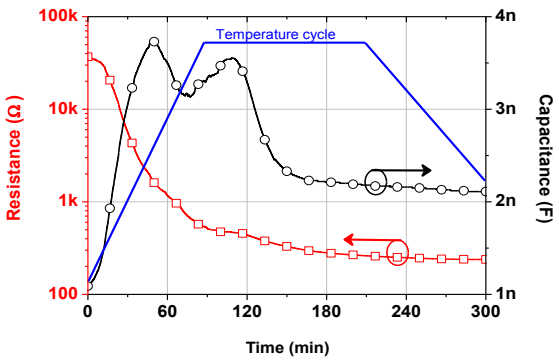
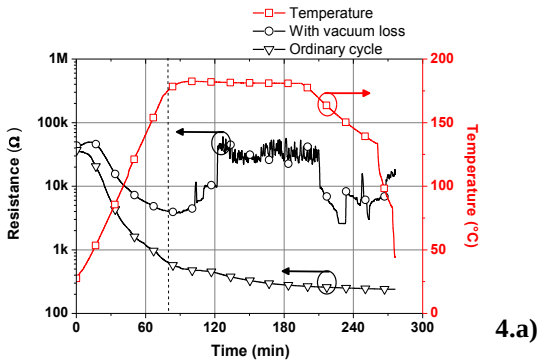


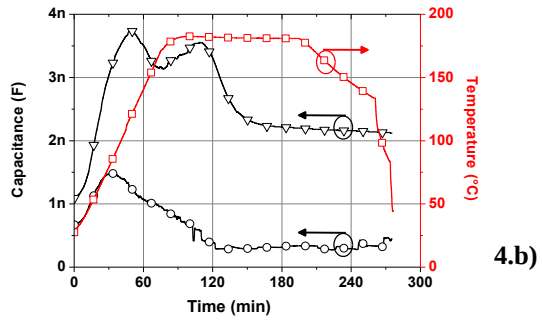
Figure 3. Changes in R_P and C_P during curing.

To prove the advantage of our measurements, we caused vacuum loss when curing. This incident is visible on the values of R_P and C_P (figures 4.a and 4.b). After curing, there is also a large difference between the values obtained in the curing with an incident and those measured under normal curing condition. Thus we have obtained, in the case with an incident, a resistance around 18 kΩ against 220 Ω in ordinary or normal curing.

This loss of vacuum, caused by a bad layup, limits material compaction during liquefaction and it manifests itself by the high value of the resistance (less percolation points) or low value of the capacitance (more polymers between fibers). Therefore, it is possible to make a cure monitoring or to use a standard (benchmark) to determine the quality of a curing cycle.



4.a)



4.b)

Figure 4. Loss of vacuum detection using R_P (4.a) and C_P (4.b) measurement during curing.

6. CONCLUSION AND OUTLOOK

Thanks to a simple and robust instrumentation, with flexible thin electrodes, an electrical impedance spectroscopy was carrying out inside of carbon composite material T700/M21. These electrodes associated with an acquisition system allow tracking material's behaviors during curing in an oven. Over time evolution of the measured electrical parameters R_P and C_P is according to different states of the material. Then, a loss of vacuum was detected by a large gap of these electrical parameters. In fact, in manufacturing, incidents during layup or curing can cause errors in the matrix/fibre or voids volume fractions, or even structural defects (delamination, imperfect ply-drop etc.). Therefore this monitoring can be used to control or to optimize the manufacturing processes of composite materials.

Furthermore, after curing these flexible electrodes facilitate access inside of material and can be used, either for monitoring the health of composite material during the phases of conditioning and service, or to access to nanoparticles that can be added in polymer.

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