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Morphological and mechanical studies of surface treated mica reinforced high density polyethylene

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ABSTRACT

Morphological studies of high density polyethylene (HDPE) reinforced by mica flakes has been investigated. The mica flakes were surface treated by silane and titanate coupling agents. Effects of these treatments on the polymer-mica interface is discussed. It is found that surface treatment, particularly silane treated composites, have better adhesion between the reinforcing agent and the polymer matrix. This effect can only be detected when the mica concentration is higher than 20 weight percent. This interfacial adhesion could not be seen across the surface of the sample but only when the measurements were made parallel to the surface.

INTRODUCTION

Inorganic fibres and flakes effectively reinforce thermoplastics, enhancing their performance charateristics and improving their cost/performance ratio. Such reinforcement often gives lower cost thermoplastic resins, which have better performance than even the more expensive resins (1-7). The morphology of polymer composites play very crucial role in determining their end use properties. The micro-structure of reinforced thermoplastics are bit more complex than those of non-reinforced materials. The diversity of the composites are due to the complex structure of these micro-structures. The morphology of these materials could be manipulated to optimize their properties. The processing parameter could also have very profound effect over the morphology. The chain orientation is varied by addition of flakes. The degree of crystallization is changed due to nucleation effects of flakes.

Internal stresses are also function of various processing factors (8,9). The purpose of the present studies is to present the morphological and mechanical results of high density polyethylene (HDPE) reinforced with mica flakes.

EXPERIMENTAL

Materials: The polymer used was a high density polyethylene (HDPE) supplied by Union Carbide (DGDY6089), having an weight average molecular weight of 680,200 and Z molecular weight of 812,000. Its melt index was 0.8g/10 min. and density of 0.955 g/cm^3.

Mica flakes used had an average particle size of 200 mesh. Coupling agents used in surface treatment were α-amino propyl-triethoxysilane (A-1100 of Union Carbide) and alkoxy trimethacryl titanate (Kenrich Petrochemicals, KR-33DS).

Surface Treatment of Mica-Flakes: Mica flakes were silane treated by using dilute solution method (1). Solution was prepared by adding 2g silane/100g mica mixture to 150mL of solvent comprising 10:90 ratio of water to methanol. The pH of the solution was adjusted to 5, with acetic acid. To complete the hydrolysis of organo-silane, the solution was agitated for about 15 minutes. Now the reinforcing agent was added to the solution, mixed for 10 minutes and oven dried for 24 hours at 110°C. The organo titanate was added directly to the polyethylene melt at 2% concentration.

Sample Preparation: The homogeneous composite samples were prepared by melt blending of polymer resin and reinforcing agent in the mixing head of "Brabender" plasticoder at 200°C, adjusted to a rotation speed of 50 rpm, for 15 minutes. For each surface treatment four compositions with 20, 30, 40 and 50 weight percent were prepared. The films were made by using a hot press at 40 MPa and 200°c for 5 minutes. Films were quenched in cold water before cutting them in to strips of different dimensions. The thickness of these films was adjusted to 0.5 - 1.0mm, having a cross section of 3mm and length of 60mm.

Microscopic Studies: The morphology of the mica flakes and the fracture surface of mica composites was examined in a JEOL - Scanning Electron Microscope, IS M-280. The samples were coated with gold/pladium.

Mechanical Properties: Stress-strain measurements were performed on an Instron Tensile Tester at room temperature at an extension rate of 10 mm/min. For dynamical mechanical measurements, a Rheovibron visco-elastometer was used at 110 Hz.

RESULTS AND DISCUSSION

Size of the seived and unseived mica flakes is given *in Table 1*. Seiving of the mica flakes results in better uniformity, better length and consequently higher L/D (length/diameter) ratio.

TABLE 1. Dimensions of Mica Flakes.

	UNSEIVED FLAKES	**SEIVED FLAKES**
Form	**Ellipsoidal**	**Ellipsoidal**
Average length (μ^n)	45	55
Average Thuckness (μ^n)	2.1	2.1
L/D Ratio	22	27

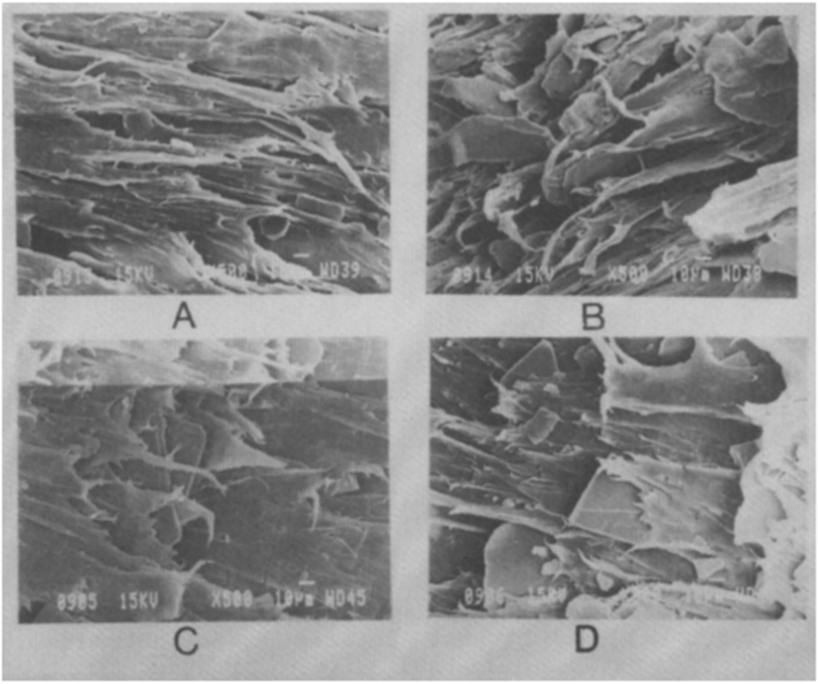

Fig.1: SEM photographs of mica-HDPE composites, a) 20% untreated, b) 40% untreated, c) 20% silane treated, d) 40% silane treated.

Fig.1 shows a micrograph of a mica-HDPE composite with and without silane surface treatment. The filaments were capillary extruded and fractured under tension. It is seen that the dispersion of mica flakes in the untreated composite is heterogeneous, with patches of higher mica concentration at some places. This poor dispersion is better shown by mica increasing the content to 40 weight percent. The silane treatment gives quite an improvement in the mica flakes particularly on their surfaces. The aggregate sizes are decreased by decreasing of void spaces.

The effect of titanate treatment on the dispersion of mica is shown in Fig. 2(a,b). The adhesion between the flakes and matrix is less evident than in the case of silane treatment. It is observed in the SEM photographs that when the mica concentration is low, the polymer matrix is deformed and exists in the form of elongated filament. However, when concentration of mica is increased, the deformation of the polymer matrix decreases. To study the mica-polymer interface, electron microscopic measurements were made parallel to the surface of samples. Fig.2(c,d) show, SEM photographs of 50 weight percent mica-HDPE sample. There is hardly any evidence of adhesion between the mica and polymer interface.

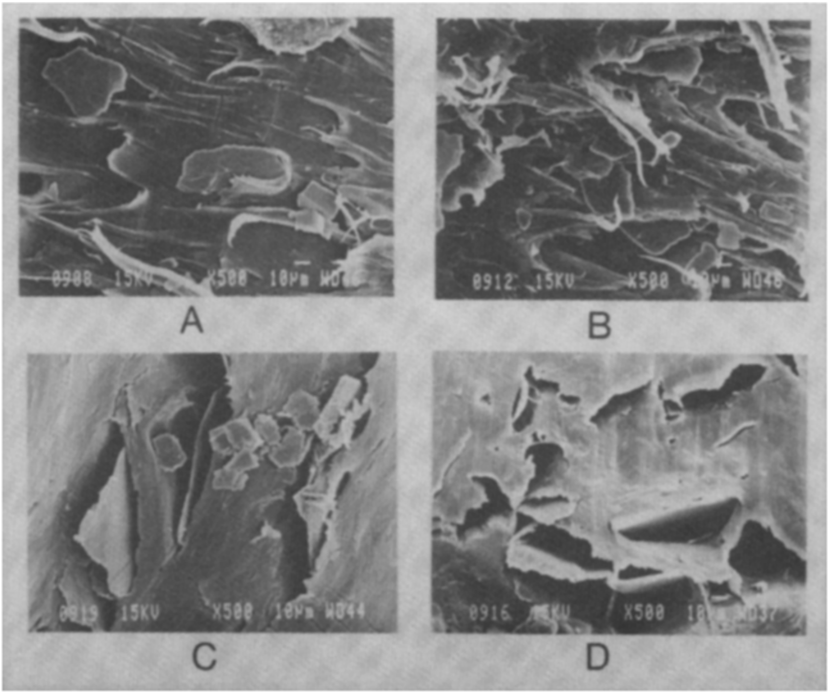

Fig.2: SEM photographs of mica-HDPE Composites, a) 20% titanate treated, b) 40% titanate treated, c) 50% untreated, parallel to the surface, d) 50% silane treated parallel to the surface.

When the same experiment was repeated with silane treated composite, on the contrary a strong adhesion was observed between the mica and polymer: it is seen that even though the polymer surface is deformed, it remains attached to the mica.

Typical stress-strain data, obtained at room temperature, of mica-HDPE composites, untreated and treated with silane and titanate are presented in Table 2. The results indicate that for untreated composites, the modulus increases almost linearly with the filler concentration. The ultimate strength decreases or remains constant while the elongation at rupture decreases with filler concentration. The drop in strength of filler reinforced unmodified polymers have already been observed by other investigators (4,10). This strength reduction is attributed to poor bonding between the hydrocarbon matrix and the filler. Table 2 shows that the effect of the two coupling agents are very similar where the role of these coupling agents as bonding promotor is evident. By comparing the silane and titanate treatment it can be seen that use of titanate yields lower modulus and ultimate strength values than those treated with silane, but much higher than untreated ones. The titanate we believe, behaves as a lubricating agent and causes a breakdown of the aggregates in response to the applied stress, resulting in an increase in elongation. Silane on the other hand, increases the adhesion between the mica and the polymer matrix resulting an increase in modulus as well as strength. It is to be noted however that when a different formulation of titanate was used, it acted as adhesion promoter rather than lubrifying agent (1). The improvement of the tensile properties of surface treated mica-composites can partly be explained by the better adhesion between the matrix and the renforcement. Another important factor is flake orientation (4,6). Coupling agents may favor the dispersion of the filler and reduce the size of agglomerates.

TABLE 2 Mechanical Properties of HDPE-MICA composites

MICA %	**UNTREATED**			**SILANE**			**TITANATE**		
	E MPa	**G MPa**	**ε %**	**E MPa**	**σ MPa**	**ε %**	**E MPa**	**σ MPa**	**ε %**
0	787	33	823	787	33	823	787	33	823
20	1390	25	398	1401	36	470	1395	32	400
30	1852	28	269	2075	41	327	1830	36	317
40	2778	27	117	2930	44	215	2818	39	201
50	3627	29	77	4017	47	107	3807	40	133

E= Modulus, σ= Ultimate strength, ε= Break Elongation.

The molecular motions in the solid state and their effect on the thermo-mechanical properties of polymers are best described by dynamic mechanical analysis. Fig. 3 presents typical loss tangent data from dynamic mechanical experiments for HDPE-20% mica composites. It is seen that with addition of fillers, in the absence of surface treatment, the tan δ peak increases. This is praticle friction. Surface treatment decreased the damping peak, we beleive, by decreasing particle-particle interaction. As a result better dispersion is achieved and agglomerates are broken down.

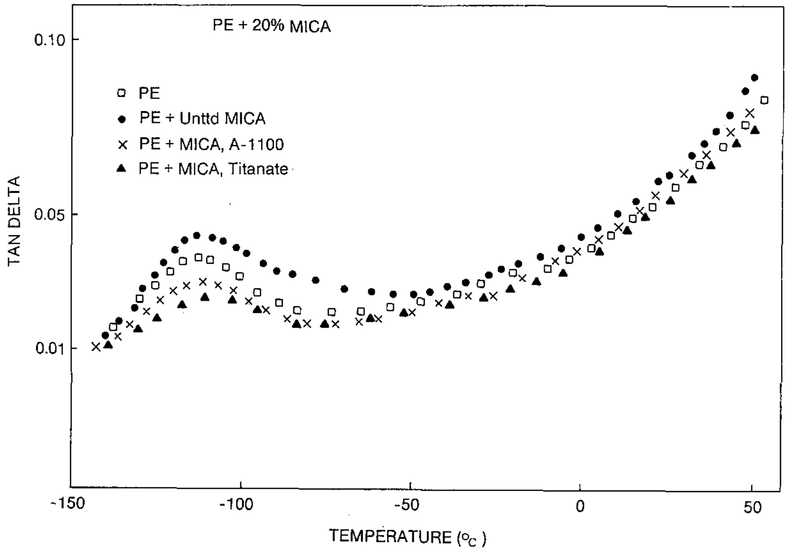

Fig.3: Temperature dependence of the dynamic loss tangent (tan δ) of HDPE-Mica composites.

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