

# Fabrication of Polymer Composite Enhanced with CNTs-aggregated Fiber via an Electric-field-assisted Method

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**Abstract**—Due to the challenges associated with utilizing arranged carbon nanotubes to reinforce bulk polymer, herein a scalable method of preparing CNTs-fiber-enhanced composite is proposed. The electric-field-assisted method based on regulated frequencies improves the alignment of carbon nanotubes in the polymer, addressing the issue of uniformly distributing CNTs with varying types and sizes in the composite matrix. Results showed a CNTs-aggregated-fiber structure in the composite matrix, which is markedly different from the random arrangement. Such a structure can notably improve the mechanical and electrical properties of the polymer. Specifically, the composite achieves 57.2% higher flexibility than the original pure polymer, with a small reduction of elastic modulus. The effects of time, frequencies, and curing time are investigated to regulate the arrangement of CNTs with various qualities. Such a solution-based method can be adapted to a wide range of polymers and is scalable for the efficient production of large-area composites for further applications, such as in electronic device packaging.

**Keywords**—carbon nanotube fiber, CNTs-reinforced composite, electric-field-assisted method, polymer composites

## I. INTRODUCTION

Carbon nanotubes (CNTs) are nanomaterials with a special atomic structure, consisting of sheets of graphene rolled into cylindrical tubes, giving them outstanding mechanical, thermal, electronic, and biological properties. Because of these unique properties, CNTs are widely used in many technical fields. Since their discovery by Sumio Iijima [1], CNTs have been extensively studied for their unique properties such as thermal conductivity and high electrical conductivity [2]. In 1996, Fishbine [3] discovered for the first time that CNTs can be electrically polarized in an electrostatic field, thereby inducing electrostatic dipole moments to arrange CNTs. On this basis, Oliva et al. [4] proposed a dynamic CNTs network formation model under the AC fields, and systematically expounded the migration process of CNTs. Dielectrophoresis (DEP) at a fixed frequency for the alignment of CNTs in polymers inadequately utilizes CNTs' responses to various frequencies, usually leading to agglomeration and compromised composite properties [5, 6].

To address these challenges, this paper proposes an electric field-assisted method for the scalable preparation of CNTs-reinforced composites. The process involves dispersing CNTs in the oligomer solution using ultrasound and then applying AC and DC electric fields of different frequencies to

arrange the CNTs to form CNT-aggregated fibers. The AC-DC electric field-assisted method could form the structure in one step, avoiding the quality differences caused by applying AC and DC separately while multi-step forming [7]. It also offers a significant advantage over traditional direct arrangement techniques by utilizing variable frequency control to precisely guide CNT alignment. CNTs are connected to each other in the same direction at high frequency, and CNTs fibers are dispersed and arranged longitudinally at low frequency, which simplifies the process and reduces production costs. This in-situ approach ensures the creation of consistent, high-quality fiber structures, which are essential for the mechanical integrity of the composites. Moreover, the method's scalability and cost-effectiveness make it highly suitable for large-scale manufacturing, providing a versatile solution for a variety of polymers and applications. This solution-based method can be applied to a variety of polymers and can be extended to the efficient production of large-area functional composites.

The selected material system combines the advantages of conventional composite resins and CNTs composites. Conventional composite resins usually have mechanical properties such as flexibility. In this study, SU-7206 oligomer solution was used as a good UV-curable protective coating. CNTs were dispersed by ultrasonic dispersion and arranged using an electric field, and finally, a fiber structure was achieved [8]. However, when conventional composite resins are used for electronic component packaging, their heat dissipation and dielectric properties are often inferior to those of existing epoxy molding compounds.

CNTs-reinforced polyurethane acrylates (C-PUA) are then prepared using an in-situ UV radiation curing process. The synthesized composites exhibit well-distributed fiber composite structures, which improve mechanical and electrical properties while maintaining great flexibility. The effects of different process parameters were studied, and the arrangement of CNTs of different masses was investigated.

## II. EXPERIMENTAL SECTION

### *Dispersion of CNTs in polymer solutions*

CNTs typically exhibit adsorption and aggregation tendencies due to static electricity and intermolecular forces. Therefore, ultrasonic dispersion is essential to prevent agglomeration when dispersing them in various systems. In a 1mg/ml solution, 0.1 mg of CNTs (60-100 nm in diameter, 3-

15  $\mu\text{m}$  in length, sourced from Macklin, China) is dispersed in 0.5 ml of SU-7206 (from Soltech, China), combined with 0.46 ml of (5-Ethyl-1,3-dioxin-5-yl) methyl Acrylate (stabilized with MEHQ, from Macklin, China), and 0.04 ml of the photoinitiator OMNIRAD 819 (from Bide, China). This dispersion process is conducted using an ultrasonic machine at 35°C for 5 minutes.

#### DEP alignment and polymer curing

The experimental setup is shown in Fig. 2a. It includes a 50x high-voltage amplifier (ATA-2000, Aigtek Ltd., China) to amplify signals from a function generator (SOG1062K, SIGLENT, China), while an oscilloscope (SIGLENT, China) monitors the 1/100 output. Two ITO-coated glass pieces serve as electrodes for DEP, with the prepolymer solution and its mold placed between them. Initially, an alternating potential of  $138 V_{\text{RMS}} + 5 V_{\text{DC}}$  (Formed an E-field for  $4.6 \times 10^3 \text{ Vac/cm} + 16 \text{ Vdc/cm}$ ) at 1 kHz is applied through the electrodes for 10 minutes. The frequency is then switched to 10 Hz without changing other settings. The complete fabrication procedure for the CNTs-reinforced polymer composite involves several steps. First, the polymer is pumped into the container until it is fully filled. Next, specific AC and DC electric voltages are applied from a high-voltage amplifier to arrange the CNTs. This process prepares the distributed fiber polymer solution with arranged nanotubes. The prepolymer is then cured using 400 nm UV light from the top while maintaining the specific AC and DC voltages for 10 seconds. After curing, a fully cured polymer with a distributed fiber polymer composite bulk is obtained. Finally, the bulk material is removed from the mold cavity.

#### Characterization and mechanical tests

Optical microscopy images of the bulk materials are captured using the Metalloscope (Olympus BX53M, MA). Nanoindentation, or Depth-Sensing Indentation (DSI), is used to evaluate the mechanical properties of the materials. This technique measures parameters such as load-displacement curves, elastic modulus, hardness, fracture toughness, strain-hardening effects, viscoelastic behavior, and creep resistance at the nanoscale. The nanoindentation statistics are collected using a Nanoindentation apparatus (TI-750L Hysitron, US) in Depth Control Mode.

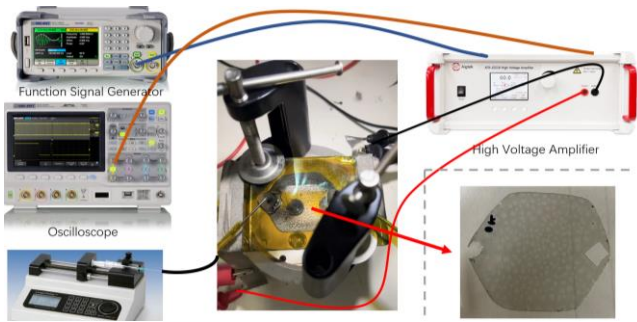


Fig. 1. Experimental setup and macro image of aggregated fiber C-PUA composite bulk

### III. RESULT AND DISCUSSION

#### Characterization

CNTs arranged under an electric field of  $4.6 \times 10^3 \text{ Vac/cm} + 16 \text{ Vdc/cm}$  with varying frequency and time exhibit different arrangements within the composites (Fig.2). Randomly dispersed CNTs exhibit agglomerations due to Van der Waals interactions (Fig.2 i). It is noteworthy that CNTs

tuned at a high frequency of 1 kHz for 10 min and then at a low frequency of 10 Hz for 10 min showed an aligned structure in the composite matrix, specifically, whisker-like black lines connecting the two ends of the agglomeration were manifested under an optical microscope (monochrome light source,  $450\times$  magnification). In comparison, random-aligned CNTs show only agglomerations without any formation of fibers. Observing the nanofiber-line of DEP-aligned CNTs by adjusting the focal plane between the bottom and top surfaces reveals larger agglomerations around the bottom surface. The bottom surface shows a higher fiber diameter in the direction of decreased potential. This disparity arises from the dielectrophoretic alignment process, whereby polarized CNTs migrate towards regions of lower electric potential energy under the influence of a direct current electric field. This phenomenon leads to increased agglomeration density at the bottom surface, influenced by the dielectric characteristics of CNTs and interactions with the solvent [1]. This implies that employing different frequency steps in the alignment process can effectively generate more orderly aggregated CNT structures compared to randomly dispersing CNTs within the polymer.

At high frequencies (1 kHz), CNTs tend to form end-to-end chains due to dipole-dipole interactions. Reducing the frequency to 10 Hz, induced charge electro-osmosis (ICEO) becomes dominant, creating a quadrupolar flow that pushes CNTs apart, favoring individual vertical alignment instead of tip-to-tip chaining [1,2]. The alignment torque from the electric field can be measured by observing dipole polarization, following the formula

$$U(h) = U \sin^2(h) \quad (1)$$

where  $U$  is the applied potential difference and  $h$  is the dipole angle relative to the field. This formula integrates into the theory of electrodynamics, encompassing both the electrostatic potential of CNTs in all frequency limits and their dipole polarization characteristics [3].

At low frequencies, the electrostatic potential  $U_{MW}$  of CNTs is given by:

$$U_{MW} = \frac{p_{em} L^3 E^2}{12} \ln\left(2 \frac{L}{D}\right) \left(1 - \frac{1}{1 + \frac{4}{3} \ln^2\left(\frac{L}{D}\right)}\right) \quad (2)$$

where  $U_{MW}$  is the electrostatic potential of the CNT (390V),  $p_{em}$  is the dielectric constant ( $\sim 8$ ),  $L$  is the length of the nanotube,  $D$  is the diameter, and  $E$  is the root-mean-square electric field strength [1]. This alignment torque, balancing electric field and viscous flow, aligns CNTs vertically until equilibrium is reached [4]. These CNTs fibers are formed through end-to-end or "side-by-side" connections, facilitated by Van der Waals interactions and potential bonding.

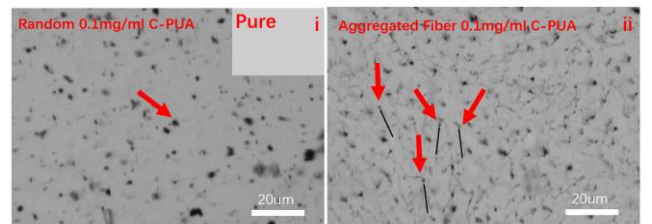


Fig. 2. Optical images of i) random-aligned 0.1mg/ml C-PUA showing CNTs aggregated into large dark particles, with a top-right inset image of pure polymer; ii) DEP-aligned 0.1mg/ml C-PUA showing clear fiber-line structures surrounding with small dark particles of CNTs.

## Mechanical properties

As an electronic component packaging material, its high strength and flexibility can ensure the reliability of the internal circuit operation of electronic components. CNTs exhibit enhanced thermal conductivity which may also be an advantage. Nanoindentation is employed to investigate the mechanical properties, specifically the elastic modulus and hardness of the materials. The bulk material is tested using the depth mode of a nanoindentation machine, as illustrated in Table 1. While pure polymer exhibits the highest elastic modulus and hardness, flexibility is a significant factor in operating conditions of electronic devices with severe vibration or impact.

The addition of CNTs to the polymer reduces the efficiency of the photocuring process due to their absorption of UV energy, resulting in a decrease in the polymer's elastic modulus [6]. The DEP-aligned 0.1mg/ml C-PUA with aggregated fiber approaches the elastic modulus of the pure polymer but experiences a 57.2% reduction in hardness. However, the fiber composites demonstrate a higher elastic modulus and improved flexibility compared to any other CNT polymer composites previously mentioned [12], suggesting enhanced local energy dissipation and greater flexibility. This effect arises because CNTs hinder UV light from curing the prepolymer [6]. Among the tested polymers, random-aligned 0.1mg/ml C-PUA is the more affected (65.6% lower), with DEP-aligned 0.1mg/ml C-PUA showing less impact (13.1% lower). In DEP-aligned 0.1mg/ml C-PUA, well-distributed CNTs fibers withstand pressure better than surrounding polymer molecules, reinforcing the elastic modulus.

Nanoindentation testing is utilized to assess the mechanical properties of materials. The bulk is fixed on the machine base, and the properties of the bulk are assessed by inserting the nanoprobe to a depth of 1000um using the depth mode, perpendicular to the CNT fiber surface. The nanoindentation results across different sample locations are consistent, indicating material uniformity even when specific regions like fibers are tested. Enhanced mechanical contact between CNTs contributes to end-to-end reinforcement in the longitudinal direction [13]. Dispersed CNTs derive benefits from their interactions with polymer walls, including van der Waals forces, which reinforce the elastic modulus of the composite material [14]. However, imperfect arrangement of CNTs, particularly within CNTs fibers, can result in structures resembling poorly aligned Random CNT polymers, posing challenges for localized material performance. Despite these complexities, overall composite performance aligns with the described characteristics.

TABLE I. ELASTIC MODULUS AND HARDNESS (NANOINDENTATION)

Num	Elastic Modulus and Hardness		
	Sample Type	Elastic Modulus (MPa)	Hardness (MPa)
1	Pure Polymer	719.79	19.4
2	Random-aligned 0.1mg/ml C-PUA	247.59	4.03
3	DEP-aligned 0.1mg/ml C-PUA	626.04	12.58

For Pure polymer, random-aligned 0.1mg/ml C-PUA, DEP-aligned 0.1mg/ml C-PUA.  
Nanoindentation TI-750L Hysitron, US

## IV. CONCLUSION

In conclusion, the fabrication of aggregated fiber 0.1mg/ml C-PUA through an electric-field-assisted method offers a promising approach for enhancing mechanical and electrical properties in electronic device packaging. This method effectively addresses the challenge of achieving uniform CNT distribution and alignment within polymer matrices, crucial for optimizing composite performance. By utilizing two-frequency electric fields, we successfully created well-organized CNTs fiber structures that significantly enhance flexibility while maintaining a competitive elastic modulus compared to pure polymers. Nanoindentation results confirmed consistent material properties across various sample locations, underscoring the uniformity and reliability of these composites. The enhanced mechanical contact and reinforced interactions between CNTs and polymer walls further contribute to the composite's robustness and durability. These advancements position DEP-aligned C-PUA as promising materials for applications requiring high strength, flexibility, and reliable electronic component packaging. Future research may focus on scaling up production and exploring additional applications in advanced electronic devices. The application of these DEP-aligned C-PUA in electronic component packaging ensures high strength and flexibility, crucial for maintaining reliable operation under severe conditions such as vibration and impact. Enhanced thermal conductivity and mechanical properties make them ideal candidates for improving the performance and reliability of internal circuitry in electronic devices. Therefore, these composites hold great potential for advancing electronic packaging technologies, offering solutions that meet stringent performance requirements while maintaining scalability in production.

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