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Influence of the polymer molecular weights on the electrical properties of Poly(vinyl alcohol) – Poly(ethylene glycols)/Graphene oxide nanocomposites

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ABSTRACT

Graphene has unique electrical properties at room temperature properties with a flexible 2D layer and stronger material over discovered and use to enhance the electrical properties of materials. This study investigated the influence of two important factors the molecular weights (Mw) of polymer and the concentration of nanofillers on the electrical properties of samples. Hybrid nanocomposites were fabricated from polyvinyl alcohol (PVA) and polyethylene glycol (PEG) with three different molecular weights (4 k, 8 k, and 20 k) using developed mixing-sonication-solution methods. This method was successfully exhibited a fine homogeneity and good dispersion of both polymers and nanofillers in the matrix as displayed by using an optical microscope (OM). The effect of the addition of graphene oxide on the alternating electrical properties showed a reduction in the dielectric constant and the isolation loss of the polymer compounds (PVA-PEG-GO) with increasing the frequency of the electric field and the loss of isolation increased. It was also shown that the increase of electrical conductivity is noticeable with samples by increasing the molecular weight of PEGs to the highest at 20 k Mw. Increasing the polymer molecular weight in addition to the contribution of GO revealed an extraordinary improvement of the electrical properties that bring promising and performance materials for electronic, photovoltaic devices, heterojunction, solar cells, and energy storage systems, other substantial applications.

Selection and peer-review under responsibility of the scientific committee of the 3rd International Conference on Materials Engineering & Science.

1. Introduction

Polymers are promising that changed the world's concept of industrial and replaced most of the materials with plastic materials and diverse applications. It also entered all industries and various optical, electronic, and applications such as photovoltaic cells and optoelectronic, including medical industries [1,2]. Despite that, polymers are considered isolated and weak mechanically as well as in most other properties due to defects in the structure. To overcome these drawbacks on polymers, nanocomposite technology is one of the significant methods applied. It is successful to develop several properties such as electrical, physical, thermal and mechanical, etc. [3] using an impressive nanofiller. In last decay, graphene is considered an important factor and best fillings that bring amazing and unique properties. It is strongly affecting the polymer nanocomposites [4] and developed many applications [5]. Graphene oxide (GO) has large surface areas in addition to several and varied functional groups, and other unique properties as one of the most impressive materials [6]. It easy to interact and compose with a wide range of materials that are exhibited to significant development and manipulated all the material properties [7].

Poly(vinyl alcohol) (PVA) and poly(ethylene glycol) (PEG) have significant properties as biodegradable polymers with low toxicity and biocompatibility [8,9]. These unique polymers have many significant functional groups that increase their ability to compatible with other materials, nanofillers and polymers, etc, that make them attractive materials for scientists, engineering, and researchers [10,11]. PVA and/or PEG are investigated with a range of other materials, polymers, and filers, etc. to overcome their weakness or developed their properties for several or specific applications.

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Chengjie Li et. al. [12] used a solution method to exam the influence of the loading of GO of the PEGs with different molecular weights. The result showed to improve the mechanical and thermal properties, where the higher molecular weight of PEG (20 k) deposited a strong hydrogen bonding interaction with the GO nanosheets. Other groups [13] focused on the effect of the various concentration (from 0.1 to 0.3 g) of GO to synthesis PVP/PVA-GO to improve the optical properties. The energy gap and the absorption edge are improved to 4.28 eV and 4.45 respectively, with increasing the concentration of GO. A similar conclusion was reported by another investigation by Fahad H. Falqi et al. [14]. They reported significant improvement in the thermal stability of the PVA-PEG polymer after the addition of the various ratio of graphene from 0.1 up to 1 wt%. Even a lower values of molecular weight of PEGs (200, 800, 2000, and 6000) g mol^{-1}, this factor displayed significant influence with the GO as a biosensor to determine the profenofos for food as reported by Xiong Jin'en et al. [15].

Despite several investigations [12–15] to discover these materials with various factors, the effect of molecular weight still has a lack of knowledge and does not fully understand the interfacial interaction and relation between the molecular weights with low loading ratio of GO. However, the effect of molecular weight and substantial band gap alteration of graphene oxide on the electrical properties. For example, the energy of the bandgap of the PEG-PVA as nanocomposites has become very interesting recently [16,17] and an important factor that not fully understand and covered [18]. To overcome this issue and reduce the gap of knowledge in this area, graphene oxide is considered an interesting nanofiller able to bandgap of the materials that could bring significantly enhance its applications, such as ozone treatment [16] and optical properties [19].

This investigation aims to understand the impact of the interfacial interaction between the effect of both the molecular weights of PEG in the PVA-PEG blend with the additional low loading ratio of GO on the structural and the electrical properties of the PVA-PEGs matrix. Developed the solution-sonication-acoustic method applied to fabricated the PVA-PEGs/ GO-based- hybrid nanocomposites and characterized using Fourier transforms infrared (FTIR), X-ray diffraction (XRD), Optical Microscope (OM).

2. Experimental parts

2.1. Materials

Poly(ethylene glycol) (PEG) with (4 k, 8 k and 20 k) g $mole^{-1}$ were supplied from Ltd company, Indi, whereas poly(vinyl alcohol), (with purity 99.8% and melting point 230 k), was purchased from Panreac company, Spain. Sigma-Aldrich Company, UK was provided the graphite powder with particle size $\leq 40\ \mu m$, which used to synthesis the GO, hydrogen peroxide, potassium permanganate ($KMnO_4$), sulfuric acid (H_2SO_4) with the analytical grade, 99.8%, hydrochloric acid (HCl, 35%) and sodium nitrate ($NaNO_3$).

2.2. Preparation of GO

Applying Hummer methods [20] on graphite to synthesized GO nanosheets following the same procedure and full details with characterizations in our previous studies [2,21].

2.3. Purification of polymer mixture and hybrid nanocomposites

Several hybrid nanocomposites were prepared, 1.5 g of poly(vinyl alcohol) was dissolved in 50 ml of distilling water at 70 °C for 1 h using a magnetic stirrer. 0.5 gm from the three molecular weights of polyethylene glycol (PEG) were dissolved in 50 ml of distilling using a magnetic stirrer for ½ hour. Then PVA was mixed with each molecular of PEG to get three polymer blends, the same procedure was applied to prepare three hybrid nanocomposites after the addition of GO to the polymers matrix. Where 0.1 g of graphene oxide was suspended in distilled with sonicated for ½ hour to get a full dispersion before mixed with the polymer matrix. All the six samples, blended polymers (PVA-PEGs) and hybrid nanocomposites (PVA-PEGs/ GO) were placed in a petri-dish under air for 96 h at room temperature for fully dry, as revealed in Table 1.

2.4. Characterization

Fourier transforms infrared (Bruker, Germany), Optical Microscope (Nikon – 73,346 from Olympus), LCR meter (HiTESTER 3532-50, HIOKI, India) were used to characterize the films of both blended polymer and nanocomposites.

3. Theoretical part

The Arrhenius equation is used to calculate the electrical conductivity (σ), which varies exponentially with temperature (T) [22].

$$\boldsymbol{\sigma_{dc}} = \boldsymbol{\sigma_o} \exp\left(-\boldsymbol{E_a}/\boldsymbol{KT}\right) \quad (1)$$

Where K and E_a refer to the Boltzmann constant and the activation energy, respectively. The dielectric constant (ε') of (PMMA-PVA/ GO) nanocomposites can be considered using the following equation [23].

$$\boldsymbol{\varepsilon}' = \frac{C_p}{C_o} \ldots \quad (2)$$

C_P means the parallel capacitance and C_o is the vacuum capacitor.

The dielectric loss (ε") is the dissipated energy as heat due to the absorbed the electric energy through the material that when the dielectric material is exposed to the A.C voltage, It can be calculated for (PMMA-PVA/GO) nanocomposites using the following equation.

$$\boldsymbol{\varepsilon}'' = \boldsymbol{\varepsilon}' \boldsymbol{D} \quad (3)$$

D means the factor of dispersion of (PMMA-PVA/GO) nanocomposites. The conductivity of A.C electrical (σ_{AC}) was determined by applying the following equation.

Table 1
Summarized the polymer mixtures and polymer hybrid nanocomposites.

Sample ID	Concentration g					Dry method
	PVA	PEG 4 k	PEG 8 k	PEG 20 k	GO	
PVA-PEG 4 k	1.5	0.4	–	–	–	At room temperature under air for 96 h
PVA-PEG 4 k/GO		0.4	–	–	0.1	
PVA-PEG 8 k		–	0.4	–	–	
PVA-PEG 8 k/GO		–	0.4	–	0.1	
PVA-PEG 20 k		–	–	0.4	–	
PVA-PEG 20 k/GO		–	–	0.4	0.1	

$$\sigma_{AC} = \omega\varepsilon_o\varepsilon'' \ldots \quad (4)$$

ω: Angular frequency ($\omega = 2\pi f$,ε_o means the vacuum permittivity and σ_{AC}means the generated temperature measurements in the isolated materials. That is caused by the charges vibration or dipoles rotation in their positions, as a consequence of the alternating of the field [24].

4. Results and discussion

The FTIR spectra of the PVA-PEGs blends films and hybrid nanocomposites with the wavenumber were displayed in Fig. 1. The characteristic function group of GO that at 3283 cm^{-1} stretching of OH, 1720 cm^{-1} of C = O, 1620 cm^{-1} of C = C skeletal ring stretching, 1360 cm^{-1} of -C–H bending and 1040 cm^{-1} of C–O–C epoxy stretching functional groups, that is agreed with structure of the GO in agreement with the literature [20,21]. Both PVA-PEGs blend polymer films and PVA-PEGs/ GO hybrid nanocomposites presented the functional group of all the components with some shifting or increasing in the intensity such as 3283 stretching and 1720 cm^{-1} and 1360 cm^{-1} of GO, 3219 symmetric stretchings of $–CH_2$, 1341.57 cm^{-1} of OH, – CH asymmetric and C – O Stretching vibration of PVA and 2917 cm^{-1} asymmetric stretching $–CH_3$, 1466 cm^{-1} of –CH blending, 1240 – 1106 cm^{-1} of C–O–C epoxy stretching and 961 $cm^{-1}$of C–C stretchings of PEGs, respectively [25].

These presented peaks exhibited a strong interfacial interaction between the molecular chains of PVA/PEG such as the ether group of polymers with C = O and the C–H group of GO. Hydrogen bonding is the major factor that affects and interacts between the GO nanosheets and PEG, where the hydrogen bond led to an improvement in the interfacial interactions between polymers and GO [26]. Where, the intermolecular hydrogen bonding between the polar functional groups, such as –OH and COOH of GO with ether group such as C–O–C sites of PEG chain interact [27]. All these peaks proved the successful fabrication of nanocomposites.

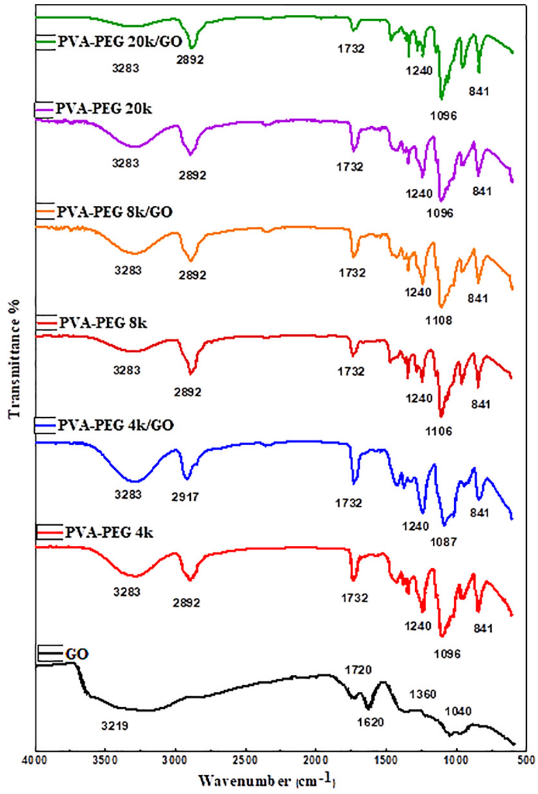

Fig. 1. FTIR spectra of PVA-PEGs films and PVA-PEGs/GO nanocomposites.

Fig. 2 illustrates the images of the PVA-PEGs films and (PVA-PEGs/GO) nanocomposites with 400X magnifications using optical microscopy. The blended polymer films show good interaction due to the fine homogeneous between the polymers, whereas the addition of graphene oxide nanosheets showed better and fine dispersion with increasing the molecular weight of PEGs from 4 k to 20 k. Moreover, the nanocomposites contained the 20 k of PEG exhibited the best homogeneity between polymers and good homogeneity of the GO in the matrix. This supports the previous finding and supports the FTIR spectra that showed better interaction of the polymer with increasing the Mw and GO, in agreement with other researchers' findings [19].

Fig. 3 shows the change of electrical conductivity of PVA-PEGs blend polymers and (PVA-PEGs/GO) hybrid nanocomposites with the frequency. The A.C. electrical conductivity notably improved with increasing the frequency because of the interpolarization that occurs at low frequencies, as well as the transmission of charge carriers by the process of hopping. The higher value of conductivity was reduced after 10^6 Hz, whereas after that it was rescued by increasing the frequencies to high values. The incorporation of GO displayed an important role in all nanocomposites to improve the conductivity in comparison with blend polymers. Also, the increase in the molecular weight showed another increase where the nanocomposites including 20 k PEG exhibited the best results compare with other nanocomposites included 4 k and 8 k of PEGs.

Fig. 4 illustrates the value of the dielectric constant with the frequency of PVA-PEGs blend polymers and PVA-PEGs/GO hybrid nanocomposites. The results showed a reduction in the results with increasing frequency. The effect of the low loading ratio of graphene oxide displayed a significant contribution to the value of the dielectric constant. Another finding that noticed the dielectric constant results improved with the increase in the molecular weight of PEGs from 4 k to 20 k, additionally to the contribution of GO. This could due to the increase in the value of the dielectric constant is to the formation of a network of graphene oxide nanosheets within the polymer matrix. The nanocomposites exhibited better results in comparison with their related polymer blend due to the contribution of GO. Meanwhile, increasing the molecular weight of PEGs to 20 k showed another improvement in nanocomposites compared with other nanocomposites.

The dielectric loss as a function of the frequency of the (PVA-PEG-GO) nanostructures is shown in Fig. 5. At lower frequency, the insulation loss value is high, and it decreases with the increasing frequency of all the blend polymers. This is due to the decrease in the contribution of interpolarization when increasing the frequency. Whereas, this behavior was significantly changed with the contribution of GO. GO contribution caused an increase in the value of dielectric loss with increasing the frequency to 10^3 Hz after that the values were reduced but even that is stilled higher than blend polymers. This is the highest absorption of scope for an Omnidirectional. This absorption is due to the Maxwell - Wickner phenomenon which was caused by the emergence of a C current. Due to the difference in the dielectric constant and the conductivity of the phases in the superposed material. This electric current has the same phase as the magnetizing field. When the frequency is increased to (10^3 Hz), the loss in the dielectric is nearly constant (PVA-PEG-GO). This is due to mechanisms of other types of polarization that occur at higher frequencies.

Another improvement in the results was related to increasing the molecular weight of PEGs to 8 k then 20 k of PEGs in the nanocomposites. The best values were presented to the nanocomposites included higher molecular weight at 20 k of PEGs in comparison with the other nanocomposites in agreement with the other above results.

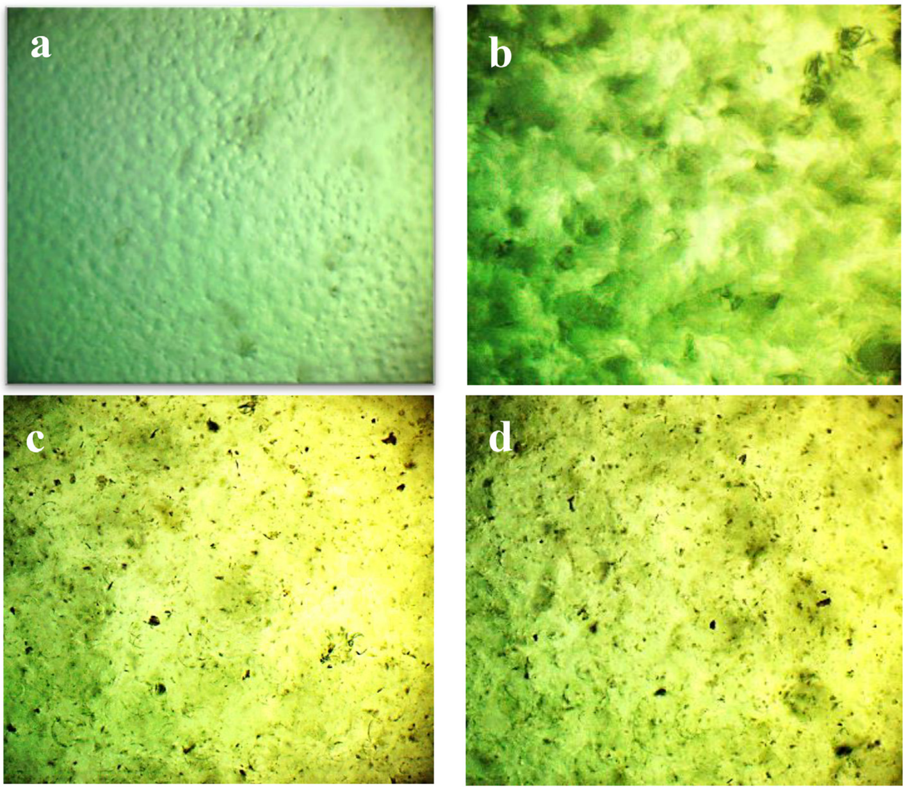

Fig. 2. The optical microscopy images with 40X magnification of (a) PVA-PEG films, (b) of PVA-PEG 4 k/GO, (c) PVA/PEG 8 k/GO, and (d) PVA/PEG20k/GO nanocomposites.

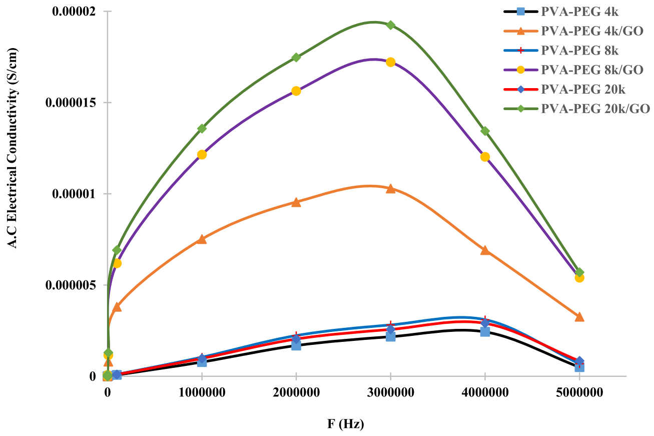

Fig. 3. The A.C. electrical conductivity with the frequency of PVA-PEGs blend polymers and PVA-PEGs/GO hybrid nanocomposites.

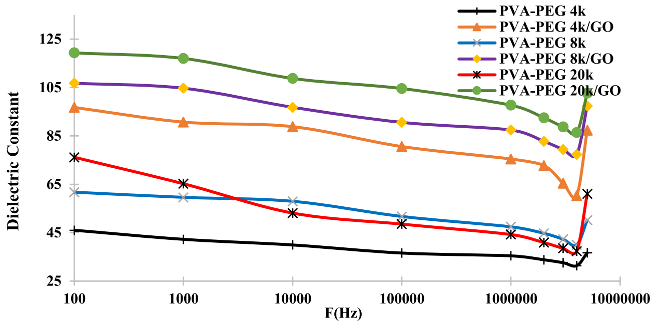

Fig. 4. The dielectric constant with the frequency of PVA-PEGs blend polymers and PVA-PEGs/GO hybrid nanocomposites.

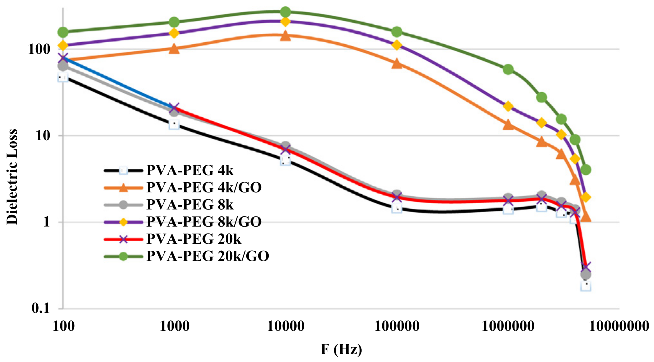

Fig. 5. The dielectric loss with the frequency of PVA-PEGs blend polymers and PVA-PEGs/GO hybrid nanocomposites.

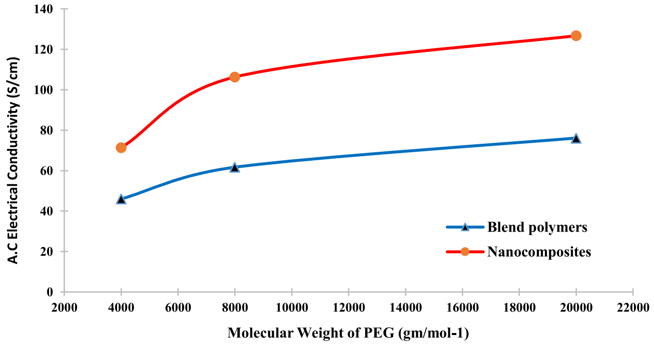

Fig. 6. The A.C Electrical Conductivity with the Molecular weight of PEG in the PVA-PEGs blend polymers and PVA-PEGs/GO hybrid nanocomposites.

This is due to the increase in charge carriers by increasing the molecular weight of PEGs, in agreement with other findings [6].

The change in the A.C. electrical conductivity of PVA-PEGs films and PVA-PEGs/GO hybrid nanocomposites with increasing the molecular weight of PEG at a frequency of 1000 kHz is illustrated in Fig. 6. It was noted that the A.C. electrical conductivity enhanced with increasing the molecular weight of PEG in the blend polymers, whereas the contribution of GO showed significantly improved A.C. electrical values compared with blend polymer films. This increase is due to the effects of the vacuum charge, where the ions are in the form of groups or clusters separated from each other when the molecular weight was small.

While, the conductivity increases with increasing partial weight (PEG) as a result of increasing the ionic charge carriers and the formation of a connected network of PEG-GO ions within the superposed material, and thus the conductivity increases with increasing the partial weight. Where the key to achieving stretchable high-mobility transistors is the transition molecular weight of a given polymer semiconductor that is practically useful, besides, the conjugation length of the net polymer may also affect the conductivity; the longer conjugation length, the lower energy required to excite charge carriers [18].

5. Conclusions

The important finding that increasing the molecular weight of PEG in nanocomposites presented a significant improvement in the conductivity results, especially the higher molecular weight of 20 k in comparison with other molecular weights in other nanocomposites. Moreover, the contribution of GO shows another important improvement in comparison with other nanocomposites, where increasing the PEGs molecular weight led to an increase in the interfacial interaction with the functional group into GO surface such as the ether group of PEGs with C = O and C–H in the GO surface. The nanosheets of graphene oxide are in the form of clusters or groups separated from each other, and then the dielectric constant is almost constant. Therefore, it is believed that the slight increase in the value of the dielectric constant is due to the increase in charge carriers by increasing the concentration of graphene oxide, which increases the polarized charges. The value of the dielectric constant is increased. And these results are consistent with the researcher's findings. This finding could open promising materials for wide and various applications as several applications as electronic devices, solar cell applications, and energy storage systems.

CRediT authorship contribution statement

The corresponding author '**Ehssan Al-Bermany**' was responsible for supervision and Writing - review & editing, also was responsible for ensuring that the descriptions are accurate and agreed upon by all authors. The first author '**Shurooq S. Al-Abbas**' was responsible for conceptualization, designing and conducting methodology section of the manuscript. The second author '**Rusul A. Ghazi**' was responsible for data curation and Formal analysis. The third author '**Athmar K. Al-shammari**' was responsible for investigation and software. The fourth author '**Nisreen R. Aldulaimi**' was responsible for resources. The fifth author '**Ali R. Abdulridhaa**' was responsible for validation. The sixth author '**Sameer H. Al-Nesrawy**' responsible for writing - original draft.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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