

## The mechanical and thermal properties of PE/CNC nanocomposite

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### Abstract

Polyethylene (PE) nanocomposites containing Cellulose Nano Crystal (CNC) were prepared via melt blending in a twin screw extruder. The evaluation of PE/CNC nanocomposites was studied for food packaging applications. The nanocomposites were investigated by thermal, mechanical analyses. Addition of CNC particle while having no effect on melting and crystallization temperature had intensive effect on the degree of crystallinity of the polyethylene. It is shown that due to the weak interfacial bonding between CNC and PE matrix, a decrease in yield stress and modulus of elasticity was observed for PE/CNC nanocomposites. According to the x-ray diffraction analyses, incorporation of the CNC into PE matrix induced a preferred crystalline orientation for PE crystallites.

**Keywords:** Cellulose Nano Crystal; Food Packaging; Nanocomposite; Permeability; Polyethylene.

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### INTRODUCTION

One of the main applications of nano-composites is active packaging. active packaging has been successful in shelf-life extension of food and subsequently, reduction of food waste for post-harvest interval [1]. Active packaging can interact with packaged food inside packaging and extend the shelf-life. All of these actions are conducted via the active component inside the packaging walls. In this regards, nanotechnology has large contribution in active packaging developments [1-3]. In case of active nano-packaging, the added nano-object/nanostructure normally by interacting with the contained materials leads to improved food preservation and shelf life. Various types of nano-additive have been employed in active packaging such as organo-modified clay, Nano-Zinc oxide, Nano-TiO<sub>2</sub>, crystalline nano cellulose, and nano-porous materials, i. e. Zeolite. In addition to shelf-life extension, barrier properties [4], mechanical properties [5], thermal properties. Among these nano additives, cellulose nano crystal (CNC) has potential application for

development of innovative packaging, as well as the enhancement of conventional packaging properties [6-8]. The CNC can act like physical shield to block the oxygen transmission into the packaging [1, 2] and consequently, increases the shelf-life of fresh food [9]. Obviously, it has direct effect on the shelf life of foods and fresh produces. However, a few works have been conducted on the effect of using CNC on thermal, mechanical of polymeric packaging [2]. Cellulose has high mechanical strength, flexibility, durability, low density and sound damping performance properties [2]. The mechanical properties, reinforcing capabilities, low density and biodegradability of Nano sized cellulose make it an ideal candidate for polymeric packaging [1, 10-13]. Various research studies were conducted on utilizing CNC in polymeric food packaging. Considering that a high number of studies have been carried out on polymer/CNC nano composites, there is still lack of knowledge involving addition of CNC in polymers applied in food packaging. In this study nano composite with different cellulose Nano particles content

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Table 1. The CNC nanoparticles characteristics [InnoTech, Alberta, Canada].

Color	density (g.cm <sup>-3</sup> )	Particle diameter (nm)	Particle length (nm)	Morphology	Specific area (m <sup>2</sup> .g <sup>-1</sup> )	Onset of degradation Temperature (°C)
White	1.6	5-15	100-200	Needle	200-300	250

(0.5, 1, 2 and 5 wt%) in LDPE was used in order to compare the Nano crystal cellulose effect on matrix properties and some particular properties of materials required to be used in food packaging, i. e. mechanical, physical properties, water vapor and oxygen transmission rate and antimicrobial properties will be particularly analyzed.

## EXPERIMENTAL

### Materials

Low density Polyethylene (LDPE) with a melt flow index (MFI) of 1.7 g.(10 min)<sup>-1</sup> (230°C/2.16 kg) and density of 0.96 g.cm<sup>-3</sup> (23°C) was purchased from Maroon Petrochemical Company, Iran. Dried needle like nano-cellulose (CNC) particles were prepared from InnoTech, Alberta, Canada. As shown in Table 1, the CNC particle is two dimensional nano-scale particulate. Maleated polyethylene (MA-PE) was supplied by Karangin Company, Iran. MA-PE was used as compatibilizer and was added to all of PE/CNC nanocomposites in 1wt.%.

### Preparation of the nanocomposites

Cellulose nano crystal (CNC) particles were physically mixed with the polymer granules including pure and maleated polyethylene. Then, the mixture was processed in a twin extruder ( $\Phi=19$  mm, L/D=40) (a co-rotating twin screw extruder ZSK, Germany).

The nanocomposites with different proportions of the PE, CNC, were prepared through a 1-step blending and melt-compounding process in a twin extruder under the following conditions:

- Processing temperatures: 180 to 200°C,
- Screw speeds: 100-160 rpm, and
- The prepared pellets were dried at 80°C for 24 h in a vacuum oven to remove the absorbed water prior to manufacturing the films.

Table 2 summarizes the formulation of nanocomposites prepared in this study. The pure PE was used as a reference material.

The prepared compounds with different amount of CNC contents were converted into polymeric films via a Brabender film blow machine. The melt temperature was 200°C. The prepared film had the width of 20 cm and thickness of 20 nm.

Table 2. Nanocomposites with different CNC content.

Sample designation	CNC (wt%)
N0 <sup>1</sup>	-
NC1	0.5
NC2	1
NC3	2
NC4	5

<sup>1</sup>pure PE

### Characterization

#### Mechanical properties evaluation

The modulus of elasticity and yield stress of pure PE and the nanocomposites were measured according to ASTM D638. The tests were performed on a universal Hounsfield frame (H10KS) at 5 mm/min crosshead speed. A 100SC extensometer was incorporated to determine modulus of elasticity[14].

#### Differential Scanning Calorimetry

Crystallization behavior of pure polyethylene and PE/CNC nanocomposites were studied using a modulated differential scanning calorimeter (MDSC). About 3-10 mg of each sample was heated up from 25 to 200°C at a heating rate of 10°C/min. The sample was then held for 5 min at 200°C to eliminate thermal history of the sample, induced during film blowing process, before being cooled to room temperature [15]. The melting and crystallization behaviors were determined from the first heating scan. The melting temperature was also defined as the maximum of the endothermic melting peak. The degree of crystallinity of the samples was calculated from the ratio of measured enthalpy to the enthalpy value for 100% crystalline PE, taken as 290 J/g. All operations were performed under the nitrogen atmosphere in order to prevent oxidation [16-22].

#### Scanning Electron Microscopy (SEM)

The shape and surface characteristics of the PE/CNC nanocomposites were analyzed by a scanning electron microscope (Mira 3T scan), under vacuum conditions and at a voltage of 30 kV. Samples were mounted on the aluminium stub and photomicrographs of the powders were taken. Before that, a thin layer of gold was applied over the samples to make them electrically conductive,

using a cool sputter coater (BAL-TEC SCD 005, Switzerland) for 100 seconds at a current of 30 mA.

### X-Ray Diffraction

To determine the effect of CNC incorporation on crystalline structure and crystal orientation of polyethylene, Wide-angle X-ray diffraction (XRD) analyses were applied using a Zayes 3003PTS. An acceleration voltage of 30 kV and a current of 30 mA were applied using Cu K $\alpha$  radiation. To distinguish the effect of CNC on crystal orientation of polyethylene in longitudinal and transversal direction of film blow film, the XRD diffraction was conducted in two directions of the films.

## RESULTS AND DISCUSSION

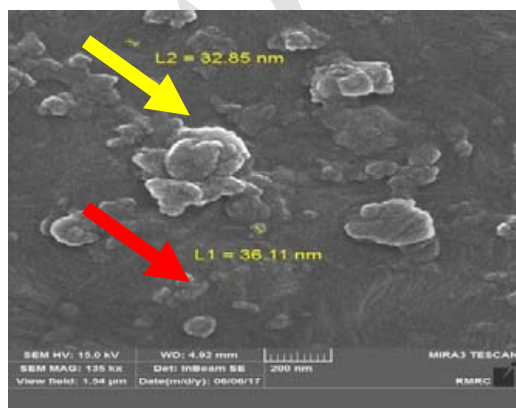
### Morphological study

Fig. 1(a) shows scanning electron microscopy image of polyethylene nanocomposite containing CNC particles. The particle-size is less than 100 nm and as shown the CNC particles have been well-distributed in LDPE matrix (Red arrow). However, there is some agglomerated CNC particles in nanocomposites matrix (Yellow arrow). Fig. 1(b) shows a high magnification SEM image of a CNC agglomerated particulate. As illustrated, the particulate contains large amount of needle like nano cellulose crystals. It was expected that in ideal condition, every needle like crystal be exfoliated in the polymeric matrix. However as shown in Fig. 1(a), a bunch of needle shape crystals make a nano-sized particulate in polymeric matrix.

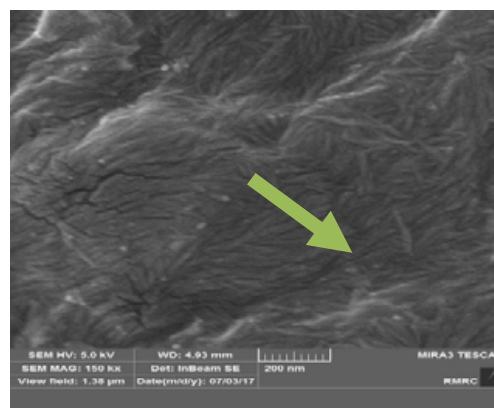
Fig. 2 illustrate XRD graphs of neat polyethylene and nanocomposites containing different loading

of CNC particles for transverse and longitudinal conditions (for the films). In these graphs the effect of incorporation of CNC into polymeric matrix on crystallite orientation can be studied. The low density polyethylene has diffraction peaks for (110) and (200) planes in  $2\theta=22.5^\circ$  and  $37.5^\circ$ , respectively. The pure CNC has diffraction peaks for (110), (200) and (004) planes in  $2\theta = 17^\circ, 22.5^\circ$  and  $35^\circ$ , respectively. As shown in Fig. 2(a) by incorporation of 0.5wt.% CNC into neat polyethylene the (200) diffraction peak of LDPE has been diminished in transverse diffraction. In this condition, according to the Fig. 2(c), an increase for (110) and (200) intensity peaks should be observed in longitudinal direction[16]. However, a slight increment can be realized for the intensities (Fig. 2(b)). It means that the CNC particulates induces a preferred crystallite orientation in LDPE matrix. In other words, the (200) plans of LDPE crystallites are aligned in transverse direction. This preferred orientation might be responsible for change of mechanical properties as well as permeability of LDPE/CNC nanocomposites in different direction. For the case of LDPE/CNC nanocomposites diffraction peak of (004) for CNC is not present in the graphs. The absence of the (004) peaks for PE/CNC nanocomposites is a proof for this preferred orientation for CNC particles as well as polyethylene crystallites [4, 6].

Thus, as shown, there is an overlap between diffraction peaks of (110) plans for LDPE and (200) plans for CNC particles ( $2\theta = 22.5^\circ$ ). Therefore, it makes it difficult to interpretation of the LDPE crystallites behavior after incorporation of CNC particles into the matrix.



(a)



(b)

Fig. 1. (a)The nanocomposites containing CNC nanoparticles (b) high magnification image of a CNC agglomerated particle.

*Effect of CNC particles on crystal orientation of PE*

Crystal orientation of polyethylene nano-composite specimens evaluated quantitatively using ratio of I(110) to I(200) obtained from X-ray diffraction peaks. where I(hkl) indicates the intensity of corresponding diffracted plane. As clearly seen in Table 3, the crystal orientation was affected by the CNC incorporation in longitudinal direction significantly and slightly in transverse direction. It implies that, generally, CNC induces preferred orientation of crystallites in film blown

machine. This might be due to needle like shape of particulates.

*Thermal properties*

The result of Differential scanning calorimetry (DSC neat polyethylene and LDPE/CNC nano-composites is summarized in Table 4. As shown, the degree of crystallinity of LDPE increases for incorporation of 0.5 wt.% CNC into LDPE. However, for the case of nanocomposites containing 1 and 2 wt.% CNC the degree of crystallinity has been

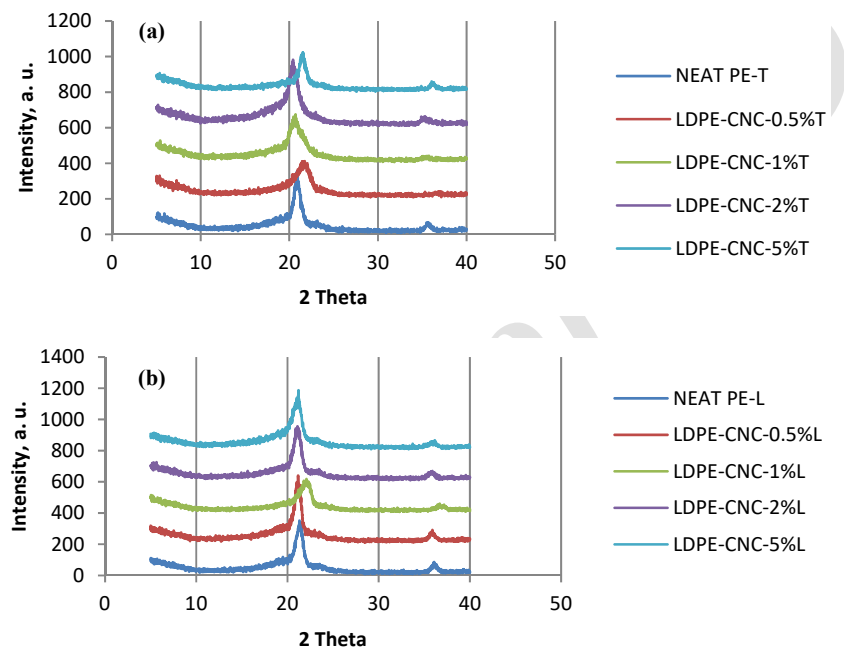


Fig. 2. XRD pattern of neat polyethylene and nanocomposites with different CNC loadings (a) Transverse (b) Longitudinal.

Table 3. Crystallite, Orientation, I (110)/I (200).

Specimen	CNC (wt%)	Crystallite Orientation	
		I <sup>t</sup> (110)/I <sup>t</sup> (200)	
		Longitudinal	Transverse
N0	0	4.23	2.07
NC1	0.5	4.83	1.72
NC2	1	3.96	2.02
NC3	2	5.48	2.16
NC4	5	5.77	1.59

Table 4. Differential scanning calorimetry results of polyethylene and nanocomposites samples.

Sample	T <sub>m</sub> (°C)	T <sub>c</sub> (°C)	X <sub>c</sub> (%)	Tan α (rate of nucleation)
Neat PE	111.43	96.24	12	38.4
NC1	111.45	96.3	13.4	41.4
NC2	112.25	96.86	7.7	21.3
NC3	111.95	97.55	6.5	17.2
NC4	110.66	97.57	15.7	56.6

decrease about 40% in comparison to neat LDPE. Reduction of the degree of crystallinity can be due to the lack of interface capability between the polymer matrix and the CNC particulate [6]. In other words, the CNC particles behave like impurity inside the polyethylene matrix. Interestingly, for the case of LDPE containing 5wt.% CNC particles the degree of crystallinity increases dramatically and even more than neat polyethylene [3, 6]. This may contribute to the preferred orientation of the CNC in the film blowing machine. In fact, the high loading of CNC into the matrix and making constrain during the melt process in preferred orientation and cause an increment in degree of crystallinity [6, 17].

The degree of crystallinity is consistent with the data of rate of nucleation for the neat LDPE and LDPE/CNC nanocomposites. The lower rate of nucleation ( $\tan \alpha$ ), the lower degree of crystallinity.  $\tan \alpha$ , where  $\alpha$  is the slope of the crystallization peak of DSC curve, has been introduced as a characteristic of spherulite nucleating rate [23]. The effect of incorporating different amount of nanometric calcium carbonate on spherulite nucleating rate is summarized in Table 4. From data shown in Table 4, it can be concluded that incorporation of CNC into PE matrix led to an initial increase, decrease and again increase in nucleating rate of that spherulites. Reduction of nucleating effect occurred in the case of PE containing 1 and 2 wt.% that may be due to particles aggregation. It is noteworthy that aggregation of particles, i. e. being more prone at higher concentrations, reduces the surface of CNC particles in contact with polyethylene and thus, reduces the appropriate sites for nucleation of spherulites [2].

It is also concluded that addition of CNC into

polyethylene does not have any significant effect on crystallization and melting temperature of PE/CNC nanocomposites.

According to Gibbs-Thompson concept which is used to calculate lamellae thickness using Eq[1], the higher the melting point, the higher the lamellae thickness of the polyethylene spherulites.

$$L = \frac{2\sigma_e}{\Delta H_f \rho_c} \frac{T_m^0}{T_m^0 - T_m} \quad (1)$$

where  $\sigma_e$  is the fold surface energy,  $\Delta H_f$  is the enthalpy of fusion for infinity large crystal and  $\rho_c$  is the density of fully crystalline polyethylene.  $T_m$  is the melting temperature of sample and  $T_m^0$  is the equilibrium melting temperature of polyethylene. Therefore, the lamellae thickness was not affected by incorporation of CNC into polyethylene matrix [16]. It means that there is no compatibility between polyethylene crystals and Cellulose crystals.

#### Mechanical properties

Tensile properties of pure PE and PE/CNC nanocomposites is illustrated in Fig. 3(a) and (b). The calculation is according to the following equation:

$$E_c = \frac{E_p E_f}{E_p \Phi_f + E_f \Phi_p} \quad (2)$$

where  $E_c$ ,  $E_p$  and  $E_f$  are modulus of nanocomposite, matrix and filler, respectively,  $\Phi_p$  and  $\Phi_f$  are the volume fraction of matrix and nanoparticles, respectively. As expected from Eq. 2, addition of CNC will increase modulus of elasticity of PE/CNC nanocomposites. However, the

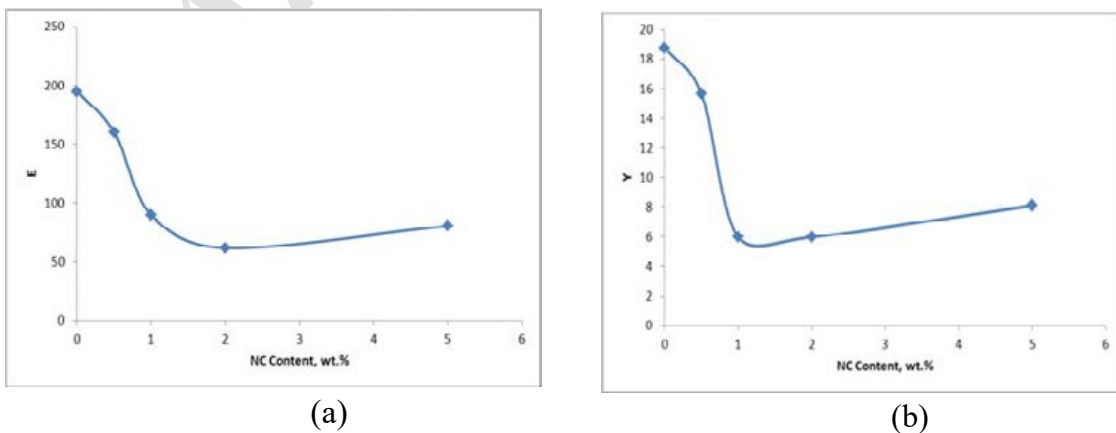


Fig. 3. (a) Modulus of elasticity, (b) Yield stress of pure PE and PE/CNC nanocomposites.



measured values shows decrease of modulus of elasticity of PE/CNC nanocomposites, as well as the yield stress[10]. For interpretation of this behavior two counter balance effects should be considered, the nucleating and the reinforcing effects of CNC. It is shown, in Table 4, that the degree of crystallinity of nanocomposites decreased, because of existence of CNC[6, 9,18]. Obviously, a decrease in degree of crystallinity decreases the modulus and yield stress of the polymer. However, reinforcing effect of the nano-filler will introduce an increment in the modulus of nanocomposite, Therefore, both factors, i. e. decrease of degree of crystallinity of the polymeric matrix and the effect of interfacial adhesion of reinforcement particle, induces decrease of modulus of elasticity of the nanocomposites. However, an increment of the modulus of elasticity for the case of PE containing 5 wt.% is contributed to the increase of degree of crystallinity of the polyethylene. This behavior emphasizes that the contribution of the degree of crystallinity on the modulus of elasticity of the PE/CNC is much more than that of the effect of the non-compatible reinforcement. Both the debonded CNC particles as well as low degree of crystallinity are responsible for yield stress drop for PE/CNC nanocomposites. The fine spherulitic structure also intensifies this yield stress reduction. . In regards to debonding of the CNC nanoparticles, the yield stress reduction can be contributed to two mechanisms: (i) the cavity effect in the polymeric matrix, and (ii) the crazing of the polymeric matrix. The former is the cavity made by the CNC particle induced by existence of the CNC particles and also weak interface between the matrix and the CNC particles. The latter is due to the shape of the CNC particles, i. e. needle-like, which can trigger the crazing phenomenon at the yielding point.

Again, the yield stress increment for the case of PE containing 5wt.% CNC can be contributed to the increase of degree of crystallinity and/or crystalline orientation in nanocomposite samples

[5, 12].

The results of MFI analyses of neat PE and nanocomposite CNC are summarized in Table 5. The MFI of polyethylene increases via incorporation of CNC into the matrix. It means that the CNC particles can facilitate melt flow of the LDPE. In fact, the needle like particles slips on each other and cause an ease of flow (Fig. 4).

*Shelf life extension of fresh produces via utilizing LDPE/CNC nanocomposites*

Contrary to food products, agricultural products enter new lifecycle after being harvested and separated from the tree branch. In the new cycle, agricultural products (fruits and vegetables) breathe so that oxygen consumed, dioxide-carbon, ethylene and moisture arise from itself. In this condition, each food product should consume all oxygen around itself and change it to dioxide-carbon, ethylene and moisture (hypothesis: oxygen gas outside of the package cannot penetrate into the package space) [3]. But, in real condition, oxygen gas passes through polymeric wall and penetrates into the package. Also, dioxide-carbon gases, ethylene and moisture can pass through polymeric wall and get released. It should be mentioned that penetrability rate of each gas is different on the basis the type of polymer. It means that once the same amount of oxygen enters the package, the same amount of dioxide-carbon or ethylene is not release from the package [4].

Due to these conditions, it is necessary to precisely control the penetrability of these gases.

Table 5. MFI results of neat PE, and nanocomposites CNC samples.

Sample	MFI (gr.10 min <sup>-1</sup> ) (190 °C, 2.16 kg)
Pure PE	1.69
NC1	1.72
NC2	1.75
NC3	1.78
NC4	1.79



Fig. 4. Slippage of needle like CNC on each other and facilitation of melt flow of LDPE.

When the space inside a package is devoid of oxygen and dioxide-carbon only remains in this space, anaerobe bacteria grows in such environment and agricultural product will quickly spoil. If penetrability of oxygen to inner space of the package is not limited, agricultural product will firstly absorb it and will quickly spoil [4]. Secondly, the presence of excessive oxygen removes nutrition like vitamins, fats, ... and other substances. On the other hand, in the condition of free absorption, inside the package will be full of ethylene gas. For plants and agricultural products, ethylene gas acts as growth hormone in human and causes the food product to be ripe. However, the excessive presence of the gas causes the agricultural product to ripe early. In packaging the agricultural products, is important to use a special combination of gases to add to the durability time of any food and agricultural products [1].

In food packaging, to have a maximum shelf-life a certain gas mixture inside the packaging is required. Therefore, active packaging and concept of "selective gas permeability" can assist to prepare the certain gas mixture inside the packaging. The existence of CNC nanoparticle as well as certain degree of permeability of different polymers for different gas molecules (for instance the permeability of CO<sub>2</sub> is five times greater than O<sub>2</sub> molecules) may induce a selective permeability. In this sense, the resulting increase in the speed of diffusion allows the maintenance of particular food in better conditions and consequently improves of its shelf life. The approach can be done for agricultural products by Nano-cellulose

To increase the durability of food and agricultural products, appropriate conditions of gas combination of atmosphere inside the packages can be made for desired product. To do so, it is necessary to control the passing of desired gases from package wall [3].

## CONCLUSIONS

Based on the results obtained, the following conclusions can be drawn:

- Although addition of CNC does not affect the melting and crystallization temperatures, the degree of crystallinity decreases dramatically for the nanocomposites containing less than 5wt.%. Interestingly, the degree of the crystallinity of PE increase for the case of nanocomposites containing 5 wt.% which may be contributed to the constraint induced by

the CNC particles. It's worth to state that trend of the nucleation rate of the polyethylene spherulite is in good agreement with degree of crystallinity trend.

- It was shown that incorporation of CNC into polyethylene matrix induces a preferred crystalline orientation which may influence the mechanical properties and gas permeability.
- Due to incompatible interface between CNC particles and polyethylene matrix, mechanical properties decline has been observed. In this regards, the change in degree of crystallinity has more impact on the mechanical properties in comparison to reinforcement particles.

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## CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this manuscript.

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