

Effects of Magnetic Alignment and CNT Agglomeration on Reinforcing Fracture Toughness of Polymers

RICARDO BRAGA NOGUEIRA BRANCO, KOHEI OYAMA
and NAMIKO YAMAMOTO

ABSTRACT

Reinforcing composite materials with carbon nanotubes (CNTs) has the potential to improve mechanical and/or multifunctional properties due to their nano-size. Research has been done on using CNTs to reinforce the interlaminar strength of carbon fiber reinforced composites (CFRPs), but most of the previous work is about randomly oriented carbon nanotubes. Currently, one of the main challenges regarding CNT integration into polymers is mitigating their agglomeration and controlling their dispersion in the polymer matrix. By aligning CNTs with an external field, more tailored structure control can be achieved, and a better understanding of how CNT agglomeration and dispersion relate to external field application and CNT concentration is needed. In this work, we studied the effects of magnetic field magnitude, CNT concentration, and matrix viscosity on CNT agglomeration and morphology. We measured the fracture toughness reinforcement of epoxy-CNT nanocomposites at various CNT concentrations (0.1 vol.% and 0.5 vol.%), magnetic field magnitudes (no field, 180 G, and 300 G), and matrix viscosities (older epoxy-hardener system with higher viscosity and newer epoxy-hardener system with lower viscosity). Our results demonstrated that aligning CNTs with a magnetic field can lead to extra reinforcement when compared to using randomly oriented CNTs if the field magnitude, CNT concentration, and matrix viscosity are selected accordingly. The maximum fracture toughness reinforcement achieved with the higher viscosity epoxy-hardener system (~72%) was with 0.5 vol.% of CNTs with a 180 G field, whereas the maximum reinforcement with the lower viscosity epoxy-hardener system (~62%) was observed for the samples fabricated with 0.1 vol.% of randomly oriented CNTs. COMSOL simulations were also conducted to understand the behavior of CNT agglomeration and alignment at different field magnitudes and CNT concentrations, and were compared with the experimental results. To maximize CNT reinforcement, more work needs to be conducted to address the challenge of CNT agglomeration and dispersion control in a polymer matrix, such as a more in-depth study of how different field magnitudes affect fracture toughness improvement and new methods to improve CNT dispersion.

INTRODUCTION

Nanofillers offer a way to reinforce the interlaminar properties of CFRPs due to their larger surface area and higher specific mechanical properties than micro-sized fillers. In addition, nanofillers provide reinforcement without negatively affecting in-plane properties of composites [1]. Among various types of nanofillers, CNTs exhibit advanced mechanical properties (specific strength of 1036 - 2300 MPa/(g/cm³) and specific stiffness of 75 - 350 GPa/(g/cm³)) [2], hence are the subject of intense investigation for use in reinforcing composites. Due to their high aspect ratio (~500 to 4000) [3], large surface-to-volume ratio, and high crystallinity, CNTs provide significant improvement in mechanical properties of CFRPs, such as in fracture toughness (from 9% [4] to 68% [5] for 0.1 wt.% and from 15% [6] to 152% for 0.5 wt.% [5]).

Major challenges currently associated with integrating CNTs into CFRPs for reinforcement are effective CNT dispersion and CNT morphology control. First, good CNT dispersion within a polymer matrix is essential since agglomeration negatively affects reinforcement capability [4–9]. To improve dispersion, CNT functionalization is the most common method [10,11], which also leads to improved interfacial bonding between the nanotubes and the matrix [10,12]. Second, controlling CNT morphology within the matrix is important to tailor material properties; for example, by aligning CNTs within the interlaminar epoxy layer of CFRPs, an improvement in interlaminar fracture toughness is expected due to a more prominent effect of crack deflection and crack bridging by the CNTs. CNT alignment using a magnetic field is a major topic of investigation in this conference paper. Other types of alignment methods were considered, including electric field and gas flow. Electric field alignment with a direct current leads to filler migration towards electrodes, therefore causing non-homogenous dispersion [13], and electric field alignment using an alternating current lead to CNTs aligning with field lines [14]. Electric field alignment, however, introduced problems such as adding electrodes to the sample and dielectric breakdown in insulating epoxy. Gas flow alignment is not possible with higher viscosity matrices such as epoxy [15]. Alignment via a magnetic field is both contactless and quick [14]. Even though studies have been conducted on magnetic alignment of CNTs [16,17], the research is limited. Here, we explored its effects on CNT structuring within a matrix at various field magnitudes and different CNT concentration.

In this study, polymer nanocomposites (PNCs) containing randomly oriented and magnetically aligned CNTs were tested for their fracture toughness and characterized post-fracture to assess CNT morphology without carbon fibers as the first step to valuate effectiveness of CNT implementation. CNT-epoxy samples were prepared with two CNT volume fractions (0.1 vol.% and 0.5 vol.%), varying magnetic field magnitudes (no field, 180 G, and 300 G), and two matrices with two different viscosities. Samples at each condition were tested for fracture toughness based on the ASTM 5045 standard. CNT morphology on the fracture surface was investigated using SEM imaging. CNT alignment quantification was attempted using Raman spectroscopy. COMSOL simulations were performed to model CNT alignment behavior at the different sample conditions.

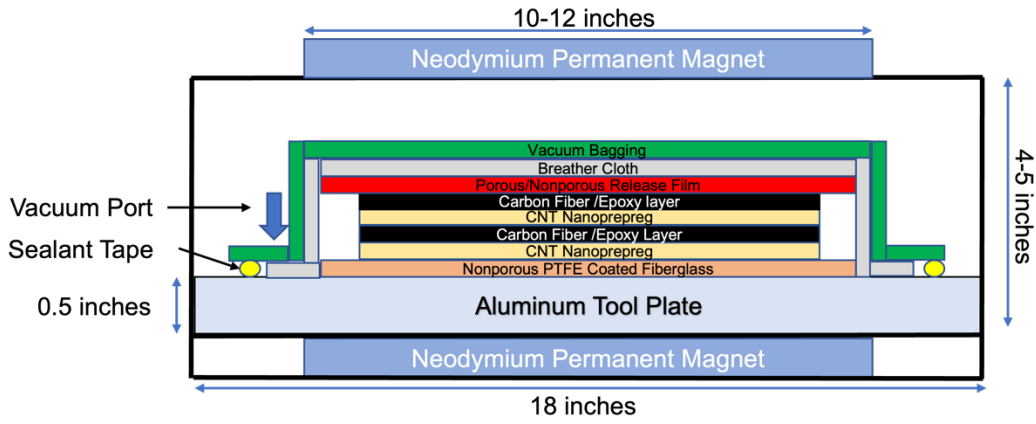


Figure 1. Schematic of the out-of-autoclave process of the stack consisting of nanoprepreg and dry woven carbon fiber layers.

The knowledge obtained from this work will be critical to develop CNT-reinforced CFRPs by effectively using the correct CNT concentration at the right magnetic field magnitude to mitigate the effects of nanotube agglomeration while tailoring CNT assembly within the matrix to maximize interlaminar fracture toughness reinforcement. Out-of-autoclave (OOA) processing of CFRPs with partially cured (B-staged) layers infiltrating the dry fibers is being studied in conjunction with this work to help achieve the goal of fabricating CFRPs with magnetically aligned CNTs with the OOA preparation method (see Figure 1).

METHODS

CNT Preparation

The carbon nanotubes were synthesized in house by chemical vapor deposition (CVD) with the same process previously used in our lab [18]. The resulting CNTs were $\sim 200 \mu\text{m}$ in length, measured using SEM (Thermo Fisher FESEM Verios). From TEM (FEI Titan), the average number of walls was observed to be 3, and the inner and outer diameters were measured to be $6.75 \pm 0.91 \text{ nm}$ and $9.50 \pm 1.68 \text{ nm}$, respectively. A CNT density of 0.822 g/cm^3 was calculated from Equation (1), which is equivalent to the equation for CNT density derived in [19]. The assumptions for this calculation were that each CNT wall possess the same specific surface area of a sheet of graphene ($s = 1315 \text{ m}^2/\text{g}$) [19], the walls are spaced equally, and the inner volume was included for the total volume calculation. After synthesis, CNTs were coated with an 80 nm layer of nickel, based on previous work [14,20], using the same electron beam evaporator machine used for the substrate coating.

$$\rho_{MW} = \frac{2n}{sd_{out}^2} [d_{in} + d_{out}] \quad (1)$$

CNTs are naturally diamagnetic [2,21,22], but Ni-coated CNTs were shown to be ferromagnetic [10], which was necessary in the case of this study since we magnetically aligned the CNTs, and we needed a filler with higher magnetic susceptibility. After CNTs were synthesized and coated with the nickel layer, they were functionalized to

improve dispersion in the polymer matrix and bonding strength in the filler-matrix boundary. The covalent functionalization process through diazotization was chosen based on previous work [12,14]. To eliminate excess solution, a filtering process detailed in [18] was used, which consisted of flowing the solution in a syringe (Advantec MFS Pp25 Syringe Filter Holder) with a membrane of 0.2 μm pore size (Foxy Life Sciences Hydrophobic PTFE EZFlow Membrane Disc Filter, 25 mm diameter) to retain the functionalized CNTs. Dimethylformamide (DMF) was used to release the CNTs from the membrane into a separate beaker. After the filtering process, the solution was centrifuged with DMF until the solution was clear. Finally, the CNTs were centrifuged with acetone twice to eliminate the DMF. The functionalized Ni-coated CNTs were characterized using optical microscopy in a DMF solution, and their average agglomerate length was measured to be $14 \pm 4 \mu\text{m}$ and $3 \pm 4 \mu\text{m}$, respectively.

PNC preparation and testing

The CNT-epoxy nanocomposites were fabricated using the same process for all samples, only varying the CNT concentration, magnetic field magnitude, and epoxy system viscosity. The chosen matrix system was a 100:26.4 mixture of aerospace grade epoxy (EPON 862 resin, $\rho_{\text{epoxy}} = 1174 \text{ kg/m}^3$) to hardener (EPIKURE W, $\rho_{\text{hardener}} = 1019 \text{ kg/m}^3$). Samples were made with both 0.1 vol.% and 0.5 vol.% of CNTs, and for each concentration, samples were fabricated with randomly oriented CNTs, CNTs aligned with 180 G, and CNTs aligned with 300 G. For all these conditions, fabrication was done with both an old, more viscous, epoxy-hardener system (opened for more than two years), and a newer, less viscous, epoxy-hardener system (opened for less than six months). With these parameter variations, we aimed to assess the effects of CNT concentration, magnetic field magnitude, and matrix viscosity on fracture toughness reinforcement.

The three-point bending test was conducted following the ASTM 5045 standard to obtain the fracture toughness data for all samples. Using an aluminum mold, samples were set to follow the dimension relations of the standard [23] (30.8 mm in length, 7.0 mm in width, 3.5 mm in thickness) and a notch (1.1 mm in depth and 1 mm in width). A natural crack had to be introduced in the notch location by lightly tapping the sharp edge of a razor blade to generate a natural crack (see Figure 2a). In addition, the length of the notch and the natural crack, which is the total crack length (a), had to be measured using an optical microscope (Olympus BX51WI, 5x) to follow the standard's rule that a/W , where W is the sample width, must be between 0.45 and 0.55. Such single edge notched bending (SENB) samples were tested using an electro-mechanical universal test system (MTS Criterion, 1 kN load cell, 100 Hz acquisition rate, 10 mm/min loading rate).

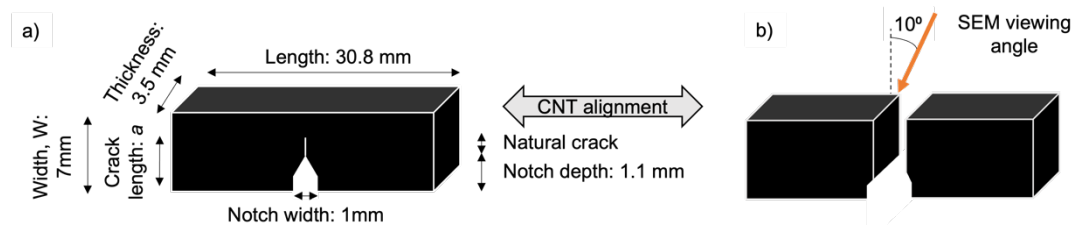


Figure 2. a) Three-point bending specimen dimensions and b) SEM viewing angle of fracture surface.

Sample Characterization and Simulations

After PNC samples were used for the three-point bending test, they were characterized with Scanning Electron Microscopy (SEM) and Raman Spectroscopy. First, SEM analysis was performed to assess CNT morphology on the fracture surface. The sample inspection was done at an angle to capture CNT content on the surface as shown in Figure 2b. Second, Raman spectroscopy (Horiba LabRam Vlabnir) was conducted as an attempt to characterize the degree of CNT alignment within the sample. It has been shown [24] that the G peak magnitude in the spectra increased when the laser polarization direction matched with the CNT alignment direction. In our test, however, results were inconclusive since the G peak magnitude remained virtually constant with varying laser polarization directions.

COMSOL simulations were conducted to assess the agglomeration and alignment behavior of CNTs under magnetic field application. By using ellipsoid particles in a 2D mesh, simulations were performed for both 0.1 vol.% and 0.5 vol.% of CNTs at both 180 G and 300 G magnetic field magnitudes. A model for the viscous forces and torques on ellipsoid particles was used [25], and the interpretations from the different simulated cases will be discussed in detail in the following section.

RESULTS AND DISCUSSION

Fracture Toughness Data

Fracture toughness data was gathered for samples fabricated with an old, more viscous, epoxy-hardener system (opened for more than two years) and with a new, less viscous, epoxy-hardener system (opened for less than six months). The baseline value was calculated by averaging results from at least three samples, as per the ASTM 5045 standard. The fracture toughness with the old epoxy system was measured to be $0.428 \pm 0.027 \text{ MPa}\cdot\text{m}^{1/2}$ whereas that for the new epoxy system was $0.551 \pm 0.119 \text{ MPa}\cdot\text{m}^{1/2}$. A higher crosslinking density for the new epoxy system is the likely explanation for this discrepancy since an older epoxy system oxidizes over time [26]. The value provided by the manufacturer for the EPON 862 and EPI-W system with the same mixing ratio is $0.811 \text{ MPa}\cdot\text{m}^{1/2}$ [27]. Lidston et al. [28] also observed different fracture toughness values ($0.650\text{--}0.850 \text{ MPa}\cdot\text{m}^{1/2}$) for their baseline samples when compared to the value provided to the manufacturer, and this difference can be explained by the variations in the natural crack introduced by tapping the notch with the razor blade, as required per the ASTM 5045 standard.

For the old resin-hardener system (see Figure 3), all samples containing CNTs were observed to provide fracture toughness reinforcement over the baseline, but not linearly with increasing CNT volume fraction. Samples prepared with 180 G field application, the fracture toughness increased with increasing CNT content, with the maximum reinforcement ($\sim 72\%$) obtained for samples with 0.5 vol.% of CNTs. On the other hand, samples fabricated with no magnetic field application showed no significant variation in fracture toughness with changing CNT concentration. Finally, when a 300 G field was applied, the fracture toughness decreased with increasing CNT content. Regarding fracture toughness reinforcement with varying magnetic field for the same CNT concentration, we can see that the reinforcement is not significantly changed for the

samples with 0.1 vol.% of CNTs. For the samples with 0.5 vol.% of CNTs fracture toughness improved when a 180 G field is applied, and then decreased with a higher field of 300 G. This nonlinear behavior in the trends can be attributed to CNT agglomeration, which is exacerbated with both higher CNT concentration and higher magnetic field magnitude.

For the new resin-hardener system (see Figure 4), no significant fracture toughness reinforcement was observed with either 180 G or 300 G magnetic field application for both 0.1 vol.% and 0.5 vol.% of CNTs. Conclusive reinforcement is only seen for the samples of randomly oriented CNTs, with the largest improvement being of ~62% for the samples with 0.1 vol.% of CNTs. With 0.5 vol.% of CNTs, possibly due to higher CNT agglomeration, reinforcement was significantly lower (~17%). Varying the magnetic field application resulted in different reinforcement behaviors depending on the CNT content. For 0.1 vol.% of CNTs, there is a decrease in toughening with increasing magnetic field, whereas for 0.5 vol.% of CNTs, the reinforcement is virtually negligible for all samples. It is possible that with the new epoxy-hardener system, due to its lower viscosity, CNTs agglomerated with more ease than the samples with the old epoxy-hardener system. This result suggests that a lower magnetic field magnitude provides maximum reinforcement capability for a lower viscosity matrix. Additionally, such lower reinforcement was observed with the new epoxy-hardener system since the baseline sample had a higher fracture toughness value.

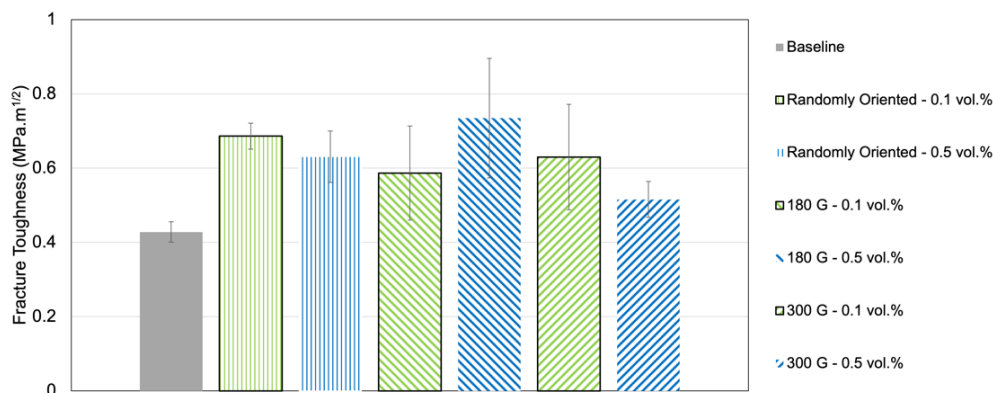


Figure 3. Fracture toughness data for samples fabricated with the old epoxy-hardener system with varying CNT concentration and magnetic field magnitudes.

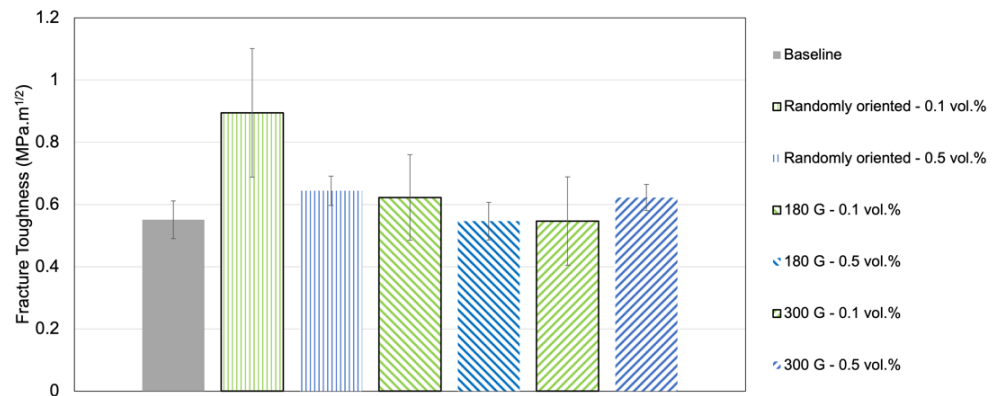


Figure 4. Fracture toughness data for samples fabricated with the new epoxy-hardener system with varying CNT concentration and magnetic field magnitudes.

SEM Imaging

With increasing magnetic field magnitude, the density of the CNT agglomerates was observed to increase for both epoxy systems and at different CNT concentrations. As we can see from the SEM images in both Figure 5 and Figure 6, increasing the magnetic field strength led to denser agglomerate regions. For example, in Figure 5a (no field) CNTs appear to be spaced more evenly than CNTs in Figure 5c (300 G), despite both images showing clear epoxy-rich regions. This trend is also seen in Figure 6, where the CNT agglomerate regions become more packed as the field increases.

The trends observed with the SEM assessment agree with the trends obtained from the fracture toughness data for samples made with the old epoxy-hardener system and the new epoxy-hardener system. First, for the case of samples made with the old epoxy-hardener system, the highest fracture toughness improvement was with the 180 G field application and 0.5 vol.% of CNTs. From the SEM images (see Figure 5), we can see that at 180 G field (see Figure 5b), the CNTs seem to be better dispersed and with smaller epoxy-rich regions than in the cases with no field application (see Figure 5a) and with 300 G (see Figure 5c). Second, in the case of samples fabricated with the new epoxy-hardener system, the highest fracture toughness reinforcement was observed with the samples with randomly oriented CNTs. The SEM images (see Figure 6) also show agreement with the data trend, since the CNT agglomerate regions are observed to be more agglomerated at higher fields than with no field application. Hence, in this case, the negative effect of CNT agglomeration in fracture toughness reinforcement outweighed the benefits of having aligned CNTs in the matrix. This again demonstrates the fine tuning required of various parameters, including matrix viscosity, CNT concentration, and magnetic field magnitude, to maximize reinforcement capability.

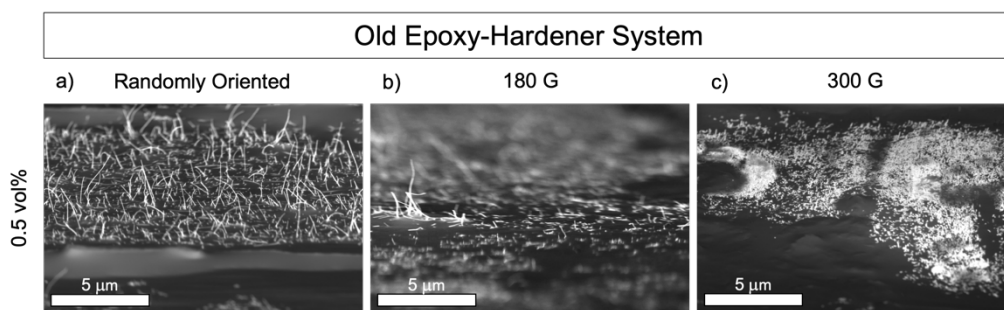


Figure 5. SEM images showing the fracture surface of PNCs prepared with the old epoxy-hardener system, with 0.5 vol.% of CNTs, with a) no field, b) 180 G, and c) 300 G.

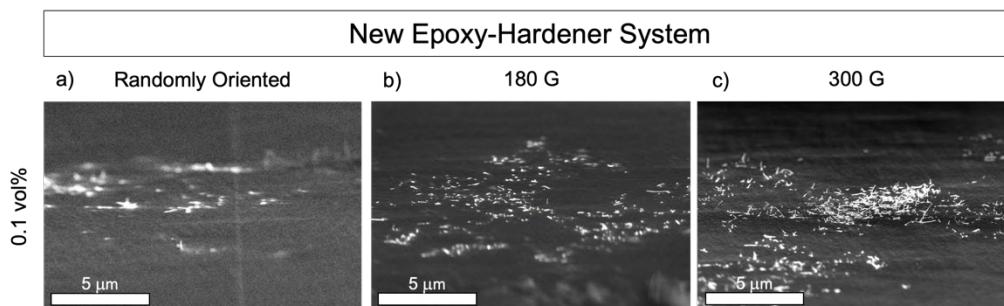


Figure 6. SEM images showing the fracture surface of PNCs prepared with the new epoxy-hardener system, with 0.1 vol.% of CNTs, with a) no field, b) 180 G, and c) 300 G.

COMSOL Simulations

COMSOL Multiphysics simulations were conducted to gain insight about CNT agglomeration behavior and magnetic field alignment control, and then compared with the experimental trends. To model CNTs in COMSOL, ellipsoids were used since the analytical solutions for viscous forces and torques are available [25]. The ellipsoid particles had aspect ratio of 7.5 and volume of $62.8 \mu\text{m}^3$, where the volume was obtained from optical microscopy measurement of over 100 CNT agglomerate regions. The simulation area was set as a 2D square plane of $340 \mu\text{m}$ side length having $42.5 \mu\text{m}$ infinite domain layers. Using a MATLAB code, initial particle orientation and location were set randomly. The ellipsoid material was set as Ni to simulate the Ni-coated CNTs, with the surrounding fluid having viscosity of 70 cP and density of 998 kg/m^3 to represent the EPON 862/EPI-W resin-hardener system used in the experiments. After the meshing was complete, magnetic fields of 180 G and 300 G were applied to the COMSOL model for both 0.1 vol.% and 0.5 vol.% of CNTs.

Simulation results (see Figure 7) displayed similar trends with those from experimental data and SEM imaging: more agglomeration occurring at either the higher 0.5 vol.% of CNTs or higher magnetic field of 300 G, and more localized alignment with the combination of smaller volume fraction of 0.1 vol.% of CNTs and 180 G magnetic field application. For the samples with 0.1 vol.% of CNTs, it was observed that at 180 G, CNTs did not assemble into an agglomerate at 0.2 s. On the other hand, the first CNT agglomerate formed at only 0.082 s when a 300 G field is applied. For the samples of 0.5 vol.% of CNTs, CNTs assembled faster for the case of 300 G field application. For the samples with 0.5 vol.% of CNTs, application of both 180 G and 300 G fields led to faster assembly time when compared to the 300 G field case at the lower 0.1 vol.% of CNTs, demonstrating that both a larger field magnitude and a larger CNT concentration contribute to faster agglomeration time.

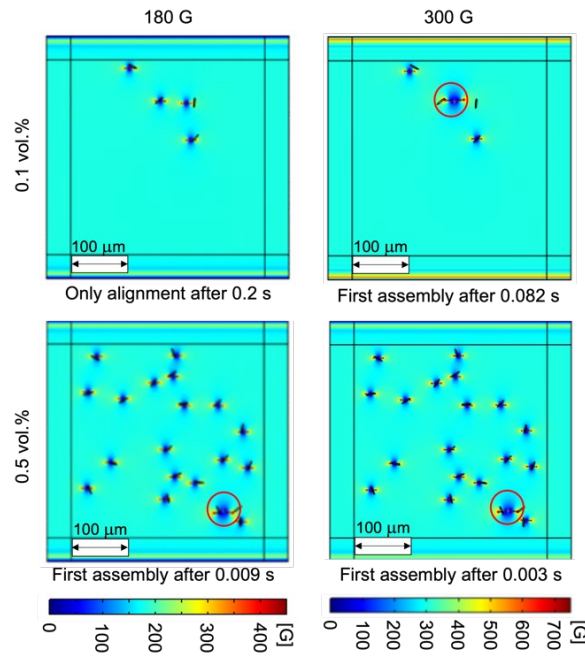


Figure 7. COMSOL simulations representing CNT alignment at 180 G and 300 G fields at both 0.1 vol.% and 0.5 vol.% of CNTs; the red circle indicates the moment a pair of ellipsoids first assemble.

Comparison with Previous Work

Fracture toughness data of CNT-reinforced epoxies from this study was compared with the results in literature [4–8,28–34]. The results from the reviewed literature all contained samples containing randomly oriented CNTs, except for the study conducted by Lidston [28] which had aligned CNTs in the samples. In addition to fracture toughness data measurements, many authors also characterized their samples using SEM [4–8,29,30,32–34] and observed CNT toughening mechanisms originating from CNT pullout and/or crack bridging. Toughening mechanisms, including CNT pullout, were also observed in this work. In contrast to other authors, we also assessed CNT morphology based on varying magnetic field magnitude and not only varying CNT concentration.

First, fracture toughness data from this work was compared with the data in literature where the CNTs were functionalized. Among the samples with functionalized CNTs, this work showed the best fracture toughness improvement percentage for both 0.1 vol.% (~62%) and 0.5 vol.% (~72%) of CNTs (see TABLE I). Relatively lower fracture toughness of baseline samples and aligned CNTs without much agglomeration are possible reasons why the reinforcement in this work is higher than that achieved by others. A common trend was observed with the samples of functionalized CNTs: fracture toughness improvement increases with increasing CNT fraction in a nonlinear fashion. This nonlinearity was also observed by other researchers such as Torabi [33] (see TABLE I), where fracture toughness increases and starts to reach a plateau at the larger 1 wt.% concentration.

Second, in addition to the past work about functionalized CNTs, the past work with pristine CNTs without functionalization was summarized and compared. The largest fracture toughness improvement observed from the reviewed literature was by Bisht [5] (0.1 vol.% of CNTs: 68%; 0.5 vol.% of CNTs: 152%). Compared with this work, our reinforcement percentage for 0.1 wt.% of CNTs was not significantly higher, but the toughening achieved for 0.5 wt.% of CNTs was over 200%. This high toughening reinforcement, without functionalization, can be attributed to well dispersed CNTs since their lengths are smaller (1-3 μm) than in other work (~10-30 μm [4,8,29,33,34] and ~14 μm in this work), and to an effective dispersion method of ice bath sonication [5]. Regarding the fracture toughness reinforcement trends with increasing CNT concentration, fracture toughness increases and then decreases after a certain CNT fraction value (> 0.5 wt.% [5,8], > 0.6 wt.% [4], > 3 vol.% [28]), as seen in TABLE I. This trend can be explained by understanding that a higher CNT concentration leads to more agglomeration, which in turn hinders reinforcement after a certain threshold.

TABLE I. FRACTURE TOUGHNESS DATA COMPARISON WITH PREVIOUS WORK.

	CNT morphology	CNT mass fraction (%)	CNT Functionalization	Test method	Fracture toughness of baseline [MPa.m ^{1/2}]	Fracture toughness improvement (%)
This study (old epoxy system)	Randomly oriented	0.08 (0.1 vol.%)	Covalent diazotization	Three-point bending	0.428	60.30
	Aligned at 180 G					37.08
	Aligned at 300 G					47.15
	Randomly oriented	0.48 (0.5 vol.%)				47.41
	Aligned at 180 G					71.66
	Aligned at 300 G					20.49
This study (new epoxy system)	Randomly oriented	0.08 (0.1 vol.%)	Covalent diazotization	Three-point bending	0.551	62.4
	Aligned at 180 G					12.89
	Aligned at 300 G					-0.80
	Randomly oriented	0.48 (0.5 vol.%)				16.95
	Aligned at 180 G					-0.82
	Aligned at 300 G					12.96
Chandra et al. [30]	Randomly oriented	0.5	Amino-functionalized	Three-point bending	1.720	17.44
Benra et al. [31]	Randomly oriented	0.5	Elastomer covalently bonded	Three-point bending	0.450	45.00
Cha et al. [7]	Randomly oriented	1	Noncovalent	Three-point bending	1.010	96
		2				93
		3				100
Xu et al. [32]	Randomly oriented	0.7	None	Three-point bending	0.670	38.80
Torabi et al. [33]	Randomly oriented	0.1	Functionalized (type not specified)	U-notched semicircular bend	1.340	15.67
		0.3				25.37
		0.5				32.89
		1				35.07
Zeinedini et al. [4]	Randomly oriented	0.1	Not specified	Three-point bending	0.800	8.54
		0.3				15.85
		0.5				24.39
		0.6				26.83
		0.7				21.95
Bisht et al. [5]	Randomly oriented	0.1	None	Three-point bending	2.500	68.00
		0.2				100.00
		0.3				128.00
		0.5				152.00
		1				44.00
Lidston et al. [28]	Aligned	(3 vol.%)	None	Three-point bending	~ 0.800	12.50
		(9 vol.%)			~ 0.850	-5.88
		(11 vol.%)			~ 0.650	0.00
Opelt et al. [6]	Randomly oriented	(0.15 vol.%)	None	Compact tension	0.870	17.24
		(0.5 vol.%)				14.94
		(1.5 vol.%)				3.45
Gojny et al. [34]	Randomly oriented	0.1	Amino-functionalized	Compact tension	0.650	21.54
		0.3				23.08
Ayatollahi et al. [8]	Randomly oriented	0.1	Not specified	Three-point bending	1.619	15.19
		0.5				26.31
		1.0				19.21

Discussion on Magnetic Field Alignment

From the experimental results in this study (see TABLE I), it is possible to see that applying a magnetic field to align CNTs can lead to fracture toughness reinforcement over a sample with randomly oriented CNTs. The positive effect of magnetic field application in this work is seen on the case of samples prepared with the old epoxy system (higher viscosity) with a 180 G field and 0.5 vol.% of CNTs, which had a ~72% reinforcement over the baseline. However, using a higher field magnitude (300 G) at the same conditions yielded ~20.5% reinforcement. Moreover, using a resin system with lower viscosity (new epoxy-hardener system) in fact decreased fracture toughness by ~1% compared to the baseline. Hence, a tradeoff exists between the level of CNT alignment, which improves toughening effects, and the level of CNT agglomeration, and hinders reinforcement. This tradeoff depends on the magnetic field strength, CNT concentration, and matrix viscosity.

Our hypothesis on how to maximize fracture toughness reinforcement using magnetic field application is that a lower magnetic field magnitude is needed with decreasing matrix viscosity or with increasing CNT concentration. To prove this hypothesis, however, more tests must be done at different magnetic field magnitudes. The 180 G field application proved to be beneficial in increasing the samples' fracture toughness with the old epoxy system (higher viscosity) with 0.5 vol.% of CNTs, but fracture toughness decreased or stayed virtually constant for the samples made with the new epoxy system (lower viscosity). Moreover, it is expected that a lower field magnitude than 180 G would maximize reinforcement for samples made with the new epoxy system. In addition, other changes to the current processing method can potentially yield positive effects in maximizing reinforcement. The COMSOL simulations showed that the alignment time is on the order of less than a second for CNTs to begin aligning, which is much faster than the time during experiments dedicated to aligning the CNTs (30 minutes). Moreover, either reducing the layer of the nickel coating on the CNTs or the time allotted for alignment could mitigate agglomeration effects due to the magnetic field application. However, this would have to be confirmed experimentally as part of possible future work.

CONCLUSIONS AND FUTURE WORK

In this work, PNC samples were prepared following the ASTM 5045 standard to gather fracture toughness data of samples at different CNT concentrations (0.1 vol.% and 0.5 vol.%) with different magnetic field magnitudes (no field, 180 G, and 300 G); these conditions were replicated for both an old epoxy-hardener system and a new epoxy-hardener system to assess the effects of viscosity change in CNT morphology and fracture toughness reinforcement. The highest fracture toughness improvement for samples made with the old epoxy-hardener system (higher viscosity) was of ~72% with a concentration of 0.5 vol.% of CNTs at a 180 G field magnitude. On the other hand, for the new epoxy-hardener system (lower viscosity), the highest reinforcement was observed at ~62% for samples made with 0.1 vol.% of CNTs with no field.

Although sample characterization via Raman spectroscopy was inconclusive, SEM characterization of the fracture surface provided insight in agreement with trends observed from the experimental data. The samples with more dispersed CNTs

corresponded to samples with higher fracture toughness reinforcement. A higher magnetic field application was observed to lead to more CNT agglomeration in most cases. The COMSOL results agreed with experimental observations since a higher field magnitude also corresponded to more agglomeration in the simulations. Other factors such as CNT concentration and matrix viscosity must also be considered to maximize reinforcement potential. We observed through SEM imaging that CNTs tend to agglomerate as the CNT concentration increases and as the matrix viscosity decreases. This directly corresponds to why the highest fracture toughness improvement with the new epoxy-hardener system was achieved with no field application and at 0.1 vol.% of CNTs, since this lower viscosity system allows CNTs to agglomerate more easily. Furthermore, we hypothesize that for the new epoxy-hardener system, a magnetic field lower than 180 G would maximize reinforcement due to its lower viscosity compared to the old epoxy-hardener system, which had maximum reinforcement with the 180 G field and 0.5 vol.% of CNTs.

In this work, as opposed to previous studies, we sought to gain insight about how varying magnetic field magnitudes affect CNT reinforcement of PNCs with varying CNT concentrations and different matrix viscosity. We concluded that a fine tuning of magnetic field magnitude, CNT concentration, and matrix viscosity is essential to maximize toughening effects. Magnetic alignment of CNTs does improve fracture toughness reinforcement given that the field magnitude is adjusted based on CNT content and matrix viscosity.

Inserting magnetically aligned CNTs within the interlaminar region of CFRPs can potentially reinforce the mechanical properties of composites by improving the fracture toughness of the in-plane layer, as shown in this work. Future work includes assessing the combined effects of introducing magnetically aligning CNTs into CFRPs through infiltration of dry fibers and placement in between prepreg layers. Concurrent to this study, a former lab member conducted work on fabricating CFRPs with B-staged epoxy layers made with an OOA process. Knowledge gained from both studies will be applied to fabricate CFRPs with magnetically aligned CNTs.

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