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Article

Multiwalled Carbon Nanotubes Polylactide Composites for Electrical Engineering—Fabrication and Electrical Properties

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Abstract: In the article, both the processes of manufacturing highly homogeneous polylactide resin/multiwalled carbon nanotubes (CNTs) and their properties are described. Regarding the application of carbon nanotubes polymer composites, one of the most important problems to solve is obtaining good dispersion of the filler in the polymer matrix. Preparation of polylactide/multiwall CNTs composites by quick polymer solidification and freezing the state of the dispersion of the nanotubes in the polymer solution is described. The method we used employs an increase in viscosity (carried out rapidly) of the sonicated polymer solution containing the CNTs by spray deposition. Good dispersion of the nanotubes is confirmed by electron microscopy. The obtained nanocomposites exhibit a low percolation threshold for electrical conductivity (above 0.25% by weight). The described method leads to obtaining an electrical conductive surface on virtually any material and reduces the small amount of an expensive filler (CNTs) needed to achieve good electrical conductivity. Furthermore, the carbon nanotubes used in the fabrication process of the composites were obtained using the liquid-source chemical-vapor deposition (LSCVD) synthesis method.

Keywords: polylactide; conductive composites; carbon nanotubes; polymer composites; organic electronics

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1. Introduction

Polymers are materials that are used today almost everywhere, in every aspect of human lives and in every field of industry. The reasons are the lower prices of these materials in comparison with metals, and lower manufacturing and processing costs. Pure polymers are (the vast majority) insulators and thus can be used as insulating materials, electric-device housing and packaging. In order to use polymers as electrically conductive materials, there is a need of using conductive fillers. In the literature, one can find much information on the use of conductive fillers to obtain electrically conductive polymer composites. There are reports of the use of metal nanoparticles as the fillers: nickel [1], copper [1,2] or silver [3] as well as carbon fillers, such as amorphic carbon [4], carbon fiber [4,5], graphene or carbon nanotubes [6–8]. In the case of metal nanoparticles, the percolation threshold was of the order of 4 to 8% by weight, for soot or carbon fiber of 4% by weight. In the case of carbon nanotubes, the span is significant and ranges from 0.0025% to 11% by weight, depending on the manufacturing technique, the type of carbon nanotubes and the type of polymer matrix. [9] A summary of the values in question is set out in the table below (Table 1) [10–13].

The composites can be used either as static electricity or electromagnetic waves shielding or in organic conductive wires. In the publication, polylactide (PLA) was used. Polylactide is one of the most important biodegradable and biocompatible polymers. It can be produced from renewable resources [19] and it is environmentally friendly. It exhibits good physical properties, which can be further altered by filling with various additives. These features make PLA an attractive alternative for synthetic plastic materials of petrochemical origin, which degrade slowly even after hundreds of years. In this paper, we present methods of preparation of nanocomposites with good dispersion of carbon nanotubes (CNTs).

Table 1. Percolation threshold of electrically conductive polymer composites.

Polymer Matrix	Preparation Method	Filler	Percolation Threshold [wt %]	Reference
PVC	Powder compaction	Ni	8.5	[1]
		Cu	5	
EVA	Compression moulding	CCB (carbon black)	14	[14]
		CF (carbon fiber)	7	
		MWCNT (multiwalled carbon nanotubes)	3	
Phenolic resin	Compression moulding	Graphite	40	[15]
		Expandable graphite	12	
		Carbon fiber	10	
Epoxy resin	Solution mixing/stirring	MWCNT	0.0025	[16]
Polystyrene	Solution mixing/stirring/film casting	MWCNT	$\leq$3	[17]
Poly(ethylene terephthalate)	Solution mixing/compression moulding	MWCNT	0.9	[18]

Since their discovery [20], carbon nanotubes have aroused great interest among researchers as a material itself and for many other applications, such as a polymer-composite filler. The main reason for that is the properties of the CNTs, such as high thermal and electric conductivity and excellent mechanical properties [21–23]. To be able to use the properties of the CNTs, it is necessary to obtain a proper dispersion in the polymer matrix, which is a problem for the material in question and indeed the biggest disadvantage. Moreover, uniform good dispersion of the filler particles in the polymer matrix is the most important parameter in terms of composite performance. The CNTs used during experiments were synthesized in the lab using the LSCVD synthesis technique [24]. Carbon nanotubes come from the synthesis in the form of the "carpet", which causes aggregation and the formation of agglomerates. In order to use carbon nanotubes in nanocomposites, it is necessary to remove agglomerates and use techniques that lead to obtaining a good dispersion. There are several methods to obtain the dispersed CNTs, and due to further planned experiments involving spraying the layer of the composite on a target surface, we used an ultrasonic treatment of the CNT solution. Even though it is one of the most effective methods, the dispersion state is not stable in the case of using organic solvents and the CNTs re-aggregate quickly. Surfactants such as SDS (sodium dodecyl sulfate) [25] or other ionic compounds can be used, but their drawbacks include an unwanted modification of composite properties, as these additives remain in the material after the solvent evaporates. The best possible method to prevent the re-aggregation consists of the effective use of ultrasonication and freezing of the state of uniform dispersion. This method is spray—the drying technique described in this paper, which also leads to obtaining the highly homogeneous nanocomposite, suitable for use in electrical engineering, and provides evaporation of the solvent. The method presented also allows the application of an electrically conductive composite on virtually any solid surface, which was the goal of the experiments and is what makes possible the use in electrical application.

2. Methods and Results

Polylactide (PLA) used as the polymer matrix was provided by Cargill-Dow. It contains 95.9% of L-lactide and 4.1% D-lactide. Reagent grade trichloromethane ($CHCl_3$)

was used as a solvent for polylactide and was provided by Chempur Poland. The same reagent was used to obtain a dispersion of carbon nanotubes, before adding the PLA granules to the solution.

The multiwalled carbon nanotubes, used as a filer in the nanocomposites studied in this paper, were synthesized in our laboratory. The synthesis method chosen was liquid-source chemical-vapor deposition (LSCVD). For the synthesis process, a three-zone furnace with individual temperature controller for each zone and valves controlling the flow rates of the carrying gases were used. That setup allowed setting and controlling the precise temperature during the synthesis. The method choice was dictated by the ease of change of the key synthesis parameters, such as the catalyst solution concentration, the flow rate of the carrying gases, and the temperatures of both the catalyst solution evaporation and the deposition (CNT growth). Alternating these conditions influences the material parameters of the outcoming carbon nanotubes, such as the average length, the purity or the synthesis efficiency [26–28].

For each synthesis, the deposit material was examined using a thermogravimetric technique applied to the CNTs coming from the synthesis in the form of the carpet, which can be seen in the scanning electron microscope (SEM) image—Figure 1.

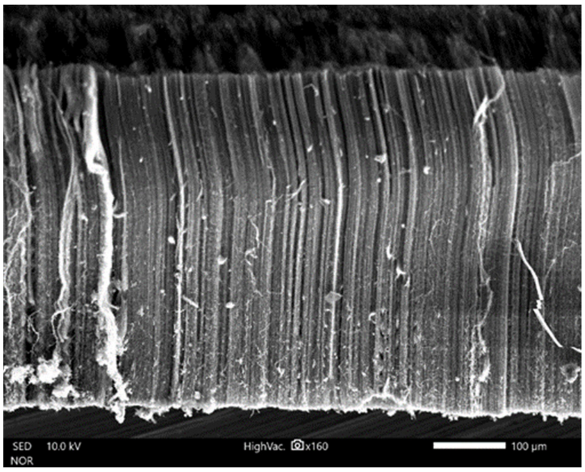

Figure 1. Carbon nanotubes in form of the "carpet"—raw material from synthesis.

CNT carpet is made of many single carbon nanotubes grown during the synthesis perpendicular to the carpet's base. The CNTs in that form are not suitable as filler material due to being entangled during the synthesis process, which is visible in Figure 2.

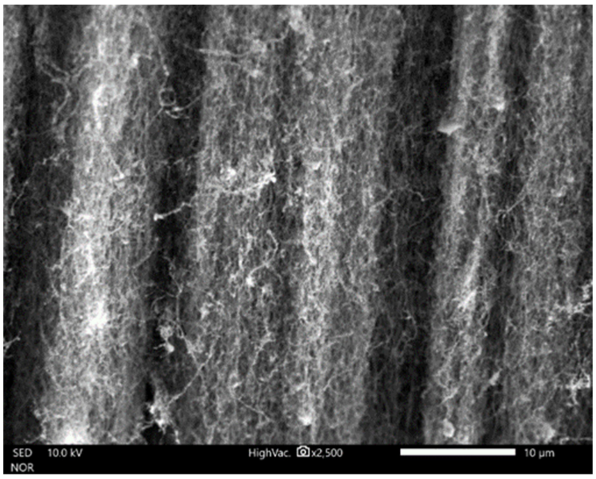

Figure 2. Magnification of a side of the "carpet" showing multiple entangled bundles—SEM image.

There are also many CNT bundles and agglomerates that are not visible until the moment of creating the solution. That causes the need to employ proper techniques leading to obtaining individual disentangled CNTs (proper dispersion state).

One of the most effective dispersion methods and also widely used is preparing the solution of the carbon nanotubes in solvent having appropriate properties. The previously mentioned water and surfactant-added solution was not an acceptable option due to the problem of adding ion compound to the system. As a consequence, additional removing of the water residue from the composite is required, which is very difficult and unsuccessful and could influence the electrical and mechanical properties of the obtained material. The other problem was the solubility of the polylactide matrix, which was the second important factor determining the choice of the solvent; thus, for the experiments, reagent grade trichloromethane ($CHCl_3$) was chosen. It also has satisfactory CNT dispersion parameters, with a Hansen solubility parameter of 18 [29].

The dispersion time for each specimen was 90 min. The dispersion effectiveness examination process was two-stage. The first one was an initial examination using a scientific-grade optical microscope working in transmission mode. Subsequently, after the sonication ended, the droplet of $CNT/CHCl_3$ solution was placed between the slide glass and the coverslip. Observation on this scale gives us a much better picture of the dispersion, but we are not able to observe the individual nanotubes, but rather the "ropes" formed from them, where several nanotubes joined by van der Waals forces are visible as a long object, often with a diameter of microns down to a few microns. The possibility of observing smaller objects using the described technique is not possible due to the wavelength: the length of the light used for observation must obviously fall within the range of visible radiation, i.e., $\lambda = 400 \div 700$ nm.

The initial state (about 1 min after dispersion) is shown in Figure 3a and the state after 30 min in Figure 3b.

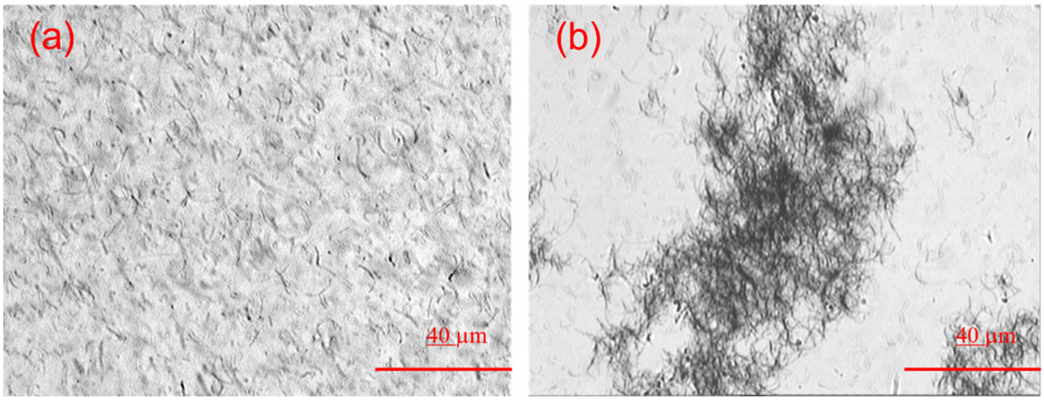

Figure 3. CNT solution in trichloromethane (**a**) 1 min after the end of the ultrasonication, (**b**) 30 min after the end of the ultrasonication.

The experiments prove that the re-aggregation process is very fast; it is only a matter of minutes if the solution can be used for the fabrication of the composites. Determination of the proper ultrasonication time with a light microscope was followed by scanning electron microscopy—the second stage. A scanning electron microscope Jeol JSM200 was used. While SEM does not give the information on a big scale, the technique allows the detection of bundles and micrometer-scale agglomerates of the CNTs. Prior to the examination, a droplet of the solution was placed on the Si wafer and left till the solvent evaporated.

From what is visible on the SEM micrograph (Figure 4), there are no CNT bundles or agglomerates. Having been separated in the dispersion process, carbon nanotubes can be observed. That experiment led to optimizing the minimal time, thus guarantying a uniform dispersion and the maximum amount of the CNTs in the solution, which allowed the disentanglement of the bundles and agglomerates disintegration.

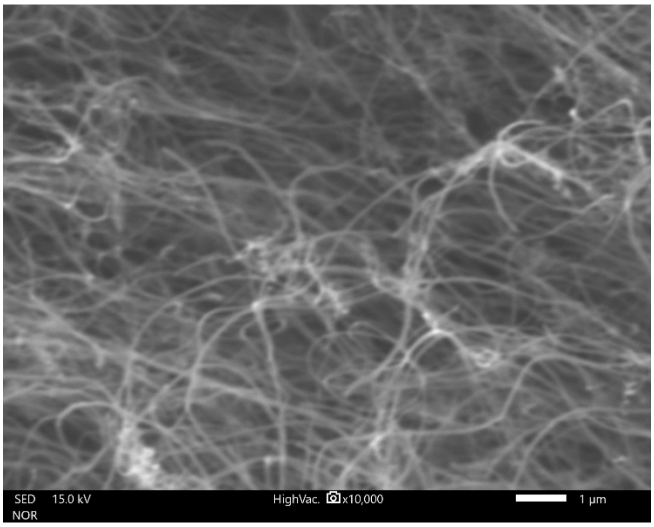

Figure 4. SEM image of carbon nanotubes on Si wafer, after dispersion in $CHCl_3$ (90 min).

The next step in the fabrication of the composites was adding the PLA granules to the solution and ultrasonication until the polymer has been fully dissolved. Adding polylactide directly to the dispersed CNTs also causes the rise in viscosity and acts similar to the surfactants in terms of dispersion stability, though the effect is not as long-lasting.

$CHCl_3$ is a good solvent of PLA, and it can be effectively removed from the polymer during the spray-drying process. There is no need of using a heating target or preheating solution in order to remove the solvent from the polymer/carbon nanotube thin film created in the spray-drying casting technique. Moreover, the thickness of the obtained composite layer can be easily controlled by the amount of carrying gas used in the process and the number of sprays. As the carrier gas during the spraying process, atmospheric air was used. Other gases can also be applied, but the choice was dictated by the ease of applying the composites to fabrication process in an industry environment, as it allows the usage of multiple sources of air and pressure change. Another important feature of the described composite manufacturing process is a rapid rise of viscosity due to solvent evaporation that allows achieving the desired dispersion freezing effect.

The PLA/CNT solution was sprayed on the glass surface, forming the nanocomposite foil. Prior to the electrical examination, similar to what was previously described, the microscopic two-stage process was performed. The additional examination was conducted for each sample with a thermogravimetric technique—TA Instruments Q50 apparatus. In the case of the examined specimens, both light and electron microscopy investigations are possible. The obtained foil is semi-transparent and the CNTs act as the gray filter. As was the case with observing the solution, it is not possible to observe the single carbon nanotubes, but the detection of the agglomerates can be carried out. The next step is the composite SEM imaging. Figure 5 shows the CNTs in the PLA matrix.

Figure 5 reveals the formation of a conductive array of carbon nanotubes in the polymer matrix. The CNTs form conductive paths, which enable the effective discharge of the electric charge coming from the microscope beam. Typical for most polymers is the need of applying the conductive layer at the surface prior to the SEM examination, which makes the dispersion analysis possible. The lack of charging effects also indirectly confirms the manufacturing of the electric conductive composites. Both the amount of the CNTs and the presence of no solvent in the composite were confirmed using a thermogravimetric technique (TGA) in the case of each sample before using the electrical analysis. The result obtained for one of the examined specimens is presented in Figure 6. The apparatus used was a TA Instruments Q50.

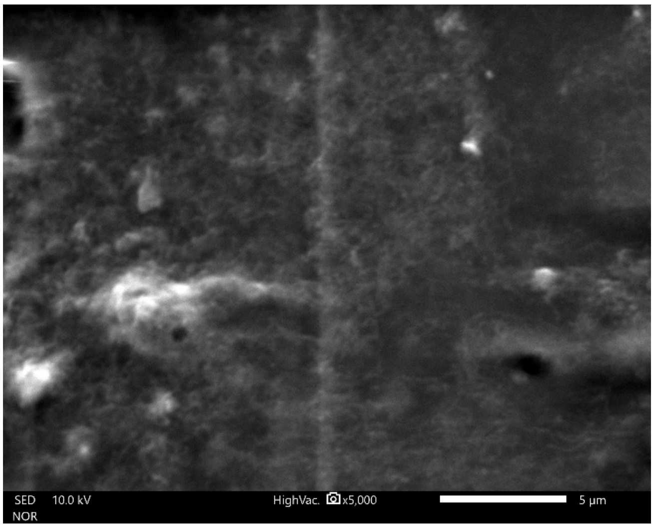

Figure 5. SEM image of the surface of the PLA/CNT nanocomposite foil (2% by weight content).

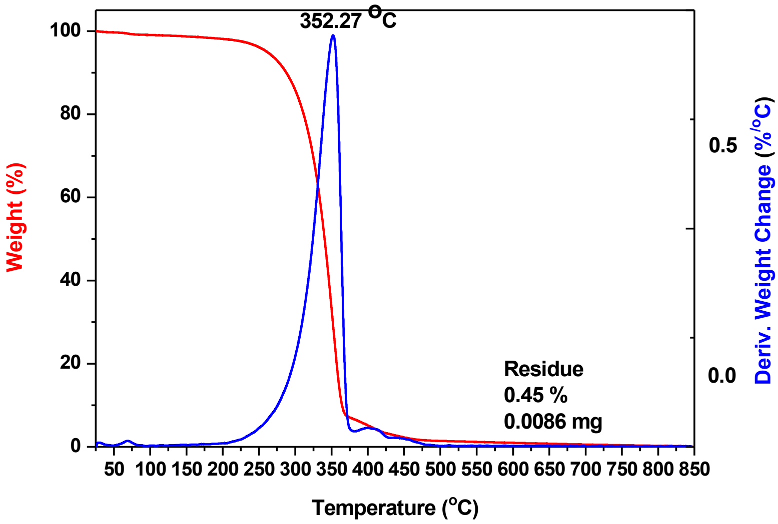

Figure 6. TGA graph of carbon nanotubes/polylactide composite; Residue is 0.45%.

TGA analysis shows the negligible presence of a solvent, since the peak corresponding to its presence is almost not observable (less than 0.5% by weight for all examined specimen), which confirms the effective evaporation of the $CHCl_3$ during the spraying process. Moreover, during the spraying process, the substrate was not heated, and the solution was at room temperature. Heating would improve the solvent evaporation, but one of the key assumptions in the development of the composite fabrication technique was to develop the simplest possible process in terms that are industry-friendly.

The electrical conductivity of the obtained nanocomposites was examined for the multiple weight contents of the CNTs. The range of the carbon nanotube content was 0 (neat polymer) to 10% by weight. Pure PLA, like the majority of polymers, is an insulator. By producing composites with a conductive filler, we created a percolation network in the material through which charges can flow. The more conductive "paths" formed in a material, the higher its conductivity. By constructing a diagram of the dependence of the specific conductivity of the samples depending on the weight content of CNTs in the matrix, we obtained the information about the filler needed to obtain the electrically conductive material. The result is shown in Figure 7.

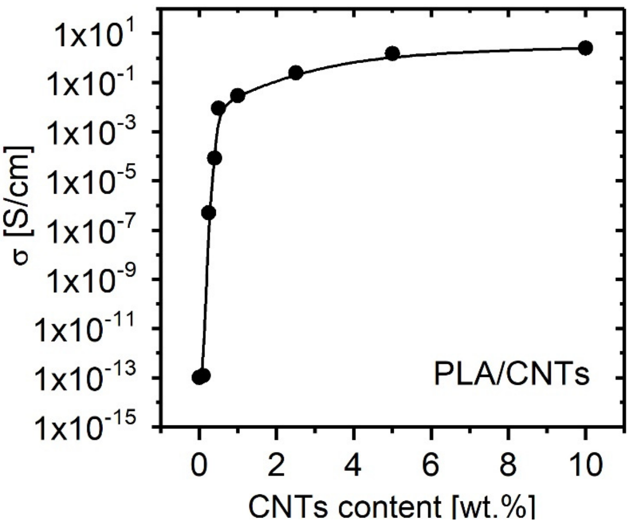

Figure 7. Percolation curve plot—PLA/CNT nanocomposites specific conductivity vs CNT weight content.

An increase in specific conductivity is visible from the conductivity value of the neat matrix itself, i.e., about 10^{-13} S/cm to 2.7 S/cm for the highest tested content, i.e., 10% by weight, where the plateau is reached. All the conductivity results on the graph are the mean values of the five samples, obtained using the four-electrode method, to minimize a possible error based on the specimen material defects. In order to determine the percolation threshold for the obtained composites, the percolation theory was used, and the calculations were based on Equation (1) [30–32]:

$$\sigma_{(p)} = \sigma_0(p - p_c)^t, \tag{1}$$

where p is the filler content, p_c is the percolation threshold and t is the critical exponent, corresponding to the dimensionality of the conductive network and usually having values of 1 for the two-dimensional network and 1.65 for the three-dimensional. According to the above dependence, it is possible to determine the percolation threshold p_c and the critical exponent t. Determining these values is possible after plotting the conductivity dependence plot s versus $p - p_c$ on the log- log scale. The resulting plot is shown in Figure 8.

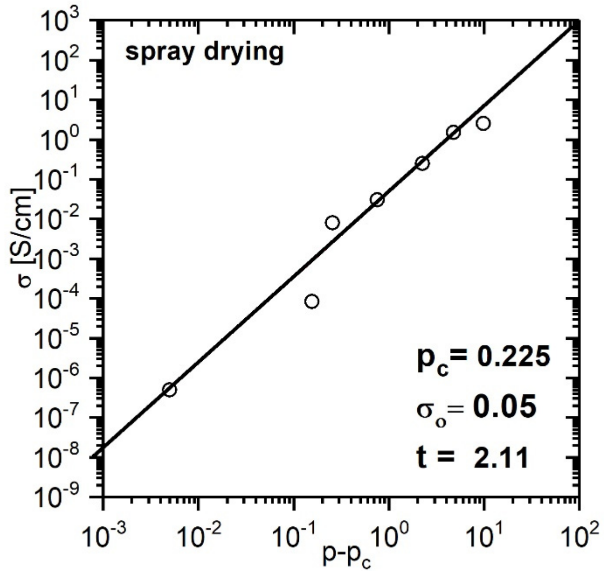

Figure 8. PLA/CNT nanocomposites specific conductivity in constant electric field vs. $p - p_c$.

According to the calculation, the percolation threshold value is 0.225% by weight and the critical exponent is 2.25, which indicates a 3-D array. The p_c is the calculated percolation threshold. According to the model assuming the random distribution of the filler particles in the polymer matrix, the pc value depends on the dimensionality of the filler particles and the aspect ratio D/d, where D denotes the diameter and d corresponds to the length [33].

3. Discussion

The developed method of carbon nanotube dispersion and preparation of PLA/CNT nanocomposites allows for the effective use of ultrasound treatment in order to disintegrate agglomerates of carbon nanotubes and obtain electrically conductive nanocomposites with high homogeneity and good dispersion of filler particles in the matrix. Furthermore, the method is easily applicable for different polymers by changing the solvent used. The obtained composites can be applied directly on the target surface without the need of using other subsequent methods such as injection molding. There are many works in the area about PLA/CNT nanocomposites. What is different and makes our work unique is the method of application (spray drying), which makes it highly “industrial friendly” and easy in terms of using expensive equipment. The percolation threshold is quite lower than in most of the current works, since the dispersion of the filler is achieved due to rapid viscosity rise. The combination of those factors has an advantage compared to the two- or three-stage process mentioned in Table 1 and proves the possibility of commercial applications.

The polylactide used in the experiments is one of the “green polymers”, meaning that it is obtained from biodegradable resources and is itself biodegradable. Hence, the obtained material is more environmentally friendly compared to polymers of a petrochemical origin.

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