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Improvement of the Aerospace Composite Manufacturing Process using MWCNTs Fibres

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Abstract

In this work, a novel sensing polymer manufacturing technique that allows controlling the resin front advance during a Vacuum Assisted Resin Infusion (VARI) process is presented. Macroscopic fibres made of Multiwall Carbon Nanotubes (MWNTs) integrated in different materials fabrics, such as glass, carbon and Kevlar fibres were manufactured to monitor the resin front advance. This front was monitored by measuring their electrical resistance due to the impregnation of the resin in the fibres. Due to their high selectivity and sensitivity, these sensors may be used in manufacturing processes in which no visual inspection can be performed, such as in RTM.

1. Introduction

Operation and sensing techniques used in infusion and RTM processes which are the most common out-of-autoclave processes for polymer composites manufacturing, have been improving substantially in the last years. They have the aim of satisfying more and more the needs of certain profitable industry sectors such as the automotive, space and aircraft ones [1]–[4] which demand particular materials properties for their applications. Properties such as lightweight and low density, low level of porosity and consequently of voids and defects, moderate cost, excellent corrosion resistance, high ductility (except thermosets), and sometimes high toughness and transparency are characteristic of polymer composites and can be only achieved if several manufacturing parameters such as pressure, fabrics compaction, temperature and fluid front velocity are rigorously monitored and controlled.

Moving forward to these automated systems in polymer composites industry, and more in particular when using out-of-autoclave techniques, accurate and the least invasive sensing techniques are aimed to be integrated inside the same manufacturing process. The most common sensors are the fibre optics [5], [6], capacitive [7], piezo-resistive [8], temperature [9], pressure [10] and ultrasound [11] ones that give information of the temperature, pressure and position of the resin front advance. In most cases they are incorporated between the fabrics inside the mould in RTM or inside the vacuum bag during infusion processes.

In addition, fibre-polymers [4][12] are highly valued in industry for their excellent mechanical properties which are inherited by their corresponding composites. They can be more easily integrated in the fabrics than other sensors and may modify much less the laminate structure. Some of the most common fibres used are the ones of carbon, glass and aramid (i.e. Kevlar). Due to the relevance of these three fibre-composites in industry, carbon, glass and Kevlar fabrics were tested in this paper under typical infusion conditions by measuring the electrical resistance change of Multiwall Carbon Nanotubes (MWCNTs) yarns [13].

One of today's most efficient fibres used as smart sensors are the Carbon Nanotubes (CNTs) [14]–[16] and more specifically the multiwall ones (MWCNTs) due to their excellent mechanical (high strength and stiffness) [17] and electrical properties (high conductivity and piezo-electricity) [15]. In fact, they were chosen in this paper as the most efficient to be used for sensing. Moreover, they can give information about the role that the different mechanical and thermal variables play in the RTM (Resin Transfer Moulding) and/or infusion manufacturing process. Furthermore,

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they allowed locating the resin front advance in time and measuring the proper physical magnitudes needed to have high quality laminates at the end of the process. Dry fabrics such as glass, carbon and Kevlar fibres were tested and VARI using resin Derakane was carried out. Resin was injected through the inlet at atmospheric pressure in a vacuum bag that covered the fabrics and that was placed on a one-sided mould. Its front advance moved in the direction of the pressure negative gradient wetting them in a laminar flow regimen while the initial vacuum produced inside the hermetic bag was displaced.

The paper is structured as follows: Firstly, the materials and technology used in this work are presented. After, results obtained are discussed. The contribution of the metallic welding between the MWCNT sensors yards and the DAQs electrodes to the CNTs electrical resistance signal measured for different pressure cycles was studied. A LVDT (Large Variation Displacement Transducer) was placed on the different materials fabrics in order to determine the contribution of the MWCNT compaction due to the vacuum pressure cycle. To conclude, a VARI experiment was carried out with two MWCNT yards placed along the fabrics in the direction of the flow and placed symmetrically at both sides of the center so that the effect of these external factors commented above to the resistance change signal measured during the infusion were measured and discriminated. The results obtained in this paper will have a great impact when fabricating smart fibre sensors as flexible electronics for infusion monitoring and sensing, and will allow controlling the fluid front advance in infusion and RTM processes in order to obtain high performance laminates with very good mechanical properties and low level of defects.

2. Materials and methods

In order to determine the best candidates of CNT yarns as resin front advance sensors, high oriented MWCNTs and low-oriented MWCNTS fibres fabricated at IMDEA facilities were evaluated. MWCNT yarns change in resistance was measured. After and inside a vacuum bag, different types of fabrics were placed on a metallic sheet in order to study the infusion process. However, with the purpose of studying the welding effect, no fabrics were used in this case (Figure 1). Finally, and with the goal of accurately knowing the resistance variation produced by the polymer impregnation in the CNTs during the infusion process, the same type of infusion commonly performed in a typical industrial manufacturing process (VARI: Vacuum Assisted Resin Infusion) was performed. After obtaining resistance change measurements, the contribution of these external factors (weld contacts, vacuum pressure and fabrics compaction) were subtracted to the signal obtained during the infusion and the electrical resistance change due to the resin impregnation on the fabrics was obtained. Resistance, fabric thickness and pressure values were monitored on line using a Labview interface that connected Quantum MX840A DAQ to the different sensors.

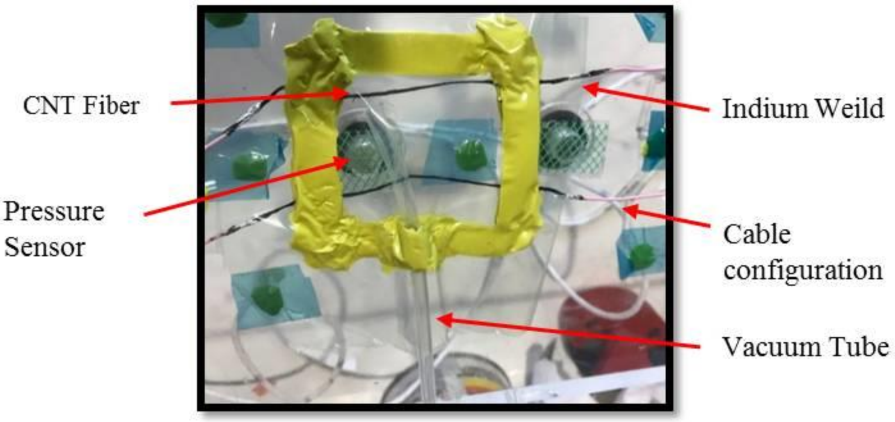

Figure 1. Configuration with electrode contacts neither inside vacuum bag nor in the fabrics

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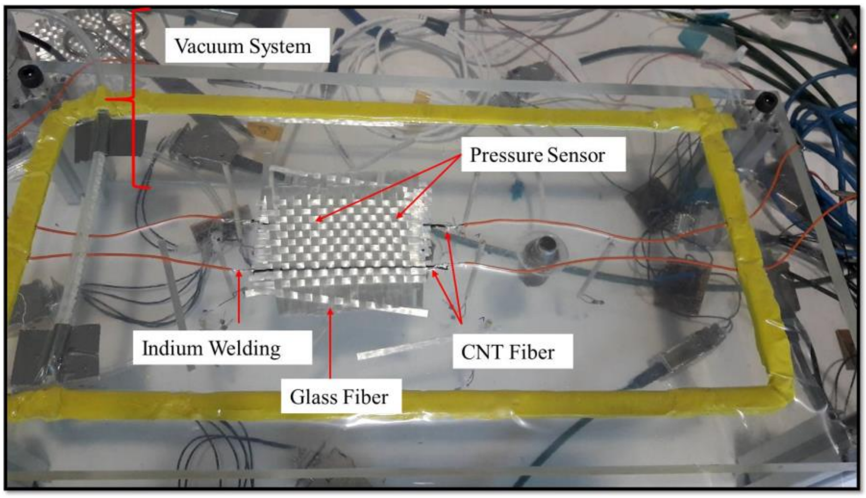

Figure 2. Vacuum bag configuration with CNT fibres and Glass fibre layers.

In this work, a conventional infusion configuration at room temperature was carried out. Resin employed was Derakane 8084 without catalyst agent. Vacuum pressure was generated using a vacuum electrical pump with polyurethane tubes connected to a hermetic plastic bag in the outlet. Resin went out through this outlet placed in the opposite side to the inlet in the direction of the flow and it was taken from a vessel supporting atmospheric pressure. Between the vessel outlet and the electrical vacuum pump, a tramp was used as resin container. Finally, in order to determine the resin front advance position vs time, a scale was drawn on the top of the bag and a video camera was placed above the mould in order to film the resin front advance during the infusion process (see Figure 3) and to compare it with the CNTs yarns resistance variation vs length. These CNT yarns were placed between the fabrics (one yarn between the 1st and the 2nd fabrics and the other between the 2nd and the 4th ones). In addition, two Omega absolute pressure sensors (see gray circles in the photo below, the left one is the pressure sensor 1 and the right one the pressure sensor 2 that is far away from the inlet) were positioned at a quarter and at three quarters respectively of the total vacuum bag length from the beginning of the fabrics. The direction of the flow is from left to right in the figure below and its laminar flow is due to the difference in pressure between the inlet and the outlet.

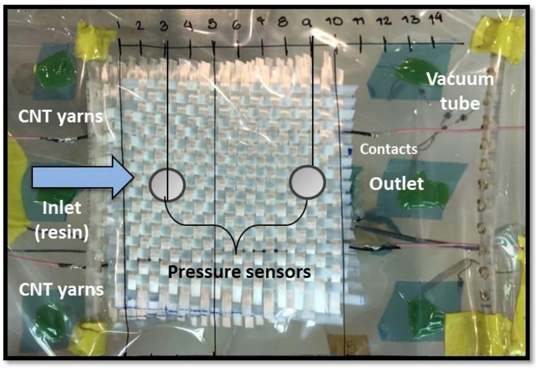

Figure 3. Infusion set-up with the CNT yarns and pressure sensors inside the vacuum bag

3. Results and discussion

3.1 Selection of the best MWCNT

Very similar change in resistance was shown between highly oriented MWCNT fibres and low oriented ones when a drop of butanol was released from a pipette and dropped the yarns. However, highly oriented MWCNT yarns are more difficult to fabricate, are weaker and can be easily broken. This makes highly oriented MWCNTs the best candidates

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for controlling the front advance during an infusion experiment. It was measured that low oriented MWCNT yarns electrical resistance change was of 10.4 % and highly oriented ones MWCNT 8.6%.

3.2 Effect of the weld contacts in the CNT yarns

As it can be seen in the Figure 4, there is an initial high drop in the electrical resistance when vacuum is produced at the beginning of the infusion. The reason for this drop is because of the welding between CNT fibres and the electrodes cables.

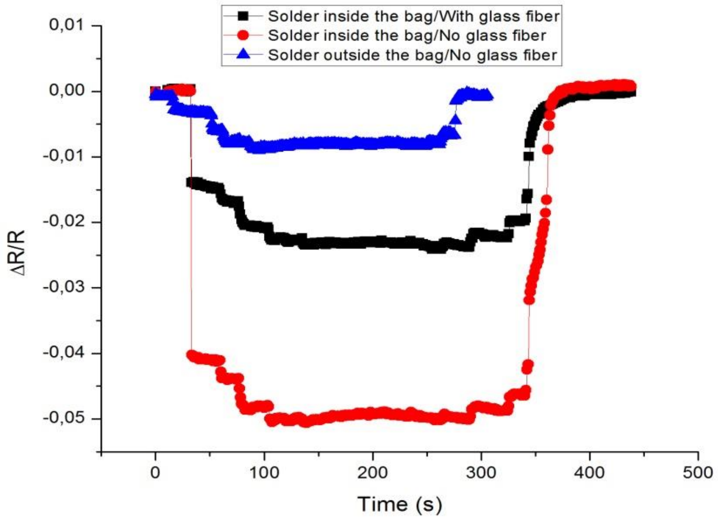

Figure 4. Behavior of the CNTs electrical resistance with different electrodes welds configurations and for different pressure values.

3.3 Effect of the vacuum pressure in the CNT yarns

In order to study the contribution of the vacuum pressure in the CNT yarns using different fabrics, several vacuum cycles were applied to four different types of fabrics in order to evaluate the behavior of the CNT electrical resistance to vacuum pressure. The procedure followed to apply the vacuum pressure variation consists of two parts: a positive one, going from the atmospheric pressure to -1000 mbars and a second part going in the reverse direction: from such vacuum value to the atmospheric pressure again.

As it is shown in Figure 5, a hysteresis cycle in the electrical resistance variation for each of the pressure cycle followed is observed. A very similar shape hysteresis behavior was measured when using both types of glass fibre and Kevlar fabrics. Glass fibre was the one with less change in resistance and smaller hysteresis. This is because of its lower of piezo-resistivity that is highly related to its lower conductivity. This difference is higher when these resistance values are compared with carbon fibre.

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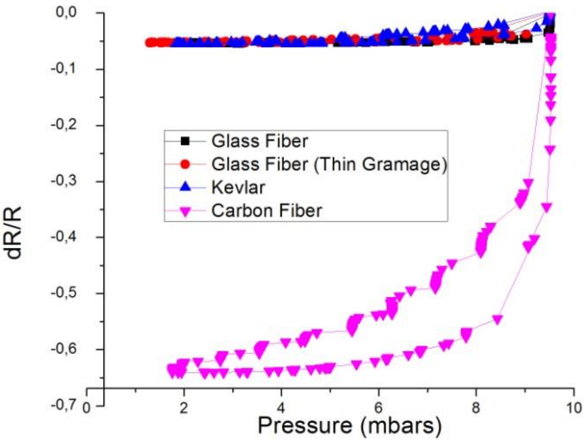

Figure 5. Pressure vs resistance variation curve for the four textiles used: Kevlar, Carbon fibre, and two types of Glass fibre.

3.4 Effect of the fabric compaction on the CNT yarns

In order to determine the effect that difference in pressure and consequently, fabrics compaction has in the CNTs electrical resistance, a LVDT (Large Variation Displacement transducer) was used. The LVDT is an electromechanical device that transforms the vertical mechanical movement into an electrical signal and gives information about variations in the thickness of the fibres and fabrics.

It is shown in that CNTs electrical resistance change is more affected by carbon and normal grammage glass fibre fabrics than when using Kevlar and low grammage glass fibre. In case of glass fibre fabrics, CNT fibres are more affected by the vacuum pressure using normal grammage fibres than with low grammage ones.

3.5 Vacuum assisted resin infusion process

Once external factors such as welding, vacuum pressure and fabrics compaction effects on the CNT yarns electrical resistance were studied, vacuum assisted resin infusion (VARI) was carried out in order to test the CNT yarns as potential sensors to detect the resin front advance. Furthermore, with the aim of controlling and automating this resin front advance and reducing the porosity and number of voids, several experiments was performed to dismiss the external factors described above and that can interfere in the electrical resistance measurements, so that at the end spatial position of the resin front advance in the fabrics is measured.

In the Figure 6 different regions of electrical resistance variation related to the different physical processes that were playing during the infusion is shown. These regions are the following:

1. Fabrics and CNT yarns are at Atmospheric Pressure.
2. Beginning of the vacuum pressure.
3. Beginning of the injection. Resin starts to flow. First contact between CNT yarns and resin.
4. The first pressure sensor is wetted by the resin. There is a pressure and electrical resistance increase detected by the pressure sensor 1 and the CNT yarns respectively.
5. The first pressure sensor is completely wetted by the resin and one of the CNT yarns saturated.
6. The resin gets in touch with the second pressure sensor: external pressure carried by the resin is detected by the pressure sensor 2. Electrical resistance in the right CNT yarn continues growing until it is saturated.

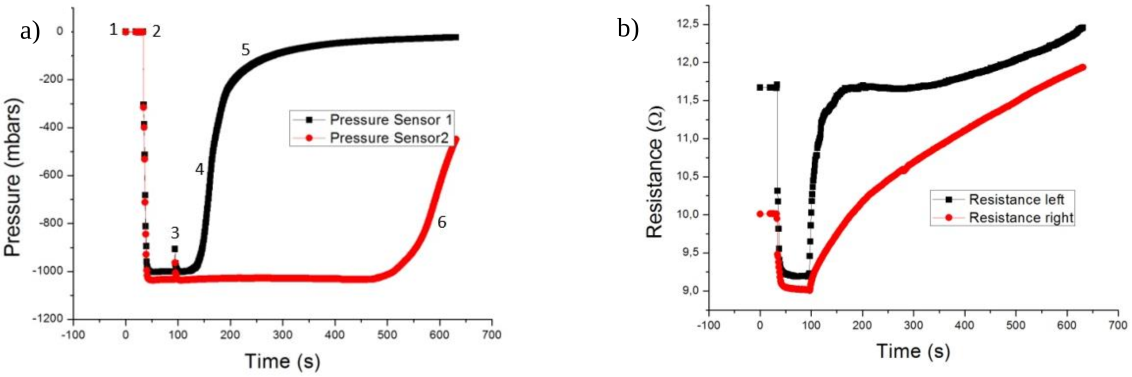

Figure 6**:** Pressure and resistance vs. t time for an infusion process of Glass fibre and Derakane resin

As shown above, electrical resistance of both CNT yarns is reduced when the vacuum pump is turned on and the system passes from atmospheric to vacuum pressure. This electrical resistance drop is mostly due to the indium welding contact. Then, electrical resistance slightly decreases as the CNT fibres are slowly being wetted by the resin. In case we focus on the variation of electrical resistance vs. pressure (Figure 7), instead of plotting the nominal one versus time, an electrical resistance increase is observed when the resin wets the yarns and again a hysteresis cycle can be noticed.

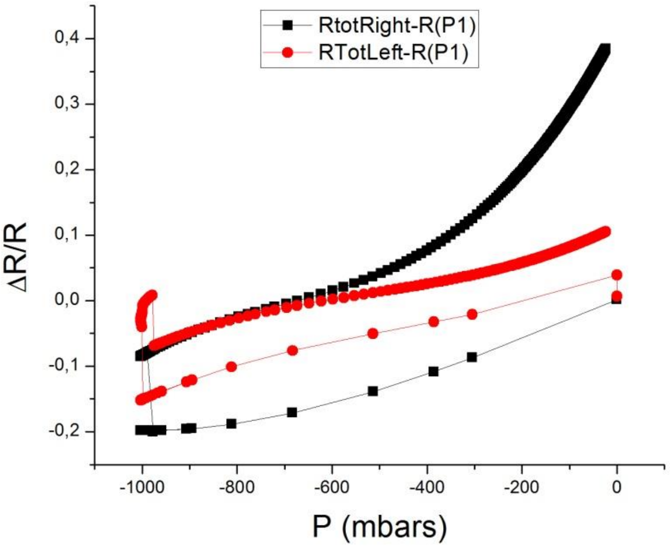

Figure 7: Variation of the change in the CNT yarns electrical resistance vs pressure in the infusion process for the pressure sensor 1

In addition, the resin front advance as a function of time was studied making use of a video camera placed above the infusion table. Results are in agreement with the Darcy's law and are consequence of the capillarity effect. It was seen that at the beginning of the experiment the resin flowed at higher speed than at the end of the test [18].

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3.6 Total contribution of the external factors and the resin impregnation

In order to know the real contribution of the resin impregnation in the fabrics to the CNT yarns electrical resistance, the change in the electrical resistance due to the welding effect and to the fabrics compaction was subtracted to the CNT yarns electrical resistance measured during the infusion process. The variation of the electrical resistance ΔR (Ω) to these external contributions measured is very low: 0.03 Ω for the left and 0.05 Ω for the right CNT yarn.

After subtracting all the external elements that blurred the real signal due to the impregnation of the resin in the fabrics, the new electrical resistance change signal was obtained and it was very similar to the one obtained in Figure 7 when the infusion experiment was performed.

The procedure followed in this paper allowed understanding how much external factors such as welding electrical contacts, vacuum pressure and fabrics compaction affected to the CNT yarns electrical resistance variation during the infusion. As it was described above, CNT yarns change in resistance are not considerably changed due to the resin impregnation in the fabrics.

4. Conclusions

In this paper MWCNT yarns were used to monitor the resin front advance along glass, Kevlar and carbon fibre. VARI (Vacuum assisted resin infusion) using Derakane was the type of infusion performed and MWNTs electrical resistance change was on-line monitored while the resin was impregnating the fabrics. Variables affecting the MWCNTs electrical resistance such as the electrical contacts between the CNTs and sensing electrodes, the fabrics pressure change and their compaction were studied and analyzed. As a result, a hysteresis plot either of the fabric compaction or the MWCNT yarns electrical resistance versus the pressure supported by the fabrics was obtained. It was concluded that these three external factors were not strong enough to modify the MWCNT yarns electrical resistance owed to the fabrics resin impregnation during the infusion. These results make the MWCNTs yarns excellent sensors to determine the position of the resin front advance vs time without the necessity of using neither any visual inspection nor producing any modification in the laminate structure.

References

[1] G. Kopp, E. Beeh, R. Schšll, A. Kobilke, P. Stra§burger, and M. Krieschera, “New Lightweight Structures for Advanced Automotive Vehicles–Safe and Modular,” *Procedia - Soc. Behav. Sci.*, vol. 48, pp. 350–362, 2012.

[2] X. Ning, J. Dang, X. Yue, and J. Yuan, “Properties analysis of novel composites for space robots,” *Polym. Compos.*, vol. 35, pp. 564–569, 2013.

[3] S. Gholizadeh, “A review of non-destructive testing methods of composite materials,” *Procedia Struct. Integr.*, 2016.

[4] A. Mouritz, “Aerospace materials: past, present and future,” in *Introduction to Aerospace Materials*, Woodhead Publishing, 2012, p. 640.

[5] N. Gupta and R. Sundaram, “Fibre optic sensors for monitoring flow in vacuum enhanced resin infusion technology (VERITy) process,” *Compos. Part A Appl. Sci. Manuf.*, vol. 40, no. 8, pp. 1065–1070, 2009.

[6] C. J. Keulen, M. Yildiz, and A. Suleman, “Multiplexed FBG and etched fibre sensors for process and health monitoring,” *J. Reinf. Plast. Compos.*, vol. 30, no. 12, pp. 1055–1064, 2011.

[7] J. S. Kim and D. G. Lee, “Analysis of dielectric sensors for the cure monitoring of resin matrix composite materials,” *Sensors Actuators, B Chem.*, vol. 30, no. 2, pp. 159–164, 1996.

[8] S. Konstantopoulos, E. Fauster, and R. Schledjewski, “Monitoring the production of FRP composites: A review of in-line sensing methods,” *Express Polym. Lett.*, 2014.

[9] M. Hübner and W. Lang, “Online Monitoring of Composites with a Miniaturized Flexible Combined Dielectric and Temperature Sensor,” *Proceedings*, vol. 1, no. 10, p. 627, 2017.

[10] M. Kahali Moghaddam, M. Salas, I. Ersöz, I. Michels, and W. Lang, “Study of resin flow in carbon fibre reinforced polymer composites by means of pressure sensors,” *J. Compos. Mater.*, vol. 51, no. 25, pp. 3585–3594, 2017.

[11] W. Stark and W. Bohmeyer, “Non-destructive evaluation (NDE) of composites: Using ultrasound to monitor the curing of composites,” in *Non-Destructive Evaluation (NDE) of Polymer Matrix Composites: Techniques and Applications*, 2013, pp. 136–181.

[12] M. R. Sanjay, P. Madhu, M. Jawaid, P. Senthamaraikannan, S. Senthil, and S. Pradeep, “Characterization and properties of natural fibre polymer composites: A comprehensive review,” *Journal of Cleaner*

DOI: 10.13009/EUCASS2019-308

Production. 2018.

[13] J. J. Vilatela, R. Khare, and A. H. Windle, “The hierarchical structure and properties of multifunctional carbon nanotube fibre composites,” *Carbon N. Y.*, 2012.

[14] S. Luo, Y. Wang, G. Wang, K. Wang, Z. Wang, C. Zhang, B. Wang, Y. Luo, L. Li, and T. Liu, “CNT Enabled Co-braided Smart Fabrics: A New Route for Non-invasive, Highly Sensitive &large-area Monitoring of Composites,” *Sci. Rep.*, 2017.

[15] J. J. Vilatela and R. Marcilla, “Tough Electrodes: Carbon Nanotube Fibres as the Ultimate Current Collectors/Active Material for Energy Management Devices,” *Chemistry of Materials*, vol. 27, no. 20. pp. 6901–6917, 2015.

[16] J. J. Vilatela and a H. Windle, “A Multifunctional Yarn Made Of Carbon Nanotubes,” *J. Eng. Fibre. Fabr.*, 2012.

[17] M. Al-Bahrani, Z. J. Gombos, and A. Cree, “The mechanical properties of functionalised MWCNT infused epoxy resin: A theoretical and experimental study,” *Int. J. Mech. Mechatronics Eng.*, 2018.

[18] J. C. Fernández-Toribio, A. Íñiguez-Rábago, J. Vilà, C. González, Á. Ridruejo, and J. J. Vilatela, “A Composite Fabrication Sensor Based on Electrochemical Doping of Carbon Nanotube Yarns,” *Adv. Funct. Mater.*, vol. 26, no. 39, pp. 7139–7147, 2016.