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ORIGINAL ARTICLE

A Novel Approach to Fabricate Carbon Nanomaterials–Nanoparticle Solids through Aqueous Solutions and Their Applications

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Abstract
A better understanding of the bonding and aggregation processes occurring between carbon nanomaterials and metal oxide particles in aqueous solutions is important in the development of novel nanosolids for applications in the areas of sensor development, highly conductive paint, nanotube alignment, polymer composites, Li-ion batteries, and many other areas. The current investigation reviews these processes and presents a detailed description of the aggregation processes occurring between carbon nanomaterials and metal oxide particles (metals) in various aqueous solutions. The results indicate that the charge attraction between the particles results in a strong homogeneous bonding that occurs within the aqueous solution and for the first time demonstrate and describe the aggregation process of these nanoparticles. The relative importance of many parameters that impact the aggregation process is identified and discussed, and guidelines for controlling the aggregation process are presented. This is a simple and cost-effective process to manufacture a novel nano-solid based on carbon nanomaterial and metal oxide. In addition, the process is easy to scale up and optimize. The methodology could lead to many significant applications as well as commercialization.

Keywords Nanosolid · Aggregation technique · Nanofluids · Charge attraction · Nanoparticles · Nanomanufacturing

1 Introduction

Carbon nanomaterials and metal oxide nanoparticles have attracted significant consideration for the last two decades because of their attractive mechanical, electrical, and thermophysical properties. For example, carbon nanotubes (CNTs) have a thermal conductivity (TC) of 3000 W/mK [1, 2], an electrical conductivity of more than 4000 S/cm [3], a tensile modulus of 1 TPa, and a tensile strength of 50–150 GPa [4, 5]. As a result, these nanomaterials have been applied in several applications with the purpose of improving the overall system performance. To further develop nanocomposites with improved properties, such as the electrical and TC, CNTs have been mixed with different types of polymers. Historically, epoxies have been mixed with CNTs to improve the mechanical and thermophysical characteristics of the composite. Tensile strength and Young's modulus of the resulting new material have demonstrated an improvement as high as 34% and 60%, respectively [6]. The results obtained by Paiva et al. [7] showed that Young's modulus of SWNT/poly(vinyl alcohol) (PVA) composite has improved by 55% when only 5 wt% of functionalized SWNTs has been used to fabricate the composite. Also, an increase in the tensile modulus of more than 130% for MWNTs/PVA composites at values as low as 2 wt% nanotube loading has been reported [8]. Similarly, CNTs, CNFs, graphene, and metal oxide NPs have been used to prepare nanofluids, a fluid containing well-dispersed nanoparticles, in an attempt to create fluids with high TC [9, 10]. The resulting highly conductive nanofluids can be used in numerous applications, for example in cooling purposes in heat transfer equipment

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such as nuclear reactors, radiators, vehicles, and electronics applications. Several investigations have been conducted in the past several years designed to evaluate the fabrication of highly conductive nanofluids with very high TC properties [11–13]. One of the early examples of nanofluid fabrication has been reported by Hong et al. [14], in which 0.05 wt% CNTs was shown to increase the TC of the fluid by 10%.

A review of the recent research conducted using CNTs and metal oxide NPs to enhance the mechanical, thermal, and electrical properties of composites and the TC of nanofluids indicates only modest improvement in the thermophysical and mechanical properties for many cases. For example, for most nanofluids, the TC improvement is not significant enough for practical uses. The concomitant rise in viscosity of the fluids results in a poor dispersion because of the use of high weight percentages of CNTs or NPs and is the principal constraint limiting the practical implementation of nanofluids in thermal management applications [15]. For the case of nanocomposites, the dispersion issue and lack of alignment were the principal impediments to the enhancement of the mechanical and the electrical/thermal properties [16]. As a result, the scientific community has tried to identify other solutions by which the mechanical, electrical, and thermophysical properties can be improved at low weight percentages, without increasing the viscosity of the fluids. A number of different approaches have been pursued; one of the most common is the alignment of CNTs or nanoparticles in aqueous solutions to enhance nanofluids' TC. However, this approach is only applicable for magnetically sensitive nanomaterials. A limited number of metal-oxide nanoparticles such as Fe_2O_3, Fe_3O_4, $NiFe_2O_4$, $CoFe_2O_4$, etc. Fe_2O_3 and CuO have been evaluated for the nanofluids' TC enhancement. Furthermore, the impact of alignment on the TC of nanofluids has also been investigated. A 0.4 vol% of Fe_2O_3 NPs was found to increase the TC of water by 20%, while CuO NPs improved the TC by only 12%. By applying an external magnetic field to align the NPs, the TC could be increased by 83%. This significant improvement is attributed to the improved alignment of the Fe_2O_3 nanoparticles [17]. Although the alignment of magnetic metal oxide nanoparticles in aqueous solutions is relatively easy, the alignment of carbon nanomaterials such as CNTs, CNFs, and graphene is more complicated, as they do not typically possess magnetic properties. An interesting solution for this problem was first introduced by Hong et al. [18] in which SWNT's alignment in water was achieved by attaching Fe_2O_3 NPs to the SWNTs by using sodium dodecylbenzene sulfonate (NaDDBS) surfactant as binder and then applying a magnetic field. In this work and in related references [19–26], it was observed that the aggregation process could be accomplished in an aqueous medium, in the presence of SDBS surfactants. The presence of SDBS is crucial to help obtain good dispersion for the CNTs and promote the attraction of the nanoparticles through the electrostatic interaction between the sulfonic head of the surfactant that has a negative charge, due to the dissociation process in water and the partially positive nanoparticles [27]. However, previous studies have focused only on the properties of fluids. As a result, all of the components (carbon nanomaterials, surfactants, and metal oxides) were in the aqueous state and could not be removed or reduced. In most circumstances, surfactants are needed, but are not helpful to the physical properties of the mixture. As a result, there is a need to develop a new method to use surfactants to fabricate uniformly aggregated solids containing carbon nanomaterials and metal oxides for applications such as composites, battery electrodes, etc., but keeping the surfactant to a minimum.

In the following investigation, a new concept for fabricating novel solids based on carbon nanomaterials and metal oxides through aqueous solutions is developed and their use in several important applications is proposed and discussed. The goal of this work is to gain a comprehensive grasp of the bonding and aggregation processes that occur between CNMs and metal-oxide nanoparticles.

2 Investigative Approach

Graphene was purchased from Cheap Tubes Inc. and NaDDBS and Fe_2O_3 nanoparticles were purchased from Sigma-Aldrich and used as received. Ultrasonication was performed with a Branson Model 450 Digital Sonifier with a ½" disrupter horn. Scanning electron microscopy (SEM) images were obtained using a Zeiss Supra40 variable-pressure field emission SEM with an Oxford AZtecEnergy advanced system. Transmission electron microscopy (TEM) images were obtained using a JEOL JEM-2100 LaB6 transmission electron microscope equipped with a high-resolution (HR)-style objective-lens pole piece.

The manufacturing process is shown in Fig. 1. Briefly, SDBS was mixed with CNTs and dispersed in deionized water using ultrasonication. Then the nanoparticles were added into the mixture and dispersed again. After this, vacuum filtration was used to separate the fabricated aggregated

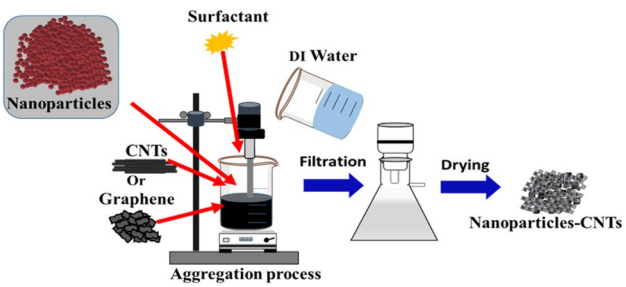

Fig. 1 Synthesis scheme for the aggregated nanoparticles (CNTs)

solids. Finally, a vacuum oven was used to dry the aggregated solids. The solution was filtered and dried in a vacuum oven. The same procedure was followed for the fabrication of Fe_2O_3-CNTs using CNTs in the place of graphene.

3 Concept Detail Discussion

Several different types of nanoparticles were used to coat the carbon nanomaterials through the aggregation process. For example, Fe_2O_3 nanoparticles were aggregated on the surface of CNTs as shown in Fig. 1. This approach is not applicable in the case of non-aqueous solutions, such as epoxy, because SDBS is soluble only in aqueous solutions. Therefore, we fabricated the aggregated nanoparticle CNTs in a water solution first. Figure 2a shows an SEM image of the CNTs that were used in the aggregation process. As is shown, the CNTs are agglomerated and interconnected with each other. To better observe the CNTs, a TEM image was acquired as depicted in Fig. 2b. The TEM image indicates that the CNTs are MWNTs as the diameter of the tubes is larger than 20 nm. The TEM image (Fig. 2c) suggests that Fe_2O_3 NPs have an average particle size between 25 and 50 nm. Furthermore, the Fe_2O_3 crystals are shown in the figure with a hexagonal structure. Figure 2c (inset) shows the lattice fringes and the space is around 0.37 nm. In addition, the crystallinity of the Fe_2O_3 NPs is further confirmed by the diffraction pattern acquired from TEM analysis, as depicted in Fig. 2d.

The aggregation of the nanoparticles on the surface of the carbon nanomaterials is attributed to the electrostatic interaction that occurs between the nanoparticles and the surfactant. The attraction between NaDDBs surfactant and CNTs is strong because of the π–π interaction between the benzene ring of the NaDDBs and the surface of the CNTs. In addition, the repulsive force between the head of the surfactant, which is the sulfonic group in the NaDDBS case, improves the deagglomeration of the CNTs [27]. As shown in Fig. 3a, the head of the NaDDBS surfactant dissociates in water into two parts, the Na cation part, and the sulfonic anionic part. The surfactant's tail that has the hydrocarbon chain and the benzene ring is adsorbed at the surface of the carbon nanotube, whereas the negatively charged sulfonic group attracts the partially positively charged iron oxide nanoparticles. The carbon nanotube solution has a pH equal to 6.15 at which the magnetic sensitive ɤ-Fe_2O_3 nanoparticles have a positive zeta potential charge (Fig. 3b) that interacts with the negative charge.

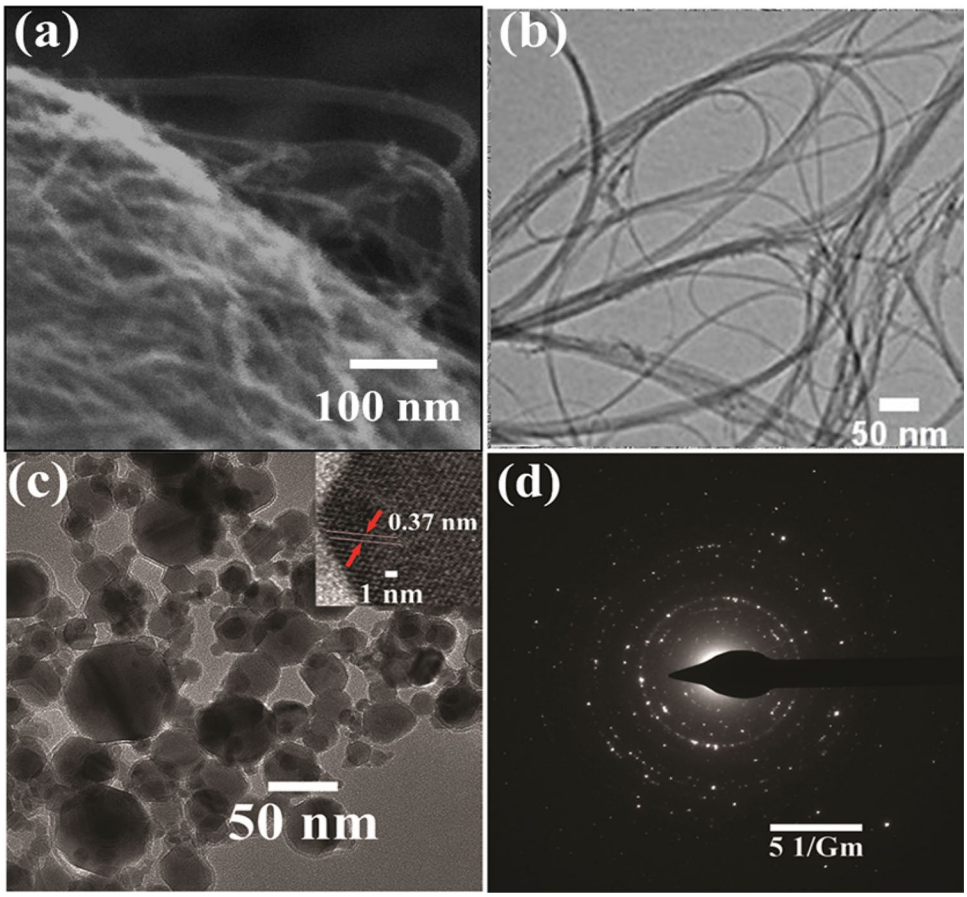

Fig. 2 **a** SEM image showing many CNTs entangled together. **b** High-resolution TEM image showing CNTs. **c** Low magnification TEM image of ɤ-Fe_2O_3 nanoparticles. **d** Diffraction pattern obtained from TEM analysis and it shows the crystalline properties of the ɤ-Fe_2O_3 particles [28]

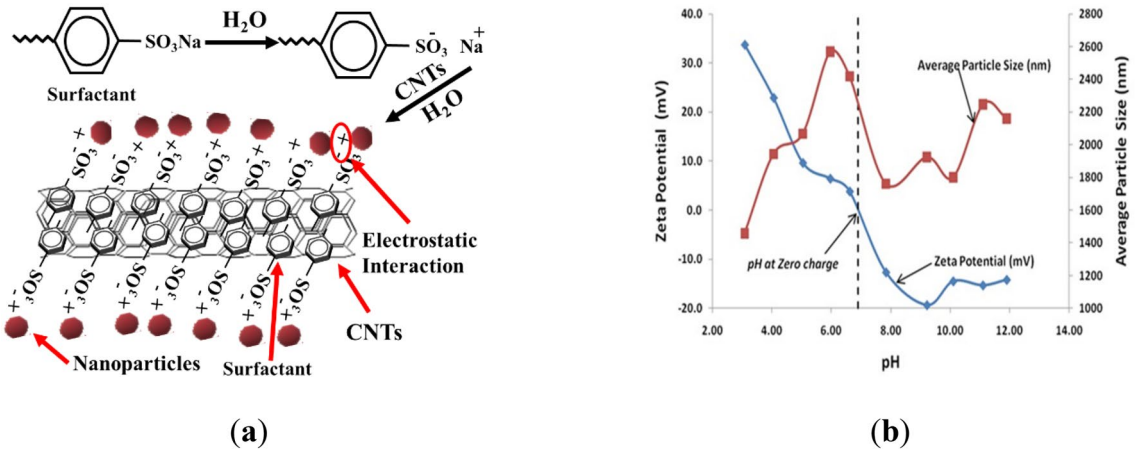

Fig. 3 **a** The separation reaction of NaDDBS in aqueous solution and the electrostatic interaction between nanoparticles and CNTs. **b** Zeta potential and average particle size vs. different pH values [17]

The SEM image in Fig. 4a shows the CNTs with aggregated Fe_2O_3 nanoparticles on the tube's surface. This was also observed when TEM images of the MgO nanoparticles were reviewed. The TEM image in Fig. 4b shows the aggregations of the MgO nanoparticles along the carbon nanotubes. Figure 4c shows the thermogravimetric analysis (TGA) of the aggregated Fe_2O_3-CNTs. It is apparent here that at 100–200 °C, some of the NaDDBS surfactant and other impurities have been burned off, with the vast majority of the CNTs having been burned completely at 600 °C. Moreover, the TGA analysis shows the presence of approximately 38% Fe_2O_3 nanoparticles. Figure 4d, showing the XPS, confirms the presence of the ϒ-Fe_2O_3 nanoparticles in the composite.

In summary, all of the images and experimental results indicate that the concept works. Carbon nanomaterials and nanoparticles can be aggregated uniformly in the nanoscale. This method could also be extended to non-carbon

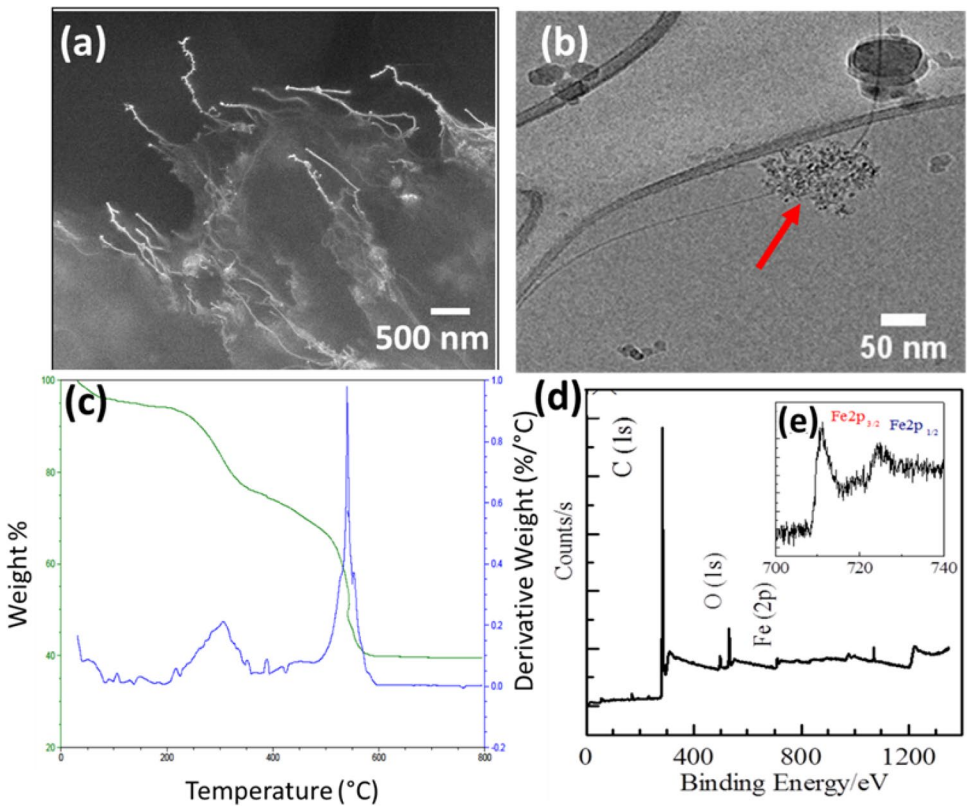

Fig. 4 **a** SEM image showing the aggregation of Fe_2O_3 nanoparticles on carbon nanotubes. **b** TEM image showing the aggregation of MgO nanoparticles on carbon nanotubes. **c** Thermogravimetric analysis (TGA) of Fe_2O_3 nanoparticles aggregated on CNTs. **d** The XPS spectrum of ϒ- Fe_2O_3 nanoparticles aggregated on CNTs. **e** High-resolution XPS spectrum of iron (2p) bands of the Fe_2O_3 nanoparticles aggregated on CNTs

nanomaterials. The nanoparticles used could be metal oxides, but also could be comprised of elemental metals, metalloids, metal alloys, metal sulfides, metal seleniums, and/or a combination thereof. In addition, this method is simple, cost-effective, and easy to be applied in large-scale applications.

4 Applications

The novel carbon nanomaterials-nanoparticle solids fabricated from aqueous solutions have many applications.

4.1 Energy Storage

Recently, an investigation of aggregated nanoparticles-CNMs for lithium-ion battery applications was done by Christensen et al. [29]. In this study, the same aggregation concept was used to attach both Fe_2O_3 and silicon nanoparticles to different carbon nanomaterials in a uniform dispersion and attachment process. Surfactants were used along with dispersion and attachment agents. An anode was fabricated from Fe_2O_3/graphene/surfactant. The anode demonstrated a capacity of over 1000 mAh/g with a stable charge/discharge process of 21 cycles and evidence of relatively little capacity reduction. The Si materials exhibited an ultrahigh capacity of approximately 4000 mAh/g. The ability of Si nanoparticles to attach to CNF, SWCNT, and graphene by this method is quite significant in the development of this technology. Cycling data showed a very high capacity, over 1000 mAh/g [29]. Figure 5 is SEM images for (a) Fe_2O_3/CNF/surfactant, (b) Fe_2O_3/SWNT/surfactant, (c) Si/CNF/surfactant, and (d) Si/SWNT/surfactant.

4.2 Magnetic Enhancement Materials

4.2.1 Magnetic Composites

Magnetic composites are very important and have numerus applications, such as magnetic recording media water purifications, energy storage, and high-density data storage. Therefore, the aggregation process of magnetic nanoparticles such as Fe_2O_3, CrO_2, or spinel structure compounds like $MnFe_2O_4$, $CoFe_2O_4$, and $NiFe_2O_4$ is also important [30–32]. The results obtained by Neupane et al. in [22] suggest that the coercive field of SWNTs coated with Fe_2O_3 nanoparticles was twice that of pristine SWNTs (Fig. 6a and b).

4.2.2 Magnetic Sensitive Mats

In a previous investigation, the magnetic properties of carbon nanostructure (CNS) mats were investigated. SWNTs were mixed first with surfactants and Fe_2O_3 using the aggregation process explained earlier and then dispersed with CNS using the tip sonicator. This was followed by a vacuumed filtration technique that was used to fabricate the CNS mats. A series of characterization techniques were conducted to study the effects of using Fe_2O_3 on the fabricated CNS mats. For example, XPS (Fig. 7a) has been used to confirm the presence of the γ-Fe_2O_3 nanoparticles in the composite,

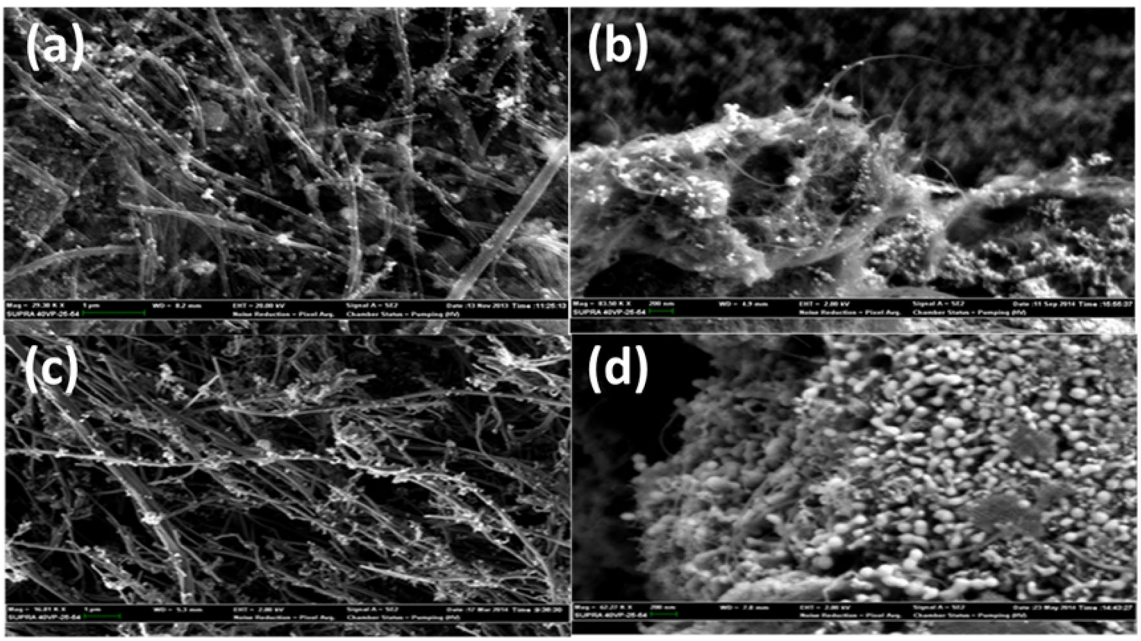

Fig. 5 SEM images of **a** Fe_2O_3/CNF/surfactant, **b** Fe_2O_3/SWNT/surfactant, **c** Si/CNF/surfactant, and **d** Si/SWNT/surfactant [29]

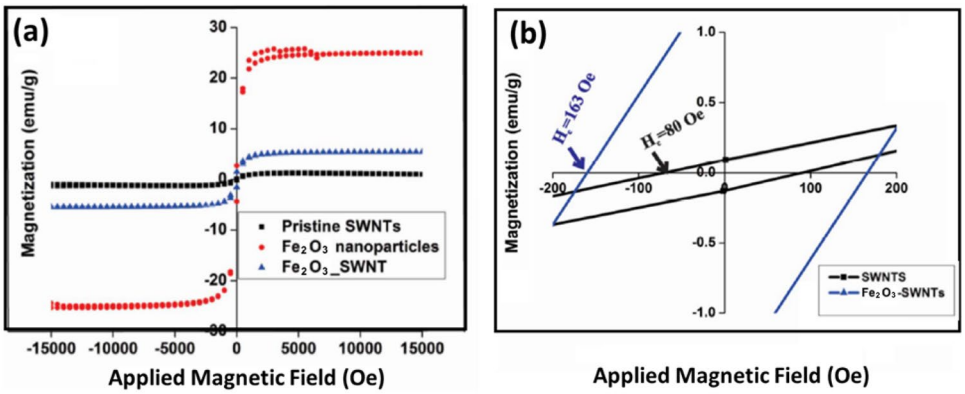

Fig. 6 **a** Magnetization (*M*) versus applied magnetic field (*H*) for pristine SWNTs, Fe_2O_3 nanoparticles, and SWNTs coated with Fe_2O_3 nanoparticles. **b** Enlarged *M*–*H* curves for pristine SWNTs and SWNTs coated with Fe_2O_3 nanoparticles [22]

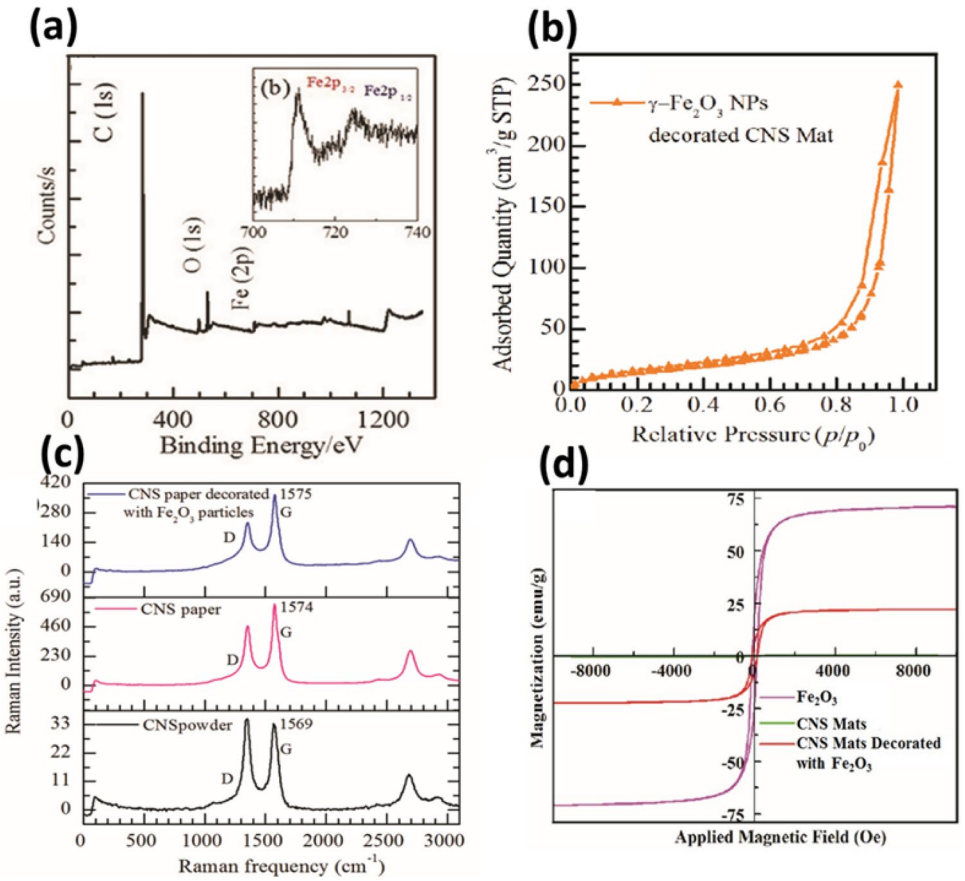

Fig. 7 **a** XPS for CNS-ϒ-Fe_2O_3 (*inset* shows high-resolution XPS spectrum of iron 2p bands). **b** Nitrogen adsorption–desorption isotherm of ϒ-Fe_2O_3 NP-decorated CNS mats. **c** Raman spectra of as-fabricated CNS mat. **d** Magnetic properties of ϒ-Fe_2O_3 NP-decorated CNS mats [27]

which were added to the CNTs during the fabrication process and attached at the surface of the CNT. As shown, the peaks of carbon and oxygen and characteristic iron peaks are apparent. Figure 4e presents a high-resolution spectrum that clearly indicates the locations of the Fe (2p3/2) and Fe (2p1/2) peaks at 713 eV and 725 eV, respectively. The spectrum peaks are in agreement with the values reported for ϒ-Fe_2O_3 nanoparticles in the literature. In addition, the Brunauer–Emmett–Teller (BET) analysis was conducted to better understand the surface area of the fabricated mats (Fig. 7b). According to nitrogen adsorption and desorption measurements, the CNS mats possess low BET-specific surface areas of 55.8 m^2/g and the DFT (density functional theory)-calculated total pore volume of 0.0263 cm^3/g.

Raman analysis illustrated in Fig. 7c indicates the existence of the two common peaks for CNTs; *G* peak and *D* peak, reduction in the *ID*/*IG* ratio from 1.06 to 0.65 is noticed, because of washing out the impurities during the filtration process. In addition, the presence of Fe_2O_3 has decreased *ID*/*IG* ratio further. Furthermore, vibrating-sample magnetometer (VSM) study was done to understand more about the magnetic properties of the fabricated

materials as shown in Fig. 7d, where the results gained from three samples, pristine carbon nanostructures (CNS) mats, CNS mat with ϒ-Fe_2O_3 and ϒ-Fe_2O_3 showed that pristine CNS mats are not magnetic. However, the aggregation of the ϒ-Fe_2O_3 NPs occurred on the CNS in creation of magnetic sensitive CNS mats. The improved magnetization of ϒ-Fe_2O_3 NPs covered CNS mats can be attributed to the large quantity of Fe present within the carbon foam. Indeed, the saturation magnetization of CNS mats is expected to increase with an increased quantity of ϒ-Fe_2O_3 nanoparticles. As a result of using the Fe_2O_3 nanoparticle aggregation approach, the electromagnetic wave absorption has been improved, as shown in Fig. 8.

4.3 Lubricants and Nanogrease

Nanogrease is another important application for the aggregated nanoparticle CNTs. Several publications have appeared in recent years discussing the fabrication of nanogreases using aggregated nanoparticle CNTs with high thermal and electrical conductivity properties [33–42]. A study by Younes et al. concluded that SWNTs form a strong 3D net structure because of the strong Van der Waals forces and this is the main reason behind the creation of stable SWNT nanogrease, as shown in Fig. 9. In addition, this study demonstrated that the attachment of the iron oxide at the outer face of the CNTs prevents the formation of the 3D network structure [37]. Furthermore, the results indicate that the TC for SWNTs grease at 10 wt% has not changed regardless of the strength of the magnetic field or its application duration time, because of the strong Van der Waals forces between the SWNTs, which hinder movement and resist the magnetic field [43] (Fig. 10).

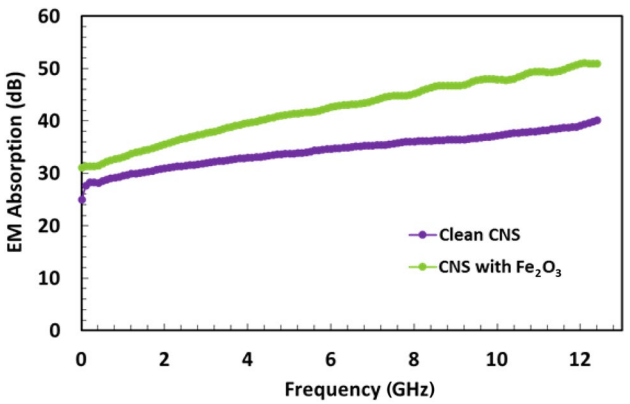

Fig. 8 Electromagnetic wave absorption of carbon nanostructures mat and aggregated Fe_2O_3 carbon nanostructures mat [28]

Fig. 9 Image of SWNT grease [37]

4.4 Polymer Composites

Recently, several authors have used the aggregated nanoparticles-CNT process to fabricate nanocomposites with improved mechanical properties and thermal and electrical conductivity. Luan et al. used aggregated Fe_2O_3-CNTs and studied Fe_2O_3-CNTs alignment's influence on the mechanical properties of PVA [21]. Liu et al. used the same technique to study the effect of the alignment on the tensile strength and TC of epoxy composite [32]. Recent work was performed by Younes et al. that reported using Fe_2O_3-CNTs to fabricate a special type of Buckypapers with magnetic properties that can be used in many applications [27, 44].

Luan et al. [21] used the aggregation approach to prepare a magnetic SWNTs PVA composite. It was found that using a pair of magnets has an obvious impact on aligning SWNTs within the composite, which ultimately improves the composite mechanical properties. A sample prepared with SWNTs and 5 wt% of Fe_2O_3 NPs was placed between two magnets for 60 min and left to cure. The tensile strength increased to 106 MPa because of the alignment of the SWNTs, as seen in Fig. 11.

4.5 Nanofluids

The idea of using the aggregation process in the area of nanofluids is to increase the TC of CNTs or graphene nanofluids. The aggregation of magnetic nanoparticles on the surface of CNTs or graphene makes them sensitive to the magnetic field. Then, by applying a magnetic field, the aggregated nanoparticles-CNT align with the magnetic field. This alignment improves the TC [18, 45–49]. However, in the solution form, the aggregates may fall apart because of the presence of water.

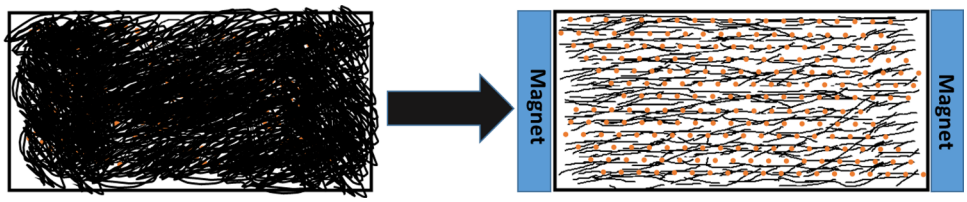

Fig. 10 Impact of magnetic field on the alignment of the SWNTs/Fe_2O_3 [32]

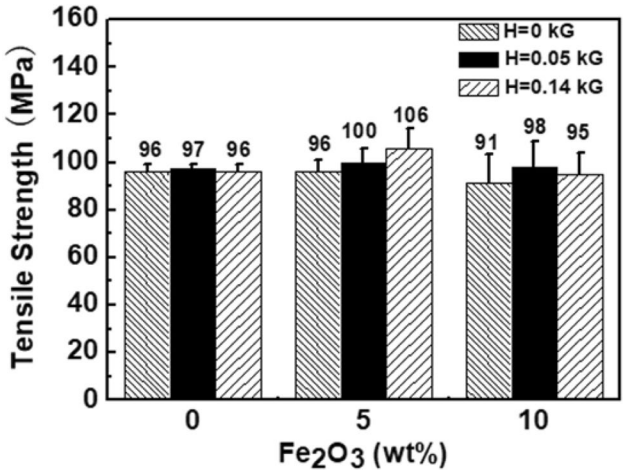

Fig. 11 Mechanical properties of SWNTs PVA composite [21]

4.6 High Thermal Performance Materials

Liu et al. [32] has used the same aggregation approach to fabricate magnetic sensitive SWNTs epoxy nanocomposite. Tip-sonicator and SDBS surfactant were used to ensure good dispersion of the SWNTs in aqueous solution. After 10 min of the dispersion process, Fe_2O_3 was added and the solution was dispersed for an additional 10 min. Finally, the solution was vacuum-filtrated and dried in an oven at 80 °C overnight. The dried SWNTs-Fe_2O_3 was then dispersed again in an epoxy solution to fabricate the composite. Subjecting the composite to a magnetic field during the curing procedure was found to effectively align the SWNTs-Fe_2O_3, which led to an enhancement in the mechanical property and the TC of the composite, as it is depicted in Figs. 12 and 13.

5 Conclusions

Carbon nanomaterials, including CNT, graphene, CNFs, and nanoparticles, such as Fe_2O_3 and SiO_2, have been aggregated homogeneously at the nanoscale in fluids in order to form new nanosolids. These nanosolids have been shown to exhibit interesting physical properties that may be suitable in a number of applications. The detailed characterization of these nanosolids indicates that the charge attraction between the particles results in a strong, homogeneous bonding process occurring within the aqueous solution, and demonstrates the nanoscale aggregation process.

This method can not only be used for CNT, graphene, and CNF materials, but also can be extended to non-carbon nanomaterials. The nanoparticles also could be comprised of elemental metals, metalloids, metal alloys, metal sulfides, metal selenium, and/or a combination thereof. The method presented here is simple, cost-effective, and can be easily adapted to large-scale applications, once the manufacturing process has been optimized.

The results and applications presented here clearly indicate the veracity and applicability of the process/concept, and while initiated as an investigation of nanofluids it has led to the development of novel nanosolids, which have the potential for significant commercial value and considerable economic impact. Continued investigations may result in additional interesting physical properties that can be used in a number of new and emerging technologies.

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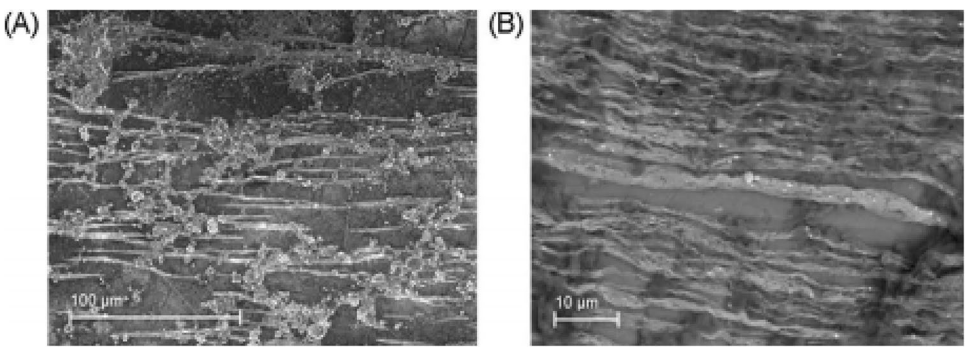

Fig. 12 SEM image of 0.017 wt% SWNT, 0.017 wt% Fe_2O_3, and 0.17 wt% SDBS with magnetic field. **a** Scale bar 100 µm, and **b** scale bar 10 µm [18]

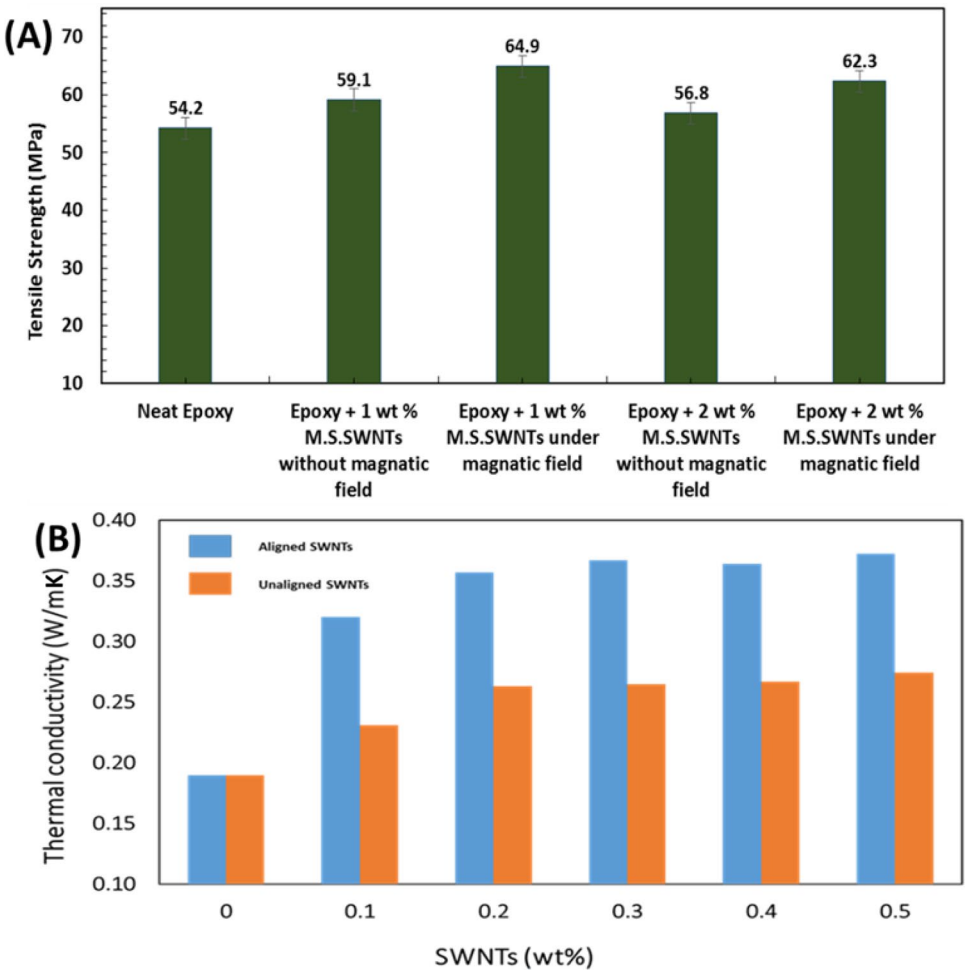

Fig. 13 **a** Comparison of the tensile strength of epoxy nanocomposites. **b** TC for different wt% of SWNTs nanocomposites [32]

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Compliance with Ethical Standards

Conflict of interest The authors declare no conflicts of interest.

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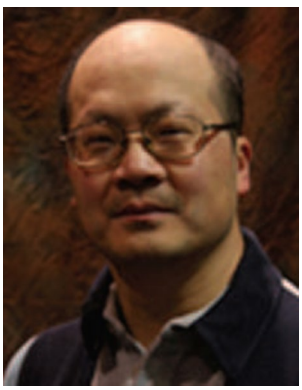

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