

**STRUCTURES AND MECHANICAL PROPERTIES OF TALC AND CARBON BLACK REINFORCED HIGH DENSITY POLYETHYLENE COMPOSITES : EFFECTS OF ORGANIC AND INORGANIC FILLERS**

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**ABSTRACT**

Organic filler like carbon black (CB) and inorganic filler like talc (T) with 0, 0.5, 1.0, 10, 20 and 40 wt% were separately loaded in high density polyethylene (HDPE) by the extrusion moulding method at 160°C. Then, different sets of filler loaded HDPE composites were prepared using the compression moulding technique, and their structures and mechanical properties were characterized. The pure HDPE sample, as examined by the X-ray diffraction (XRD) technique, showed orthorhombic structure, which did not change either with filler types or with their concentration. The only variations found in the structure are the changes of crystallinity and crystallized size that depend on both types of fillers and their concentrations. Incorporation of CB in HDPE emphasizes the crystallinity and crystallized size more than that of T. The tensile strength of the composite decreases with the increase of both types of fillers, and this decrease is explained on the basis of Nielson model, which basically describes a poor interaction between filler and HDPE matrix. An increase of Young's modulus of 350% is observed with the increasing CB and T contents, representing an increase of the stiffness in the materials. Flexural strength increased with the increase of CB content but decreased with the increase of talc content. Although the microhardness was observed to increase with both types of fillers, the hardness value was 80% higher for CB loaded-composites than that of T at 40 wt% filler content. These findings strongly indicate that the compatibility of HDPE is better with organic filler than with inorganic one.

Key words: X-ray diffraction, Tensile strength, Young's modulus, Flexural strength

**INTRODUCTION**

Polyethylene has been one of the most extensively studied synthetic plastics in polymer science and engineering, principally because it is chemically simple and it has an increasing number of applications (Lin *et al.* 2005). Polyethylene (PE) is a thermoplastic that finds extensive application as films, sheets, bottles and containers, pipes and tubes,

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and in wire insulation and cables (Yasmin *et al.* 2004). Although, two types of PE *viz.* high density polyethylene (HDPE) and low density polyethylene (LDPE) with corresponding densities in the range 0.94 - 0.96 and 0.91 - 0.94 g/cm<sup>3</sup> as synthesized by conventional polymerization techniques are available, scopes of its application in wide spectrum of engineering requirements are still underway (Zhang *et al.* 2004), demanding further research on this material.

Various types of additives such as antioxidants, fillers, UV-absorbers and flame retardants are mixed with PE to meet desired requirements. Among these additives, filler reinforced polymer composites find potentials over traditional engineering materials (Jancar *et al.* 1999). Fillers have an important role in modifying various properties of polymers. Inorganic and organic fillers are reported to improve mechanical, thermal and electrical properties of some crystalline polymers, when they are used as reinforcing agents (Psarras *et al.* 2003). Of inorganic fillers, talc is having a huge demand for synthesis of polymeric composites due to its excellent blending nature, thermal resistance, superior electrical resistance, chemical inertness and smooth greasy feel. Of organic fillers, carbon black (CB) have gained much attention in fabrication of polymeric composites, because CB reinforced composites are being exploited in many industrial and domestic appliances including self-regulating heating elements, current switching, fluid sensor, thermal controller, capacitors and electromagnetic interference shielding (Zhao *et al.* 2003). However, research works found on filler reinforced polymer composites are mostly based on isotactic polypropylene (Akinci 2009, Mina *et al.* 2010), though a few research works on filler loaded PE composites have been published (Ulutan *et al.* 2000, Yuan *et al.* 2003, Dang *et al.* 2003, Luyt *et al.* 2006). In addition, homogeneous dispersion of fillers in polymer matrix by melt blending has also been a challenging task. To overcome this problem, multiple extrusion method has been followed in the present study to improve blending between HDPE and filler.

Moreover, organic and inorganic fillers loaded HDPE composites prepared at the same processing methods can reveal useful scientific information about the influence of these two different fillers on the performance of the resulting composites. Therefore, inclusion of talc as an inorganic filler and CB as organic filler in HDPE with low and high contents of filler has been performed at the same processing methods in order to observe their effects of structures and mechanical properties.

## MATERIALS AND METHODS

Granular-shaped high density polyethylene (HDPE) was purchased from BASF, Germany. The fillers, such as carbon black (CB) and talc (T) were purchased from the local market. Different sets of composites were prepared by mixing a low (0.5, 1.0 wt%) and a high (10, 20 and 40 wt%) content of CB and T with HDPE separately. The

constituents of each set of composites were consecutively triple extruded at 160°C to improve the dispersion of the fillers in polymer by an extruder (Model No. LME-230). Then, desired samples were produced at 160°C and a pressure of 60 kN using a compression moulding machine. CB and T loaded HDPE composites are here-in-after referred to as HDPECB and HDPET, respectively. A pure HDPE, a series of HDPE composites prepared with 0.5, 1.0, 10, 20 and 40 wt% CB contents are abbreviated as HDPECB0.5, HDPECB1, HDPECB10, HDPECB20, HDPECB40 and a series of HDPE composites with 0.5, 1.0, 10, 20 and 40 wt% T contents are referred to HDPET0.5, HDPET1, HDPET10, HDPE20 and HDPET40. X-ray diffraction studies of the HDPECB and HDPET composites were performed by an X-ray diffractometer (Model JDX-8P, JEOL Ltd., Tokyo, Japan) using  $\text{CuK}_\alpha$  radiation of wavelength,  $\lambda = 1.5418 \text{ \AA}$ . These samples were then subjected to mechanical measurements such as tensile and flexural testing using a universal testing machine (Model No. H10KS), as according to the ISO standard (ASTM Designation 1991, Sjorstrom 1981). Microindentation test using a square-based pyramid indenter of diamond was also performed upon the application of a given load at a constant loading-time of 6 sec (Calleja *et al.* 2000). Microhardness ( $H$ ) is conveniently measured from the following mathematical relation (Mina *et al.* 2004):

$$H = K \frac{P}{d^2} \quad (1)$$

where,  $K$  is called a geometrical factor equal to 1.854. The unit of  $H$  is measured in megapascals, if  $P$  is in Newtons and  $d$  is measured in meters.

## RESULTS AND DISCUSSION

*Structural analyses:* The XRD patterns, having  $2\theta$  range of 5 - 55°, for pure HDPE and CB reinforced HDPE composites with 0.5, 10 and 40 wt% CB content are shown in Fig. 1. The crystal structures of polyethylene were found to be orthorhombic, monoclinic and hexagonal, depending on the processing conditions as reported (Bunn 1953, Bevis *et al.* 1971, Basset 1976). However, in the observed profiles one strong peak appearing at 21.5 and two moderately strong peaks at 23.9 and 36.2° show inter planner spacing of 4.132, 3.707 and 2.481 Å, respectively. Analyses of these peaks correspond to the (110), (200) and (020) lattice planes and indicate that HDPE used in this investigation shows orthorhombic structure (Yijian *et al.* 2005). Some small peaks are also apparent at angles larger than 40°. On the other hand, CB shows a strong peak at 26.4°, representing the planes (100) of distance 3.375 Å (Sang *et al.* 2006). On the other hand, the XRD profile of HDPECB0.5 is almost similar to that of HDPE, and those of HDPECB10 and HDPECB40 are the combinations of the peaks of HDPE and CB. The peak position does not change at all, while the intensity and width of each peak is different, depending on the

samples. These suggest that there is a change of crystallinity and crystallite size in the samples. The XRD patterns for pure HDPE and HDPE-talc composites with 0.5, 10 and 40 wt% talc content are shown in Fig. 2. Several peaks from talc are clearly apparent. Of these, important ones appearing at 9.2, 19 and 28.8° correspond to the lattice plane distances of 9.609, 4.669 and 3.099 Å, which have been indexed as (002), (020) and (006) planes, respectively (George *et al.* 1940, Gang *et al.* 2004). On the other hand, the XRD profile of HDPET0.5 is almost similar to that of HDPE, and those of HDPET10 and HDPET40 are actually the blends of the peaks of HDPE and talc. The peak position remains unchanged, although depending on the samples the intensity of each peak is different with slight modification of peak width. These suggest that there may be a change of crystallinity and crystallite size in the samples. The XRD patterns for pure HDPE and HDPE-talc composites with 0.5, 10 and 40 wt% talc content are shown in Fig. 4. The intensity of the peaks observed corresponding to (110) and (200) planes for each sample are analyzed, and the data of crystallinity and crystalline thickness are respectively evaluated using the following formulae:

$$f_c = \frac{I_c}{I_c + \eta I_a} \quad (2)$$

and

$$t = \frac{k\lambda}{B \cos \theta} \quad (3)$$

where,  $f_c$  is the crystallinity,  $I_c$  is the crystalline intensity,  $\eta$  is a constant equal to 1.235 for polyethylene (Wunderlich *et al.* 1973),  $I_a$  is the amorphous intensity,  $t$  is the crystallite size,  $k$  equals 0.9, (Han *et al.* 2008),  $\lambda$  is the wavelength of X-ray,  $B$  is the breadth of the full-width at half maximum of the diffraction peak measured in radians and  $\theta$  is the Bragg angle. The value of  $f_c$  and  $t$  are summarized in Table 1. According to the XRD results, the pure HDPE indicates the orthorhombic crystalline structure that remains unaltered by the existing processing conditions and the addition of fillers of whatever their amounts and origins. Although the crystalline structure of HDPE is unchanged by the inclusion of fillers, the crystallinity and crystal size increase with the increase of reinforcement, and this increase is larger for CB than for talc. Similar phenomenon was noticed in recycled HDPE/clay hybrid and HDPE/wood composites (Lei *et al.* 2007, Yong *et al.* 2007). It can be noted that the peak position of two HDPE peaks in this study are slightly shifted to higher angle, which can be assigned to the unit cell distortion of HDPE by filler inclusions.

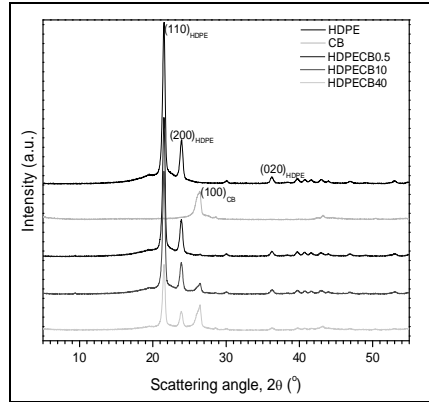


Fig. 1. XRD profiles of pure HDPE, CB and HDPE/CB composites.

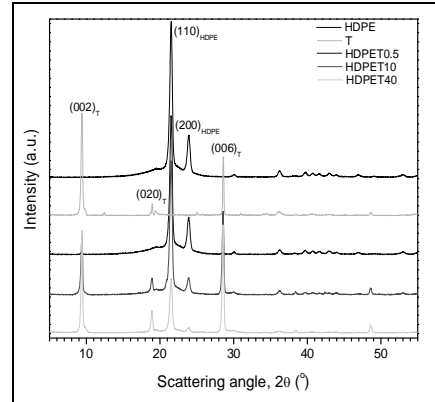


Fig. 2. XRD profiles of pure HDPE, T and HDPE/T composites.

## MECHANICAL PROPERTIES

*Tensile strength:* The tensile strength (TS) as a function of filler content for the pure HDPE and HDPE/CB composites are shown in Fig. 3. The TS values for pure HDPE, HDPE/CB0.5, HDPE/CB10, HDPE/CB20, HDPE/CB40 are, respectively found to be 23, 23.5, 20.5, 18.5 and 17 MPa, and those for HDPE/T0.5, HDPE/T10, HDPE/T20, HDPE/T40 are respectively found to be 19.7, 16, 15.8 and 11.6 MPa.

**Table 1. Crystallinity and crystallite size of pure HDPE as well as HDPE/CB and HDPE/T composites with 0.5, 10 and 40 wt% filler contents.**

Samples	Composites formulation	Crystallinity	Crystallite size (nm)
HDPE	HDPE	0.65	17.6
HDPE/CB	HDPE/CB0.5	0.66	20.6
	HDPE/CB10	0.67	28.2
	HDPE/CB40	0.71	33.0
HDPE/T	HDPE/T0.5	0.66	18.2
	HDPE/T10	0.68	19.2
	HDPE/T40	0.69	20.0

The TS value decreases with the increase of filler content in the composite. However, this decrease, when compared between HDPE and HDPE/CB40, is 5% and between HDPE and HDPE/T40 is 11%. This suggests that the compatibility HDPE is better with organic filler than with inorganic one. A number of researchers have documented TS variations with filler content in various polymers, where they reported

that TS decreases with filler content. Some of them explained this fact using the following Nielson model (Nielson 1974):

$$\frac{\sigma_C}{\sigma_P} = (1 - \phi^{\frac{2}{3}})A_Q \quad (4)$$

where  $\sigma_C$  and  $\sigma_P$  denote the tensile stress of the composite and the polymer matrix, respectively;  $\phi$  represents the weight fraction of the filler;  $A_Q$  accounts for the adhesion quality between polymer and filler interface. According to the above formula if a discontinuity in stress transfer to polymer-filler interface occurs, then  $A_Q$  increases with the addition of filler, resulting in the decrease of TS. Therefore, the Nielson model is better suitable to explain the observed TS decrease with the increase of filler content.

*Young's modulus:* Young's modulus ( $Y$ ) indicates the stiffness of a material and its variation with filler contents is shown in Fig. 4. It is clear from the results that the  $Y$  value rapidly increases from 0 to 0.5 wt% CB content after which it gradually increases. While the neat HDPE shows the  $Y$  value of 320 MPa, the HDPECB40 and HDPET40, respectively shows these values of about 1440 and 1600 MPa, indicating a  $Y$ -increase for the composites of 350 and 400% from the pure HDPE. Thus, the addition of fillers increases, which may be attributed to the increasing crystallinity of the samples, suggesting that the composites show better performance than the pure HDPE.

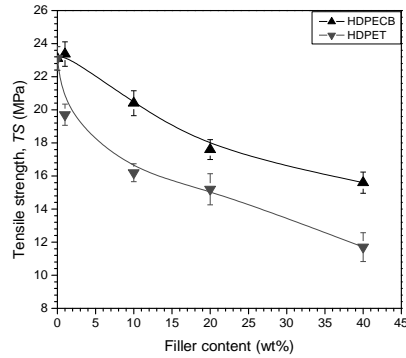


Fig. 3. The Change of TS values for HDPECB and HDPET composites.

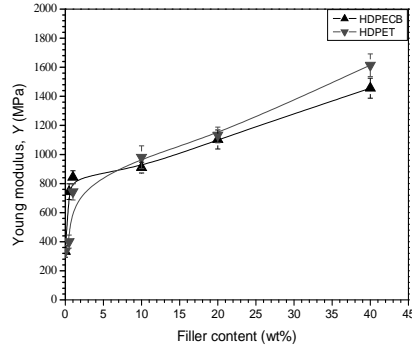


Fig. 4. A comparison of Young's modulus for HDPECB and HDPET composites.

*Flexural strength:* Flexural strength (FS) for the pure HDPE and HDPECB composites are shown in Fig. 5. The FS increases with the increase of CB content in the composite. Contrary to this, FS decreases with the increase of talc content in the composite. However, the maximum increase of FS occurring at 40 wt% CB content is 78%, and the maximum decrease of FS occurring at 10 talc content is 18%. These findings strongly designate that the CB reinforced HDPE shows superior mechanical performance to talc loaded HDPE.

*Tangent modulus:* Tangent modulus ( $G$ ) demonstrates the stiffness of a material and its variation with filler content is shown in Fig. 6. It is clear from the results that the  $G$  value gradually increases from 0 to 40 wt% CB content. Thus, the addition of fillers increases the stiffness of the resulting composites. This can be inferred by the increase of stiffness, which may be attributed to the increasing crystallinity of the samples, thereby suggesting that the CB reinforced composites show better performance than the talc loaded composites.

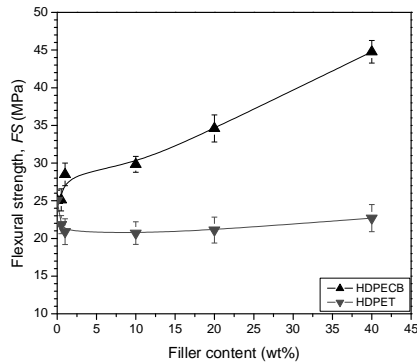


Fig. 5. A comparison of FS values of pure HDPE, HDPECB and HDPET composites.

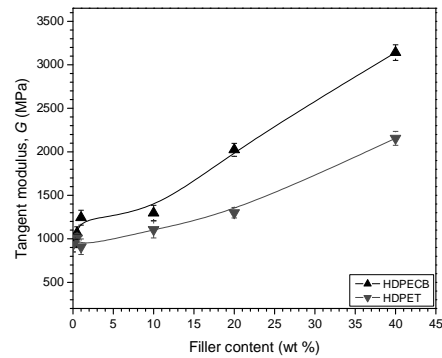


Fig. 6. The change of tangent modulus,  $G$ , with respect to CB and talc contents.

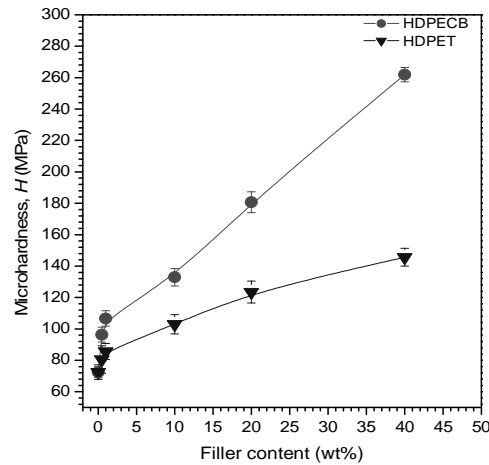


Fig. 7. The dependence of microhardness ( $H$ ) on CB and talc contents.

*Microhardness:* In microhardness measurements, both elastic and plastic deformations occur in the sample after the application of a load. The  $H$  values for neat HDPE, HDPECB40 and HDPET40 are measured to be 72, 144 and 260 MPa, respectively. Thus, the maximum hardness increase for HDPECB40 and HDPET40 from HDPE is about 262

and 101%, respectively. The increase of H complies with the similar increase of Y for increasing filler content, clarifying a good interfacial adhesion between filler and polymer in the microscopic scale. It is noteworthy that both H and Y values provide mechanics at a very low strain, whereas tensile strength reflects information rather in a macroscopic scale, because the latter is characterized by plastic deformation occurring in the material. Since CB loaded composite exhibits 80% higher hardness value than talc loaded composite at 40 wt% filler content, the compatibility of the organic filler with polymer matrix seems to be good enough in comparison to the inorganic filler. The observed results show a better micromechanical performance of the composites, where the influence of CB on the compatibility with polymer matrix is superior to that of talc (Fig. 7).

## CONCLUSION

The structure of prepared samples as examined by the XRD technique reveals the orthorhombic structure. The TS decreases with the increase of filler content in the composite. The maximum increase of Young's modulus is observed for the composites of the highest content of CB and T by about 400 and 360%, representing the increase of stiffness in the fabricated material. The maximum increase of G for HDPECB and HDPET from pure HDPE is about 224 and 122%. A significant increase of microhardness is observed for CB loaded composites as compared to the talc loaded composite. These findings strongly indicate that the compatibility of HDPE is better with organic filler CB than with inorganic talc, and the CB reinforced HDPE composites show better mechanical performances to talc loaded ones.

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## REFERENCES

- Akinci, A. 2009. Mechanical and structural properties of polypropylene composites filled with graphite flakes. *Arch. Mat. Sci. Engg.* **35**(2), 91-94.
- ASTM Designation. 1991. F 417-78, Standard Test Method for Tensile and Flexural Strength (Modulus of Rupture) of Electronic-Grade Ceramics. *Annual Book of ASTM Standards*.
- Basset, D.C. 1976. Chain-extended polyethylene in context : A review. *Polymer* **17**: 460- 470.
- Bevis, M. and E. B. Crellin. 1971. The geometry of twinning and phase transformations in crystalline polyethylene. *Polymer* **12**: 666-684.



- Bunn, C. W. 1953. *Fires from Synthetic Polymers*. Hill, R. (ed.) Amsterdam, Elsevier, pp. 240-300.
- Calleja, F.J. and S. Fakirov. 2000. *Microhardness of Polymers*. Cambridge University Press.
- Dang, Z., L. Fan, Y. Shen, C. Nan and S. Zhao. 2003. Study of thermal and dielectric behaviour of low-density polyethylene composites reinforced with zinc oxide whisker. *J. Therm. Anal. Calo.* **71**: 635-641.
- Gang, Wu, Bianying, Wen and Shaohua, Hou. 2004. Preparation and structural study of polypropylene-talc gradient materials. *Polym. Int.* **53**: 749-755.
- George, L., Clark, Herbert and D. Rhodes, 1940. Practical Evaluation of Commercial Rubber Carbon Blacks by X-ray diffraction. *Ind. Eng. Chem. Anal. Ed.* **12**(2): 66-71.
- Han, G., Y. Lei, Q. Wu, Y. Kojima and S. Suzuki. 2008. Bamboo-Fiber Filled High Density Polyethylene Composites: Effect of Coupling Treatment and Nanoclay. *Polym. Environ.* **16**: 123-130.
- Jancar, J. 1999. *Advances in polymer sciences : Mineral fillers in thermoplastics.* **139**, Springer, Berlin/Heidelberg.
- Lei, Y. Wu, Q. Clemons, C. Yao, F. and Xu Y. J. 2007. Preparation and properties of recycled HDPE/clay hybrids". *Appl. Polym. Sci.* **103**: pp. 3056-3063.
- Lin, Y., W. Du, D. Tu, W. Zhong and Q. Du. 2005. Space charge distribution and crystalline structure in low density polyethylene (LDPE) blended with high density polyethylene (HDPE). *Polym. Int.* **54**: 465-470.
- Luyt, A. S., J. A. Molefi and H. Krump. 2006. Thermal, mechanical and electrical properties of cooper powder filled low density and linear low density polyethylene composites. *Poly. Degrad. Stab.* **91**:1629-1636.
- Mina, M. F. and S. Akbar. 2004. Versatile applications of microindentation test in the characterization of polymers and their blends. *Bangladesh J. Phys.* **1**(1): 84-94.
- Mina, F. M., M. A. Haque, M. K. H. Bhuiyan, M. A. Gafur, Y. Tamba and T. Asano. 2010. Structural, Mechanical and Thermal Studies of Double-Molded Isotactic Polypropylene Nanocomposites with Multiwalled Carbon Nanotubes. *J. App. Poly. Sci.* **118**: 312-319.
- Nielson, R. 1974. *Mechanical Properties of Polymers and Composites*. Marcel Dekker: NewYork.
- Psarras, G. C., E. Manolakaki and G.M. Tsangaris. 2003. Electrical relaxations in polymeric particulate composites of epoxy resin and metal particles. *Compos. A.* **34**: 1187-1198.
- Sang-Young Lee, Soon-Yong Park and Heon-Sik Song. 2006. Lamellar crystalline structure of hard elastic HDPE films and its influence on microporous membrane formation. *Polymer* **47**: 3540-3547.
- Sjorstrom, E. 1981. *In : Wood Chemistry: Fundamentals and Applications*. Academic Press, London. pp. 169.
- Ulutun, S. and M. Gilbert. 2000. Mechanical properties of HDPE/magnesium hydroxide composites. *J. Mater. Sci.* **35**: 2115-2120.
- Wunderlich, B. 1973. *Macromolecular Physics*: Vol. 1, Crystal structure, Morphology and Defects. Academic Press. New York and London.
- Yasmin, A. and I. M. Daniel, 2004. Mechanical and thermal properties of graphite platelet/epoxycomposites. *Polymer* **45**: 8221-8229.
- Yijian, Lin, Weichuan Du, Demin Tu, Wei Zhong and Qianguo Du. 2005. Space charge distribution and crystalline structure in low density polyethylene (LDPE) blended with high density polyethylene (HDPE). *Polym. Int.* **54**: 465-470.
- Yong Lei, Qinglin Wu, M. Craig, Clemons Fei Yao and Yanjun Xu. 2007. Influence of nanoclay on properties of HDPE/wood composites. *J. Polym. Sci.* 3958-3966.

- Yuan, Q., W. Jiang, L. An, R. K. Y. Li. and Z. Jiang. 2003. Mechanical and thermal properties of High-Density Polyethylene (HDPE) Toughened with Glass Beads. *J. Appl. Poly. Sci.* **89**: 2102-2107.
- Zhang, X. M. and S. Elkoun. 2004. Oriented structure and anisotropy properties of polymer blown films: HDPE, LDPE and LLDPE. *Polymer* **45**: 217-229.
- Zhao, Z., W. Wu, He E. and Z. Chen. 2003. The conduction mechanism of carbon black - filled poly(vinylidene fluoride) composite. *Mater. Lett.* **57**: 3082-3088.

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