COVER SHEET

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Title: **Tensile Properties of Unidirectional Polymer Composites Reinforced by Aligned Carbon Nanotube Yarns**

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ABSTRACT

Carbon nanotubes (CNTs), as they possess outstanding mechanical properties and low density, are considered as one of the most promising reinforcements in composite structures. Due to their capability of transferring loads, CNTs in long continuous forms such as yarns and tapes can withstand 20 times as much load as steel can do at the same weight. In this research, carbon nanotube yarns were wound onto an aluminum plate using a custom-built fixture to fabricate a unidirectional strip. Then, by brushing epoxy resin on the strip and laminating four layers, the unidirectional CNT reinforced epoxy resin composite beam specimens were produced. The mechanical properties of the unidirectional CNT-reinforced composite (CNTRC) were determined using standard tensile tests. This study presents a method for manufacturing CNTRC out of CNT yarns, determining the CNTRC's Young's modulus as well as the tensile strength, and obtaining its strain field via digital image correlation (DIC) method. It is observed that the pressure due to sandwiching of the aluminum plates during the manufacturing process leads to nonuniformity of the specimen in the width along midspan of the longitudinal direction which results in the specimen's not being perfectly unidirectional. This phenomenon can cause the matrix cracking in tensile test and reduce the ultimate tensile strength up to 78% in comparison with perfectly unidirectional specimens. However, the Young's modulus of such composites is comparable with those obtained from previously existing research. Also, Results from DIC showed the possible failure prone areas in the specimens, as it presents a up to 64% difference between the highest and lowest strain in the tensile loading direction through the specimens. This study will serve as a foundation for future research involving CNT composites, particularly the use of their high anisotropy to produce auxetic composites with large negative Poisson's ratios.

Keywords: CNT yarns; Unidirectional CNT composites; Filament winding; Mechanical properties

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INTRODUCTION

Carbon nanotubes (CNTs), due to their exceptional mechanical, electrical, and thermal properties, are attracting growing attention in the research community [1–4]. The mechanical properties associated with the CNT composites are influenced by the type and amount of CNT utilized in the composite. One of the emerging types of CNTs is aligned CNT yarns—made by spinning and twisting CNT into long continues yarns [5].

CNTs were first discovered by Iijima in 1991 [6]. They have been considered as the new generation of reinforcements in composite structures [7–9]. CNT composites are usually constructed by integrating CNTs with various matrices, such as metals [10], ceramics [11], and polymers [12]. There are various studies on the characteristics of composite reinforced by chopped CNTs. For instance, in the paper by Zare [13], a model was proposed to calculate the tensile strength of such composites. In another research, Shokrieh *et al.* [14] investigated the effect of adding multiwall CNT to polyester composites. They demonstrated that adding 0.05 wt% of CNT can improve the ultimate tensile strength by 45% compared to specimen without this reinforcement. Also, Hussein *et al.* [15] conducted a research on the effect of adding multi-walled CNT as well as carbon fiber in an epoxy resin composite on the mechanical and thermal properties of such composites. They reported that adding multi-walled CNT to this composite can elevate the impact energy absorption of epoxy composites by 14%. Chen *et al.* [16] utilized multi-walled CNT along with glass fiber to reinforce the core layer of a sandwiched structure. They showed that the interlaminar toughness of the specimen can be increased by 124% by adding the reinforcements to the core layer in comparison with the unmodified sample.

Unlike chopped CNTs, CNT yarns can substantially enhance the load-bearing capabilities of the composites, leading to enhancement in the toughness, tensile strength, as well as fatigue resistance of composites [17–21]. Additionally, the anisotropy of such CNTs make them an ideal choice for manufacturing auxetic composites [22]. In this regard, Fan and Wang [23], using a shear deformable beam theory, investigated the low-velocity impact on the CNT yarn composites. The beam in this study is designed to have a negative Poison's ratio, *i.e.,* auxeticity. Also, Kim *et al.* [24] measured the mechanical properties of aluminum rings covered via CNT yarn composite. They applied dynamic as well as static loadings on the structure. In this study, it has been shown that although adding CNT yarns over the aluminum ring increases the weight by 11%, it can improve the load bearing by over 200%. Additionally, the mechanical characteristics associated with CNT yarn composites were investigated [25]. In this study, they examined the impact of different winding tension on the behavior of the composite. They presented that the tensile strength of CNT composites is 69% of that of CNT yarn itself. Also, Barber *et al.* [26] analyzed the deformation as well as the failure strength of composites made with CNT yarns and polymer. They utilized X-ray to characterize the specimens and showed that sharp gradients and the accumulation of the resin are two major reasons for failure.

Up to now, various studies have been conducted on chopped CNT composites to investigate their mechanical characteristics. However, integrating CNT yarns into matrices to make composite beams and plates is yet to be explored. Thus, in this research, using a custom-built winding device, the CNT yarns were wound over a plate to make CNT strips. Next, by applying epoxy resin as the matrix, unidirectional CNT yarn composites with four layers, *i.e.,* $[0]_4$, were manufactured. The CNT composite strips were analyzed via microscope before and after tensile testing. By testing the specimens via a tensile tester, the mechanical properties of unidirectional CNT composites were obtained and presented. This article will be used as a basis for our future studies associated with auxetic CNT composites, specifically, using the high anisotropy of such composites to make auxetic composites with large negative Poisson's ratios [3, 23, 27, 28].

METHODOLOGY

Specimen Fabrication

The MIRALON CNT yarn made by Huntsman with a diameter of 150 µm (0.6 g/cm^3 density, 240 MPa tensile strength, and 4.9 GPa tensile modulus) was used to fabricate unidirectional CNT reinforced epoxy resin composites. The CNT yarns were wound over an aluminum plate, for which the rotary motion of the winding process was controlled by a DC stepper motor. The distance between adjacent CNT fibers was controlled via a linear motor and a controller. By coordinating the rotary motion of the aluminum plate and the translational motion of the feeder mounted on the linear motor, aligned CNT strips with controllable distance between adjacent CNT fibers can be fabricated. The layout of the motors, controllers, CNT feeder, and aluminum plate is shown in Fig.1. The West System 105/206 epoxy resin was manually brushed onto each layer of the aligned CNT strip prior to winding the next layer. This process was repeated until four layers of laminated CNT/resin composites were made. It is worth mentioning that the tension in the yarns, which was reported as a critical factor that affects the resulting mechanical properties [25], was controlled by the friction between the feeder and roller. After that, the laminate was sandwiched between two additional aluminum plates and placed in a vacuum bag for 24 hours for curing. Then, four coupon specimens with a dimension of 40 mm by 9.5 mm were cut out of the original laminate (100 mm by 11 mm) with waterjet cutting.

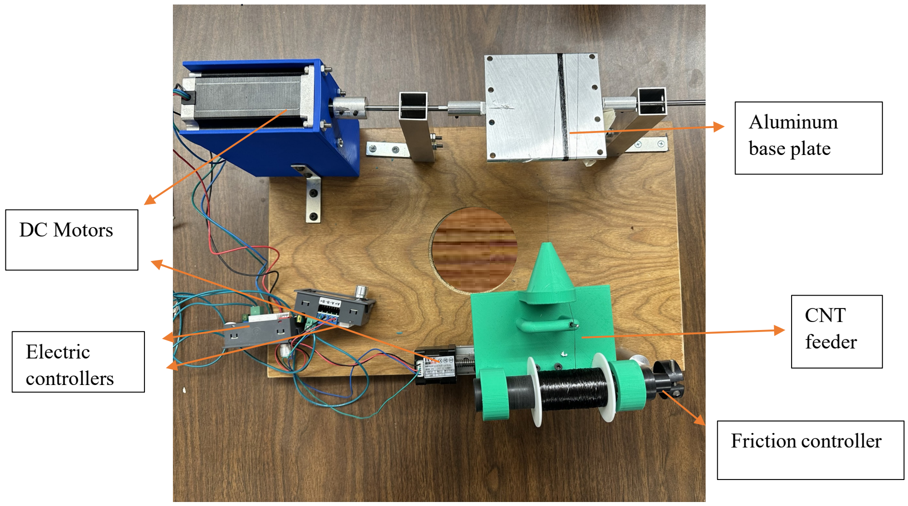


Figure1. Photograph of the custom-built aligned CNT yarn winder, including DC motors, controllers, feeder, and aluminum plate.

Specimen Characterization

Figure 2 shows the cured laminate on the aluminum plate. It can be seen, the CNT-reinforced composite (CNTRC), due to the pressure from the vacuum bag and different friction—the friction between the plate and CNTs at both ends are higher than that in the middle of the plate, has different widths in different locations, resulting in the nonuniformity of the specimens.

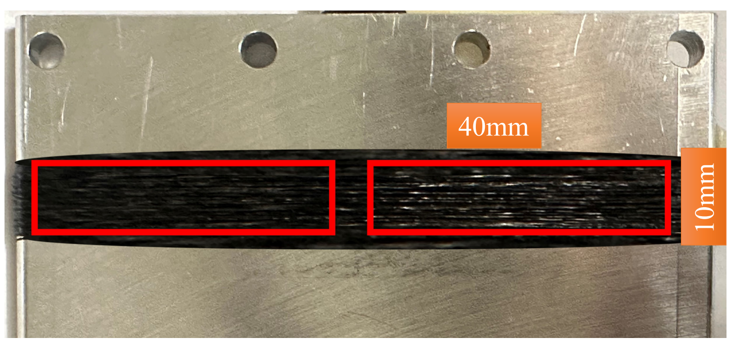


Figure 2. Photograph of the cured CNT composite laminate wound over the aluminum plate.

Also, in order to make sure the specimen was aligned unidirectionally and check for possible defects, a digital microscope, Hirox KH-8700, was employed to examine the CNT/epoxy resin composite specimens. The obtained images from the digital microscope are presented in Fig. 3. As it is observed, the CNTs are aligned together to form a unidirectional composite structure. Additionally, it is observed that there are some spots where the resin has not completely infiltrated, causing the specimen to be dry and porous. These dried regions can be considered as a bult-in crack in the specimen, as they formed a void in the longitudinal direction of the specimen. Also, the regions with proper impregnation of resin can be seen as well.

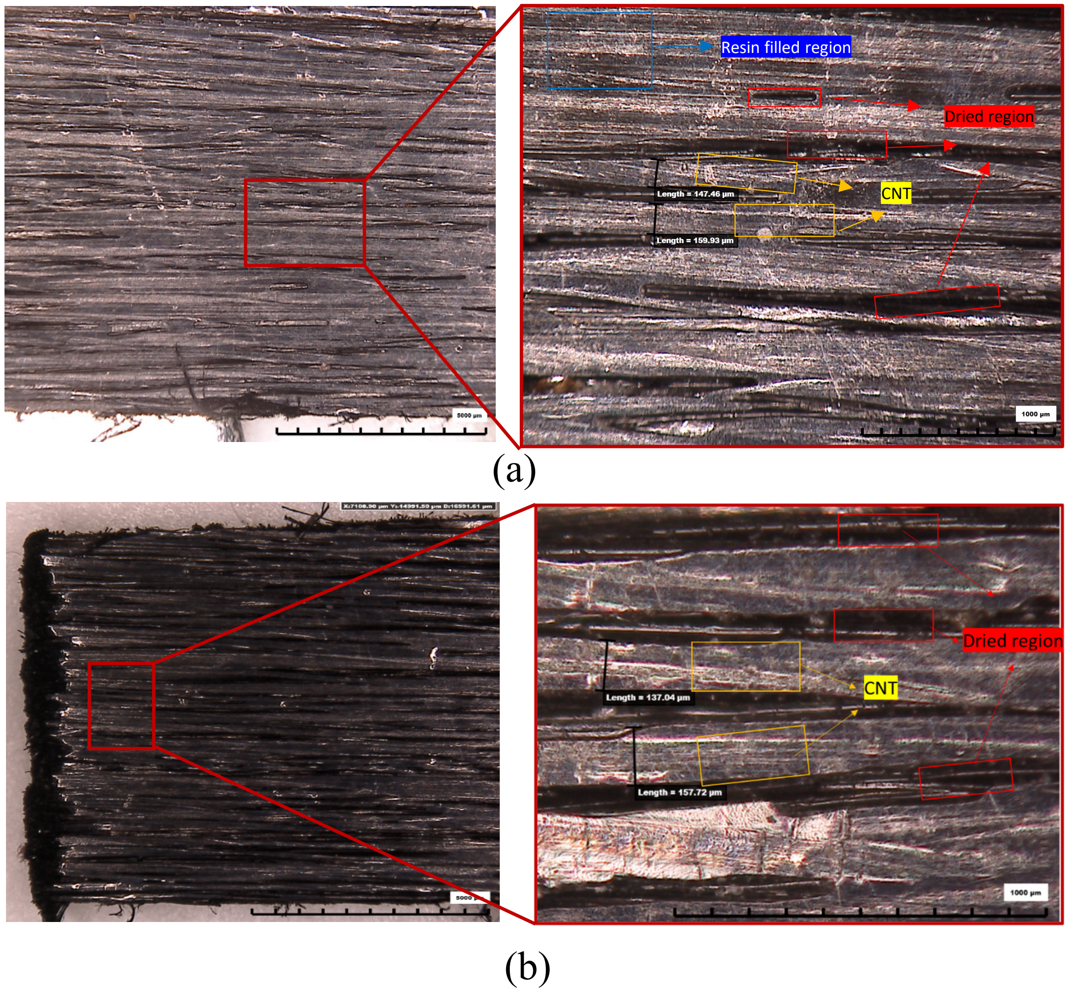
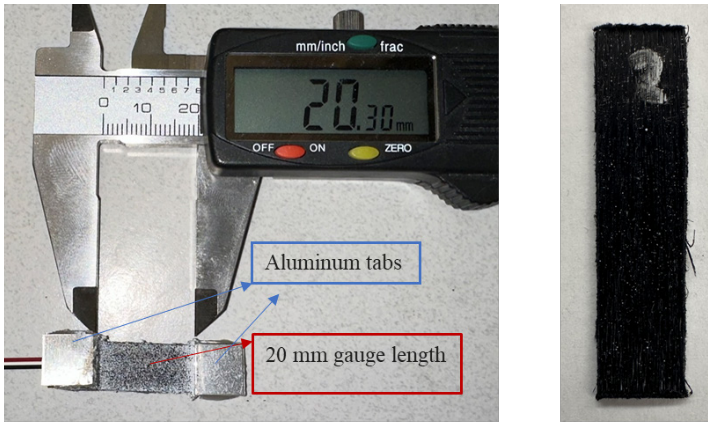


Figure 3. Digital microscope images of the CNT composite specimens: (a) sample #2 (b) sample #3.

Experimental Procedure

The tensile test specimens were prepared by bonding four aluminum tabs of 10 mm by 10 mm to the two ends of the coupon specimens. These specimens were used for tensile tests using an MTS testing machine. To capture the strain in the specimens, both digital image correlation (DIC) and strain gauge were utilized. Figure 4 shows the prepared speckle pattern for DIC. Then, by taking the video during the tensile process, the deformations in the specimens can be recorded and used for DIC analysis via the MATLAB toolbox, *i.e.*, DUODIC [29]. After the tensile tests, each specimen was characterized to identify the failure modes.



(a) (b)

Figure 4. (a) DIC pattern, aluminum tabs, and gauge length on the specimens, and (b) the specimen before bonding tabs and applying DIC pattern.

RESULTS AND DISCUSSION

The obtained tensile properties, including the Young's modulus, tensile strength, and failure strain are shown in Table 1. Our results are compared with those by [25], in which they reported that the Young's modulus and tensile strength of CNTRC are 42.02 ± 4.48 GPa and 873.12 ± 32.64 MPa, respectively, for the CNT/EPON^{TM} 828 composite laminate with two unidirectional layers. Although the Young's modulus is comparable, there exists a large difference between the obtained tensile strength and those in [22]. Two possible reasons for such a difference are: (i) the nonuniformity in the width along the longitudinal direction of the specimen resulted from the vacuum compression of the caul plates which cause the specimens to be not perfectly unidirectional and (ii) the voids due to regions that are not fully impregnated by the epoxy resin during the manufacturing process, as shown in Fig. 2, working as a bult-in crack. It is possible that the nonuniformity and the high porosity of the specimens could have led to more imminent failure of the specimens and hence the lowered tensile strength of the specimens.

Table 1. Calculated Young's modulus, tensile strength, and failure strain for each specimen

Specimen #	Young's modulus (GPa)	Tensile strength (MPa)	Failure strain (%)
1	66.20	187	0.37
2	41.85	236	0.49
3	57.60	435	0.41

Figures 5, 6, and 7 show the strain field ε_{yy}, photograph of the fractured specimens, digital microscope images of the fractured regions, and the microscopic images from specimens 1, 2 and 3 before testing, respectively. It should be mentioned that part (c) of Figures 5, 6, and 7 are the regions in which the failure occurred taken before applying tabs, painting the specimen, and tensile tests. These figures show that the major failure mode in all three specimens was matrix cracking, instead of CNT fiber failure, which also explains the lower tensile strength of the prepared CNTRC specimens in comparison to those reported in [25], as the major failure mode reported were the CNT fiber failure. The reason why that the matrix cracking occur is the fact that the original laminate was not uniform in the width in longitudinal direction, which leads to the specimens not being perfectly unidirectional. It can also be concluded that the nonuniformity in the width along the longitudinal direction and voids causes the specimen to be under nonuniform strain during testing, leading to the matrix cracking failure mode of the CNTRC.

Additionally, the DIC results demonstrated that the specimen #1, #2, and #3 on the right side, where the fracture occurred, are under strain. In other words, these region for specimen #1, #2, and #3 has 30%, 64%, and 34% higher strain compared to regions which are under lowest strain, respectively. Generally, from Figs. 5(c), 6(c), and 7(c), it can be observed that some of the CNT yarns were cut in half and not connected to both ends of the specimens. Specifically, Fig. 5(c) demonstrated that due to the fact that fibers are not aligned perfectly unidirectional, some part of CNTs at the edges are cut which made the specimen prone to failure in those parts. This incident is more observable in specimen #3, *i.e.,* Fig.7(c), in which the specimen failed in the exact same spot that CNT yarn is cut and pulled out. Given this, the load bearing capability of the specimens are significantly reduced, thereby causing the tensile strength to be dramatically reduced. Also, it can be seen that the tensile strength and the Young's modulus of specimen #2 is much higher than the other specimens, as only a small portion of the yarns are cut and the rest of the specimen stayed intact to bear the applied load. In addition to this, specimen #2 failed by matrix cracking towards the center of the laminate, which can be explained by Fig. 6(c), *i.e.,* the microscope image taken before the tensile testing, where the region with a large void due to the lack of resin impregnation acted as the initiation spot of the matrix cracking.

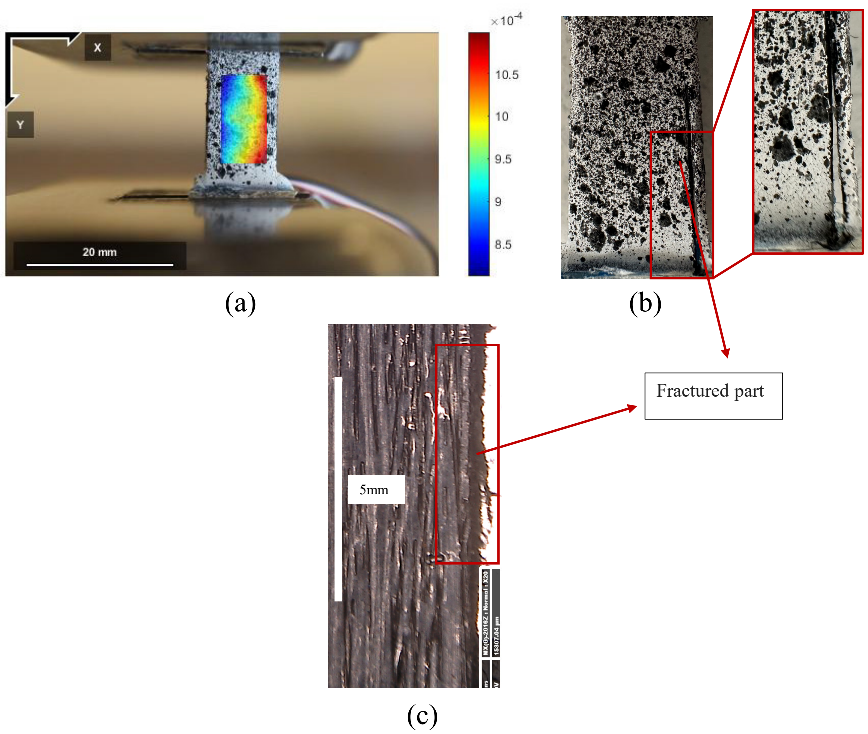


Figure 5. (a) the DIC strain (ε_{yy}), (b) the fractured specimen, and (c) microscopic image before tensile test for specimen #1.

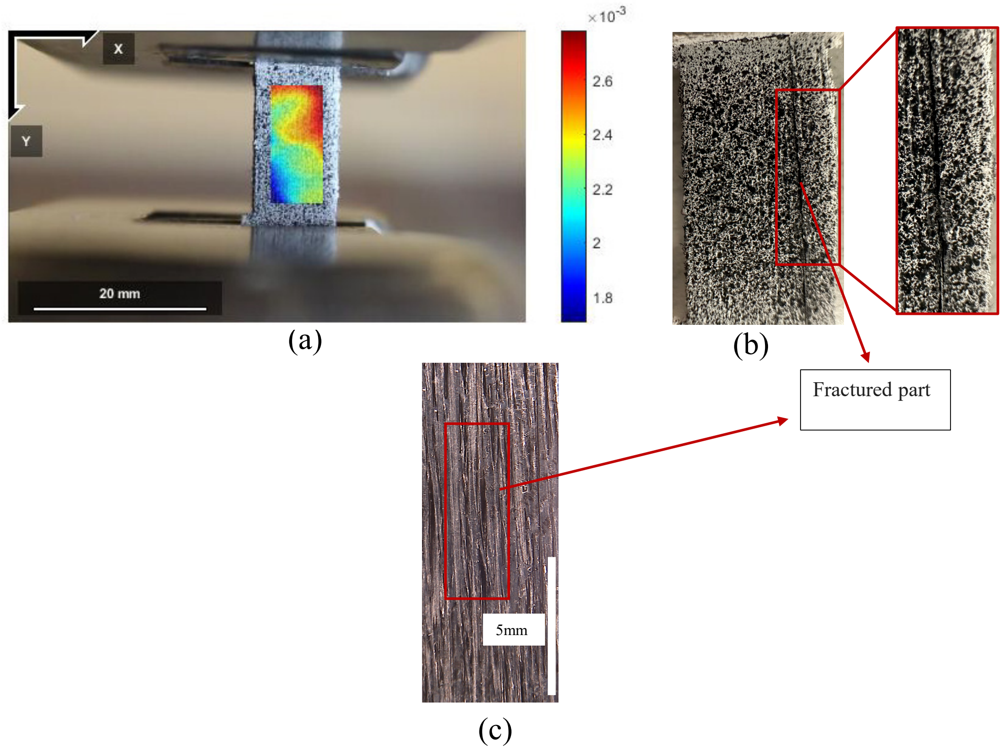


Figure 6. (a) the DIC strain (ε_{yy}), (b) the fractured specimen, and (c) microscopic image before tensile test for specimen #2.

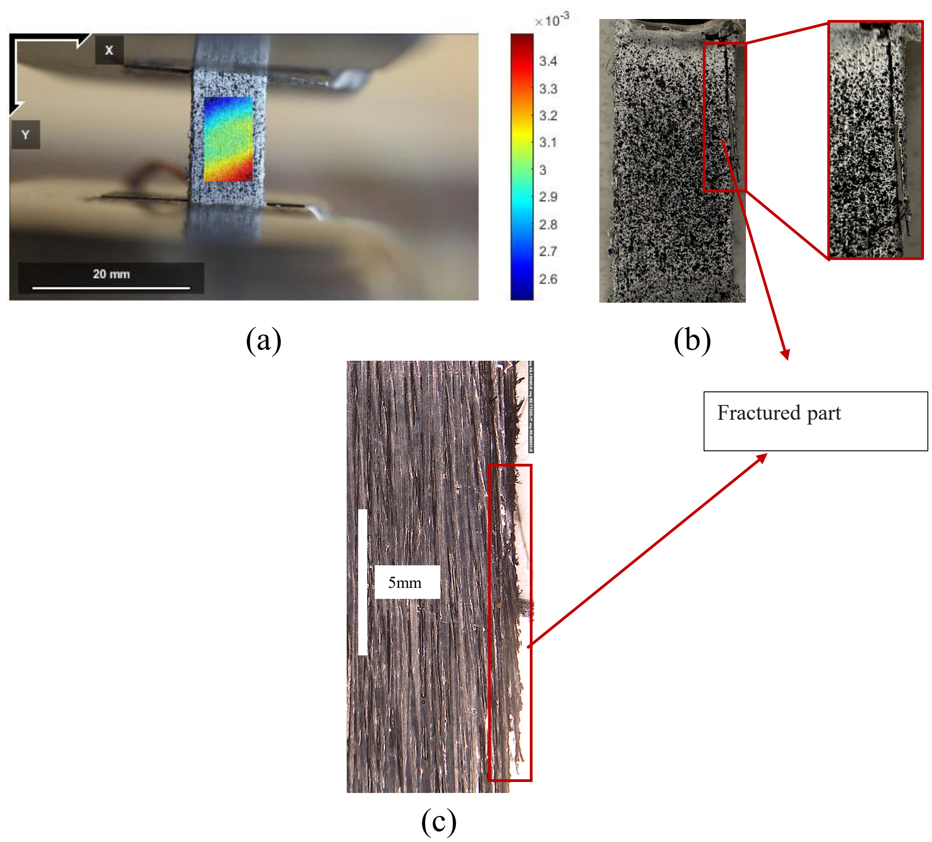


Fig.7. (a) the DIC strain (ε_{yy}), (b) the fractured specimen, and (c) microscopic image before tensile test for specimen #3.

CONCLUSION

The unidirectional CNT/epoxy resin composite laminate specimens with four layers were fabricated using a custom-built fixture for winding the CNT yarns. The specimen was meticulously examined via microscopic images to identify voids and the alignment of the adjacent yarns. Then, using tensile tests, the Young's modulus as well as the tensile strength of prepared CNTRC specimens were obtained. Also, through DIC, the strain field associated with each specimen was presented. The results indicate that the flaws during the manufacturing process, causing the yarns to be curved along the longitudinal direction, led to the specimen's not being perfectly unidirectional. This resulted in the nonuniform strain distribution and matrix cracking failure of the specimens under tension. This can lead to diminishing of the specimens' tensile strength by up to 78% in comparison with an existing study, of which the tensile strength of CNT yarn composites was reported. By looking at the DIC results, the failure prone area can be identified as the strain distribution is nonuniform through the specimen and has up to 64% difference in between high and low strain regions in the tensile loading direction. Due to nonuniformity of the CNT yarns, some of the yarns are cut or narrowed down at the sides of the specimens. Also, it is conceivable that, in the specimens that are more uniform, higher ultimate tensile strength and modulus can be achieved since the yarns bear the applied load and not the matrix. In order to achieve a better CNT reinforced composite, the voids and nonuniformity must be controlled by optimizing the manufacturing process, after which the presented fabrication process can be utilized to manufacture unidirectional CNT yarn prepreg and can be employed to make laminated CNT composites with the desired orientation to produce high negative Poisson's ratios.

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