



HAL
open science

Online characterization of fiber-reinforced thermoplastic composite deconsolidation

Luc Amedewovo, Steven Le Corre, Basile de Parscau Du Plessix, Laurent Orgéas, Arthur Lévy

► **To cite this version:**

Luc Amedewovo, Steven Le Corre, Basile de Parscau Du Plessix, Laurent Orgéas, Arthur Lévy. Online characterization of fiber-reinforced thermoplastic composite deconsolidation. 20th European Conference on Composite Materials, ECCM20, Jun 2022, Lausanne, Switzerland. pp.462-469. hal-03762013

HAL Id: hal-03762013

<https://hal.science/hal-03762013>

Submitted on 26 Aug 2022

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

ONLINE CHARACTERIZATION OF FIBER-REINFORCED THERMOPLASTIC COMPOSITE DECONSOLIDATION

Luc, Amedewovo^{a,b}, Steven, Le Corre^a, Basile, de Parscau du Plessix^b, Laurent, Orgéas^c, Arthur, Lévy^a

a: Université de Nantes, CNRS, Laboratoire de Thermique et Energie de Nantes, LTeN, UMR6607, 44000 Nantes, France – steven.lecorre@univ-nantes.fr

b: IRT Jules Verne, Chemin du Chaffault, 44340 Bouguenais, France

c: CNRS, Université Grenoble Alpes, Laboratoire 3SR, 38000 Grenoble, France

Abstract: *Deconsolidation occurs in a fiber-reinforced thermoplastic composite when it is heated above a certain temperature. During heating in manufacturing processes (for example in welding, AFP, thermostamping, ...), when low pressure is applied, porosities can appear in different forms (bubbles, delaminations, etc.). Various works have been carried out in the literature to understand this phenomenon. In most of these works, deconsolidation is characterized by post-mortem analyzes or thermomechanical analyzes (TMA). However, these methods do not highlight deconsolidation mechanisms. In order to overcome these limitations, a new device for deconsolidation characterization is developed. It makes it possible to characterize online and continuously the deconsolidation of fiber-reinforced thermoplastic composite laminates. First results on moisture and residual stress effect on deconsolidation, were obtained on aerospace grade composite material (Carbon fiber/PEKK laminates).*

Keywords: Fiber-reinforced Thermoplastic composite; Deconsolidation; Porosity; Real time Characterization; Thickness Variation

1. Introduction

In recent years, the demand for fiber-reinforced composite materials has continued to grow in the aeronautical sector. The interest of composite materials lies in their specific mechanical properties (mechanical property/density ratio) which are better, compared to metals or other families of materials. This property of composite materials gives the possibility of considerably reducing the weight of structures in order to reduce gas emissions. In aeronautics, fiber-reinforced thermosetting matrix composites are the most widely used today. Fiber-reinforced thermoplastic composites are a better alternative to address assembly-related issues encountered with thermosetting composites, e.g., with the possibility of being welded [1][2].

However, during thermoplastic prepregs manufacturing processes involving fusion (continuous welding, thermostamping, AFP, ...), when a low pressure is applied, the deconsolidation of the composites occurs during heating leading to detrimental porosities [3][4]. Given the negative effect of porosities on the mechanical performance of parts [5][6], a better control is necessary to prevent or minimize deconsolidation. The conditions for the porosities appearance and the mechanisms that govern their growth is to be investigated.

Two major hypotheses exist in the literature to explain deconsolidation mechanisms. On the one hand, some authors suggest that the deconsolidation is due to a relaxation of the residual stresses initially present in the composite plates after their consolidation [4] [7] [8]. On the other

hand, other authors explain the deconsolidation by the diffusion, under the effect of temperature and pressure, of volatiles (including water) dissolved in the laminates during their storage or during the prepregs manufacture [3][9][10].

In most of these works, deconsolidation is characterized by *post-mortem* analyzes (thickness measurements before and after deconsolidation, micrographs, ...). However, they do not inform on the *in-situ* mechanisms involved during heating. Another method used to characterize deconsolidation is thermomechanical analysis (TMA) [10][11], allowing *in-situ* (continuous) characterization of deconsolidation during the heating phase. However, standard TMAs requires samples of small dimensions (few mm only) which are not representative of the size of the composite structures.

In this work, a novel TMA device has been developed (section 2) and validated (section 3) for the continuous and online characterization of the deconsolidation of large high-performance fiber-reinforced thermoplastic matrix composite laminates carbon/PEKK (section 4).

2. Experimental setup

The developed device, called CODEC bench (COMposite DEconsolidation Characterization), allows continuous and online monitoring of the deconsolidation of a composite plate subjected to a temperature variation, representative of a manufacturing process. The characterization is done by continuous measurement of the composite thickness which is related to structural change in the composite such as porosities. The CODEC setup is shown in Figure 1.

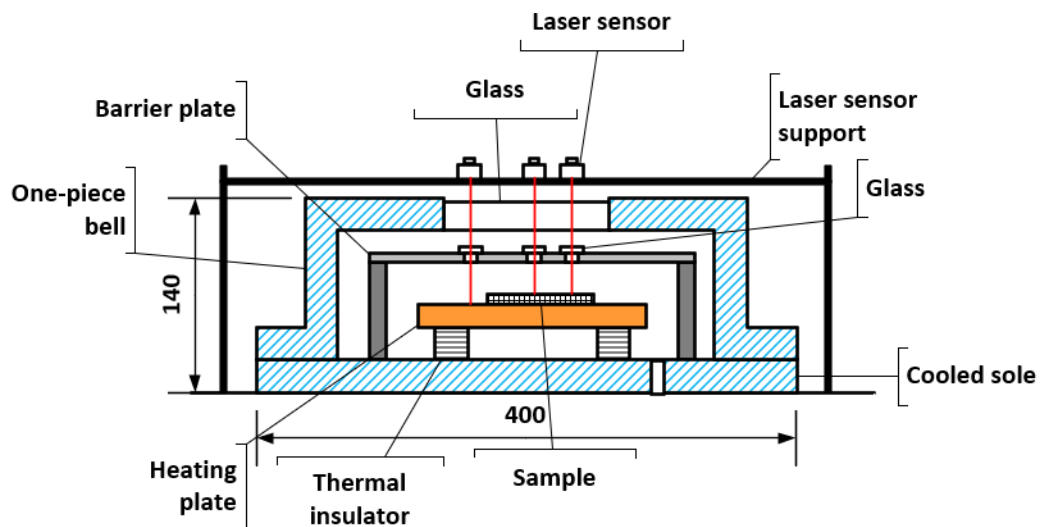


Figure 1. Scheme of the CODEC device designed for online characterization of deconsolidation of large fiber-reinforced thermoplastic composite plates, using laser sensors for thickness measurement, during its processing on a hot plate. This gives insight on the deconsolidation

The device is composed of a copper heating plate, which is placed in a closed thermal chamber (in aluminium) with a 25 mm thick borosilicate glass window. Water circulation channels are machined in the sole to cool the chamber walls. Two blocks of Pamitherm thermal insulators are placed between the sole and the heating plate, to reduce heat transfer by conduction. In order to vent the chamber during heating, a hole is machined in the sole to maintain atmospheric pressure inside.

The hot plate temperature is measured by a K-type monitoring thermocouple located in the middle of the hot plate, 1 mm underneath its surface. The thickness variation of the samples is measured using 3 Keyence IL-S065 laser sensors through the borosilicate window. The three distance measurement sensors have a measurement range of 55 to 75 mm and a linearity of $\pm 0.075\%$ of the full scale. The laser emitted by the sensors has a wavelength of 655 nm (visible light), to which the borosilicate is transparent. The maximum environmental temperature that the sensors can withstand is 50°C. For this reason, the sensors are positioned in a box located on a structure outside the chamber.

Thermocouples, laser sensors, and power controller are connected by means of modules to a single NI CompactDAQ acquisition system. The heating power is generated by cartridge heaters and regulated according to the temperature measured by the control thermocouple. The control is performed automatically using a single software developed on labVIEW. Thus, the temperature, thickness data acquisition and temperature control are synchronized.

3. Thickness variation measurement validation

In order to validate the CODEC Bench, the Coefficient of Thermal Expansion (CTE) obtained on a homogeneous material, *i.e.*, 6061 Aluminum, using a standard dilatometer (Linseis L75HS500LT), was compared with the CTE obtained with the same material on the CODEC setup.

For standard dilatometry, the test was performed on a 10 mm × 10 mm × 2.98 mm sample of 6061 aluminum which was heated at 2°C/min from 20°C to 300°C. Aluminum does not experience phase change or deconsolidation during heating. Strain as a function of temperature is plotted in Figure 2. From this curve, the linear CTE was determined along the thickness direction (Table 1).

Another test was thereafter carried out with the CODEC device, with a bigger plate of the same alloy (125 mm × 25 mm × 2.98 mm). The sample placed in the device was heated at the same heating rate as on the dilatometer up to 250°C.

Two laser sensors were used to measure the thickness variation of the sample. One sensor aims the center of the sample (sensor 2) and another 40 mm from the center (sensor 3). The last sensor (sensor 1) aims the heating plate (Figure 2.c). During the heating, the distances between the sensors and the targets (sample and heating plate) were recorded. The sample elongation was obtained by the difference between the actual distance and the initial ones. The heating plate elongation (baseline, sensor 1) was subtracted from the total elongation calculated on the sample. This operation allowed us the intrinsic sample elongation to be estimated. From these data, the linear CTE of the sample was determined along the thickness direction (Figure 2.b).

The curve obtained with the CODEC device is non-linear. This non-linearity is due to the macroscopic movements of the heating plate and air convection which disturbed the laser measurement. However, the comparison of the CTEs obtained by standard dilatometry and with the CODEC device, at the measurement points (Table 1) shows that the CODEC device is able to characterize small strains with a measurement resolution of $\pm 36 \mu\text{m}$.

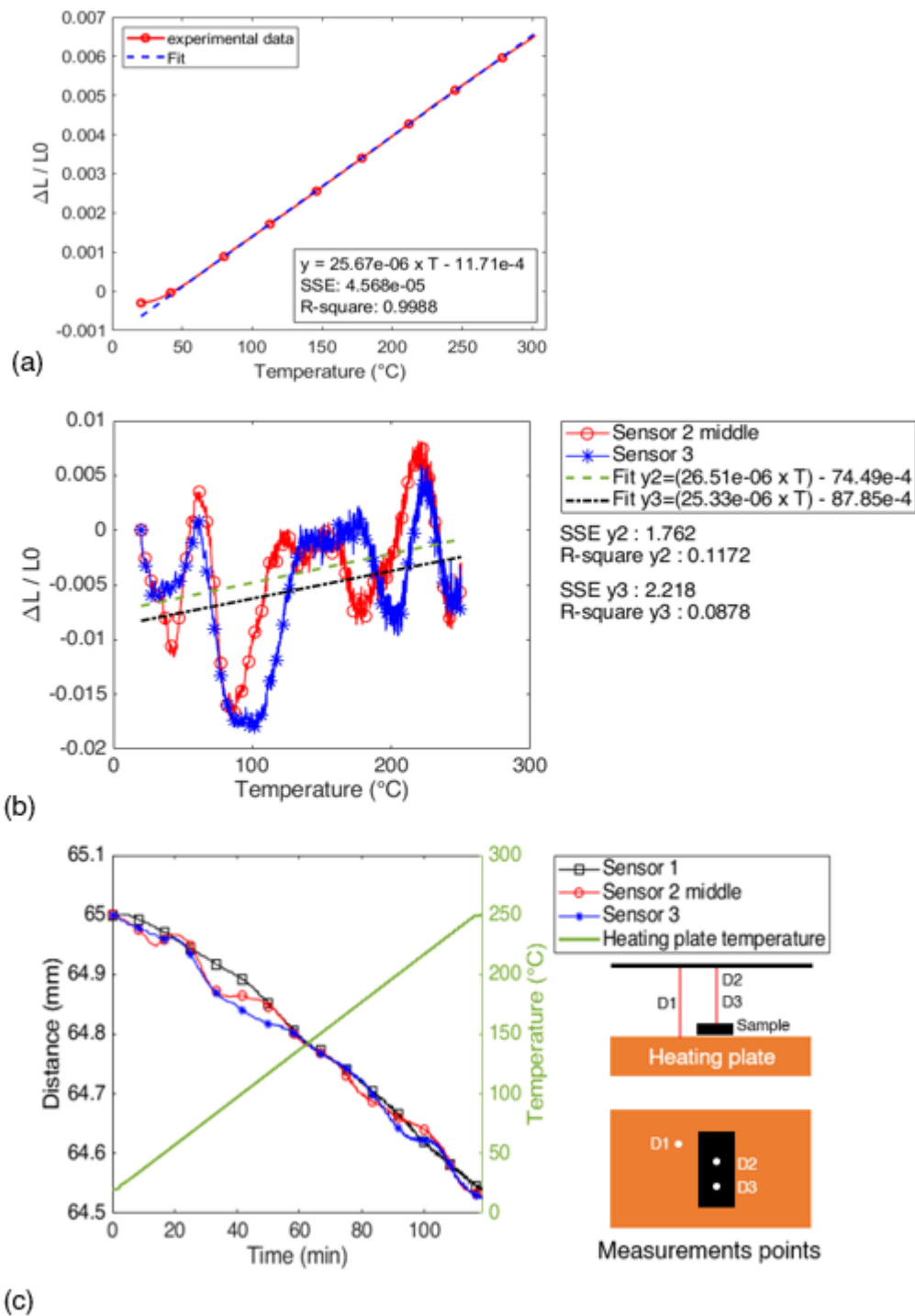


Figure 2. computed strain of a 6061 Aluminum reference material during (a) standard dilatometry (b) on CODEC device at two measurement points (c) Raw distances measured over time during ramp up on the reference material

Table 1: Comparison of CTEs obtained by standard dilatometry and on the CODEC bench.

| Dilatometry ($^{\circ}\text{C}^{-1}$) | 25.68e-06 | | |
|--|-----------|-----------|-----------|
| | Sensor 2 | Sensor 3 | Mean |
| Codec setup ($^{\circ}\text{C}^{-1}$) | 26.51e-06 | 25.33e-06 | 25.92e-06 |
| Absolute error ($^{\circ}\text{C}^{-1}$) | 0.84e-06 | 0.34e-06 | 0.25e-06 |
| Relative error (%) | 3.27 | 1.32 | 0.97 |

4. Composite deconsolidation characterization

In this section, the moisture and residual stress effect on the deconsolidation of a high-performance thermoplastic composite is investigated. Using the CODEC setup, deconsolidation tests were carried out on unidirectional (UD) laminates composed of carbon fibers (CF) and PEKK 7002 resin.

4.1 Materials

Prepregs CF/PEKK plies are supplied by Toray Advanced Composite. The plies have a fiber areal weight (FAW) of $194\text{g}/\text{m}^2$ and a theoretical thickness of 0.185 mm. The resin mass content is 34%. The glass transition temperature (T_g) and melting temperature (T_m) of PEKK 7002 are 160°C and 337°C , respectively (according to the manufacturer).

First, the plies were stacked in a mold following the same orientation $[0]_{16}$. Consolidation was then carried out on a PEI 50t press according to the following cycle: heating at $10^{\circ}\text{C}/\text{min}$ up to 380°C under a pressure of 0.1 MPa; holding of the temperature for 20 min under a pressure of 4 MPa; cooling at $10^{\circ}\text{C}/\text{min}$ at the same pressure, then demolding. 16 UD plies of $350\text{ mm} \times 350\text{ mm} \times 2.90\text{ mm}$ were manufactured accordingly. Optical micrographs on the consolidated plates validate the absence of porosities (Figure 4).

4.2 Pretreatments and testing procedure

In order to investigate the moisture and residual stress effect, the deconsolidation tests were carried out on two groups of samples: Ambient storage (AS) samples and dried samples (DS). The AS samples were cut by water jet from the consolidated plates stored in the workshop for 5 months in ambient conditions. A weight measurement after a drying test at 180°C on an $80\text{ mm} \times 80\text{ mm}$ sample, cut from the stored plates, showed that the water content of the plates is 0.013% at the end of the storage.

The dried samples were also cut from the stored plates and then dry at 180°C for 72 hours. This drying condition was defined by continuous weight measurement during drying tests at different temperatures (140°C , 180°C , 250°C). Drying at 180°C for 72 hours fully eliminated the effect of humidity without any thermal degradation of the material.

The samples dimensions for the deconsolidation tests were $125\text{ mm} \times 25\text{ mm} \times 2.90\text{ mm}$. Within the codec setup, the samples were heated at $60^{\circ}\text{C}/\text{min}$ up to 380°C and then maintained at this temperature for 5 min, follow by a cooling (natural convection between the sample and the air). This cycle is representative of typical temperature for CF/PEKK during its processing (resistance welding, thermostamping, ...). Each testing condition was repeated three times. At the end of

each test, the thickness variation was also measured by *post mortem* analysis, using a 0.01 mm precision micrometer. *Post mortem* optical micrographs were also performed on the samples.

4.3 Results and Analysis

Figure 3.a shows the evolution of deconsolidation strain $\varepsilon_d = \ln(e/e_0)$ (e_0 and e being the initial and actual sample thicknesses, respectively) vs time, during deconsolidation of the DS and AS sample. The sample strain brutal increase during the ramp up, is a signature of deconsolidation. In the absence of humidity, the maximum deconsolidation strain reaches 0.17, which is mainly ascribed to the residual stress relaxation. Indeed, when the material temperature reaches the melting temperature range, the sample begins to melt and behaves like a viscoelastic liquid. The phase change from a viscoelastic solid to a viscoelastic liquid favors the residual stress relaxation.

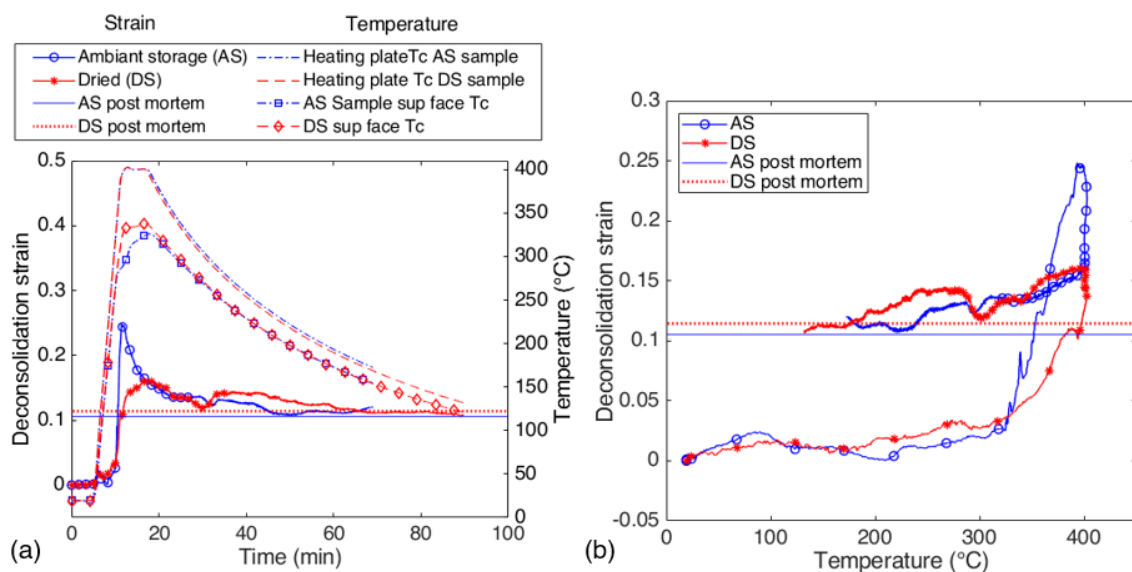


Figure 3. deconsolidation strain (a) vs time (b) vs temperature measured at the center of the AS and DS sample

Figure 3.b shows the deconsolidation strain *versus* the heating plate temperature. The sample strain behavior can be divided into 3 phases. During the first phase, the sample experiment thermal expansion. In the second phase, when the temperature reaches the melting temperature range, the deconsolidation begins and lead to an important strain increase followed by a slight strain decrease. This decrease, mostly visible in the AS samples, is probably linked to the collapse of the large porosities under gravity effect. In the third phase, the sample strain decreases due to the sample shrinkage during the cooling.

In the case of AS samples, the maximum strain observed is 0.09 higher than the DS samples maximum strain. This difference is related to the combination of the moisture effect with the residual stress relaxation. The moisture effect is linked to the water molecules diffusion in the porosities (due to the temperature increase) leading to the porosities internal pressure increase. The porosities start to grow when their internal pressure is greater than the external pressure applied by the composite. According to the small difference between the maximum strain, it can be noted that, on the material studied and under the defined test conditions, the effect of humidity is small but not negligible compared to the effect of residual stresses. The diffusion

phenomenon strongly depends on the heating rate. To be able to draw a conclusion on the predominant phenomenon, it will be necessary to carry out tests at other heating rates.

The *post mortem* optical micrographs also highlight the moisture effect. In the AS samples, micrographs show larger porosities, mainly located at the subsurface (Figure 4.b). In the DS samples, the porosities have small sizes and are homogeneously located in the middle of the sample (Figure 4.c). However, the measured *post mortem* thickness indicates a small difference between the AS sample and dried sample strains ($\Delta\varepsilon_d = 0.01$, see Figure 3.b). The *post mortem* thickness measurement solely cannot therefore explain the microstructure differences.

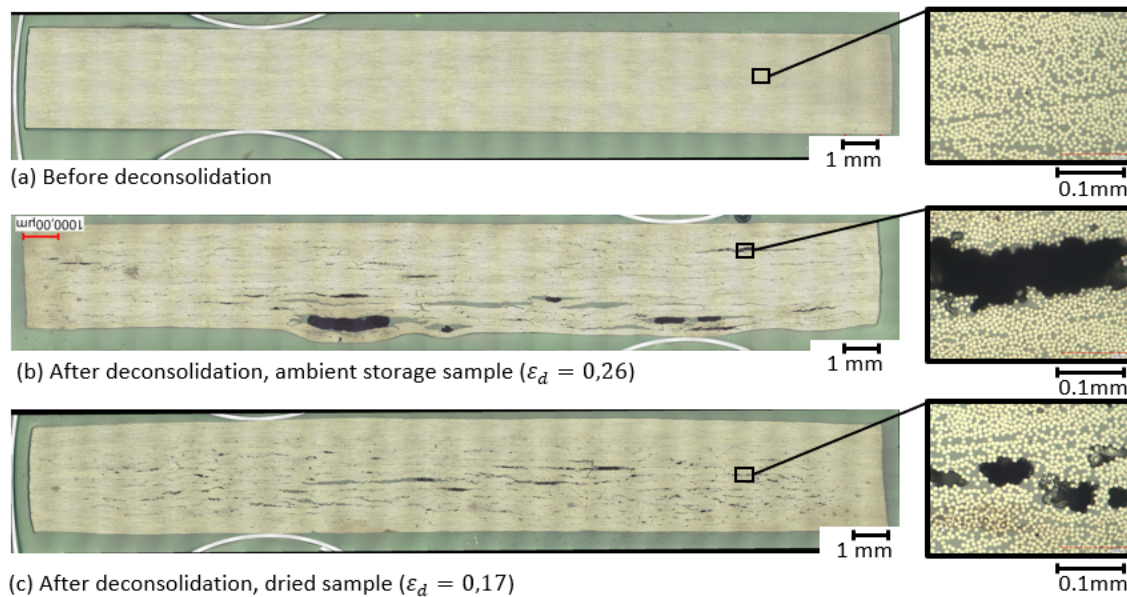


Figure 4. cross-sections micrograph of a 25mm wide sample after (a) press consolidation, material is fully consolidated (b) after ambient storage for 5 months (c) after drying at 180°C for 72 hours

Unlike the *post mortem* thickness measurement, the online deconsolidation strain characterization shows that the porosities final morphology and distribution is highly affect by the *in-situ* mechanisms during heating. The difference between the final content, distribution and morphology of the porosities can now be explained and highlighted by the deconsolidation strain history. The large porosities observed in the AS sample are due to the fast and high increase of the porosities size visible the deconsolidation strain evolution. The low porosities content observed in the DS samples is related to the slow growth of the porosities size during the heating also visible in the deconsolidation strain evolution. Thanks to the online characterization on the CODEC setup, these two observations allowed a better understanding of the moisture diffusion effect and residual stress relaxation effect on deconsolidation.

5. Conclusion

In order to better understand the mechanisms involved during deconsolidation, the CODEC device has been developed in this study. The device allows continuous and online characterization of composite laminates deconsolidation, under industrial conditions, using thickness measurement with a resolution of $\pm 36 \mu\text{m}$.

Using the CODEC setup, deconsolidation tests were carried out on a CF/PEKK composite sample dried at 180°C for 72h and sample stored in ambient conditions for 5 months. The tests results show a small (but not negligible) effect of moisture on deconsolidation, compared to residual stress effect. The deconsolidation kinetics measured correlates well with the final porosities content and morphologies observed on post mortem micrographs. This provide a better understanding of the mechanisms involved during the deconsolidation of a press consolidated thermoplastic composite laminate. In the future the codec will be updated and used to investigate the effect of heating rate and pressure on deconsolidation, in order to have a deep comprehension of deconsolidation, under different industrial conditions.

Acknowledgements

The authors would like to thank Julien Aubril and Arnaud Arrive for the help on the design and machining of the CODEC bench. The authors would like to acknowledge the funding of PERFORM project led by IRT Jules Verne (France Institute for Advanced Research and Technology Manufacturing technologies for composites, metals and Hybrid Structures) who made this work possible. Authors wish to associate the industrial partners of PERFORM project: Airbus, Daher, Safran, Latecoere, Stelia Aerospace, Clayens NP, Naval Group and Faurecia.

6. References

1. Mays GC, Hutchinson AR. Adhesives in civil engineering. Cambridge University Press. 1992.
2. Crane RL, Dillingham G. Composite bond inspection. *J Mater Sci.* oct 2008;43(20):6682-94.
3. Banea MD, da Silva LFM. Adhesively bonded joints in composite materials: An overview. *Proc Inst Mech Eng Part J Mater Des Appl.* 1 janv 2009;223(1):1-18.
4. Donadei V, Lionetto F, Wielandt M, Offringa A, Maffezzoli A. Effects of Blank Quality on Press-Formed PEKK/Carbon Composite Parts. *Materials.* 23 juin 2018;11(7):1063.
5. Saenz-Castillo D, Martín MI, Calvo S, Rodriguez-Lence F, Güemes A. Effect of processing parameters and void content on mechanical properties and NDI of thermoplastic composites. *Compos Part Appl Sci Manuf.* juin 2019; 121:308-20.
6. Liu X, Chen F. A Review of Void Formation and its Effects on the Mechanical Performance of Carbon Fiber Reinforced Plastic. :19.
7. Wan Y, Takahashi J. Deconsolidation behavior of carbon fiber reinforced thermoplastics. *J Reinf Plast Compos.* sept 2014;33(17):1613-24.
8. Brzeski M, Mitschang P. Deconsolidation and Its Interdependent Mechanisms of Fibre Reinforced Polypropylene. *Polym Polym Compos.* oct 2015;23(8):515-24.
9. Simacek P, Advani SG, Gruber M, Jensen B. A non-local void filling model to describe its dynamics during processing thermoplastic composites. *Compos Part Appl Sci Manuf.* mars 2013;46:154-65.
10. Slange TK, Warnet LL, Grouve WJB, Akkerman R. Deconsolidation of C/PEEK blanks: on the role of prepreg, blank manufacturing method and conditioning. *Compos Part Appl Sci Manuf.* oct 2018;113:189-99.
11. Shi H, Villegas IF, Bersee HEN. A STUDY OF PROCESS INDUCED VOIDS IN RESISTANCE WELDING OF THERMOPLASTIC COMPOSITES. 2015;13.