

Development of Sensing Techniques for Material State Awareness in Natural Fiber Composites

MATTHEW WEBSTER, NOUR HAKIM, LUKE POST,
KARINA HAFEEZ, DAVID WARREN, ERIC TUCKER,
SANDI MILLER and ALLISON CHRISTY

ABSTRACT

Natural fiber reinforced composites (NFRCs) offer a potential alternative to carbon-fiber composites in some aeronautics applications. Active health monitoring may enable broader use of NFRCs by providing the material state awareness needed to reduce design knockdown factors and to monitor for environmental degradation in service. This paper describes initial steps taken toward development of dielectric and acoustic in-situ sensing approaches that would allow moisture and material property changes to be monitored in real-time. Coupons of three different thermoplastic NFRC material systems were exposed to accelerated moisture absorption and changes in properties relative to baseline values were assessed. Strong correlation in both dielectric and acoustic properties with moisture level were observed, with dielectric properties showing reversible, monotonic change, while some acoustic properties changed irreversibly.

INTRODUCTION

Modern aircraft are increasingly made with carbon fiber reinforced polymers (CFRP) to reduce weight and improve efficiency. Weight reduction with CFRP can reduce fuel consumption and, thus, emissions, but introduces other sustainability issues such as production of composite constituents (e.g., carbon fiber tows and epoxy resins) and challenges with recycling CFRP at end of life. There is a need for more sustainable materials that can bolster the carbon fiber supply chain and replace CFRP in some applications to improve sustainability in both manufacturing and end of life management.

Natural fiber reinforced composites (NFRCs) offer a potential alternative to CFRP in some aircraft applications. They can be produced sustainably using bio-based resins and natural fibers (e.g., flax, hemp, jute), and can be efficiently decomposed or recycled at end of life. NFRCs have advanced in the past decade and found applications in high-performance automotive components and sporting goods industries, among others [1]. However, limitations prevent broader use in the aviation industry, especially in structural applications. For example, NFRCs are susceptible to environmental

degradation such as moisture and UV exposure that cause material property changes [2, 3]. They also have lower strength and stiffness than state-of-the-art CFRP.

Structural Health Monitoring (SHM) offers benefits for conventional aerospace materials by allowing reduced operating costs and weight. This is because SHM can decrease the safety margins (i.e., "knock-down factors") that engineers typically include during aircraft design to account for potential barely visible impact damage (BVID) or other undetected damage [3]. When applied to NFRCs, an SHM approach could reduce the required strength specifications for certain components. With lower knock-down factors needed, NFRCs (with SHM) could potentially replace CFRP (without SHM) while adding minimal additional weight. Sensors targeting specific degradation mechanisms associated with NFRCs are needed to fully realize their potential in aviation applications. Monitoring moisture and mechanical properties over the lifetime of NFRC parts could enable condition-based replacement and more accurate digital twins for understanding structural response to various flight conditions. El-Saggagh et al. studied the effects of humidity and stress state on the speed of sound in chopped fiber NFRCs [4] and found strong correlation between wave speed and both humidity and fiber fraction. Moisture and associated material property variation also influences the dielectric properties of materials as shown for glass fiber composites [5] and polymers [6], which offers another possibility for monitoring these materials.

Part of the National Aeronautics and Space Administration's (NASA's) Sustainable Manufacturing of Aircraft (SUMAC) project, sponsored by the Convergent Aeronautics Solutions (CAS) program, involves exploring health monitoring to enable broader application of NFRCs. This aspect of the project is developing sensing techniques that provide material state awareness for NFRC-specific degradation mechanisms to enable use in advanced air mobility (AAM) and other emerging aerospace applications. Sensing methods are being optimized to address two primary challenges for long-term use of NFRCs, namely moisture absorption and mechanical property variation. Bulk ultrasonic and dielectric properties of three thermoplastic NFRC material systems are studied as a function of moisture absorption in accelerated aging experiments. This work is the first step toward optimization and design of a sensing architecture for monitoring NFRCs in service. Such a sensing architecture could make NFRCs viable and cost-effective alternative to conventional aerospace materials in some applications.

MATERIALS

Flax fiber composite laminates were produced at NASA Glenn Research Center using three different bio-derived thermoplastic resins – Pond Global bio-based Pond Cycle IMF 101 HT polylactic acid (PLA), Mango Materials YOPP ST-101 poly-3-hydroxybutyrate (PHB), and Arkema Rilsan bio-based Polyamide-11 (PA-11)¹. Resin sheets were laid up with fiber mats at a mass ratio of approximately 1:1 and were pressed using parameters optimized for each material system. For the PLA and PHB laminates, eight layers of a 2x2 twill of flax (BComp Ltd.) were used as the fiber reinforcement. For the PLA composites, laminates were preheated without pressure for 15 min at

¹ Specific vendor and manufacturer names are explicitly mentioned only to accurately describe the methods used in this study. The use of vendor and manufacturer names does not imply an endorsement by the U.S. Government, nor does it imply that the specified material/equipment is the best available.

380°F, then pressed at 278 psi at 380°F for 10 minutes, pressed at 556 psi at 380° F for 5 minutes, and slowly cooled to room temperature. For the PHB composites, laminates were preheated without pressure for 10 min at 370° F, pressed at 222 psi at 370° F for 10 minutes, pressed at 444 psi at 370°F for 10 minutes, and then slowly cooled to room temperature. For the Rilsan composite, 6-layers of braided flax fiber (A&P) reinforcement was used. The material was preheated for 30 minutes at 248°F, then pressed at 100 psi at 410°F for 30 minutes, and allowed to slowly cool to room temperature. Square 0.75” x 0.75” coupons were cut from each material using a laser cutter. The edges of the specimens were sealed with epoxy prior to environmental exposure.

MEASUREMENT APPROACHES

The section below describes the measurement approaches that were used to assess the response of NFRCs during accelerated environmental exposure.

DIELECTRIC MEASUREMENT

Dielectric properties were measured using an Agilent 4192 LF Impedance Analyzer and Agilent 16451B Dielectric Test Fixture. During test, specimens were placed on the bottom plate of the test fixture and centered. The top electrode was placed on the surface and was tightened to a preset torque level for the fixture. A logarithmic sweep was performed from 1kHz to 13MHz. The dielectric constant was calculated from capacitance, C , as

$$\epsilon_r' = \frac{tC}{\pi\left(\frac{d}{2}\right)^2 \epsilon_0} \quad (1)$$

Where ϵ_r' is the dielectric constant, t is the thickness of the test coupon, d is the diameter of the guarded electrode, and $\epsilon_0 = 8.854 \times 10^{-12}$ F/m is the permittivity of free space (Figure 1).

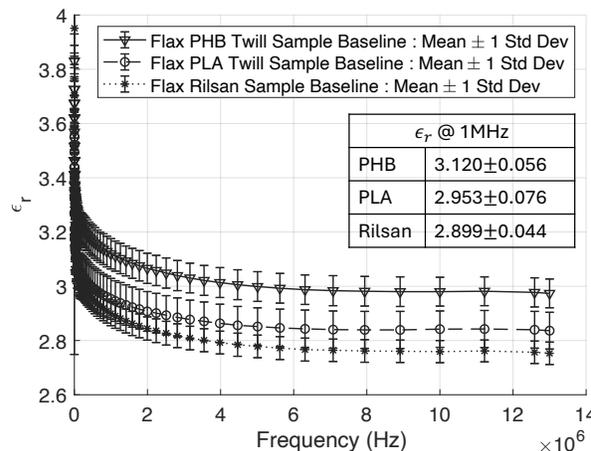


Figure 1: Baseline dielectric measurements for three NFRC materials. (Left) Dielectric test fixture used during measurement. (Right) Frequency-dependent dielectric constant.

ULTRASONIC MEASUREMENT

Ultrasonic measurements were made using a pair of 500 kHz transducers coupled to the samples using Aqualene dry couplant. The transducers were excited with a 500 kHz tone burst using a Ritec RAM-5000 high-power ultrasonic instrument and data was captured using an oscilloscope. The received waveforms were enveloped using a Hilbert transform, and the amplitude and time of arrival of the peak of the envelope were measured. Data was also collected for a reference measurement taken with only the dry couplant and no sample present (Figure 2).

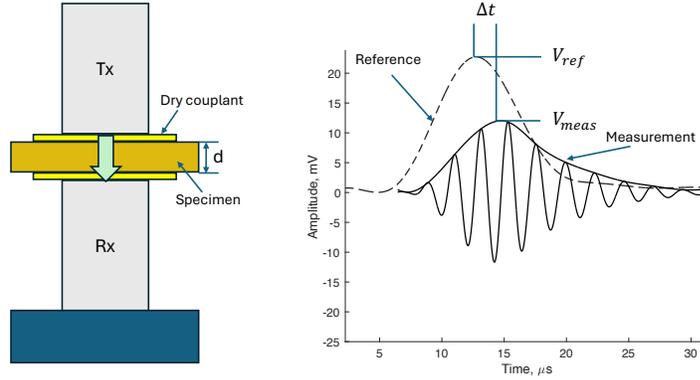


Figure 2: Ultrasonic measurement process. (Left) Illustration of the experimental setup showing the transmitter (Tx), receiver (Rx), polymer dry couplant, and specimen. (Right) Representative data showing the measurement signal, the signal envelope, and the envelope of the reference signal collected without the specimen present.

The relative change in peak time of arrival and amplitude between the reference and specimen measurement were used to calculate relative wave speed and attenuation metrics as

$$c_{rel} = \frac{d}{\Delta t} \quad \alpha_{rel} = \frac{1}{d} \ln \left(\frac{V_{meas}}{V_{ref}} \right) \quad (2)$$

Where d is the specimen thickness, Δt is the difference in peak time of arrival between reference and measurement signals, V_{meas} is the peak amplitude of the measurement signal, and V_{ref} is the peak amplitude of the reference signal.

ACCELERATED ENVIRONMENTAL EXPOSURE

Dielectric and ultrasonic measurements were made as moisture was absorbed during accelerated aging experiments. The experimental setup consisted of a temperature control chamber that was fed compressed air bubbled through a beaker of distilled water. The resulting condition inside the chamber was approximately 99% relative humidity and 77°F. Coupons were placed in the chamber for a period of 1 week and were then allowed to dry outside of the chamber for a period of 1 week in a desiccator at room temperature (approximately 72°F).

The dielectric and acoustic properties of each material were measured daily along with their thickness and weight. In addition, the density was calculated in the dry state using weight and volume measurements of materials cut from the same region as the coupons. The effective density as a function of moisture content was calculated by assuming that any change in volume was due to the addition of water in the microstructure.

RESULTS

Figure 3 shows moisture absorption in NFRC coupons as both water uptake (left) and thickness strain (right) versus cycle of exposure for the three materials. Water uptake of up to 20% by weight was observed in the PLA composite, with the PHB gaining approximately 15%, and the Rilsan gaining the least moisture at 5%. The thickness strain measurements follow similar trends, with maximum values reaching about 25% for PLA. Swelling in the thickness-direction was observed in proportion to the increase in moisture. All specimens retained approximately 2% moisture after moisture exposure and several cycles of drying, but varying levels of residual strain remained, with the most being observed in the PLA indicating potential irreversible swelling in the microstructure.

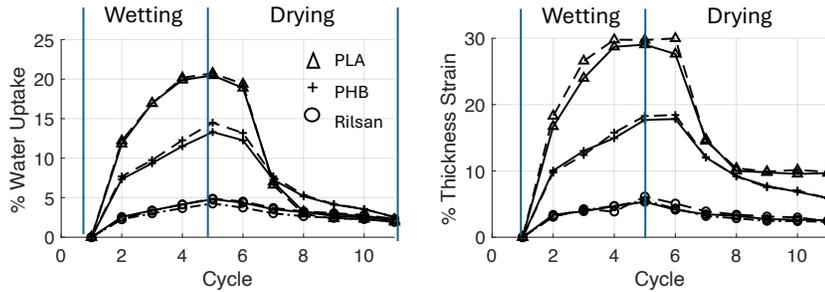


Figure 3: Wetting data for NFRC specimens subjected to one week of humidity exposure (Wetting) followed by one week of drying in a desiccator (Drying)

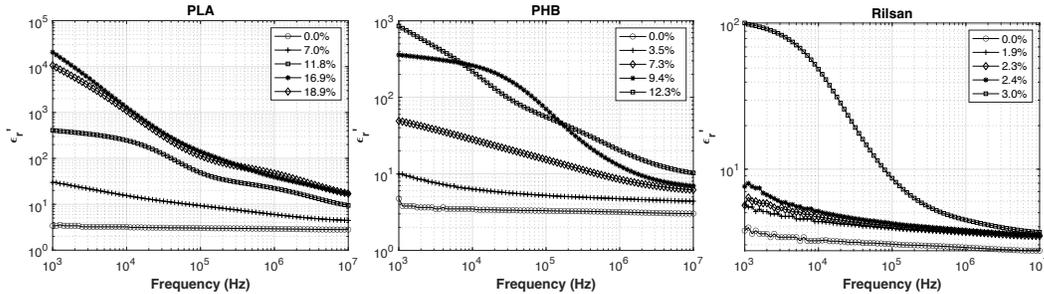


Figure 4: Frequency dependent dielectric constants measured for flax composites having PHB (Left), PLA (Middle) and Rilsan (Right)

The dielectric constant as a function of frequency is displayed in Figure 4, which increased with moisture for all three material systems. The growth in dielectric constant measured at 1MHz showed a generally nonlinear trend (Figure 5). The volume fraction of water was related to theoretical models representing the water and the composite material as capacitors in series or in parallel [7]. The volume fraction of water in the microstructure was calculated assuming that increased mass due to water absorption (Δm) lead to an increase in volume, (ΔV), given as $\Delta V = \Delta m / \rho_w$, where ρ_w is the density of water. The resulting dielectric constant under the series and parallel capacitance approximations, are given, respectively, as

$$(\epsilon_r)_{Series} = \frac{\epsilon_w \epsilon_{Comp}}{\frac{\rho_{Comp}}{\rho_w} \phi_w^m \epsilon_{Comp} + \left(1 - \frac{\rho_{Comp}}{\rho_w} \phi_w^m\right) \epsilon_w} \quad (3)$$

$$(\epsilon_r)_{Parallel} = \frac{\rho_{Comp}}{\rho_w} \phi_w^m \epsilon_w + \left(1 - \frac{\rho_{Comp}}{\rho_w} \phi_w^m\right) \epsilon_{Comp}. \quad (4)$$

Where ϕ_w^m is the mass fraction of water in the material and ρ_{Comp} is the density measured for each material system in the baseline state while ϵ_w and ϵ_{Comp} are the dielectric constants measured at 1MHz for water and the dry composites, respectively. The result of these models is also shown in Figure 5.

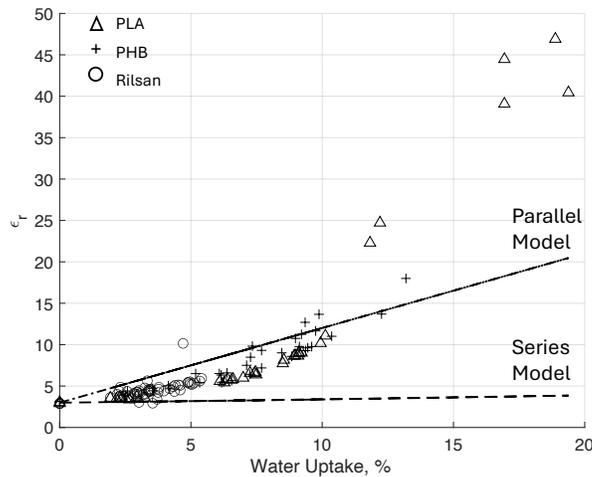


Figure 5: Dielectric constant measured at 1MHz as a function of moisture uptake overlaid with series and parallel equivalent capacitance approximations.

The data in Figure 5 follows most closely with the parallel capacitance model, indicating uniform distribution of moisture within the microstructure [7]. This is especially true for the PHB material, while the PLA material seems to exhibit much larger change in dielectric constant than predicted by either model at high moisture levels.

Ultrasonic measurements on the samples also showed significant change as a function of moisture uptake. The signal envelope from measurements taken for each of the NFRC materials at varying moisture levels is shown below in Figure 6. A clear reduction in the peak amplitude and increase in the peak time of arrival is observed as the moisture level increases. This trend is shown more directly in Figure 7, which is a plot of moisture exposure, in number of cycles, versus wave speed and attenuation of the ultrasound signal.

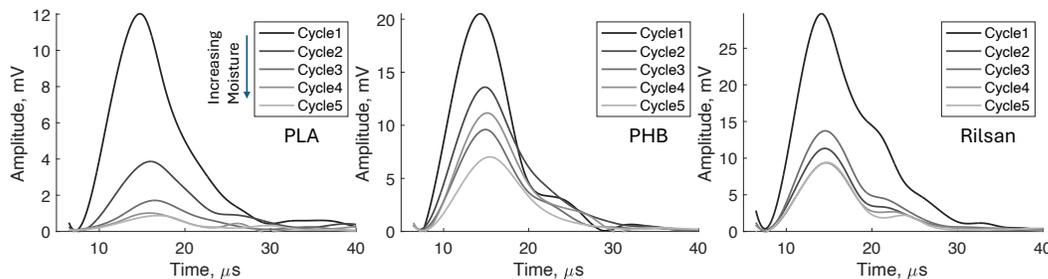


Figure 6: Ultrasonic signal envelopes during the first five wetting cycles. Each shows a reduction in amplitude and increase in peak time of arrival as moisture increases. (Left) PLA, (Middle) PHB, (Right) Rilsan.

As the material absorbed moisture, attenuation increased significantly, and the speed of sound decreased. Wave speed is influenced by both the increased density as water is absorbed and potential reduction in Young's modulus previously reported for NFRC materials exposed to moisture [2].

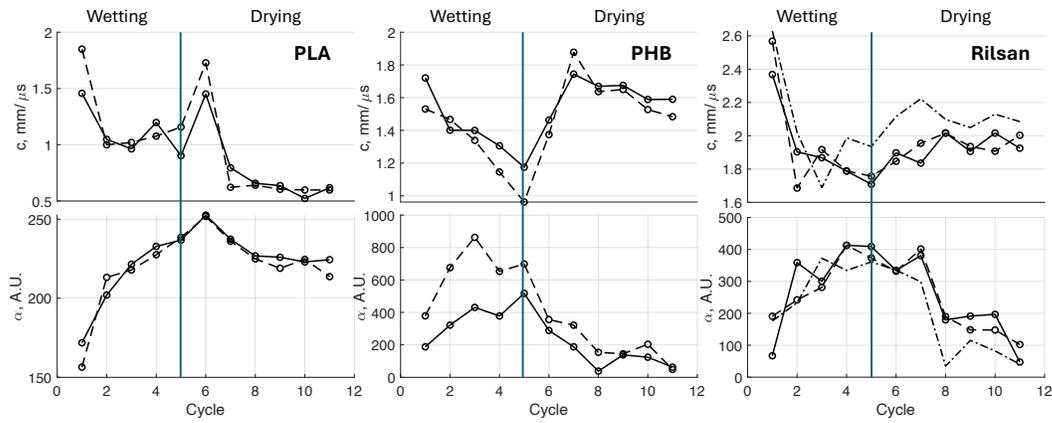


Figure 7: Relative wave speed and attenuation throughout the accelerated moisture exposure and drying cycles.

The effective change in Young's modulus in the direction of wave propagation can be estimated assuming conservation of volume and the relationship between the wave speed, the Young's modulus, and the density, ρ given by

$$c = \sqrt{\frac{E}{\rho}}. \quad (3)$$

Approximations for the Young's modulus calculated by rearranging Equation (3) (Figure 8) show values that are close to those reported for natural fiber composites in the dry state [2]. Reduction in Young's modulus of as much as 80% in PLA and 50% in the PHB and Rilsan composites are observed. While such large changes in properties could be due to damage in the microstructure [3], it is also possible that there are changes in bulk density caused by fiber swelling that is not accounted for under the assumptions applied in these calculation (i.e., that only moisture changed the material's density).

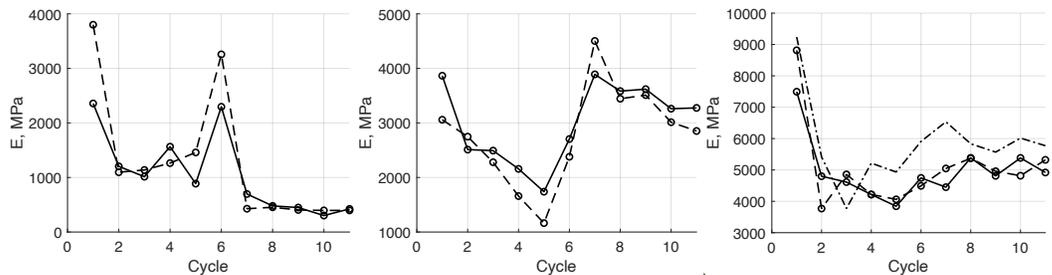


Figure 8: Young's modulus, E , approximated from ultrasonic data throughout the accelerated aging experiment.

DISCUSSION

The work presented here is a necessary first step toward development of surface-mounted sensors for monitoring degradation in NFRCs. It is shown that dielectric and acoustic properties are both sensitive to moisture uptake. The dielectric constant measured for these materials increases at a rate that is closest to the parallel capacitance mixture model, indicating a more even distribution of moisture than has been observed for some polymers [7]. In the PLA material the increase is greater than the parallel capacitor model at the highest moisture levels. Further study of the chemical composition and microstructure of the aged PLA is needed to determine the underlying cause of the high changes in dielectric properties.

Acoustic properties change significantly as NFRC materials are wetted and dried. All materials studied had an increase in attenuation and decrease in wave speed. For the PHB and Rilsan-based composites, changes in properties were mostly reversible, while the PLA showed irreversible and significant change, especially in wave speed. Due to the high levels of moisture uptake and brittle nature of PLA resin observed during manufacturing, irreversible change in wave speed could be due to resin cracking caused by fiber swelling in the matrix and increased void content around the fibers after drying [3].

This work is a first step toward a goal of developing surface-mounted sensors for material state awareness in NFRCs. Dielectric measurements made on these materials feed directly into this goal by providing input into simulations aimed at optimizing printed sensors that are under development. Likewise, the bulk wave ultrasonic measurements aid in developing an understanding of the impacts of moisture on the acoustic response (e.g., the level of attenuation observed at high levels of moisture) as well and providing an initial assessment of the sensitivity of ultrasonic approaches to the magnitudes of elastic property variation that may be expected in service. Both the dielectric moisture sensing and guided wave based ultrasonic material property characterization will be pursued in future work.

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