# EXPERIMENTAL ANALYSIS BY THERMOGRAPHY AND HEAT FLUX SENSORS OF THE CURING STAGE OF COMPOSITE PARTS MADE BY RESIN INFUSION

U. Pineda[1*], F. Bensadoun[2], E. Ruiz[2], N. Montés[1], F. Sánchez[1]

[1] *Department of Building and Industrial Production, CEU Cardenal Herrera University, C/ San Bartolomé,55 – 46115 Alfara del Patriarca, Spain.*
[2]*Chair on Composites of High Performance (CCHP), Research Centre on Plastics and Composites (CREPEC), École Polytechnique de Montréal, Department of Mechanical Engineering, P.O. Box 6079, Station "Centre-Ville", Montreal, Canada, H3C 3A7,*
*\*Corresponding author: upineda@uch.ceu.es*

**Keywords:** Degree of cure, Contact heat flux sensors, non-Contact heat flux, Thermal camera, Thermography, Resin Infusion

**Abstract**
*This paper analyses the curing process of flat composite plates composed of glass fibers and vinylester resin through two thermal sensing techniques. The heat flux generated during the exothermic chemical reaction of the resin is measured by using a conductive heat flux sensor. Simultaneously, the radiative heat generated during the exothermic process is measured by a high definition thermal camera. Based on these techniques, the degree of cure (DoC) is estimated by the heat fluxes obtained in each area of the composite piece. The advantage of using a thermal camera is that it analyses the entire composite, while the heat flux sensor only throws punctual results limited to the area of the sensor. The thermography will detect variations in the DoC that may induce part deformation or internal stresses in the composite. This work shows the algorithms used to calculate the DoC in both sensing techniques. Finally, a comparison of the cure kinetics of the resin is done by the Differential Scanning Calorimetry (DSC) method to quantify the differences between the two techniques presented in this study.*

**Keywords:** Degree of cure, DoC, contact heat flux sensors, non-contact heat flux, thermal camera, thermography, resin infusion, RI process

## 1 Introduction

Liquid Composite Moulding (LCM) is a family of processes widely used in the production of composite parts in the automotive and construction industries. In particular, the Resin Infusion (RI) manufacturing technique is used for its ability to produce large components with a good quality/cost rate in limited series [1]. The manufacturing steps includes: preforming, mould closing and sealing, mould filling by RI, curing, demoulding and finally checking the finished composite part. In this process, the vacuum pressure is applied to drive the thermoset resin into a fibrous laminate [2]. The countermould is made of a flexible material, usually a transparent bagging film, which allows the manufacturing of parts that have various complex shapes and that can be of almost any size [3]. In addition, it acquires a strong thermal contact with the preform thanks to the vacuum pressure applied. It is important to note that after the resin injection with corresponding catalyst addition, the cross-linking accelerates and the

viscosity increases. This means that the thermoset resin is in-curing stage into the mould [4]. At present, this manufacturing process still presents reliability challenges for industrial production [1], such as a good understanding of the polymerization of parts and thus, the curing stage.

The heat released during the cure can lead to significant thermal gradients and temperature peaks according to the geometry of the part. Finally, it could generate residual stress that can a significant effect on the quality and mechanical properties of the part, generating warpage or initiating stress concentration in the matrix and delamination of the composite part [5]. Furthermore, the processing temperatures need to be optimally controlled for thermal gradients to remain small and thus, optimize the mechanical properties of the finished parts. Currently, curing stages in an on-going manufacturing relies heavily on experience of the operator [1]. Usually, the curing cycles are characterized by the Differential Scanning Technique (DSC) [6], which allows the monitoring of the cure kinetics by measuring the heat flux generated during the chemical reaction. This is extensively used in the determination of cure and post-cure parameters, such as the total energy of reaction, the reaction rate, the degree of cure (DoC), etc. Although, the DSC test requires a few milligrams of resin sample, larger amounts of resin need to be used during the manufacturing process and therefore a mass effect has to be considered in the cure kinetics calculation, as it is not visible in small samples. This means that non-homogeneity of thickness along fibre laminate as well as changes of manufacturing conditions and resin handling may generate different curing behaviour [7].

During manufacturing, the reliability and efficiency of detecting the heat fluxes from any area is influenced by the geometric shape, dimension and location of the sensors. Several intrusive and non intrusive methods exist for measuring and analyzing the polymerization of local areas. To detect the changes in the physical properties of the matrix, intrusive methods require a physic contact with the mould of both the mould and the sensor; however, the non-intrusive methods do not require a physic contact. Furthermore, contact sensors take advantage of changes of optoelectronic properties [8-10], electrical properties [11-13], acoustic properties [14-16], and thermal properties [6;17-20], while non-contact techniques mainly exploit acoustic [21] and thermal [22] properties. In this work, a non-intrusive method will be used to investigate the temperature and radiative heat flux changes by using a thermal camera during the curing stage of thermoset-based composites. The purpose of this work is to develop the most simple DoC estimation throughout a new thermal imaging technique and by comparing it to a well-known contact heat flux sensor method. The thermal camera offers the flexibility and versatility to instantaneously measure the heat flux, the $T^oC$ and the DoC at anywhere in the mould. Both thermal sensing techniques will be used to evaluate the curing stage of the composite plates. To the authors' knowledge, this is the first time the DoC is simultaneously estimated together with the manufacturing of composites by thermography analysis.
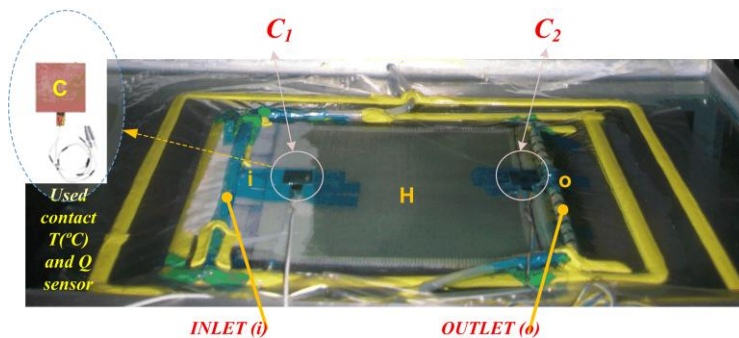
## 2. Experimentation
### 2.1 Materials
For this work, the Derakane 350-411, a vinylester thermoset resin with a viscosity of 0,350 Pa.s will be used. Before the manufacturing process, the resin is mixed with 1,25phr of MEKP-925H catalyst, 0.08 phr of 2-4 pentadione inhibitor and 0,10 phr of cobalt promoter to ensure an optimal infusion time. The resulting composite laminates are composed of two, four or six layers of 0º/90º glass fiber fabrics. The contact heat flux sensors are from Thermoflux and Captec, and the thermal camera TVS-500EXZ is supplied by NEC.

### 2.2 Experimental Set-Up

Figure 1 and Figure 2 show the setup and equipment needed for the manufacturing of squared composite flat plates, measuring 30x30 cm$^2$, and by using *Resin Infusion* manufacturing technique. The mould used in Figure 1 is placed inside a black box, as shown in Figure 3. The lower mould is a glass plate and the countermould a flexible and transparent plastic film. The viscosity of the mixture is critical because it will influence the filling time and the impregnation of the fiber tows. The plates are left to polymerize until there is no more detection of heat release by the acquisition system. The manufacturing processes are performed at room temperature conditions of 22°C and at a relative humidity rate of 50%.



**Figure 1.** Experimental Setup for manufacturing flat plates. Contact sensors (*C*) are placed near the inlet and outlet, as well as on top and under the preform.
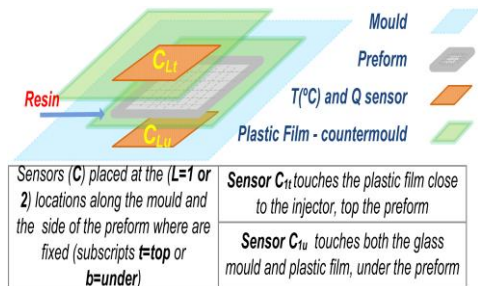
**Figure 2**. Placement of contact sensors during preforming to measure the up and down lost heat in a mould study area
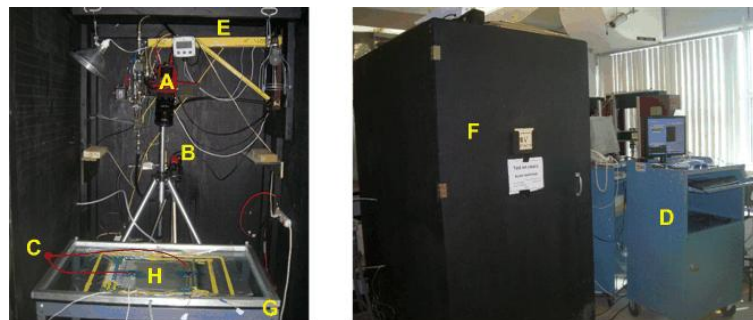


**Figure 3.** Thermal camera (A), visible camera (B), contact heat flux-T°C sensors (C), acquisition system with computer (D), temperatures sensor (E), infusion table (G), infusion mould (H).

A customized acquisition software reported in a previous study [23] is used in this work. This software has been simultaneously fed by four (4) contact sensors and thermal images from an infrared camera during the mould´s polymerization. Figure 1 to Figure 3 clearly explains the positioning of the contact sensors and the IR camera (hereinafter non-contact sensor). The IR camera is placed vertically 1,3m above the mould for an entire thermal viewing of the composite. The below "c" and "IR" subscripts are used to refer to contact sensors and thermal signals. During the tests, all the equipment has been placed inside the black box (see Figure 3) to prevent external interactions such as uncontrolled heat sources or thermal noise that may affect the abovementioned sensors response.

### 2.3 Performed Tests, Collected Data And Algorithms Via DoC Estimations

Table 1 summarizes the data collected before and after the manufacturing of the six flat plates. Two tests were performed for each *n* number of fibre layers. In Table 1, $m_p$ corresponds to the mass of the finished plate in a weight scale, *h* is the average of measured thickness in finished plates and $V_f$, the fiber volume content [24] that has been computed as shown on equation (1). Furthermore, $m_r$ stands for plate resin mass, $V_p$ for plate volume, $\rho_p$ for plate density. Finally, the amount of resin mass related to each sensed area is calculated by

equation (2); thus $m_r^c$ is the resin mass related to a contact sensor area and $m_r^{IR}$ is the resin mass related to a pixel area.

| Test | $n$ | $m_p$ (g) | $h$ (mm) | $V_f$ (%) | $m_r$ (g) | $V_p$ (cm$^3$) | $\rho_p$ (g/cm$^3$) | **Heat Flux sensor** *Sample resin mass below sensed area* $m_r^c$ (g) | **IR Camera** *Sample resin mass below sensed area* $m_r^{IR}$ (g) |
|---|---|---|---|---|---|---|---|---|---|
| 1 | 2 | 153,55 | 0,92 | 44,16 | 85,74 | 82,80 | 1,85 | 0,8574 | 1,88E-03 |
| 2 | 2 | 167,12 | 0,89 | 45,65 | 90,83 | 80,10 | 2,09 | 0,9083 | 2,00E-03 |
| 3 | 4 | 259,64 | 1,73 | 46,97 | 137,69 | 155,70 | 1,67 | 1,3769 | 3,03E-03 |
| 4 | 4 | 289,00 | 1,95 | 41,67 | 168,58 | 175,50 | 1,65 | 1,6858 | 3,70E-03 |
| 5 | 6 | 432,89 | 2,80 | 43,53 | 244,46 | 252,00 | 1,72 | 2,4446 | 5,37E-03 |
| 6 | 6 | 502,35 | 2,96 | 41,18 | 295,50 | 266,40 | 1,89 | 2,955 | 6,49E-03 |

**Table 1.** Manufacturing data from tests and calculations needed for DoC estimations.

In equation (1), $\rho_{sup}=518g/m^2$ is the area density of the preform and $\rho_{vol}=2,55*10^6g/m^3$, the volumetric density of the glass fibers.

$$V_f=\rho_{sup}*n/h*\rho_{vol} \tag{1}$$

$$m_r^k=\left(A_k*h*\rho_p\right).\left(1-V_f\right) \tag{2}$$

Where, $A_k$ is either the contact sensor area $A_c=9cm^2$ or pixel area $A_{IR}=0,0098cm^2$ on the mould. It can be seen that $h$ along finished plates varies with the $V_f$ and that $m_r^k$ is a resin mass for a sensed area. See the two last columns in Table 1

*2.4 DoC Estimation By Heat Flux Sensors And IR Thermal Camera*
The determination of DoC during an exothermic process appears in various studies as in [5;25]. This equation is shown below:

$$DoC=(1/H_T)*\int_{t_i}^{t_f} Q*dt \tag{3}$$

Where, *DoC* is defined as the ratio between the amount of heat flux $Q$ generated, at the time defined by $t_i$ and $t_f$, and the total heat of reaction, $H_T$. $H_T$ is measured by means of a DSC. This integration computes the area under the exothermal heat curve which is the amount of released energy during process. In composite manufacturing, that energy is had account to compare with $H_T$ instantaneously obtaining an instant DoC estimation.

Therefore, taking advantage the eq (3), $Q$ can be either the radiative heat flux $Q_{IR}$ computed for any image´s pixel acquired instantaneously by the IR camera or every one of the four instant heat fluxes $Q_c$ by a contact sensor. So, for subsequent comparisons of DoC by both kinds of heat data, they must be acquired simultaneously. However, they are time dependent as also sensed area dependent as mentioned [23] (e.g. pixel area or shape of the contact sensor)

In order to use these heat fluxes available for DoC estimations then, by doing a simple variation of the basic expression of the equation (3), this becomes the estimated DoC by heat radiation and denoted as **$DoC_{IR}$** as shown in equation (4)

$$DoC_{IR} = (1/H_T)* \int_{t_i}^{t_f} \left\{ \varepsilon\sigma*[T_{IR}{}^4 - T_A{}^4]/\left[ (\rho_p.h)*(1-V_f) \right] \right\} *dt \qquad (4)$$

Where, $\sigma$= **5.66704\*10-8 w m<sup>-2</sup> k<sup>-4</sup>** is the Stefan-Boltzmann´s constant. **$T_{IR}$** is the instant pixel temperature captured by thermal image. **$T_A$** is the instant surroundings temperature during each test expressed in Kelvin (k), **$\varepsilon$** is the emissivity that depends on the properties of the countermould (this may vary between zero and the unity), **$t_i$** and **$t_f$** represent starting and ending time for DoC computation. **$H_T$** is **350J/g**, this is the total heat of reaction which was measured with a Modulated- differential calorimeter M-DSC Q1000 by TA Instruments. A low pass filter is used to process these data and eliminating the noise in the thermal images.

Likewise, the equation (3) becomes the DoC estimated by the heat acquisition through the contact sensors and denoted as **$DoC_c$** as shown in equation (5). Where, **$Q_c$** **(w/m<sup>2</sup>)** is the measured heat flux by contact.

$$DoC_c = (1/H_T)* \int_{t_i}^{t_f} \left\{ Q_c/\left[ (\rho_p.h)*(1-V_f) \right] \right\} *dt \qquad (5)$$

## 3   Results, Discussion and Future works

This section analyzes three tests and the repetitions, each one of which has different **$n$** (i.e. 2, 4 and 6 layers of fiberglass). During the curing stages of the tests, the instantaneous DoC is estimated through two different thermal sensing techniques and by equation (4) and (5) (see Figure 2 and Figure 3). The DoC in neighbouring areas to the inlet and the outlet are especially studied in all tests. Figure 4 to Figure 6 show the estimated DoC as a function of time in the abovementioned mould areas.

Therefore, the blue curves estimate the DoC through two pixels, as they represent the closest position of the contact sensors (near the inlet and the outlet). In each thermal image, pixels are evaluated according to equation (4). In addition, the red curves represent the average DoC, which result from continuously computing the DoC on top and under the mould (see Figure 2).

The common feature among the six tests performed is the difference in DoC's throughout the mould, both in the inlet and outlet. The DoC is not homogeneous along the mould and physically confirmed by both thermal sensing techniques. It is observed that the variation of degree of cure is related to the thickness variation along the piece. The thickness of the piece is directly affected by the pressure gradient under RI manufacturing. In a few words, the thicker the plate is, the higher is the DoC. To a further DoC, a higher concentration of resin mass and therefore a higher heat release. The DoC in the neighbouring areas of the inlet is always higher than outlet neighbouring areas as there is more resin mass and therefore, more heat release from the inlet. Thus, it can be said that the DoC estimation method using eq (4) based on IR thermal camera, seems to be reliable, but not exact.
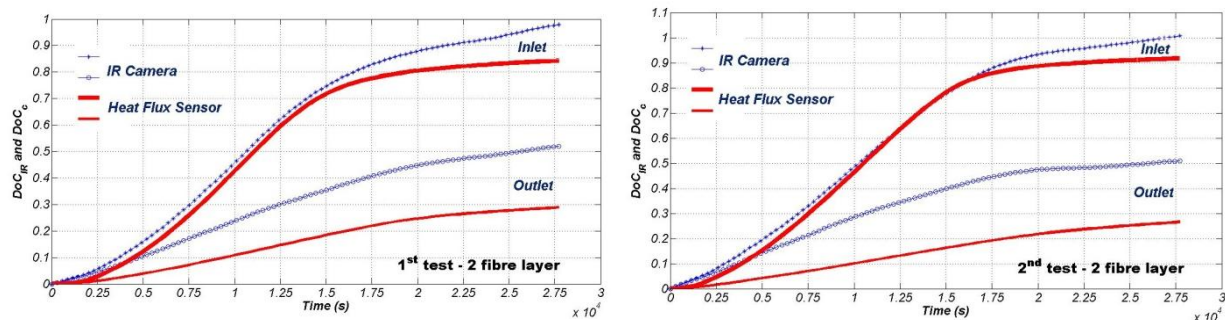
**Figure 4.** DoC reached at the inlet and outlet. Tests make under isothermal curing using (2) fiberglass layers.
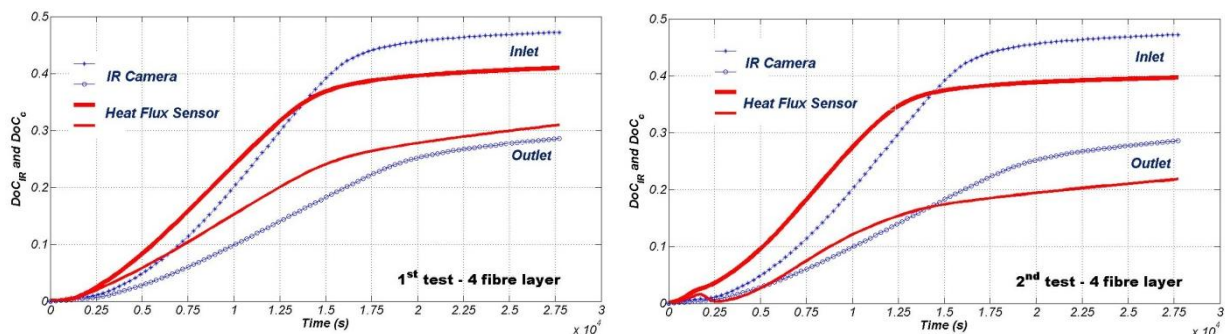


**Figure 5.** DoC reached at the inlet and outlet. Tests make under isothermal curing using (4) fiberglass layers.
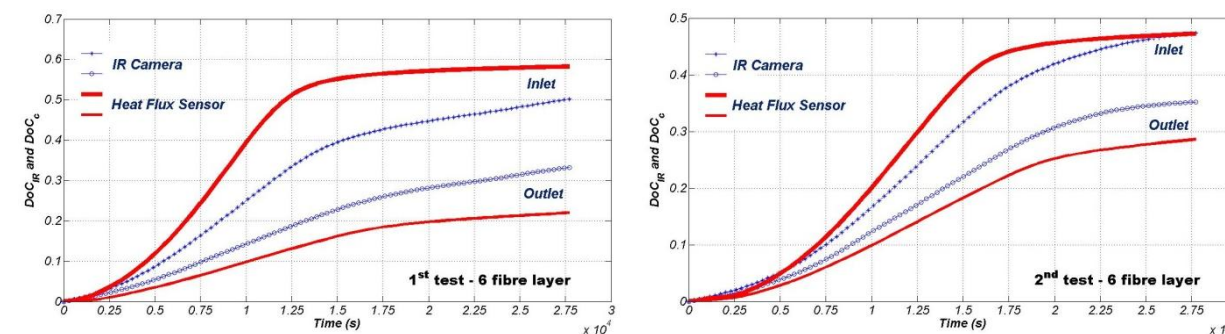


**Figure 6.** DoC reached at the inlet and outlet. Tests make under isothermal curing using six (6) fiberglass layers.

Considering the above, the accuracy of the new DoC estimation method could be improved by controlling the sources of error, which are mainly identified in variables in equation (4). First, contact sensors have the sensitivity in the order of 0.0001°C, while the sensitivity of the thermal imaging camera is 0.1°C. Thus, temperature variations smaller than 0.1°C are not detected. However, the error in the infrared measurements, especially caused by the emissivity, can be corrected by in- situ calibration or by the characterization of the manufacturing environment. Secondly, the resulting average thickness on each plate was used in all DoC thermal camera estimations. (See Table 1). Therefore, the association of the thicknesses of each piece to its thermography analysis, could improve the accuracy of the DoC estimations. Thirdly, by measuring the total energy of the reaction through thermography, instead of DSC, a higher precision might be reached. Further experiments are still been conducted to improve the understanding of the results presented.

## 4. Conclusions

In this experimental work, six composite plates were manufactured by the Resin Infusion technique. The heat flux generated during the resin's exothermic chemical reaction was measured through four conductive heat flux sensors, while the radiative heat generated during the exothermic process was measured through a high definition thermal camera.

This paper concludes that:

a) The homogeneity of DoC varies along a piece, reaching higher DoCs in areas closer to the inlet.

b) The estimation of the DoC by means of zero-dimensional sensors, such as conductive heat flux sensors, doesn't provide information about the cure levels elsewhere on the mould.

c) A new analysis method of DoC estimation by thermography allows the detection of the variations of the DoC along the piece. These DoC variations may induce part deformation or internal stresses in the composite.

d) The thermography analysis is more advantageous to the estimate the DoC, because, unlike the analysis performed by conductive heat flux sensors, it can be extended to any location on a piece.

e) The most simple DoC estimation based on thermal imaging is reliable, though not fully accurate when compared to the DoC measurement method that uses conductive heat flux sensors.

## 5. Acknowledgements

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