The fabrication and characterization of high density polyethylene composites reinforced by carbon nanotube coated carbon fibers

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\textbf{A B S T R A C T}

A simple and economical surface treatment method was developed for the first time to modify the surface roughness of carbon fibers (CF) by spray coating with carbon nanotubes (CNTs). The CNT-coated CF was then compounded, extruded and injection molded with high density polyethylene (HDPE) for hierarchical CNT-CF/HDPE composite fabrication. To evaluate the influence of CF contents and CNT coatings on the mechanical, thermal and morphological properties of the hierarchical composites, the CF content relative to the composites varying from 0 to 25 wt% and 1 wt% CNT coatings relative to CF were investigated in the research. The results showed that with the increase of CF content, all the evaluated properties including the tensile properties, flexural properties and hardness of CF/HDPE composites were effectively enhanced. Meanwhile, compared to uncoated composites, the CNT-spray coated counterparts have higher overall mechanical performance. The differential scanning calorimetry data indicated that the crystallization temperature and the crystallinity of HDPE were affected by the introduction of CF and CNT while the melting temperatures did not have apparent changes. An analysis of mechanical results combined with the morphological observation via scanning electron microscopy indicated that the interfacial bonding between CF and HDPE was improved by the incorporation of CNT.

1. Introduction

Carbon fiber (CF) reinforced polymer composites have been widely used in automotive, aerospace and construction industries by virtue of their ease of fabrication, low density, economy and remarkable mechanical performance [1]. This can be largely ascribed to the superior properties of the reinforcer CF, such as high modulus, high specific strength, and outstanding thermal and electrical conductivity [2,3]. As is well known, apart from internal intrinsic characteristics of the reinforcement and the matrix, the resultant performance of the composites is also, to a large extent, modulated by the interfacial adhesion between these two phases. A strong interfacial bonding contributes to the efficient load transfer from the matrix to the fiber, which largely avoids the occurrence of stress concentrations and enhances the overall mechanical performance [4,5]. Furthermore, the interfacial properties of the fiber-matrix entities play a crucial role in fracture toughness, compressive strength and strain to failure, fatigue resistance, impact strength and damage initiation threshold [6,7]. However, due to the non-polar and chemically inert structure with highly crystallized graphitic basal planes, the CF normally has low surface energy and poor wettability, leading to the weak interaction with the majority of polymers [8,9]. Various methods have been applied to improve the CF adhesion in the polymer matrix.

With respect to the in-plane properties of laminated composite materials, the fiber reinforcement can provide sufficient strength for many practical applications, while for the out-of-plane (i.e. through thickness) properties, the matrix usually becomes the main restraining factor for the holistic enhancement of fiber reinforced laminated composites [10,11]. Existing techniques such as z-pinning, stitching and braiding have been applied for the interfacial improvement [12]. However, these methods not only increase the additional manufacturing complexity and cost, but simultaneously compromise the in-plane strength. Therefore, fiber or matrix modifications via the introduction of nanoscale additives open up a new avenue for the improvement of interfacial properties without increasing the overall volume of the composite. Numerous researches have been carried out in recent years in developing nano-engineered hierarchical composite materials where nanoscale additives (e.g. alumina, titania and silica) are finely integrated with fibers [13–15].

Of all these nanofillers, the exploitation of carbon nanotube (CNT)—seamless cylinders of one or more layers of graphene with open or closed ends—is undoubtedly a big current interest in creating
hierarchically reinforced composite structures [16]. Based on their unique spatial structures and remarkable strength and modulus, substantial improvements for the fiber-matrix interfacial strength and matrix dominated through-thickness performance have been achieved in CF reinforced composites, such as impact strength, fracture toughness, interfacial shear strength and fatigue life [16,17]. In general, CNT engineered hierarchical composites can be fabricated via two approaches: incorporation of CNT into the matrix or direct adherence of CNT onto the reinforcing fiber surface [18]. The first approach is normally limited to low concentrations of the nanofillers. With the loading increase of CNT, the matrix will become increasingly viscose, leading to the difficulty of efficient dispersion of CNT into the matrix and sufficient filtration of the matrix into fiber reinforcement [19]. Another drawback existing in the addition of CNT into the matrix is the limited capacity of interfacial adhesion between the fiber surface and the nanoscale reinforcement [20]. Besides, the curviness and agglomeration of CNT in the matrix may reduce the effective stiffness of the nano-reinforced composites [21]. Thus, more researchers have shifted the related study to directly incorporate CNTs into fibers or fabrics. Four different strategies can be identified for the direct integration with reinforcement phase: (1) epitaxial growth of CNT from the fiber surface via chemical vapor deposition (CVD); (2) electrophoretic deposition (EPD) of CNT on the fiber surface; (3) chemical bonding between group-modified CNT and fibers; (4) spray coating of fibers and fabrics by CNT contained suspensions [22-24]. As a result, Kepple et al. reported the enhancement of interlaminar fracture toughness by around 50% via in-situ growth of functionalized CNT onto CF fabrics [25]. Via EPD technique, Yao et al. deposited CNT onto the surface of CF, spurring a significant increase (58.6%) of fiber-epoxy interfacial adhesion [26]. Sui et al. also found that the hierarchical CNT/CF/epoxy composite fabricated by continuous electrophoretic deposition method brought 33%, 10.5%, 9.5% and 4.5% increases in interfacial shear strength, interlaminar shear strength, flexural strength and residual bending strength, respectively [27].

However, some of these techniques have internal disadvantages when depositing CNTs onto fibers and fabrics. Due to high processing temperature and reaction of catalyst with fiber surface in the CVD process, both the existing sizings around fibers during their manufacture and the tensile strength of the resultant composite are damaged, let alone the less flexibility in tuning the adhesion between CNT and fibers [28]. The operational principle underlying the EPD technique also necessitates the possession of electric conductivity for the substrates, which to a certain extent limits the application of this method [29]. Furthermore, the complicated steps and cost involved in some of these techniques largely decrease the productivity and their potential for scale-up. Compared to these methods, the CNTs deposition by coating approach shows a great promise because of its easy operation and no harm to fibers (even able to heal surface defects and improve tensile properties) [30-32]. It also does not need complex chemical modifications for pristine CNT during the coating process, which is usually the prerequisite for other deposition techniques. By the spray coating method, CNT can be localized onto damage prone areas rather than deposited throughout the entire composite structure, enabling the fabrication of nano-engineered hierarchical composite systems with tunable CNT distribution for improved material performance [33,34]. Consequently, the spray coating technology for CNTs deposition onto fibrous reinforcement has the most feasibility for large-scale industrial production.

In this work, CNT with remarkable mechanical, electrical and thermal properties was employed to create a hierarchical CNT-based CF reinforced HDPE composites. As a thermoplastic polymer, HDPE has numerous merits, such as high impact strength, low friction coefficient, low density, excellent chemical resistance, and good fatigue resistance. Nevertheless, the strength and stiffness still need to be improved for the potential engineering applications. To the best of our knowledge, no related report has been published regarding the effects of CNT coating on the extrusion of CF reinforced thermoplastic composites. Via spray coating technique, CNT was deposited onto the surface of CF and then compounded into CNT/CF/HDPE composites by extrusion and injection molding (Fig. 1). The interfacial properties of the hierarchical composites were compared to the counterparts without CNT coating. In addition, the resultant nano-engineered composites were investigated in terms of mechanical, thermal and morphological characterizations.
Fig. 2. Tensile properties of CF/HDPE composites without and with CNT coatings: (a) tensile strength, (b) tensile modulus and (c) elongation at break. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Fig. 3. Flexural properties of CF/HDPE composites without and with CNT coatings: (a) flexural strength and (b) flexural modulus. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)
2. Experiments

2.1. Materials

The HDPE pellets were purchased from Qenos (Melbourne, Australia) under the tradename of Alkatan G7660. The melt flow index (190 °C, 2.16 Kg) was 0.3 g/10 min according to ASTM D1238. The mass density and average pellet size were 0.959 g/cm³ and 3 mm, respectively. The polycrylonitrile-derived CF in chopped form was purchased from ACP Composites (Livermore, USA) as the reinforcement. Its mass density and average length was 4.6 mm. The commercial functionalized MWCNTs with hydroxyl groups were manufactured by CVD technique and their average diameter was 20 nm.

2.2. Spray coating

Prior to spray coating, 1 mg/ml CNT/ethanol suspension was prepared by using an ultrasonic water bath (WUA-A09H, Thermoline Scientific, Australia) to eliminate the agglomeration of CNTs. The ultrasonication was lasted for 10 min at the frequency of 35 kHz. Due to the light weight of chopped CFs, ethanol soaking was applied to them in advance to prevent the CFs blowing during the spray coating process. Then, the ultrasonic treated CNT/ethanol suspension was loaded and spray deposited onto the surface of CFs with 1 wt% concentration by utilizing an air spray gun (Voilamart, Australia). The pressure was set at 20 psi with a flow rate of 0.2 ml/s from a 10 cm working distance. The CNT-coated CFs were dried for 24 h before the commencement of hierarchical composite fabrication.

2.3. Extrusion and injection molding

The fillers (i.e. uncoated CFs or CNT-coated CFs) and matrix (i.e. HDPE) with different ratios were fed into a laboratory scale miniature twin-screw extruder (WLG10, Xinshuo Precision Machinery, China), where the screw speed and barrel temperature were set at 60 rpm and 200 °C, respectively. The mixtures were compounded in the extruder for 5 min and then transferred to the injection molding machine (WZS10D, Xinshuo Precision Machinery, China). During the injection molding process, the pressure was maintained at 980 bars for 10 s. The cylinder temperature was set at 200 °C and the mold temperature was 60 °C. Samples for the following tensile and flexural tests were fabricated by employing corresponding molds. All these samples can be categorized into two groups.

1. Without CNT coatings: HDPE, 5% CF/HDPE, 10% CF/HDPE, 15% CF/HDPE, 20% CF/HDPE and 25% CF/HDPE
2. With CNT coatings: HDPE, 5% CNT-CF/HDPE, 10% CNT-CF/HDPE, 15% CNT-CF/HDPE, 20% CNT-CF/HDPE and 25% CNT-CF/HDPE.

2.4. Characterization

Both tensile test and flexural test for PE and composites were performed at a crosshead speed of 5 mm/min by using a universal testing machine (MTS CMT 5205, SANS, China). The tensile strength, modulus and elongation at breakage were measured in accordance with GB/T 1040 (sample size: 75 mm × 10 mm × 2 mm) while the flexural strength and modulus were measured in accordance with GB/T 9341 (sample size: 80 mm × 10 mm × 4 mm). Five parallel samples for each composition were employed in the tests above. The Vickers hardness was determined under the ambient conditions by utilizing a micro-hardness tester (HVS-1000, Jiming, China). The load of 50 gf was applied for 10 s. Data with ten replicates were measured and averaged for each sample.

The crystallization behaviors were examined by using a differential scanning calorimeter (DSC 204, NETZSCH, Germany). The granulated samples were extracted from the cross-section of extruded plates with the averaged weight of 10 mg. They were first heated from 40 to 200 °C with 10 °C/min increments, and then maintained at this temperature for 5 min so as to eliminate the influence of heat history. With the same scanning rate, these granulated samples were then cooled down to 40 °C so as to analyze the crystal temperature and crystallinity. All these samples were scanned again from 40 to 200 °C with the same increments in order to investigate their melting conditions. Nitrogen was applied as the chamber gas. According to DSC results, the crystallinities Xc of PE, CF/HDPE and CNT-CF/HDPE composites were calculated via the following equation:

\[ X_c = \frac{\Delta H_m}{(1 - w) \times \Delta H_m^0} \times 100\% \]  

(1)

where \( \Delta H_m \) is the melting enthalpy after thermal history, \( \Delta H_m^0 \) is the enthalpy of 100% crystallization of HDPE, which is 293.6 J/g, and w is the filler ratio of composites.

The surface morphology of CF and the fracture morphology of CF/HDPE composites were observed by employing a scanning electron microscope (SEM; SU-1510, HITACHI, Japan) with the acceleration voltage of 15 kV. All samples were sputter-coated with gold before analysis so as to increase the electric conductivity and SEM resolution.

3. Results and discussion

3.1. Mechanical properties

The results of tensile tests for pure HDPE, CF/HDPE composites and hierarchical CNT-CF/HDPE composites with varying CF contents were listed in Fig. 2. Compared to the pure HDPE polymer, the tensile strength of CF/HDPE composites exhibited an obvious upward trend, increasing from 29.82 MPa to 50.88 MPa by 70.62% (Fig. 2a). This result can credit to the high strength and modulus of CF as the reinforcement to transfer the stress, but yield more stress concentration. Short fiber ends at the same fiber content [37]. Both of these are detrimental to the ultimate mechanical performance. Consequently, the chopped CF utilized in this research was long chopped CF
with the average length of 4.6 mm, which exceeded the normal length of reinforced fibers incorporated into the thermoplastic composite fabrication.

In addition, the tensile strength and modulus also gradually increased for CNT-CF/HDPE composites with the rise of CF contents (Fig. 2a and b). Although the two tensile indicators increased for these two groups of composites, the values observed for CF/HDPE composites with CNT coatings were higher than those from uncoated counterparts. The introduction of CNT produced positive effect on the tensile properties by virtue of its improvement in the interfacial bonding between CF and HDPE. With the good adhesion between reinforcement and matrix, the stress transfer capacity of the composite was improved in terms of the reduction of mutual debonding. Compared to the pure HDPE, the elongation at break corresponding to the maximum displacement decreased with the increase of CF contents. It might be explained by that the ductility of HDPE matrix was offset by the high stiffness of CF. Meanwhile, after the CNT modification, the stronger adhesion at the interphase of CF and matrix further decreased this value at the same fiber content.

The flexural stiffness and strength are extremely important for composites to prevent the potential deformation and bending failure while subjecting to high load and sliding velocity. Based on this consideration, the flexural properties of pristine HDPE and CF/HDPE composites with and without CNT coatings were also explored by three-
point flexural test. As can be seen from Fig. 3, the flexural strength and modulus showed the same tendency with the tensile strength and modulus, while the tensile strength was comparatively higher than the flexural strength. With the increase of CF content, the flexural strength and modulus from both untreated and CNT-coated CF/HDPE composites increased apparently compared to the pure polymer while the hierarchical CNT-CF/HDPE composites possessed higher values than uncoated composites. When CF content reached 25 wt%, the flexural strength and modulus of CNT-coated composites climbed to 392.76 MPa and 1309.19 MPa, which are 25.89% and 19.79% higher than those from untreated counterparts. Likewise, all enhancements in flexural performance can be attributed to the high stiffness of CF and improved interfacial bonding between the reinforced fibers and the matrix.

The Vickers hardness of hierarchical CF/HDPE composites with CNT spray coatings was compared with those without coatings as a function of varying fiber contents. Higher hardness is favourable in engineering application for its stronger resistance to penetration, wear, cutting and scratching [38]. As shown in Fig. 4, with the increase of CF contents, the hardness of CF/HDPE composites exhibited an upward tendency. This can be ascribed to the increased number of linkage points with higher fiber content in the skeletal composite structure, which plays a crucial role in the improved resistance to external pressure by hindering the movement of matrix polymer chain [39]. Meanwhile, for the same content of CFs, PE reinforced by CNT-coated CFs had higher hardness values than the counterparts without CNT coatings. Compared to the composites without CNT coatings, the hardness of hierarchical CNT-CF/HDPE composites was increased by 22.67% when the CF content was 25%, which indicates that the interfacial bonding between fiber and matrix was largely enhanced. In generally, the synergistic mechanism originated from higher CF content and stronger interfacial adhesion modified by CNT coatings endowed the hierarchical composites with superior hardness.

3.2. Thermal properties

The crystallization and melting patterns of pristine HDPE, CF/HDPE composites and CNT-coated CF-HDPE composites obtained from DSC measurements were studied since fillers in a crystalline polymer matrix generally have the potential to alter thermal behaviors of the matrix. Based on DSC thermograms of the melting and cooling processes from different specimens (Fig. 5), their melting temperature (Tm), cold crystallization temperature (Tc), melting enthalpy (ΔHm) and degree of crystallization (Xc) were measured and listed in Table 1. It can be clearly viewed that Tm did not change significantly after the addition of CF, whether it was for CNT-coated or not. However, Tc in most cases for CF reinforced composites was lower than for pristine HDPE (112.21 °C), except those composites with high CF content, such as Tc for 25% CF/HDPE composite (112.58 °C) and Tc for 20% CNT-CF/HDPE composite (113.10 °C) and 25% CNT-CF/HDPE (112.89 °C) composite. This can be elaborated by the interaction mechanism existing among CNT, CF and HDPE matrix. With the introduction of long chopped CF, the friction between fibers and polymer chains can obstruct the mobility of the macro-molecular chain and thus hinder the formation of highly ordered alignment of the crystal lattice [40]. It has been reflected by the decrease of Tc and Xc for both uncoated and CNT-coated CF/HDPE composites compared to that for the pure HDPE. Simultaneously, for the CNT-coated hierarchical composites, the CNTs were mainly coated on the surface of CF and rarely dispersed in the matrix by the shear force of extrusion, which acted as the effective heterogeneous nuclei, to a certain extent, facilitating the crystal growth during the crystallization process.

### Table 1

<table>
<thead>
<tr>
<th>Samples</th>
<th>Tm (°C)</th>
<th>Tc (°C)</th>
<th>ΔHm (J/g)</th>
<th>Xc</th>
</tr>
</thead>
<tbody>
<tr>
<td>HDPE</td>
<td>140.55</td>
<td>112.21</td>
<td>207.97</td>
<td>70.83%</td>
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<tr>
<td>5% CF/HDPE</td>
<td>139.98</td>
<td>109.31</td>
<td>196.24</td>
<td>70.35%</td>
</tr>
<tr>
<td>10% CF/HDPE</td>
<td>140.67</td>
<td>110.35</td>
<td>186.69</td>
<td>70.27%</td>
</tr>
<tr>
<td>15% CF/HDPE</td>
<td>140.62</td>
<td>110.17</td>
<td>172.55</td>
<td>69.14%</td>
</tr>
<tr>
<td>20% CF/HDPE</td>
<td>141.57</td>
<td>109.49</td>
<td>159.18</td>
<td>67.77%</td>
</tr>
<tr>
<td>25% CF/HDPE</td>
<td>139.55</td>
<td>112.58</td>
<td>145.91</td>
<td>66.26%</td>
</tr>
<tr>
<td>5% CNT-CF/HDPE</td>
<td>139.09</td>
<td>110.30</td>
<td>185.07</td>
<td>66.35%</td>
</tr>
<tr>
<td>10% CNT-CF/HDPE</td>
<td>140.18</td>
<td>110.58</td>
<td>179.32</td>
<td>67.86%</td>
</tr>
<tr>
<td>15% CNT-CF/HDPE</td>
<td>140.62</td>
<td>110.74</td>
<td>170.75</td>
<td>68.42%</td>
</tr>
<tr>
<td>20% CNT-CF/HDPE</td>
<td>140.78</td>
<td>113.10</td>
<td>164.65</td>
<td>70.10%</td>
</tr>
<tr>
<td>25% CNT-CF/HDPE</td>
<td>139.93</td>
<td>112.89</td>
<td>148.49</td>
<td>67.43%</td>
</tr>
</tbody>
</table>

![Fig. 6. SEM images of (a) raw CFs, (b) CNTs, (c) and (d) CFs with CNT spray coatings.](image)
process [41]. It is well explained why $X_c$ increased to the highest value of 70.10% for the 20% CNT-CF/HDPE composite, although a slight decrease was seen for the 25% counterparts due to the inhibiting effect from CF. It should be noted, however, that the existence of CNT acted not only as heterogeneous nuclei to promote the crystal growth, but also as restriction sites to hinder the regular arrangement of polymer chains [42]. In short, the intricate mutual interplay existing in CNT, CF and HDPE contributes to the complicated thermal behaviors for the hierarchical CNT-CF/HDPE composites.

3.3. Morphological properties

The surface morphology of CF after the spray coating of CNTs was first investigated via SEM analysis. As seen in Fig. 6a and b, the surface morphologies of raw CFs were extremely smooth while the CNTs exhibited long and curvy structure with fiber-like shape. After the deposition of CNTs on the fiber surface by spray coating, the morphology of CFs with surface modification had been remarkably altered, which was shown in Fig. 6c and d with different magnifications. The initial clean and smooth surface of CFs was wrapped by randomly oriented CNTs, resulting in a much rougher structure with many protrusions. Li et al. found similar results in the surface modification of CFs, where the commercially carboxylic- and hydroxyl-functionalized CNTs were selected to coat the sized CFs by employing an aqueous suspension deposition method [17].

The fractural morphologies of CF/HDPE composites with and without CNT coatings were next examined to qualitatively analyze the interfacial bonding between fibers and matrix. The pull-out CFs from the fractural surface of fiber reinforced composites can be clearly seen from all SEM images in Fig. 7. According to Fig. 7a and b, the morphological surfaces of CFs were very clean and smooth, which not only led to the big gaps between fibers and matrix, but formed numerous deep cavities from the direct pull-out of CFs. It obviously indicated the poor interfacial adhesion existing in the uncoated CF/HDPE composites. On the contrary, the gaps between CFs and HDPE matrix became much smaller for the CNT-coated CF/HDPE composites than those of the uncoated counterparts, as were shown in Fig. 7c and d. From the SEM image in Fig. 7d, it can also be seen that the HDPE matrix was attached onto the surface of CFs. These can be attributed to the improved interfacial bonding between fiber and matrix compared to the untreated composites. By virtue of the CNT spray coating onto CFs, the increased roughness on fiber surface facilitated the local interlocking between CFs and HDPE matrix, strengthening their mutual adhesion. The enhanced frictional force at the interphase based on the increased roughness also benefited the stress transfer from the matrix to highly mechanically performed fibers, which improved the entire strength of CF/HDPE composites.

In addition, it is worth noting from Fig. 7 that with the introduction of CNTs into CF/HDPE composites, the fractural morphology at the matrix has been changed, from the relatively smooth surfaces converting to highly rough structures, which indicated the toughening effect of the matrix given by CNTs. The obvious increase of toughness in the HDPE matrix can be realized with very low content of the introduced CNTs. During the twin-screw extrusion of hierarchical CNT-CF/HDPE composites, some CNTs were peeled off from the fiber surface and dispersed into the matrix, so that the toughening of polymer matrix was generated [43]. This result further promoted the release of stress concentrations and hampered the crack propagation, which was beneficial to form a stronger interphase. All of these SEM observations in the fractural morphologies of composites well corresponded to the result from the mechanical test mentioned in Section 3.1.

4. Conclusions

As a simple and economical coating method, spray coating of CNTs onto the surface of chopped CF was first reported for modifying the morphological properties of CFs. Then, the hierarchical CNT-coated CNT reinforced composites were fabricated by employing the extrusion compounding and injection molding techniques. In order to study the mechanical properties of CF/HDPE composites after CNT coatings, tensile, flexural and hardness tests were conducted. The results showed that when the CF content reached 25%, the tensile strength and flexural strength of CF/HDPE were improved by 70.62% and 40.38% compared to the pure HDPE. After the spray coating of CNTs, the tensile strength and flexural strength were further enhanced, as well as the modulus values. Besides, the Vickers hardness of both uncoated and CNT coated composites showed an increasing trend relative to the HDPE matrix, while the coated composites have higher values than uncoated

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**Fig. 7.** Fractural morphologies of (a) and (b) CF/HDPE composites without CNT coatings, (c) and (d) CF/HDPE composites with CNT coatings.
counterparts. With the CF content rose up to 25%, the hardness of the CNT-CF/HDPE composite was increased by 22.67% compared to the uncoated composite. It can be attributed to the high stiffness and strength of CF and the improved interfacial bonding between CF and the matrix after CNT coating, which was clearly observed from SEM images. The DSC analysis also illustrated the effect of CF and CNT on the crystallization of the HDPE matrix, both of which acted as heterogeneous nuclei and restriction sites to regulate the crystal growth of matrix in the fabricated composites. In conclusion, the spray coating of CNTs on CF was demonstrated to be a promising surface modification method to fabricate the CNT-CF/HDPE composites with the improved interfacial properties to enhance the mechanical performance of the hierarchical composites.

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Appendix A. Supplementary material

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