Integrated thermo-mechanical monitoring of composites using carbon nanotube yarns

Final report

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Outline of the research conducted

The main purpose of this research was to experimentally investigate the electrical response of carbon nanotube yarns (CNTYs) under mechanical, thermal, and thermo-mechanical stimuli, and integrate them into fiber-reinforced polymer composites to assess their capabilities for structural health monitoring. The research was structured into three yearly stages, as outlined in Fig. 1.

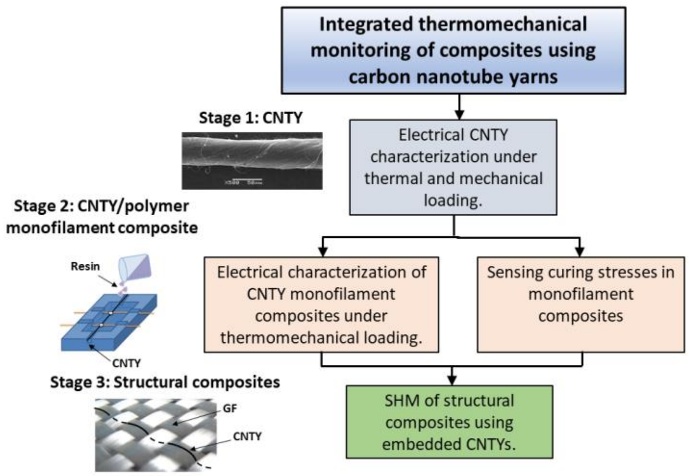

Fig. 1 Research outline, divided into three yearly stages.

The project comprises fundamental research at the individual yarn (stage 1) and single-filament composite (stage 2) levels, as well as applied research regarding the concept implementation into structural composites (stage 3). This final report encompasses the three stages, divided into sections. A no cost extension was granted such as the ONRG projected officially ends in December 2022.

1. Stage 1: Research at the individual yarn level

During the first stage of the project, physicochemical and electromechanical characterizations of the CNTY were conducted at the fundamental (yarn) level. CNTY characterizations conducted during this period included Raman spectroscopy mapping (Fig. 2a), thermogravimetric analysis (TGA, Fig. 2b), and dynamic mechanical analysis (DMA, Fig. 2c), among others. The storage (*E′*) and loss (*E″*) moduli, as well as the damping ratio (*Tan(δ)*) were measured by DMA as a function

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14. ABSTRACT
The purpose of this research is to experimentally investigate the electrical response of carbon nanotube (CNT) yarns (CNTYs, continuous fibers/yarns made by twisting thousands of carbon nanotubes) under mechanical, thermal, and thermo-mechanical stimuli, and integrate them into fiber-reinforced polymer composites to assess their capabilities for structural health monitoring under thermomechanical loading. The project was conducted from September 2019 to December 2022 achieving the proposed goals. 16 journal papers were published from the outcomes of this reseach, 15 presentations in congresses and graduation of 6 students.

15. SUBJECT TERMS

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of temperature (*T*), for several experimental conditions, see Fig. 2c. Additionally, initial research efforts were also conducted in this stage to investigate the electrical response of the yarn upon immersion into liquid epoxy and vinyl ester resins (Fig. 2d). In Fig. 2d, *T* is the temperature, *R* the electrical resistance, and the epoxy resin was cured under a temperature program, while changes in the electrical resistance of the yarn (ΔR) were monitored.

This initial characterization stage was the first step used to build fundamental property-structure relationships for understanding the subsequent experiments on composite materials, conducted in stages 2 and 3.

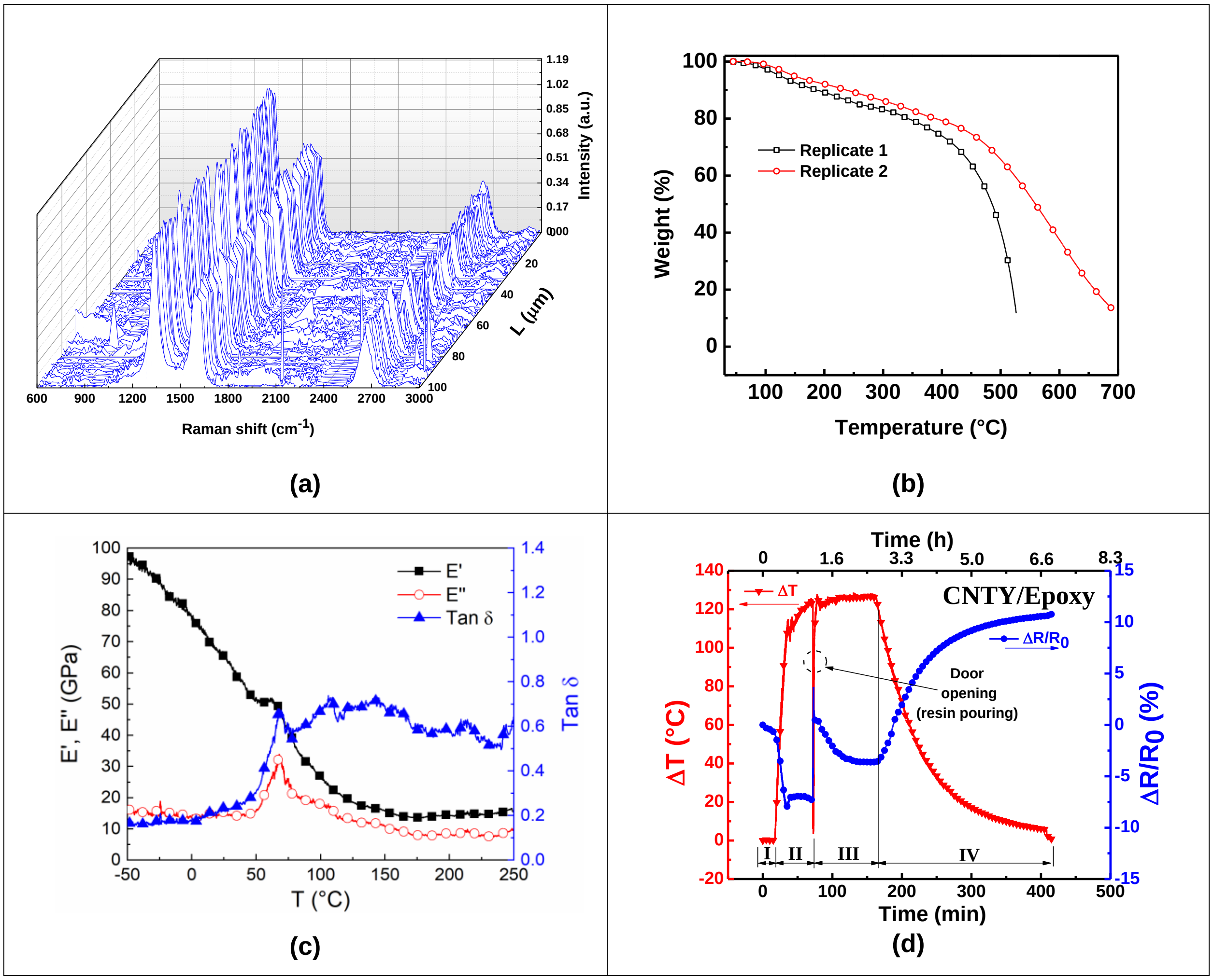
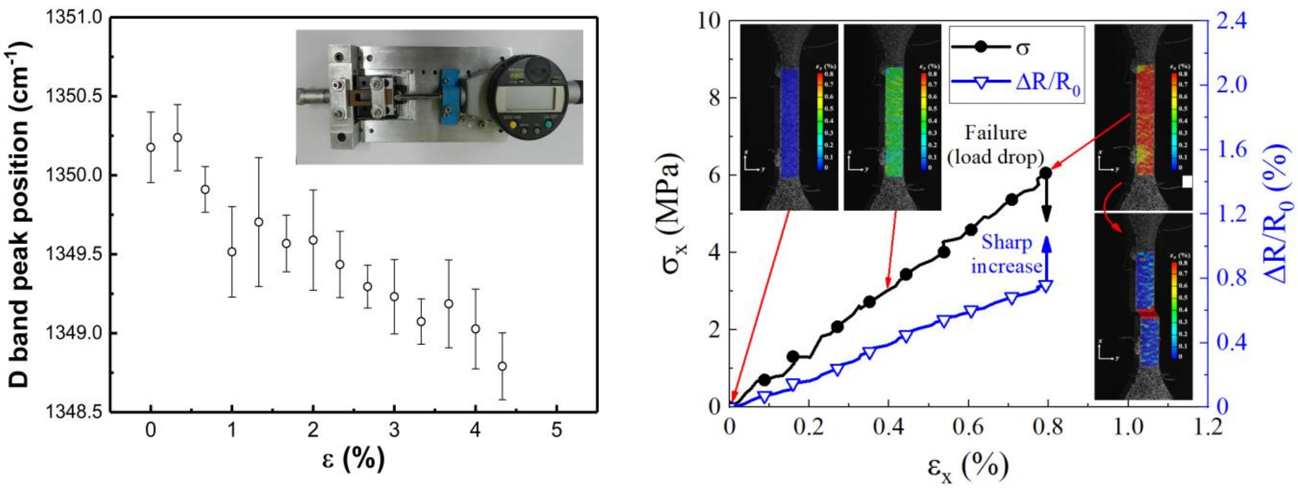


Fig. 2 Fundamental characterization of the CNTY and single filament composites. a) Raman spectroscopy, b) TGA, c) DMA, d) electrical response of the yarn upon curing of an epoxy resin.

2. Stage 2: Single-yarn composite level

Stage 2 of the project involved the finalization of the fundamental physicochemical and electromechanical characterizations of the CNTY at the individual yarn level, and continued with studies at the single-yarn (monofilament) composite level. CNTY characterizations conducted during this period included finalization of DMA testing (Fig. 2c), electrical study of the polymer curing responses (Fig. 2d), in situ Raman mapping of the individual yarn under strain (Fig. 3a), and electromechanical tests coupled with digital image correlation (DIC), Fig. 3b. For the in-situ Raman study, Fig. 3a, a special test rig was designed and constructed to apply strain (ε) to the CNTY inside the Raman equipment, while the Raman shift of the *D*-band is monitored. This Raman study generated new knowledge on the material and structural contributions of the CNTY mechanical response. Electromechanical tests such as those plotted in Fig. 3b allowed to build correlations between the mechanical stress (σ_x)-strain (ε_x) response of the coupon and its fractional change of electrical resistance ($\Delta R/R_0$).

Preliminary research efforts were also carried out in this stage to investigate the electrical response of carbon nanotube-modified fiber reinforced composites to strain and cycling loading damage, stepping into stage 3 of the project.



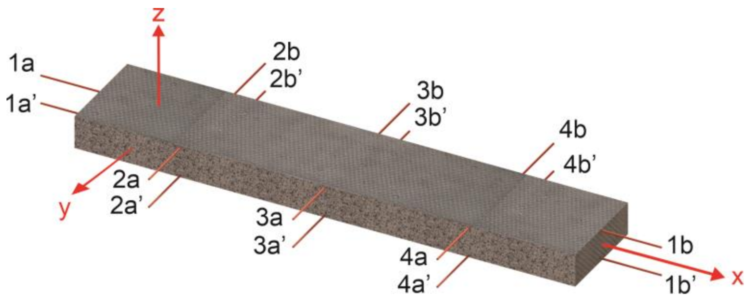
(a) (b)

Fig. 3 Individual CNTY and CNTY-monofilament characterizations. a) Stretching of the CNTY under in situ Raman spectroscopy, b) electromechanical characterization of monofilament composites using DIC.

3. Stage 3: Concept implementation into structural composites

In stage 3 (final stage) the CNTYs were integrated into polymer composites for structural health monitoring purposes. Figure 4 shows a schematic of the electrodes used in the glass fiber/CNTY/vinyl ester composite to monitor the electrical resistance (*R*) in real time during four-point bending loading of the composite. The specimen comprises 8 electrodes per surface (top surface under compression, bottom surface under tension) for measuring *R*. A multiplexing meter allows simultaneous measurement of *R* from the grid of electrodes, to construct spatial electrical maps in real time. With R_0 being the reference electrical resistance at zero strain, i.e. $R_0 = R(\varepsilon =$

0), the fractional changes of electrical resistance ($\Delta R/R_0$) were successfully used to identify and locate non-visible damage within the composite.



Measurement	Electrodes	
	Top surface	Bottom surface
1	1a – 2a	1a' – 2a'
2	1a – 3a	1a' – 3a'
3	1a – 4a	1a' – 4a'
4	1a – 1b	1a' – 1b'
5	2a – 3a	2a' – 3a'
6	2a – 4a	2a' – 4a'
7	2a – 2b	2a' – 2b'
8	3a – 4a	3a' – 4a'
9	3a – 3b	3a' – 3b'
10	4a – 4b	4a' – 4b'

Fig. 4 Glass fiber/CNTY/vinyl ester hierarchical composite and the definition of electrodes for monitoring $\Delta R/R_0$.

Figure 5 shows the electrical response of the grid of electrodes depicted in Fig. 4, as well as the corresponding DIC map of the axial strain (ε_x). The top figure corresponds to the top surface of the four-point bending specimen, which is subjected to compression. The middle figure corresponds to the bottom surface of the specimen, which is subjected to tension. The bottom figure is the DIC map of ε_x for five specific incremental loads (*A* to *E*). Point *A* corresponds to the unloaded state, point *C* to 112 MPa and point *E* to the failure stress (200 MPa). As seen from the figure, the electrical resistance response first increases with a small linear slope, due to piezoresistivity. As the load/strain increases the electrical signal ramps up. Some electrodes present larger increments in $\Delta R/R_0$ and peaks of electrical resistance close to failure, indicating that non-visible damage is occurring in the composite. The location of the electrodes which present the higher electrical resistance changes (4a-4b, 3a-3b, 2a-2b, 4a'-4b') coincide with the location of larger strain gradients indicated by DIC (bottom figure). This evidences that the electrical resistance technique can locate the position of damage within the composite. This damage can be in the form of matrix failure, fiber failure, interface and/or delamination.

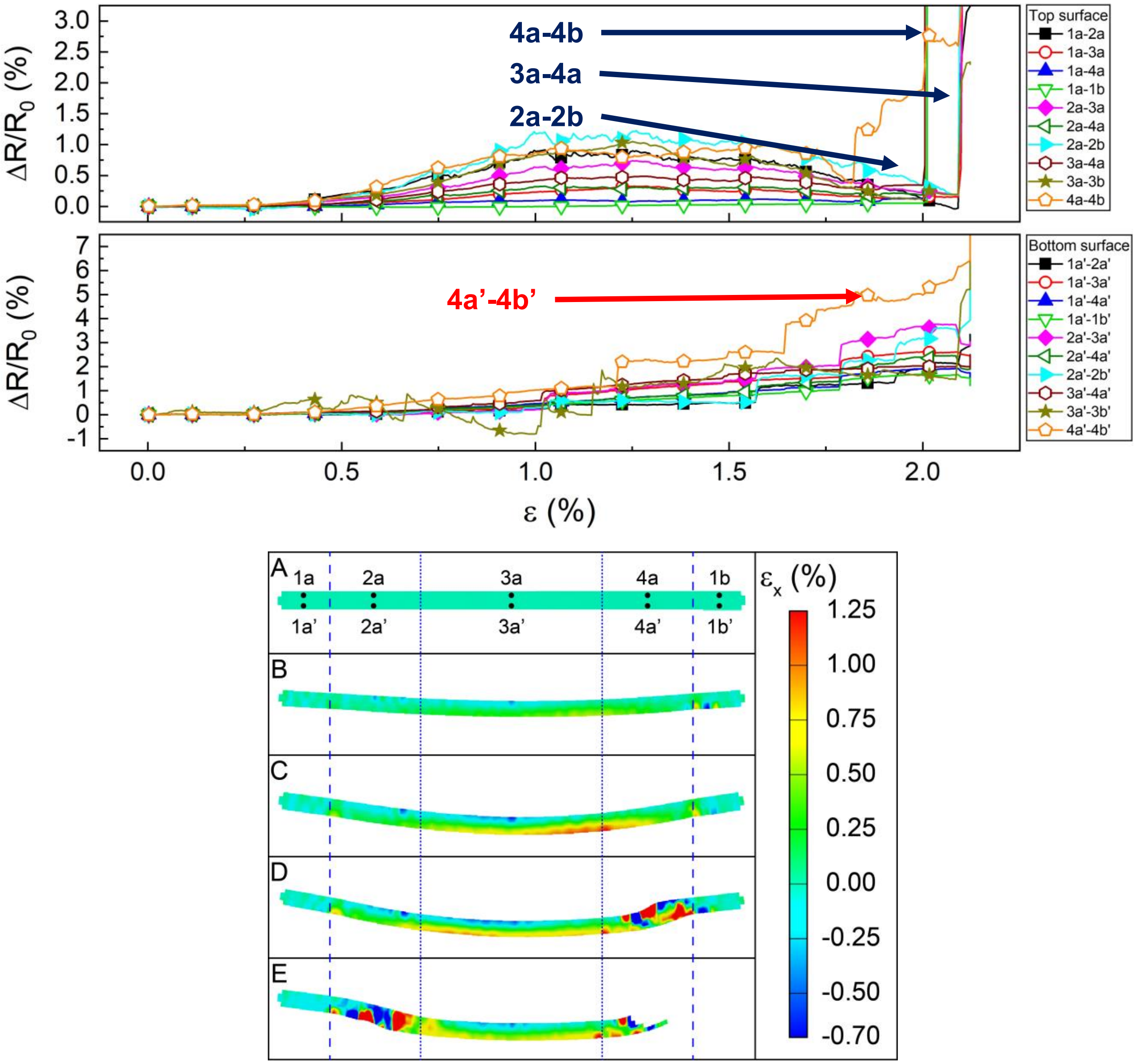


Fig. 5 Electrical resistance response of the grid of electrodes in Fig. 4 and DIC axial strain maps (bottom) for the four-point bending specimen.

4. Additional research ideas and products

Along with the research on the study of CNTYs and their implementation for monitoring the structural health of composites through the electrical response of the yarn, other (collateral) scientific ideas aroused during the research. Some of these ideas were further developed by the student members of the research group and materialized into student´s theses and research products. The additional ideas were around similar topics laid down in the project, but using different material systems. For example, work on investigation of the thermoresistive and piezoresistive sensing responses of polymer composites based on carbon nanotubes and/or graphenic sheets, as well as structural health monitoring of CNT-based hierarchical composites. The outcomes of such research were also published, as can be seen in the list of products in the following section.

5. Academic products

The research and academic products achieved by this project comprise 16 journal papers published in reputable international journals, presentations and publication in proceedings of 15 congresses and symposia, and graduation of 6 students (2 at the BS level and 4 more at the MSc level). Additionally, given that the PhD in Mexico takes at least 4 years, two PhD students who worked in the project (Abraham Balam and César Pérez) are still pursuing the PhD degree. At least two more journal papers are under preparation. All these products explicitly acknowledge ONRG support. The last slide of all oral presentations delivered in the congresses also included acknowledgement to the ONRG support. The detailed list of products follows, where an active hyperlink is included with the *doi* of the journal papers.

Published journal papers (16)

1. "Influence of polymer matrix on the sensing capabilities of carbon nanotube polymeric thermistors", A. Balam, M. Cen-Puc, A. May-Pat, J.L. Abot, F. Avilés. Smart Materials and Structures, 29, 2020, 015012. *https://dx.doi.org/10.1088/1361-665X/ab4e08*.
2. "Influence of electrode configuration on impact damage evaluation of self-sensing hierarchical composites", G. Uribe-Riestra, J. Ocampo-Bello, F. Gamboa, F. Mendoza-Santoyo, C. Pérez-López, E.A. Franco-Urquiza, M. Preud'homme, A. Castillo-Atoche, F. Avilés. Journal of Intelligent Material Systems and Structures, 3(11), 2020, 1416-1429. *https://doi.org/10.1177/1045389X20919979*.
3. Cyclic thermoresistivity of freestanding and polymer embedded carbon nanotube yarns", A. Balam, M. Cen-Puc, O. Rodríguez-Uicab, J.L. Abot, F. Avilés. Advanced Engineering Materials, 22(10), 2020, 2000220. *https://doi.org/10.1002/adem.202000220*.
4. "Electrical resistance sensing of epoxy curing using an embedded carbon nanotube yarn", O. Rodríguez-Uicab, J.L. Abot, F. Avilés. Sensors, 20, 2020, 3230. *doi:10.3390/s20113230*.
5. "Multifunctional sensing properties of polymer nanocomposites based on hybrid carbon nanostructures", R. Pech-Pisté, M. Cen-Puc, A. Balam, A. May-Pat, F. Avilés. Materials Today Communications, 25, 2020, 101472. *https://doi.org/10.1016/j.mtcomm.2020.101472*.
6. "Electro-mechanical properties of thermoplastic polyurethane films and tubes modified by hybrid carbon nanostructures for pressure sensing", C. Pérez-Aranda, Z. Valdez-Nava, F.

Gamboa, J.V. Cauich-Rodríguez, F. Avilés. Smart Materials and Structures, 29, 2020, 115021. *https://doi.org/10.1088/1361-665X/aba9e6*.

7. "Measurement of in-plane and out-of-plane elastic properties of woven fabric composites using digital image correlation", F. López-Santos, A. May-Pat, E.R. Ledesma-Orozco, A. Hernández-Pérez, F. Avilés. Journal of Composite Materials, 55(9), 2021, 1231–124. *https://doi.org/10.1177/0021998320967073*.
8. "Effect of polymer viscosity and polymerization kinetics on the electrical response of carbon nanotube yarn/vinyl ester monofilament composites", O. Rodriguez-Uicab, I. Guay, J. Abot, F. Avilés. Polymers, 13(5), 2021, 783. *https://doi.org/10.3390/polym13050783*.
9. "Electromechanical properties of carbon-nanostructured elastomeric composites measured by digital image correlation", C. Pérez-Aranda, F. Avilés. Composites Part C, 5, 2021, 100161. *https://doi.org/10.1016/j.jcomc.2021.100161*.
10. "Investigation of directional effects on the electrical conductivity and piezoresistivity of carbon nanotube/polypropylene composites obtained by extrusion", A. Balam, R.H. Cruz-Estrada, A. Castillo-Atoche, F. Avilés. Journal of Materials Science, 56, 2021, 14570-14586. *DOI: 10.1007/s10853-021-06223-3.*
11. "A comparative study of the electrical and electromechanical responses of carbon nanotube/polypropylene composites in alternating and direct current", A. Balam, R. Pech-Pisté, Z. Valdez-Nava, F. Gamboa, A. Castillo, F. Avilés. Sensors, 22, 2022, 484. *https://doi.org/10.3390/s22020484.*
12. "Simulation of mechanical response of carbon nanotube yarns under uniaxial tensile loading", A. Pirmoz, J. L. Abot, F. Avilés. Mechanics of Materials, 165, 2022, 104144. *https://doi.org/10.1016/j.mechmat.2021.104144*.
13. "Processing-structure-property relationship of multilayer graphene sheet thermosetting nanocomposites manufactured by calendering", I. Pérez, S. Flores, C. Ortiz, M. Rivero, J.E. Corona, A.I. Oliva, F. Avilés. Polymer Composites, 43(4), 2022, 2150–2162. *https://doi.org/10.1002/pc.26528*.
14. "An ultra-low-power strain sensing node for long-range wireless networks in carbon nanotube-based materials", A. Hernández-Benítez, A. Balam, J. Vázquez-Castillo, J.J. Estrada-López, R. Quijano-Cetina, A. Bassam, F. Avilés, A. Castillo-Atoche. IEEE Sensors Journal, 22(10), 2022, 9778-9786. *doi: 10.1109/JSEN.2022.3162988*.
15. "Functionalization of few-layer graphene sheets and carbon nanotubes for generation of hybrids and their effect on the piezoresistive properties of polymeric nanocomposites", C.A.

Sierra-Chi, M.A. López-Manchado, J.V. Cauich-Rodríguez, F. Gamboa, A.I. Oliva, F. Avilés. Synthetic Metals, 289, 2022, 117121. *https://doi.org/10.1016/j.synthmet.2022.117121*.

16. "Thermo-mechanical properties of carbon nanotube yarns with high energy dissipation capabilities", C. Pérez-Aranda, R. Pech-Pisté, H.J. Carrillo-Escalante, G.C. Uribe-Riestra, F. Avilés. Journal of Engineering Materials and Technology, 145(1), 2023, 011005. *https://doi.org/10.1115/1.4055540*.

Student´s thesis (6)

1. Ian Guay, "Investigation of the effect of resin infiltration on the electrical response of an individual carbon nanotube yarn". Master of Science in Engineering, The Catholic University of America, Washington, DC, EUA, May 1, 2020. Thesis directors: Jandro Abot and Francis Avilés.
2. Iván Gonzalo Pérez Alcocer, "Design, construction and service tests of a three-roll mill calendar for the dispersion of carbon nanostructures in polymeric matrices". National Technological Institute of Mexico, campus Merida, Mechanical engineering. December 1 2021.
3. Pedro Alonso Ayuso Faber, "Implementation of the digital correlation image technique and finite element analysis for the design of a tensile test on composite materials". Autonomous University of Yucatan, Engineering School (Mechatronics). January 5 2022.
4. Gabriela Carolina Uribe Riestra, "Structural health monitoring of fiber-reinforced composite materials utilizing hierarchical carbon nanostructures". Scientific Research Center of Yucatán, Polymeric Materials, Master of Science. December 3 2021.
5. Raúl Armando Pech Pisté, "Piezoimpedance of carbon nanotube yarns and its implementation in the process of thermosetting polymerization sensing". Scientific Research Center of Yucatan, Polymeric Materials, Master of Science. December 7 2021.
6. Gabriel Fernando Arana Riera, "Development of a carbon nanotube strain gage over a polymeric substrate". Scientific Research Center of Yucatan, Polymeric Materials, Master of Science. December 8 2021.

Congresses and symposia (15)

1. "Construction and characterization of a carbon nanostructured strain gage", G. Arana-Riera, F. Gamboa, M.O. González-Díaz, A. Castillo Atoche, F. Avilés. DCMS Materials 4.0 Summer School (on line), Dresden, Alemania, Aug. 17-21 2020. Oral and poster online presentation.
2. "Thermoresistive characterization of monofilament composites based on carbon nanotube yarns", O. Rodriguez-Uicab, L. Nardo, J. Abot, F. Avilés. American Society for Composites, 35th Annual Conference & ASTM D-30 Committee meeting, Stevens Institute of Technology,

NJ, USA, September 14-17, 2020. Oral presentation (online) and publication of conference paper in proceedings.

3. "Comparative study of the mechanical, electrical and piezoresistive properties of graphenic nanocomposites made with three roll mill calender and tip sonicator", I. Pérez, C. Sierra-Chi, M.A. López-Manchado, F. Avilés, XXIX International Materials Research Congress, Cancún, Mexico, August 15-20, 2021. Oral presentation (online).
4. "Design of a through-thickness tensile test for laminated composites using digital image correlation", P.A. Ayuso-Faber, F. López-Santos, C.A. Pérez-Aranda, G.C. Uribe-Riestra, M.A. Rivero-Ayala, A.A. Castillo-Atoche, E.A. Flores-Johnson, F. Avilés, XXIX International Materials Research Congress, Cancún, Mexico, August 15-20, 2021. Oral presentation (online).
5. "Carbon nanotube-based smart hierarchical composites for the development of structural health monitoring strategies", F. Avilés, C. Uribe-Riestra, R.A. Pech-Pisté, C.A. Pérez-Aranda, A.I. Balam-Mena, H.J. Carrillo-Escalante, G. Arana-Riera, M.A. Rivero-Ayala, XXIX International Materials Research Congress, Cancún, Mexico, August 15-20, 2021. Oral presentation (online).
6. "Effect of curing temperature of epoxy resin on the electrical response of carbon nanotube yarn monofilament composites", O. Rodríguez-Uicab, J.L. Abot, F. Avilés, 36th technical Conference of the American Society for Composites, Texas A&M University, College Station, TX, USA, Sept. 19-23 2021. Oral presentation (online).
7. "A computational model for the piezoresistive response of hybrid carbon nanostructured networks", A. Mora, C. Medina, F. Avilés, 36th Technical Conference of the American Society for Composites, Texas A&M University, College Station, TX, USA, Sept. 19-23 2021. Oral presentation (online).
8. "Electrical and electromechanical responses in direct and alternating current of carbon nanotube/polypropylene films fabricated by extrusion", A. Balam, R.H. Cruz-Estrada, Z. Valdez-Nava, F. Gamboa, C.V. Cupul-Manzano, M.A. Rivero-Ayala, A. Castillo-Atoche, F. Avilés. 4th Latin-American workshop on Carbon Materials (TLMC4), November 16-18 2021, IPICYT, San Luis Potosí, Mexico. Oral presentation (online).
9. "Physicochemical properties of graphenic sheets and their correlation with the electrical properties of polymeric composites", H. Canché, A.I. Oliva-Avilés, C. Sierra-Chi, I. Pérez-Alcocer, D. Aguilar-Treviño, P. Quintana-Owen, D.E. Pacheco-Catalán, F. Avilés. 4th Latin-American workshop on Carbon Materials (TLMC4), November 16-18 2021, IPICYT, San Luis Potosí, Mexico. Oral presentation (online).

10. “Property-structure relationship on the mechanics of carbon nanotube yarns”, C. Pérez-Aranda, R. Pech-Pisté, H. Carrillo-Escalante, R. Vargas-Coronado, F. Hernández-Sánchez, R. Ríos-Soberanis, J. Cauich-Rodríguez, F. Avilés. 7th International Conference on Civil Engineering and Materials Science (ICCEMS 2022), April 15-18 2022, Chiba University, Chiba, Japan. Oral presentation (online).
11. “Integration of carbon nanotube yarns into fiber-reinforced composites for structural health monitoring”, F. Avilés, G. Uribe-Riestra, P. Ayuso-Faber, M. Rivero-Ayala, J. Cauich, C. Pérez-Aranda. 7th International Conference on Civil Engineering and Materials Science (ICCEMS 2022), April 15-18 2022, Chiba University, Chiba, Japan. Oral presentation (online).
12. “Thermoresistive properties of polymer composites based on graphene sheets”, C. Medina, A. Mora, A. Balam, F. Avilés. DDC Summer School: Dimensions of Intelligence in Materials, August 22-25 2022, Poster presentation (live).
13. “Carbon nanotube yarns: structure-property relationships and their integration for electrical structural health monitoring”, F. Avilés, G. Uribe, R. Pech, C. Pérez, M. Rivero, A. Balam, G. Pool. 32nd International Conference on Diamond and Carbon Materials, Sept. 4-8, 2022. Lisboa, Portugal. Oral presentation (live).
14. “Damage assessment in hierarchical composites using carbon nanotube-modified glass fibers and carbon nanotube yarns”, G. Uribe, P. Ayuso, M. Rivero, J. Cauich-Cupul, F. Gamboa, A. Hernández-Pérez, A. Castillo, F. Avilés. International Symposium on Polymer Nanocomposites (ISPN 2022), September 28-30 2022. Lorient, France. Oral presentation (live).
15. “Electromechanical response of carbon nanotube yarns in direct and alternating currents”, R. Pech-Pisté, C. Pérez-Aranda, A. Balam, H. Carrillo-Escalante, R. Vargas-Coronado, J.V. Cauich-Rodríguez, A.I. Oliva-Avilés, F. Avilés. International Symposium on Polymer Nanocomposites (ISPN 2022), September 28-30, 2022. Lorient, France. Oral presentation (live).