ONLINE PROCESS MONITORING SYSTEMS – BENCHMARK AND TEST STUDY

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ABSTRACT: Despite the ambitions for increasing numbers of Carbon Fiber Reinforced Plastics (CFRP) parts no reductions in quality aspects can be accepted. Therefore new quality inspection methods have been taken into account. Online Process Monitoring (OPM) systems offer the opportunity to monitor process data, compare them with the ideal course of the process and draw conclusions about part quality. In this study, three commercially available OPM systems - Dielectrical Analysis (DEA), Direct Current Resistivity (DCR) and Ultrasonic Technology (US) - are discussed for use in serial production. DEA measures the change in dielectrical properties of the resin, DCR is based on resistivity changes of the resin during processing and US analyses changes in ultrasonic speed of the matrix material. All systems promise to monitor flow front arrival, in-mold viscosity and curing. Furthermore effects of aged resin and different mixing ratios and detection of toughening thermoplastics are investigated in this study. For ideal comparison of the different systems a RTM tool was designed in which the OPM sensors are installed at the same time so that the compared data are simultaneously gathered within the same experiment. Finally, the systems are evaluated with respect to the above mentioned criteria in order to support the choice of a dedicated OPM system depending on the specific case of operation.

KEYWORDS: *Resin Transfer Molding, Online Process Monitoring, Flow Front Tracking, Viscosity Monitoring, Cure Monitoring, Dielectrical Analysis, Resin Resistivity, Ultrasonic Technology*

INTRODUCTION

In aerospace as well as automotive industry an increasing number of CFRP parts has been observed driven by the need for more energy efficient vehicles. Further applications of CFRP are often limited by high costs and long cycle times. OPM systems hold high potentials to meet all these requirements by comparison of monitored process data with the ideal course of the process. Hence, part production and quality management run simultaneously so that cycle time is reduced. In a next step the production process of each part can be adapted to the specific process conditions based on gathered process data thus processing energy is saved by reduction of process tolerances and reduced cycle times. In this study currently commercially available OPM systems are tested in a benchmark study, the Dielectrical Analysis DEA 230 from NETZSCH, Optimold from SYNTHESITES, and US-Plus from PYROSENSOR.

OPM SYSTEMS

Dielectrical Analysis - DEA

In dielectrical measurements a time varying voltage is applied to the inspected material. The established electric field and the arising current depend on the ion conductivity and the dielectrical polarization of the sample. During curing of thermoset resins amount and mobility of free ions and molecules change due to the growing molecular network what can be measured by analyzing amplitude and phase lag of the voltage and current signal. Based on the changes in amplitude and phase lag the complex dielectric constant is determined [1]. In a further step ion conductivity σ can be calculated from the following equation (Eqn.1) in which ε'' is the loss factor (imaginary part of the complex dielectric constant), ε_o is the dielectric constant for vacuum and ω is the angular frequency [2]:

$$\sigma = \varepsilon'' \cdot \omega \cdot \varepsilon_o \tag{1}$$

Reciprocal value of the ion conductivity is the ion viscosity which is directly related to viscosity changes of thermoset resins [1].
As DEA measurements are based on electrical properties of the resin DEA sensors have to be insulated from electrically conductive materials like carbon fibers. Furthermore DEA measurements cannot be used for condensation polymerizations as changes in dielectrical properties of the sample are masked by the high polarity reaction products e. g. water.

Direct Current Resistivity Sensing - DCR

Optimold is based on correlations between resistivity and state of cure which has firstly been investigated for thermoset resins in the 1960s [3]. Resistivity ρ of a polymer is determined by measurement of the potential drop U across the sample and the electric current I applied to the sample. With knowledge of the distance l and the area A perpendicular to potential gradient resistivity ρ can be calculated from Eqn. 2 [3]:

$$\rho = \frac{U \cdot A}{I \cdot l} \tag{2}$$

DCR sensing is also based on electrical properties of the resin system so that the same limitations concerning insulation from carbon fibers and condensation polymerization are valid like already discussed for the DEA.

Ultrasonic Technology - US

US-Plus uses ultrasonic technology which is already well known in composite industry as non-destructive testing technique. Generally it can be divided between transmission and pulse-echo method whereas in this study only the former is relevant. In transmission measurements ultrasonic transducers are placed face to face with each other so that the bulk material in between is inspected. An ultrasonic signal is generated by a piezo-electric transducer (emitter) and coupled into the part. The transmitted longitudinal wave is detected by the second piezo-electric transducer (sensor). By measuring the time of flight of the ultrasonic signal in the inspected material and with knowledge of the part thickness the speed of sound is calculated.

Changes in sound speed correlate with changes in viscosity and cure state of thermoset resins [4]. Furthermore the attenuation of the ultrasonic signal contains information about the presence of flaws like entrapped air (porosity) or delamination in composite materials.

EXPERIMENTAL DETAILS

Test Plan

When OPM systems are used for quality inspection purposes, repeatability necessarily becomes a crucial criterion. Therefore several experiments were conducted at least two times in an identic manner for each resin system. One room temperature curing resin system - MGS® RIM 135 Infusion Resin and MGS® RIMH 1366 hardener both from HEXION - and one hot curing resin system - RTM6 from HEXCEL Composites - were used in this study.
The former was used for investigation of the OPM system's sensitivity to *flow front arrival*, detection of *race tracking* - preferential resin propagation in flow channels formed by the preform and the cavity, effects of varying *mixing ratios* and *porosity* content. The latter was used for *detection of effects of aged mono-component resins* and *thermoplastic particles.* As thermoplastic materials Polysulfone (PSU) films - 125µm in thickness from Solvay - and Polyamid (PA) fleeces - PA 1541 from Spunfab – RTM6 were applied. PSU and PA are soluble in RTM6 to a certain amount.
Flow front arrival is crucial for improved understanding of filling patterns which becomes interesting especially for complex parts. In this study flow front arrival was optically validated by using a transparent mold inserts. Detection of *race tracking* is a special case of detection of the arriving flow front localized to cavity walls. Hence, the use of the different OPM sensors close to cavity edges is proven in the race tracking trial.
Mixing ratio of the matrix material is decisive for the mechanical properties of the final part. Consequently, OPM systems should be capable of detecting deviations from recommended mixing ratios. In this study the amount of hardener was varied by +/-5% in weight resulting in ratios of 100:35 and 100:25 portions of resin to hardener. The recommended mixing ratio of RIM 135 is 100:30 and was used in the reference trials. The race tracking trial was carried out with a ratio of 100:35 in order to reduce the viscosity of the injected resin and thus enhance the race tracking effect.
As well as the correct mixing ratio, low *porosity* is essential for reaching the desired mechanical properties. In this study porosity is provoked by injection of non-degassed resin. Furthermore the mixing ratio was set to 100:25 as air bubbles aim to ascent faster in less viscous fluids.
Aging of mono-component resins leads to dumping of expensive thermoset mono component resin as the minimum viscosity for optimum infiltration cannot be guaranteed when the expiry date is excessed. Sensors for determination of the degree of precrosslinking or for the detection of the effects of aged resin during manufacture would reduce the amount of dumped material. Hence, the infiltration and curing process of RTM6 resin which was stored for 16 days at room temperature - 15 days maximum shelf life at 23°C - was monitored and compared to reference trials with fresh resin.

Test Setup and Experimental Description

A stand-alone RTM plate tool in frame construction was designed for the benchmark study and placed in a furnace for the hot as well as the room temperature curing resin system tests in order to guarantee identic process conditions. The mixed and degassed resin was injected with

constant injection pressure at 3.5 bars by means of a pressure pot which was preheated to 80°C for the RTM6 trials. RIM 135 was injected at room temperature. Before injection the tool was heated to 30°C for the RIM 135 and to 120°C for the RTM6 trials. In case of the RIM 135 experiments the progressing flow front was monitored by a camera placed above a transparent polycarbonate mold insert. Curing temperature in case of the RIM 135 trials was set to 65°C and 170°C for the RTM 6 trials. Tool Mounted Sensors (TMS) were used in case of the DCR and US measurements. The single side DCR sensor was mounted in the bottom plate whereas one US transducer was placed in every mold half. In terms of the DEA insulated disposal sensors (IDEX) were fixed onto the polycarbonate top mold by means of an adhesive tape. Additional to the OPM sensors, pressure sensors - PXM600MU-14BARGV from OMEGA - and thermocouples (Type-K) were placed at the linear in- and outlet of the mold.
The general layup of the preforms was identic for all the experiments and consisted of one insulation glass layer next to the DCR TMS and 12 layers of biaxial carbon NCF resulting in a calculated fiber volume content of 57,64%. In the experiments with thermoplastic layers one PSU film was placed between the preform and the sensors' surfaces as top and bottom layer and one layer of PA fleece was placed between each textile layer, respectively.
For the race tracking trials flow channels were provoked by cutting the preform into three intentionally too small stripes and placing metal inlays between them so that a cavity edge and a small flow channel was formed at each OPM sensor.

Process Data Acquisition

For accurate and sensitive data acquisition in terms of the DEA and the US system correct and adapted systems settings are required. Apart of the general systems setup no experimental specific settings are needed in case of the DCR system.
Due to the frequency dependency of the DEA the measuring frequency should be adapted according to the demanded process parameter. For Epoxy resin systems frequencies from 0.1-10000Hz are suggested depending primarily on the resin viscosity. With decreasing frequency the sampling time for calculation of one data point increases ranging from around 4s at 10kHz to around 40s at 0.1Hz. Furthermore each type of sensor has a preferred frequency at which comparisons between different resin systems should be made. The IDEX sensors used in this study have a preferred measurement frequency of 10Hz. As during injection resin viscosity is lowest and immediate detection of the flow front is crucial in this study the measurement frequencies were set to 10, 1 and 0.1 kHz during the infiltration stage. After curing set in, measurements with 10, 1 and 0.1 Hz were added to account for the higher viscosity levels. With increasing viscosity lower frequencies are recommended as molecules and ions are hindered in their mobility due to the growing molecular network of the polymer. Measurements at different frequencies were carried out automatically by the system one after another starting at the lowest.
For calculation of the ultrasonic speed in the investigated sample, the time of flight of the ultrasonic wave through the bare tool must be determined. Therefore measurements without placing a frame between the upper and the lower mold were carried out at distinct temperatures for the room temperature as well as the hot curing resin test setup.
Each data point of the US system represents a mean value of an adjustable minimum number of single measurements - 1000 in this study. This fact results in a minimum sampling time for one data point depending on the investigated material. In the system settings a cycle time is requested which should be longer than the minimum sampling time otherwise no data point can be calculated. As sensitivity to resin arrival should be evaluated in this study, cycle time is a crucial parameter during infiltration and was set to 1.5s which was close to the minimum

time for calculation of one data point. Before the curing cycle was started, the measurement was stopped and restarted with a cycle time of 10s in order to save memory capacity which is limited for the used system.
Additional to the digital signal - resin arrival “yes” or “no” - information about the degree of saturation of the sensed preform volume are gathered by the US system due to the transmission character of the measurement. The received ultrasonic wave is amplified so that the system’s integrated analog digital transformer is optimally exhausted. This means that received waves with lower magnitudes are more amplified than waves with higher magnitudes. As long as the preform is dry, the ultrasonic wave is almost completely reflected at the tool cavity interface and no acoustic signal reaches the receiver. With increasing saturation of the preform the acoustic impedance of the preform also increases and the arriving ultrasonic wave at the receiver is less damped. Completed saturation of the preform can hence be indicated by constant system’s amplification values.

RESULTS AND DISCUSSION

Sensitivity to Resin Arrival

All regarded OPM systems reliably detected resin arrival in every trial of the study. A clear jump in the gathered signals was registered for every system (see Fig. 1). Comparison of the taken videos and the gathered signals showed that the flow front was immediately detected by the DCR and the US system. In case of the DEA IDEX sensors around half of the sensor had to be wetted before resin arrival was signalized. This is explained with the additional glass layers between the electrodes and the carbon fibers which were necessary for sufficient insulation of the DEA sensors.

Cure Monitoring and Repeatability

Every OPM system monitored the development of the resin viscosity. As soon as the temperature was risen for the curing cycle the viscosity started to decrease until curing becomes dominant. With increasing degree of cure the systems signals also increase. End of cure for the given temperature is signalized with a constant signal as exhibited in Fig. 1.
The 100Hz as well as the 10Hz graphs of the DEA are shown in Fig. 1. Higher frequencies are suited for immediate detection of the flow front and for low viscosity stages. Therefore, lower frequencies (10Hz, 1Hz and 0.1Hz) were added in later stages of cure to account for the increasing viscosity of the curing resin.
As can also be observed in Fig. 1 minimum viscosity was detected from the different OPM systems to different times. Within all trials minimum viscosity was first signalized by the DEA, followed by the DCR and finally by the US. This might be due to the different positions of the DEA and the DCR sensor and the bulk measurement in terms of the US system.
The different mixing ratios lead to different reaction rates what was registered by every OPM system. Higher amounts of hardener lead to higher slopes meaning higher reaction rates and the other way round when the sensors signals are plotted over time compared to the ideal mixing ratio.
Almost congruent runs were observed for the same mixing ratios with respect to the DCR and the DEA system. In terms of the US system only the curves for ideal mixing ratios could be compared with each other due to the differences in the experimental setup in the race tracking and the porosity trial. Together with the reference trials with RTM6 fair repeatability can be concluded for the US system.

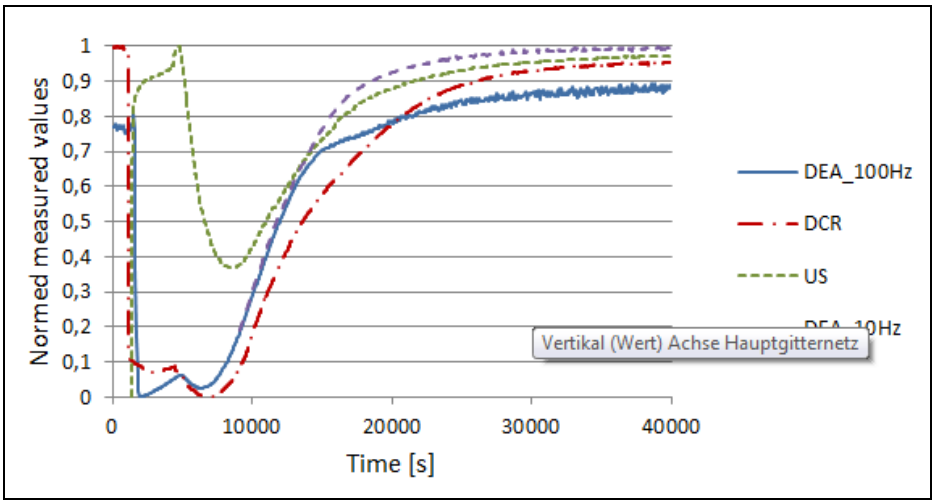

Fig. 1: Signal sequences of reference trial 1

The metal inlay in the race tracking trial lead to huge jumps in the measured ultrasonic speed so that correlations to material parameters cannot be drawn. Nevertheless US sensors can be used for detection of race tracking. The DCR TMS can be used close to cavity edges for flow front arrival as well as cure monitoring without limitation. Race tracking trials with IDEX sensors could not be performed in this study due to contradictoriness of the sensor's wires with the metal inlays.

Sensitivity to Porosity

The ultrasonic sound speed in the porosity trial was clearly lower than in the other trials. Therefore porosity measurements were carried out of this and two reference trials. Porosity and fiber volume content of the sensed volume were determined by acid digestion. Results are exhibited in Table 1 and show that the lowest porosity as well as the lowest fiber volume content was measured for the porosity trials.

Table 1: Results of acid digestion of room temperature curing resin samples

	Porosity [%V]	Fiber volume content [%V]
Reference 1	5.2	57.41
Reference 2	5.27	56.98
Porosity	0.7	53.18

Hence, the lower ultrasonic speed is not due to entrapped air in the sensed volume but due to the lower fiber volume content. Dependency of the ultrasonic speed on the fiber volume content has already been investigated in other studies in literature [4].
The results of the acid digestion from Table 1 are enforced by the gain values of the ultrasonic signals (see Fig. 2). It can be seen by the systems amplification development of the received ultrasonic wave of the different trials. The final gain values in the reference trials are similar which is in agreement to the porosity values of Tab. 1. The clearly lower porosity content of the porosity sample hence a higher degree of preform saturation is signalized by the clearly lower gain values of Fig. 2.
Concerning the DEA and the US system no differences in the porosity trial were found in comparison to other measurements.

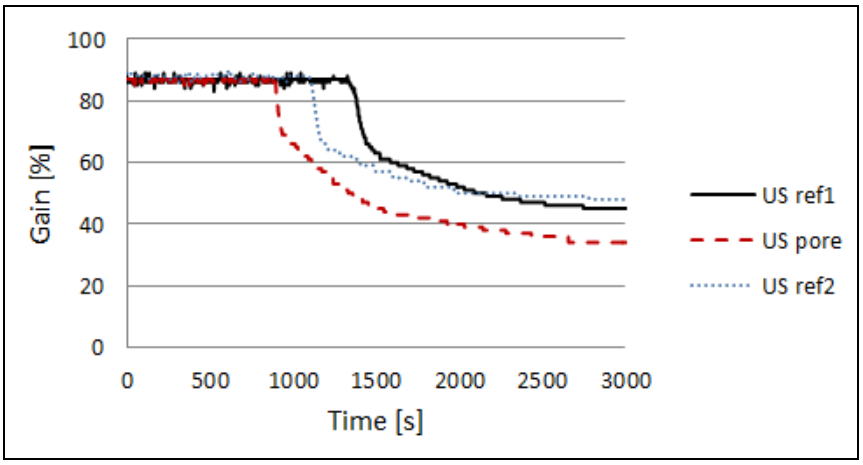

Fig. 2: Development of US systems gain

Detection of Thermoplastic Materials

Only the PSU films lead to different signals in terms of the DEA and the DCR system. Whereas the flow front and the curing reaction were monitored by the DEA, the DCR only signalized the presence of the PSU film. After resin arrival and during early stages of cure ion viscosity is about half an order of magnitude higher compared to the reference measurements and with ongoing curing the graph of the film trial converges to the reference graphs. With respect to the US system no thermoplastic materials could be observed.

Resin Aging and Repeatability

None of the regarded OPM systems could monitor any differences compared to trials with fresh RTM6 resin.
Concerning the repeatability in the RTM6 trials, best results were gathered by the DEA followed by the US. In terms of the DCR more noisy signals were measured due to presence of conductive carbon fibers. Furthermore a thin film of cured resin was observed on the sensor's surface after the last trial resulting in clearly higher resistivity values.

CONCLUSIONS

Apart from the detection of thermoplastic materials the regarded OPM systems met the expectations. Concerning the in-mold detection of aged resin no differences were observed compared to fresh resin. Either the differences were too small to be measured or they were masked by the influence of the conductive fibers. Hence, the striking differences are restricted to more general aspects. The DEA and the DCR system are limited to part surface measurements whereas the US technology gathers information from the bulk material. But available inlay sensors for the DEA and the DCR system can be used to monitor process parameters inside the part. Furthermore these sensors do not need any additional design space in comparison to the US sensors. The latter have to be mounted on both sides of the parts and must be installed at positions where the ultrasonic signals perpendicularly meet the cavity and part surfaces within certain tolerances.
Hence, as a final conclusion it can be said that the correct choice of an OPM system is mainly dependent on the surrounding process conditions like available design space or desired sensor positions and the demanded process parameters like flow front arrival or degree of saturation.

The following table summarizes important criteria for the choice of an appropriate OPM system and can be used as decision guidance.

Table 2: Overview of system characteristics

Criteria	Dielectric Analysis	Ultrasonic Technology	DC Resistivity Measurement
Resin arrival	+	+	+
Race tracking	%	+	+
Preform saturation	-	+	-
Porosity	-	+	-
In-mold viscosity development	+	+	+
In-mold quantification of mixing ratio	-	-	-
In-mold detection of aged resin	-	-	-
Reactivity	+	+	+
End of cure	+	+	+
Detection of thermoplastic film (top layer)	+	-	+
Detection of thermoplastic fleece (between layers)	-	-	-
Condensation polymerization	-	+	-
Contact free measurement	-	+	-
Signal penetration depth	low	high	low
Effort for sensor insulation from carbon	high	low	high
Design space	low	high	low
System installation effort	high	high	low
Repeatability	high	fair	fair
Frequency dependency	yes	no	no
%... No statement possible, -...impossible , +... possible			

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