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Preparation and Characterization of PMMA-HDPE and HDPE-PMMA Binary Polymer Blends

Abstract- *A comparison was made on the mechanical characteristics of binary blends polymethylmethacrylate/ high density polyethylene (PMMA-%HDPE) and vice versa. Analysis of morphology by SEM has been also accomplished. Preparation of polymer blends was performed using melt mixing method by an extruder. Tensile results showed that binary polymer blends (PMMA-%HDPE) indicated an increment in ultimate tensile strength, elastic modulus and shore D hardness compared to (HDPE-%PMMA). The blend of 95%PMMA-5%HDPE shows superior mechanical properties. SEM results indicated that the prepared blends are not fully compatible with some separated phases of the second polymer dispersed in the matrix.*

Keywords- *Polymer blends, binary blends, Tensile strength, shore D Hardness.*

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1. Introduction

Plastics are considered the most important and widely used materials in our daily life and industry for the last years. Their usage in various industrial applications is one of the reasons for this great popularity due to the tremendous range of characteristics exhibited by plastics, beside to their ease of processing. A new approach in the polymer science and technology has emerged recently. These polymers must perform under active conditions imposed by the requirements of a specific application [1-5].

Blending of polymers becomes an important approach in industrial production for tailoring products with an optimum material property. Performance and efficiency of polymer blends depend on the individual properties of polymeric components, as well as how they are arranging in space and whether the two polymers are miscible or exist as a single phase, most blends of high molecular weight polymers exist as two-phase materials. The phases morphology is of great importance in this manner and there are different morphologies exist such as dispersed spheres of polymer in the second one, co-continuous phases and lamellar structures [6]. Some studies show that uncompatiblized immiscible blend provides synergy of mechanical properties at the optimum values processing and compositional parameters [7-9].

Salih et al. made comparative study between two types of blends HDPE-%PP and (LDPE-%PP). Mechanical properties results (ultimate strength, fracture toughness, Young's modulus, hardness and creep modulus for the first blend (i.e. HDPE-%PP) was higher than LDPE-%PP. Furthermore, SEM micrographs indicated that 20%HDPE:80%PP and 20%LDPE: 80%PP were immiscible blends [10]. Blending effect was study to overcome the viscosity problem, of UHMWPE with addition HDPE via solid-state shear pulverization (SSSP) method instead of melt processing due to the poor dispersion. Results appeared that blending via SSSP leads to dramatic improvements in impact compared with other methods of preparation [11]. In addition, the researcher have been studied the effect of three types of blends with same matrix (PS) including (PS:%LDPE), (PS:%PP) and (PS:%PMMA) prepared by twin extruder. Results showed that the impact strength of blends increases with increases each of (LDPE, PP and PMMA) content [12]. The aim of the present research is preparation two groups of binary polymer blends represented by (PMMA-%HDPE) and (HDPE-%PMMA) with different weight ratios and comparing the mechanical properties for using in knee joint replacement.

2. Matrails and Methods

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I. Materials and Experimental Procedure

Polymer materials used in this study are high-density polyethylene (HDPE), provided from China, with density at 23°C (964 kg/m^3) and tensile strength at break (26 MPa), HDPE were supplied in a pellet form. PMMA in white pellet form supplied by HI Media Laboratories/ India. Product code GRM1746 with average molecular weight about 15,000.

II. Blending and pressing

PMMA-%HDPE and HDPE-%PMMA blends were mixed based on the selected ratios in Table 1 then melt processing by single screw extruder machine (general chemical industries company-Baghdad)) to form long strips of polymer blends (almost 1.5mm thickness), the extrusion parameters shown in Table 2.

A sheets of size (115×50 ×4 mm) were prepared by compression moulding technique including locating a piece of Aluminium foil to prevent adhesion, strips of extruded polymer blend (3 strips) located in stainless steel mold to have the required thickness for tests and after heating and pressing according to the parameters shown in Table 3.

III. Mechanical tests

Tensile test specimen prepared according to ASTM standard D638-87[13] computerized universal machine from Jinan Shijin Group Comp used at a constant strain rate (1 mm/min) at room temperature. All tests were repeated three times for each sample and results represent the average data of three specimens it was tested. Impact test conducted at room temperature according to ISO-179 [13], the impact test instrument model XJU-22, supplied from Time group Inc was used un notched charpy. Impact strength can be calculated from the following relationship:

$$G_c = \frac{U_c}{A} \quad (1)$$

Where

G_c: Impact strength of the material (J/m^2).

U_c: The required energy for sample fracture (J).

A: cross sectional area of the sample (m^2).

Fracture toughness can be calculated as follows:

$$K_c = \sqrt{G_c E_b} \quad (2)$$

Where:-

K_c: Fracture toughness of the sample (N.m$^{-3/2}$).

G_c: Impact strength of the material (J/m^2).

E_b:Young's modulus of the material (MPa).

Shore D hardness test used to measure the hardness of the samples according to ASTM- D-2240 [13]. Each sample has been tested 5 times and average results have been reported.

Table 1: Polymer Blends weight percentages

Polymer blends	Weight Percentages				
PMMA-%HDPE	100:0	95: 5	90:10	80:20	100:0
HDPE-%PMMA	100:0	95: 5	90:10	80:20	100:0

Table 2: Extrusion parameters

Polymer systems	Temperature (ºC)			crew speed (r.p.m)
	Zone1	Zone2	Zone3	
HDPE	140	145	140	90
PMMA	160	165	165	90
PMMA-%HDPE	160	165	165	90
HDPE-%PMMA	140	145	145	90

Table 3: The hydraulic pressing conditions

No.	Polymer systems	Temperature (ºC)
1	HDPE	130-140
2	PMMA	160-170
3	PMMA-%HDPE	140-150

4 HDPE-%PMMA 130-140

3. Results and Discussion

I. Tensile Results

Tensile inspection has been mainly achieved to investigate stress-strain behavior, Figure 1. Shows the stress-strain curve of pure polymers (polymethylmethacrylate and polyethylene), it has been observed from this figure that there is a difference in the behavior of (stress-strain) curves for each of pure polymethylmethacrylate and polyethylene. It was found that the behavior change from soft and tough for polyethylene to hard and strong for PMMA, and that related to natural of PMMA, it is generally known that the polymethylmethacrylate is more hard, brittle and stronger than high density polyethylene [13]. So PE had lower ultimate strength and higher elongation as compared to neat PMMA.

Stress-strain curve of binary blends (PMMA-HDPE) with different contents of HDPE (0, 5, 10 and 20 wt.%) and (HDPE-PMMA) with different wt.% of PMMA (0, 5, 10 and 20%) were presented in Figures 2 and 3 respectively. Figure 2 shows that the polymer blends with high percentage of PMMA had higher mechanical properties than the opposite polymer blend. In addition, there was a change in the behavior from hard and strong to soft and less strength with low percentage of PMMA in the blend. Where it was found the vice versa for the second group of binary blends (HDPE-% PMMA), the behavior of stress-strain curve change from soft and weak behavior to strong and soft with the increase wt.% of PMMA in the blend. In general, stress-strain curve behaviors of polymer blends are intermediate between their pure polymers. Still highest tensile strength and Young's modulus recorded at percentage 95%PMMA-5%HDPE as compared to samples of other wt.%.

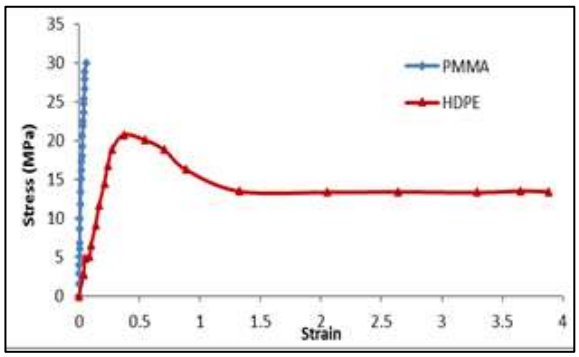

Figure 1: Stress-strain curve of pure polymers

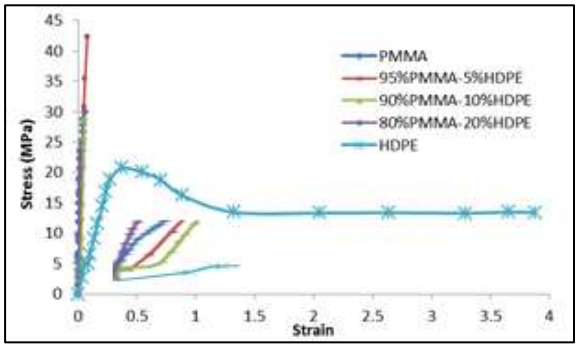

Figure 2: Stress-strain curve of polymer blends (PMMA: HDPE)

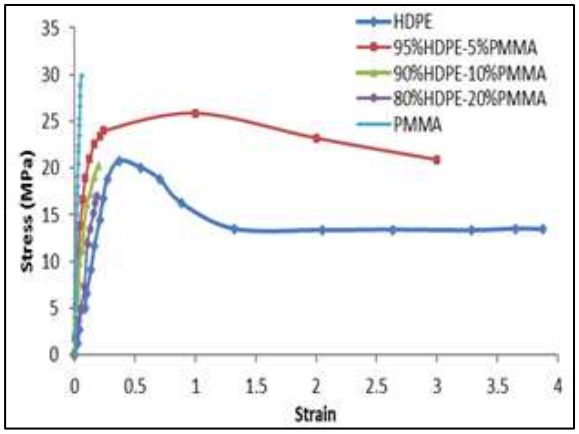

Figure 3: Stress-strain curve of HDPE-PMMA polymer blends

Ultimate tensile strength and Young's modulus of both groups of polymer blends presented in Figure 4, a and b respectively as a function of second polymer content in polymer blend, which indicate that maximum tensile strength and modulus of elasticity been at 5% of HDPE in polymer blend (PMMA-HDPE) and at 5wt.% of PMMA in the polymer blend (HDPE-%PMMA) and then decreased gradually with increasing the percentage of the added polymer. As for the elongation property, results recorded that it decrease drastically with increasing PMMA content in (HDPE:%PMMA) blend and low increasing as HDPE increased in (PMMA:HDPE) polymer blends. Whereas the fracture strength and Young's modulus value for the first groups samples larger than the second group of polymer blend, while the elongation percentage of these groups samples was found in vice versa. The above results may interpret as PMMA is stronger than HDPE, which may leads to immiscibility and incompatibility with high percentages [14] which was referred to the nature of poly (methyl

methacrylate) microstructure which has two group of methyl and methacrylate on each carbon atom of the main chain of PMMA, making considerable steric hindrance so that the polymer blend with high content of PMMA will be hard and relatively strong [12 and13].

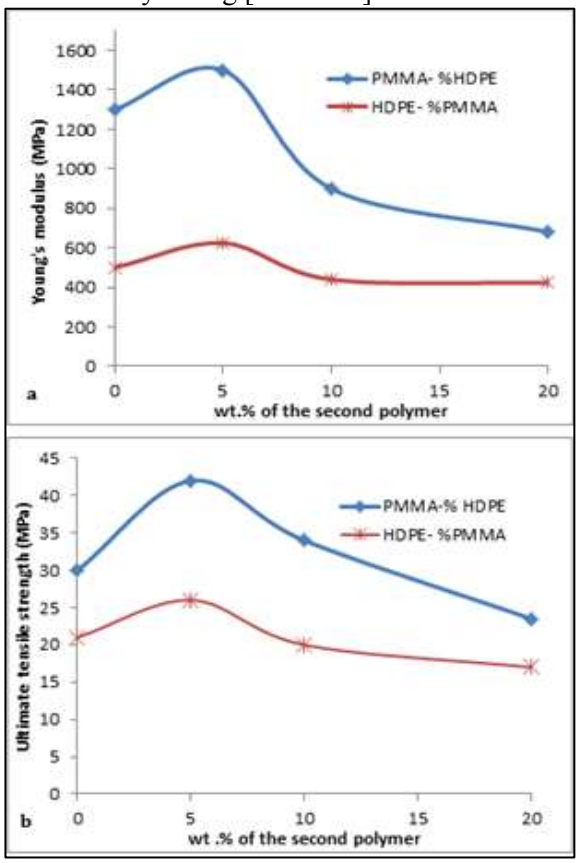

Figure 4: (a) tensile strength (b) Young's modulus of PMMA-HDPE and HDPE-PMMA polymer blends

II.Impact Results

Impact toughness is often considered the deciding factor in material selection because impact test measures the ability of polymer to withstand the load imposed upon being struck by an object at high velocity, thus it is a measure of energy required propagating the crack cross the specimen; therefore the impact properties of these samples are especially important. Figures 5 and 6 show the effect of blend ratio on impact strength and fracture toughness for (PMMA: HDPE) and (HDPE: PMMA) polymer blends respectively. Results indicated that poly (methyl methacrylate) has higher impact strength and fracture toughness compared to HDPE polymer; this may belong to the complicated structure of PMMA. As well as results showed that impact property of both neat HDPE and PMMA have been improved with the blending strategy especially with low weight ratios 5% and 10% reaching to almost 300 kJ/m^2 for HDPE: PMMA but goes down at 20wt.% while fracture toughness showed higher values at 5wt.% for both blends and goes down with increased percentage of the second polymer. Whereas the impact strength and fracture toughness values for polymer blend (PMMA: HDPE) samples higher than the second group polymer blend (HDPE: PMMA), and this due to the difference in the molecular chain structure of PMMA and HDPE. The HDPE have a linear chain structure with very little branching on the main chains (crystalline structure), so the chains can pack more closely together to increase strength, whereas PMMA has been amorphous molecular structure and that related to the presence of the meth and methacrylate groups on every other carbon atom of the main carbon chain of PMMA, so this structure has more free volume, and this rise in the relative free volume leads to the observed higher impact strength and fracture toughness values for the first groups samples [12,13].

III. Hardness Results

It is quite clear from the following results shown in Figure 7 that shore D hardness of the two groups of polymer blends (PMMA: HDPE) and (HDPE: PMMA) that PMMA is harder than HDPE and their blends. As well as it has been noticed that, polymer blends (PMMA: HDPE) has higher shore D hardness values as compared with other opposite wt. percentage polymer blends (HDPE: PMMA). Therefore, the polymer blends with high ratio of PMMA content, have harder than the others do group samples, this may be to the difference in the molecular structure. Since HDPE polymer that tend to be ductile and tough differs from PMMA, which is stiff and brittle due to high chain entanglements [13]. From this block diagram (Figure 7), it has noticed that the highest value of hardness, of polymer blends has been recorded at (95%PMMA: 5% HDPE) which is (77).

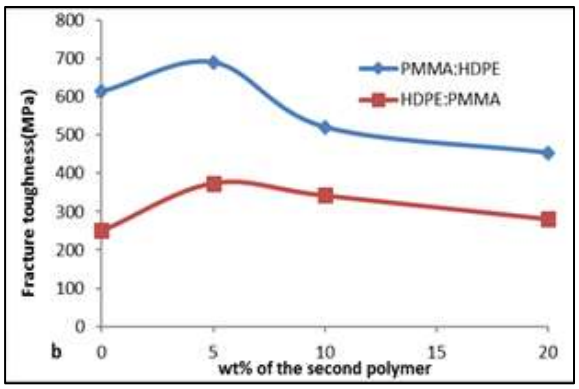

Figure 5: Impact Strength of polymer blends (PMMA: HDPE) and (HDPE: PMMA)

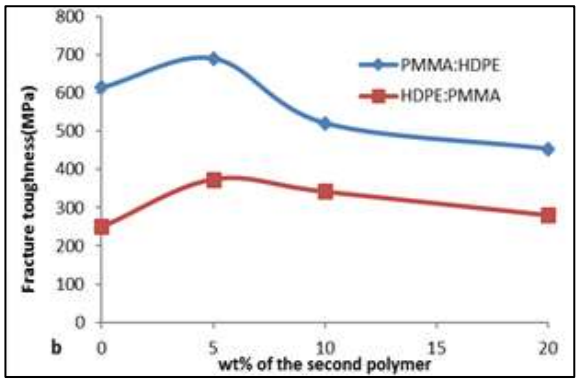

Figure 6: Fracture toughness of polymer blends (PMMA: HDPE) and (HDPE: PMMA)

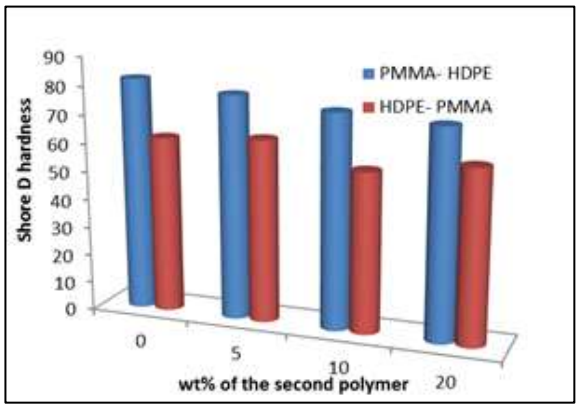

Figure 7: Shore D hardness of polymer blends (PMMA: HDPE) and (HDPE: PMMA)

IV. Morphology Results

Surface morphology changes of polymer blends evaluated by optical microscopic. Optical micrographs clearly appeared that there was difference in polymer blends morphology as shown in Figures 8 and 9 respectively. These figures show that the HDPE: PMMA are immiscible blends and incompatible in all ratios and this can be improved by droplet expansion of the second polymer in the mother matrix [16]. On the other hand the PMMA: HDPE blends are compatible and seem to be miscible at the low ratios 5wt. % while at high ratio (20 wt. %) there is immiscibility observed. Furthermore, SEM micrographs of blend fracture surfaces at different magnification were recorded in Figures 10 (a and b) and 10 (c and d) respectively, in order to correlate between (PMMA: HDPE) and (HDPE: PMMA) mechanical characteristics and the morphology, These figures show difference morphology of the two blends groups, since the fractured surface of (PMMA: HDPE) clearly showed two distinct phases – a continuous PMMA matrix and dispersed globules of HDPE phase, but with some discontinuous phase structure or so called "phase inversion". As well as it can be observed different sizes of spherical shaped HDPE particles were dispersed randomly in PMMA matrix as shown in Figure 10 (a and b), moreover incompatible immiscible blends can be characterized by the domains of one phase pulling away from the domains of the other phase resulting in a droplet-in-matrix morphology. Based on the above, immiscible polymer blends (IMPBs) may characterized by their domain sizes for their ultimate applications. If a blend's domain size is greater than 10 μm, applications include structural materials such as railroad ties and I beam, while domain sizes of less than 10 μm may referred by higher performance engineering blends [17].

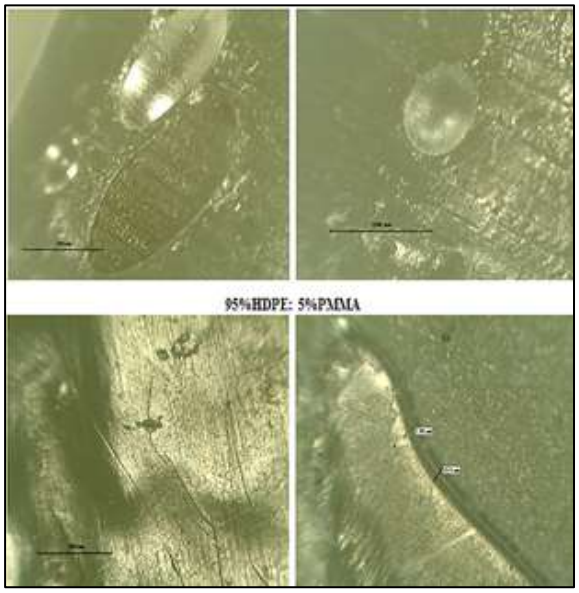

Figure 8: Optical micrographs of (HDPE: PMMA) polymer blends

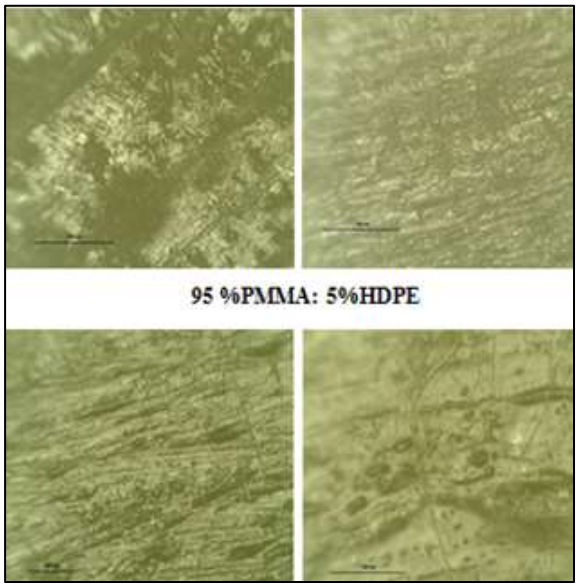

Figure 9: Optical micrographs of (PMMA: HDPE) polymer blends

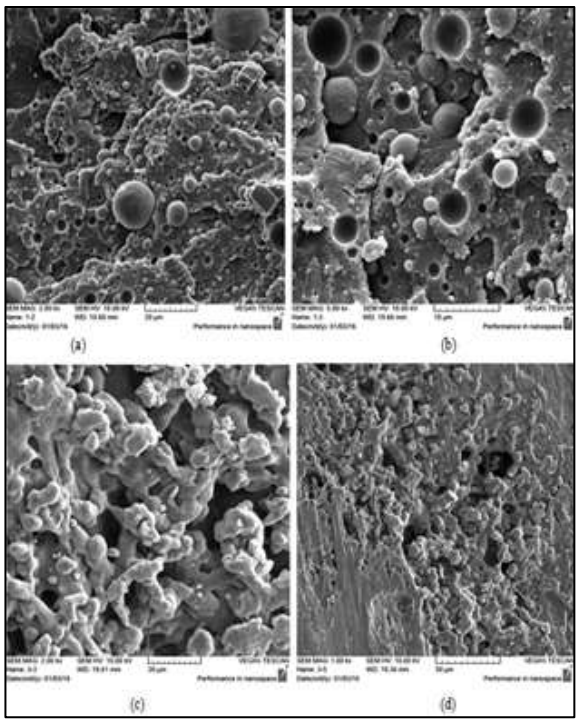

Figure 10: SEM micrographs of (a, b) PMMA: HDPE and (c and d) HDPE:PMMA polymer blends

4. Conclusion

1. Mechanical characteristics of PMMA: HDPE blends showed higher values compared to HDPE: PMMA polymer blends it may be explained by the difference in structure between PMMA and HDPE.
2. All mechanical properties (Young's modulus, ultimate strength, impact strength, fracture toughness and shore D hardness) were high at low percentage of addition (5 wt. %) for the two groups of blends, this may lead to the importance of miscibility and compatibility between the polymers blended to have the improvement in performance.
3. Optical micrographs appeared that the blends with polymethylmethacrylate matrix give miscible morphology and seem to be compatible at low wt. % HDPE, while the opposite occur in blends with high density polyethylene matrix even at low percentages of PMMA.
4. Finally, it should be clarified the importance of these two blends as biomaterials since it has high biocompatibility to human tissues. Bone cement is quite important to all orthopedic surgeons and commonly Polymethyl methacrylate (PMMA) is known as bone cement also, it is widely used for implant fixation in various Orthopedics and trauma surgery.

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