PROCESS MONITORING OF THERMOPLASTIC REACTIVE COMPOSITE MOULDING USING DURABLE SENSORS

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ABSTRACT

A dielectric monitoring system has been developed for the real-time sensing of composite manufacturing processes of reactive thermoplastic materials. In addition, optimised durable non-intrusive interdigitated dielectric sensors have been designed and manufactured for the sensing of resin arrival, viscosity changes as well as material transformation and crystallisation. The dielectric sensors receive the appropriate AC stimuli from the monitoring system while the feedback is recorded and processed by the system providing real-time material state-based information (viscosity, degree of reaction etc.). The system and the sensors were used successfully to monitor the infusion of glassfibre with CBT oligomers and their reaction to form PBT at the end of the process.

KEYWORDS: Cure/Process – Monitoring, Liquid Molding Processing, Resins/Materials – Thermoplastics

1. INTRODUCTION

In liquid composite moulding there is a real need for durable non-intrusive sensors that not only inform the production supervisor with resin arrival at some location but also provide as much information as possible of what is happening in the cavity in order to use this information in a real-time control loop and/or for quality control purposes. As the installation of such sensors is not an easy task in real production environment, it is very important that one sensor can monitor:

- Resin arrival
- Viscosity changes
- Degree of cure
- Flow front direction and speed

Previous work [1] has shown that the cure process of thermosetting resins in an autoclave processing environment can be followed in real-time and in situ by the use of the dielectric cure monitoring method. The ultimate aim is to provide intelligent process control of composite manufacturing processes, including Resin Transfer Moulding (RTM). To achieve the process control, the dielectric monitoring response has to be combined with models of thermoset cure [1, 3]. These mathematical models convert the time-temperature profile of cure process environment to the resin viscosity $\eta$ and the glass transition temperature $T_g$ and
reflect the kinetics and the molecular structure formation in the thermoset resin matrix. The importance of this approach is that the dielectric signal can provide reliably and in real-time the important process milestones: the point of resin maximum flow, the gel point and the vitrification time [2].

On the other hand for the detection of resin arrival and the monitoring of viscosity, lower cost systems are possible. In the literature several point sensors with the corresponding hardware based on DC amplifiers have been used for epoxy resins [4, 5, 6, 7, 8, 9, 10], though no commercial system is available.

In order to increase the information provided by the sensors for process control an integrated version of electrical and dielectric sensors has been developed [10] and applied in the process monitoring of thermoset and thermoplastic resins [11]. The development of this integrated flow and cure sensor has been based on the existing platform of the dielectric cure monitoring system VI-DiAMon developed at Inasco Hellas.

At the present paper some new developments on a new DC amplifier and new results on the performance of these sensors for reactive thermoplastic liquid composite moulding will be presented and discussed.

2 DIELECTRIC SENSING

Recent development in dielectric sensing utilizes interdigital electrode sensors and relies on direct measurement of dielectric properties of insulating materials, such as thermoset resin systems, with one side access. The principle relies on the application of a spatially periodic electrical potential to one surface of the Material Under Test (MUT), i.e., thermoset resin or composite material. This type of dielectric sensing takes advantage of the penetration of the electric field applied on one side (as shown in fig.1) and not through the bulk material. Thus the sensing is local and, in most cases, non-intrusive offering the potential to relate the dielectric properties (permittivity, conductivity) to chemical and structural changes in macromolecular systems. The one sided access of the test material offers the potential to eliminate the detrimental effects of conductive fibres or inclusions in the dielectric measurement of (insulating) resin systems.

Optimised durable dielectric sensors have been designed and manufactured [10,11] as shown in fig. 2. The cure sensing grid (fig. 2, left) and the mounted sensor element (fig. 2, right) receive the appropriate AC signal from the monitoring system and the feedback is recorded and post processed by the system providing real-time material state-based information.
The combination of signals produced by varying the electrical excitation frequency provides extensive information about the electrical behaviour of the material and the charged species involved within the electric field (dipoles, ions and interfaces). Since the changes in the electrical properties are usually induced by changes in the physical, chemical, or structural properties of the materials, the dielectrometry measurements provide effective means for indirect, non-intrusive evaluation of vital parameters in a variety of composite manufacturing applications.

The dielectric process monitoring system has been developed specifically to interrogate the dielectric sensors shown above. The system measurement capability has been successfully compared with off-the-self high-quality dielectric analysers in static and dynamic tests. The static tests showed the excellent performance of the system over a wide range of impedances over the frequency range 0.2 Hz to 100 kHz.

![Image](https://example.com/image.png)

**Figure 3.** Dielectric monitoring hardware (left) suitable for interrogating interdigital dielectric sensors. The custom features of the monitoring software (right) allows the real time prediction of material state information from dielectric measurements.

The use of this system for room and high temperature processed resins for everyday production proved its robustness and capabilities to detect the major process milestones: resin arrival, minimum viscosity, gel point and end of cure for thermoset matrices, as shown in Fig. 4 for a typical epoxy system for aerospace applications.
Figure 4. Evolution of the VI-DiAMon dielectric signal (points, left vertical axis), the kinetically estimated degree of cure (thick line, right vertical axis) during processing of a typical aerospace epoxy resin temperature profile (thin line, right vertical axis).

3. ELECTRICAL SENSING

The flow sensor comprises two parallel thin metallic lines (contacts) plated on an appropriate ceramic pad (fig. 5). As long as these two lines are not covered by liquid resin the resistance between these lines is close to infinity (air) whereas resistivity drops to measurable magnitudes when resin covers them. In order to measure this material’s resistivity an in-house very sensitive amplifier has been developed with a resistivity range from $10^7$ up to $3 \times 10^{10}$ Ohms. Hence, based on the signal from the sensor resin arrival and viscosity changes can be detected while measurements are repeatable and can be calibrated for quality control purposes. The hardware can scan the contacts every 0.1 s or more so a very accurate time stamp of the resin arrival at the exact contact spot can be recorded. As these contacts are very small a combination of three or four of them (fig. 5) can provide us with the local speed and direction of the flow front. Furthermore as a thermocouple is also integrated temperature is also recorded and used for the appropriate calculations of viscosity and other material state properties.

Figure 5. Durable flow (electrical) sensors (series of four).
4. RESULTS

In order to assess the robustness and the accuracy of the flow and cure sensors many trials have been executed with variable fibre volume fraction, type of fabrics for the main closed mould liquid moulding process, RTM for a single reactive thermoplastic resin.

![Figure 6. A small RTM/ mould/ sensor device for the accurate monitoring of the exact conditions during thermoplastic injection.](image)

With respect to liquid composite moulding of thermoplastic resins the monitoring of the process evolution of reactive oligomers forming to a thermoplastic matrix can be seen in fig. 7. In this figure it is observed that at three different temperatures (191, 201 and 209°C) there is a distinctive difference in the monitoring signal due to the variance in the evolution of the polymerization and the crystallisation of the polymer as has been already identified by other measurement techniques [10].
Figure 7. Isothermal injection/ fast cooling processing of dried and semi-dried CBT160 @ 191, 201 and 209 °C (semi-dried) and 190°C for fully dried.

Figure 8. Comparison between two isothermal runs of dried CBT160 @ 210 °C with and without glassfibres.
Figure 9. Comparison of three dynamic runs of dried CBT160 and glassfibres from 184 to 215 °C with variable heating rates and starting time.

Figure 10. Electrical resistance (left axis) and temperature (right axis) of the thermoplastic material against processing time (seconds) for an isothermal injection/ fast cooling reactive processing at 200 °C.

4. DISCUSSION

Valuable information of the processing status can be provided by the newly developed integrated flow and cure sensors with the relevant hardware and software. The sensors are
robust, non-intrusive and durable, providing a reliable basis for real-time control. Furthermore, it seems that they can provide valuable information on the processing status of thermosets as well as for thermoplastics. The latter needs further investigation as it shows distinctively different behaviour than thermosets.

This information will be combined with a process simulation software for the model-based control developed in parallel and will provide an intelligent system for the automatic and optimal processing of composite materials.

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