



## Effect of Grain and Calcinations Kaolin Additives on Some Mechanical and Physical properties on Low Density Polyethylene Composites

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### Abstract

In this work, a composite material was prepared from Low-density polyethylene (LDPE) with different weight percent of grain and calcinations kaolin at temperature of (850°C) using single screw extruder and a mixing machine operated at a temperature between (190-200°C). Some of mechanical and physical properties such as tensile strength, tensile strength at break, Young modulus, and elongation at break, shore hardness and water absorption were determined at different weight fraction of filler (0, 2, 7, 10 and 15%). It was found that the addition of filler increases the modulus of elasticity, elongation at break, shore hardness and impact strength; on other hand, it decreases the tensile strength and tensile strength at a break. Absorption test was carried out in water at different immersion times and different composite. The results of absorption show that it obeys Fick's law and after the addition of kaolin the amount of absorption decrease. Calcinations kaolin filler produces better mechanical properties, than grain kaolin fillers.

**Keywords:** Composite, low-density polyethylene, kaolin.

### 1. Introduction:

Materials are classified as metals, alloys, ceramics, glasses, composites, polymers, and semiconductors (1). Ceramics can be defined as inorganic crystalline materials, probably the most "natural" derived materials. They are strong, serve as good electrical and thermal insulators, often resistant to damage by high temperature and corrosive environments, but are mechanically brittle. Advanced ceramics are used in substrates that house computer chips, sensors and actuators, capacitors, wireless communications, spark plugs, inductors, and electrical insulation<sup>(2,3)</sup>. Its whiteness and plasticity make it extremely suitable for its extensive use as a filler, extender, ceramic raw material and pigment. The theoretical structural formula of kaolin is  $Al_2O_3 \cdot 2SiO_2 \cdot 2H_2O$ . It is highly refractory clay and has a melting point above 1800°C used by itself. Kaolin is white, soft, plastic clay mainly composed of fine-grained plate-like particles<sup>(4,5)</sup>.

Composites are made from different types of materials. They provide unique combinations of mechanical and physical properties that cannot be found in any single material. It is light weight, strong, ductile, high temperature resistant materials as well as hard, yet shock-resistance, cutting tools<sup>(2,5)</sup>.

LDPE is defined by a density range of 0.910-0.940 g/cm<sup>3</sup>. It is unreactive at room temperatures, except by strong oxidizing agents, and some solvent causes swelling. It has more branching, so its intermolecular forces are weak, with lower tensile strength, and its higher resilience. Also, since its molecules are less tightly packed and less crystalline because of the side branches, its density is lower<sup>(1,2)</sup>.

Many workers investigated the physico-mechanical properties of filled polyethylene with additives. Vlasova<sup>(6)</sup> studied the influence of the degree of filling and the nature of the fillers on the

properties of PE composites obtained by polymerizing ethylene on the surface of the filler particles. He found that the breaking stress and relative elongation at rupture with rise in the degree of filling diminish, while the elasticity modulus on stretching and the toughness of the polyethylene-kaolin composites rise. The scales of change in the properties of the composites with the degree of filling are determined by the nature of the filler. Tanaka and et al. <sup>(7)</sup> studied the dielectric properties of LDPE filled with aluminum silicate clay fillers by thermally stimulated currents and time domain dielectric spectroscopy. They investigated the change of the charge storage behavior and related dielectric responses by analyzing the effect of the particle size of the fillers and that of surface modification of the fillers. Dubois <sup>(8)</sup> studied the interfacial adhesion in polyethylene-kaolin composites using a stearic acid and maleic anhydride-grafted polyethylene as potential interfacial agents with the consideration of the efficiency of aminosilane-surface-treated kaolin. They found that the melt index of the enhanced interfacial filler-polymer has been decreased. They also concluded that comparing with other low molecular weight additives such as stearic acid and aminosilane, anhydride-grafted PE has very efficient in improvement the impact resistance of HDPE kaolin composites even at low contents. Privalko and Pedosenko <sup>(9)</sup> characterized the injection-molding and blow-molding grades of HDPE composites containing up to 30 Vol.% of calcinated Kaolin. Polyethylene/kaolin composites by measurements of melting temperatures ( $T_m$ ), heats of fusion ( $H_m$ ) and room-temperature densities. They showed that both  $T_m$  and the degree of crystallinity of a polymer matrix proved essentially unchanged regardless of filler content and/or presence of a custom coupling agent. Privalko et al. <sup>(10)</sup> Studied the characterization of samples of the blow-moulding grade HDPE filled with Kaolin by wide-angle X-ray scattering, micro hardness and stretching calorimetry techniques. They observed that crystallinity of the polymer matrix in the filled samples remains essentially the same as that in the neat polymer regardless of the filler content. Khazraji <sup>(11)</sup> studied the mechanical, thermal and electrical properties of epoxy resin reinforced with different clays and minerals namely bentonite, kaolinite, zeolite, selenium oxide, barium titanate and calcium carbonate. The modified epoxy resin was prepared using physical blends of this resin with 50% of additive materials, in addition to using different percentage of Iraqi

zeolite (5, 15 and 25%). It was found that the impact strength of the epoxy reinforced with 5% of Iraqi zeolite was doubled. The hardness of the sample reinforced with 5% of barium titanate was increased by 7% and the modulus of elasticity of the sample reinforced with 5% zeolite was increased by 156% compared with the non treated sample. Abed-Alhakem <sup>(12)</sup> studied the effect of Iraqi raw ceramic kaolin and boxide on epoxy resin, he showed that mechanical properties of composites filled with relatively small particles increased with increasing filler content, and decreasing filler size. Al-Neamee <sup>(13)</sup> investigated the mechanical and physical properties of PE reinforced with alumina. She found that mechanical properties increase with increase of wt% of alumina and the absorption water was decreased with the addition of filler.

In the present work, a composite material was fabricated using LDPE with different weight fraction of kaolin by using single screw-extruder to obtain material with the desired mechanical and physical properties.

## 2. Experimental:

### 2.1 Materials:

Commercial LDPE was used in this work; it was supplied by International Company for Chemical and Plastic Industries. The melt flow index and the density of the material were 20gm/10min and 0.9235 g/cm<sup>3</sup>, respectively.

Grain Kaolin has particle size of 42  $\mu$ m and density 2.23 g/cm<sup>3</sup> and melting point 1755°C. It is available in high purity and large quantity at low cost. Its chemical composition is (Al<sub>2</sub>O<sub>3</sub>=39.5%, SiO<sub>2</sub>= 46.5% and H<sub>2</sub>O=14.0%).

Calcined Kaolin has particle size of 39  $\mu$ m and density 2.53 g/cm<sup>3</sup>. It has good strength, stiffness, and available at low cost. Its chemical composition is (SiO<sub>2</sub>=59.19%, Al<sub>2</sub>O<sub>3</sub>= 32.28%, Fe<sub>2</sub>O<sub>3</sub>= 2.94%, TiO<sub>2</sub>=1.43%, CaO= 0.46%, L.O.I= 10.93%).

### 2.2 Method:

In this work, mechanical mixing was used to prepare composite materials from LDPE / kaolin using extruder machine. LDPE and kaolin were mixed with various compositions of filler (0, 2, 7, 10 and 15%). This mixture was then fed into 25mm single screw extruder. This machine is supplied with electrical heater, the materials is

melted by shearing it between the barrel and the smooth cylindrical part of the screw, finally the flat –end part of the screw act as a pump forcing the molten, composite materials out. The barrel temperature was monitored and controlled by thermostats. The die temperature was also controlled by a thermostat and was adjusted, together with barrel temperature to yield uniform output. The extrusion condition were listed in table (1). The extruder produced in the form of about 2 mm diameter monofilament was cooled in water. The monofilament produced by using a screw speed from (0-50) r.p.m. was uniform and opaque, which was cut in the form of granules of 3-4 mm length with the help of granules. The sheet was prepared by hot pressing the granules between hydraulic press at 190°C for LDPE and LDPE/Kaolin. A pressure of 10 MPa was applied for 5min to allow the composite to melt and spread out between plates. The pressure was removed and the mold sheet was quenched in water at room temperature.

## 2.3 Test Method:

### 2.3.1 Tensile Strength Test:

It can be defined as the maximum tensile sustained by the material being tested to its breaking point<sup>(2)</sup>.

$$\text{Tensile strength} = F/A \quad \dots(1)$$

where: F = force applied in N,  
A = cross section area mm<sup>2</sup>.

The tensile strengths values were determined by using tensile test according to ASTM D638<sup>(15)</sup>. Tensile properties were obtained using Zwick universal Testing Machine. The cross head speed was 10mm/min, and the standard literature for tensile test, Dumbbell -shaped was the general purpose test piece for plastic materials according to ASTM (D638), so test Specimens type 1434 with a thickness of 4mm used in this work. Elongation was measure by using Zwick machine. The measurement was carried out according to the test specification of ASTM (D638). Elongation% was calculated using eq:

$$\text{Elongation} = \frac{\text{Final} \cdot \text{gauge} \cdot \text{length} - \text{Original} \cdot \text{gauge} \cdot \text{length}}{\text{Original} \cdot \text{gauge} \cdot \text{length}} * 100 \quad \dots(2)$$

### 2.3.2 Shore Hardness Test:

Hardness is a measure of a material resistance to localized plastic deformation<sup>(1)</sup>. Shore D hardness was used to determine the hardness of the polymer materials, maximum Test Load: 50 N at Shore according to ASTM.

### 2.3.3 Impact Strength Test:

The degree of resistance of a polymeric material to impact loading is a piece of concern in some applications<sup>(2)</sup>.

Charpy test was used to determine the impact strength of the polymeric material, the samples of impact test were notched by (notch instrument) according to ASTM (D256-87)<sup>(13)</sup>. All the specimens of impact test of blend before and after reinforcement kaolin filler was standard i.e. of (10×4×55) mm. Impact strength is calculated by using the eq:

$$I.S = U/A \text{ (J/m}^2\text{)} \quad \dots(3)$$

where: I.S = impact strength (joule/ m<sup>2</sup>)

U = energy of fracture in (joule),

A = cross section area in (m<sup>2</sup>).

### 2.3.4 Modulus of Elasticity Test:

The Modulus measures the resistance of a material to elastic deformation, for linear elastic materials the stress  $\zeta$  is the related to the strain  $\epsilon$  by young's modulus. E (Hooke's law).

$$\zeta = E \epsilon \quad \dots(4)$$

Three point system was used to determine, modulus of elasticity is calculated by using the following relation:

$$E = \left( \frac{\text{mass}}{\text{deflection}} \right) \left( \frac{gL3}{48I} \right) \quad \dots (5)$$

$$I = \frac{DB^3}{12} \quad \dots(6)$$

where: I = Engineering bending momentum,  
D = width of samples (mm)  
B = thickness of samples(mm)  
G = gravity(m/sec<sup>2</sup>),  
L = sample length(mm)

$\left[ \frac{\text{mass}}{\text{deflection}} \right]$  : is the slope of linear part of mass deflection curve obtained <sup>(1,2)</sup>

**2.3.5 Absorption Test:**

Water absorption test was used to determine the coefficient of absorption by using electron-weighing type Satorius (Germany). Water tests were conducted in the temperature range (32

$\pm 2$ )°C. In order to determine the water absorption(20) we use the following equation:-

$$(m_2 - m_1) / m_1 \times 100 = \text{water absorption \%} \quad \dots(7)$$

where:  $m_2 - m_1 = \Delta m$  is the variation in weight gain with time

$m_1$ : the original dried specimen weight and from a known area we can find the water absorption for area by using the following equation:

$$\Delta m / A = (m_2 - m_1) / A \quad (\text{g/cm}^2) \quad \dots(8)$$

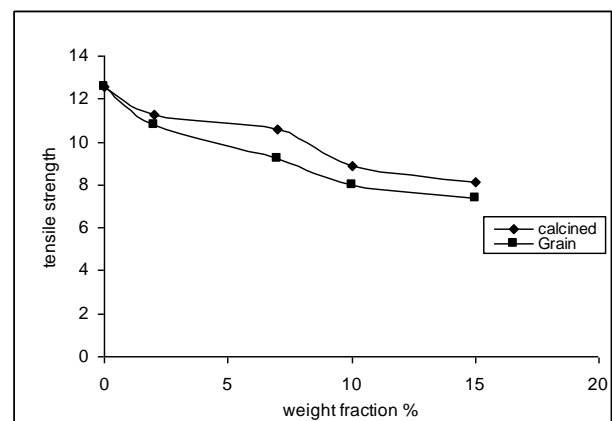
**Table 1**  
**Extrusion Parameter.**

Polymer	Température °C				Screw speed (RPM)
	Zone1	Zone2	Zone3	Zone4	
LDPE	150	170	220	190	50
LDPE/kaolin	150	190	250	220	50

**3. Results and Discussion**

Figure (1) shows the variation of tensile strength of LDPE at different wt.% of grain and calcined kaolin. From this figures, it is clearly seen that the tensile strength decreases with increasing wt% of kaolin. Growths of micro cracks, which produce sharp stress gradients, are influenced by the shape and orientation of reinforcement. Therefore, the voids and cracks do not transfer stress, making the material more compliable and there by generated increasing void space, which was responsible for stress propagation and thus lowering the material tensile strength <sup>(17)</sup>.

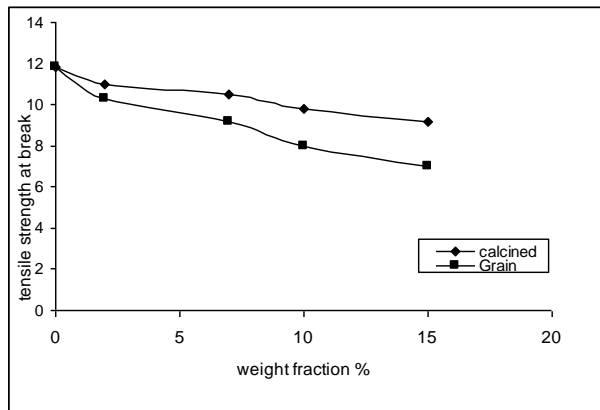
The results agree well with results obtained by Abed-Alhakem <sup>(11)</sup>. also, From this figure, it is found that the tensile strength of composite material made of (LDPE/calcined kaolin) is higher than that of composite material made of (LDPE/grain kaolin ) and this is attributed to high strength that LDPE/calcinations kaolin filler possesses. This may be due to the differences in behavior between grain and calcined kaolin that stem from their composition and structure. The polymer composites that contain the calcined kaolin having a high mullite content have improved mechanical properties compared to a similar polymer that does not contain <sup>(2,4)</sup>.



**Fig.1. The Relation Between the Tensile Strength of LDPE Sample With Different Percentage of Grain and Calcined Kaolin Filler.**

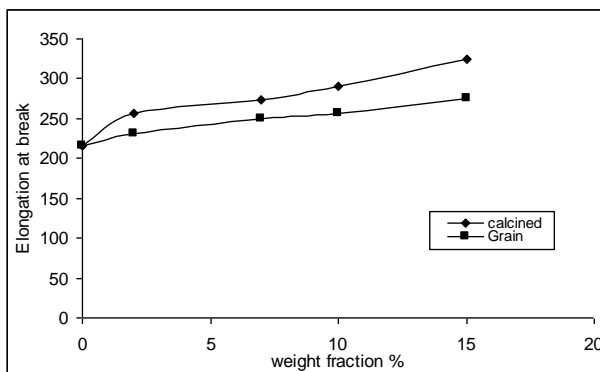
Figure (2) shows the relation between the tensile strength at break of LDPE at different wt.% of grain and calcined. From this figure it is clearly seen that the tensile strength at break decrease with increasing wt% of kaolin filler. The failure may happen to the material because of the influence of the fracture stress, which leads to crack appearing. These micro cracks collectively join to attain a macro-size which, at critical stress levels, becomes unstable, and this lead obstacles the transition of the stress. From this figure, it is found that the tensile strength at break of composite material made of LDPE / calcined Kaolin is higher than that of composite material made of LDPE / grain kaolin and being due to

LDPE/calcinations kaolin is harder and more rigid than LDPE/grain kaolin filler and also LDPE/calcined kaolin share the external stress with matrix better than LDPE/grain kaolin. This may be attributed to that, the tensile strength at break, which is effectively independent of microstructure.



**Fig.2. The Relation Between the Tensile Strength at Break of LDPE With Weight Fraction of Grain and Calcined kaolin Filler.**

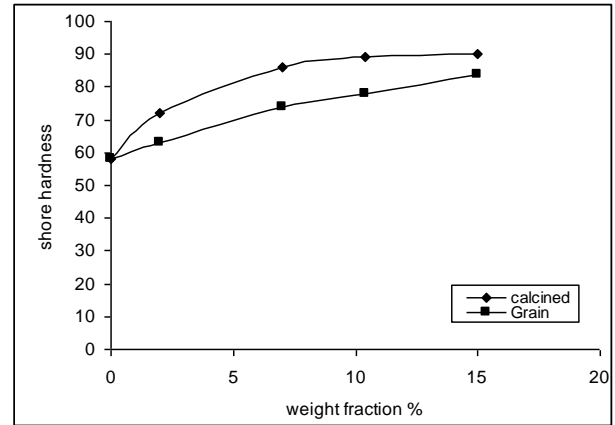
Figure (3) shows the relation between the Elongation at break of LDPE at different wt.% of grain and calcined.



**Fig.3. The Relation Between Elongation at Break of LDPE With Weight Fraction of Grain and Calcined kaolin Filler.**

The elongation at break is observed to increase on addition of kaolin filler. Experimental results are reported for the determination of the elongation of LDPE filled with and filler of various at different filler contents. From these figures it is the effect of on the weight fraction elongation at break of polymeric composite is clearly seen from the figure one can find that the elongation at break of composite material made of

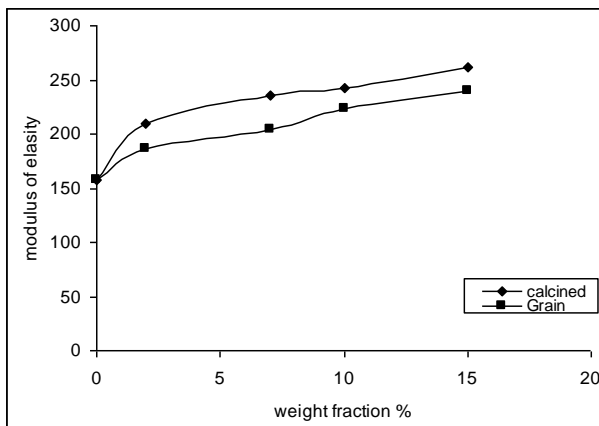
LDPE/calcined kaolin is higher than that of LDPE/grain kaolin. Figure (4) show the relation between the shore hardness and the weight fraction before and after reinforcement with filler for grain and calcined kaolin filler at different weight fraction.



**Fig.4. The Relation Between Shore Hardness of LDPE with Weight Fraction of Both Grain and Calcined kaolin Filler.**

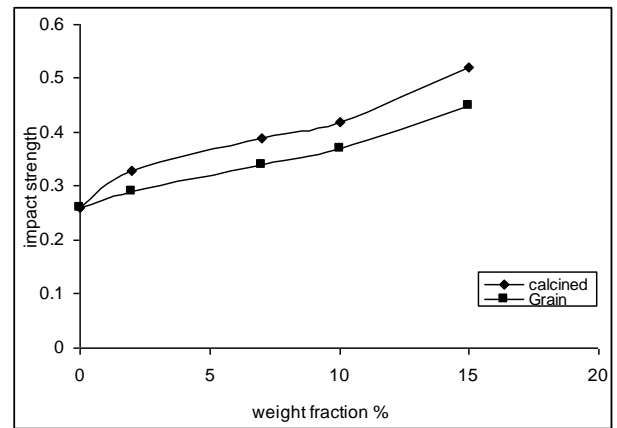
The additions of kaolin filler increase the hardness of composite material due to increase in the resistance strength of polymer to plastic deformation. This behavior may be due to the hard and high resistance to hardness of grain and calcinations kaolin filler material. The latter has a good ability to absorb the load applied by matrix LDPE and increase the resistance of matrix composed of LDPE and that will increase the binder force between the molecules of matrix with particulate reinforcement filler. The results agree well with results obtained by khazraji<sup>(10)</sup>. Figure (5) shows the effect of weight fractions of grain and calcined kaolin fillers on the tensile modulus. From this figure it is clearly seen that the modulus of elasticity increase with increasing weight fraction of kaolin filler. The particle filler agglomerates hence reduced the strain value restriction the mobility of matrix chains. The mobility of matrix chains and agglomeration of particle filler is clearly indicated by the increase in Young's modulus<sup>(18)</sup>. Also, From this figure, it was found that the modulus of elasticity of composite material made of LDPE / calcined kaolin were higher than those of the composite material made of LDPE / grain kaolin. Thus in the present study there was a good improvement of modulus of elasticity of through LDPE/kaolin the addition of particulate kaolin on modulus of elasticity. Calcined kaolin having a high mullite

content may be distributed uniformly or non-uniformly within the polymer matrix to yield desired product properties. Grain Kaolin exists naturally in the hydrous form. In the hydrous form, kaolinite minerals form crystal structures that are linked together by hydroxyl containing moieties. Hydrous kaolin may be converted to calcined kaolin containing a major amount of mullite by thermal processes. Such processes result in a dehydroxylation of the kaolin and an aggregation of the particles, and convert the crystal structure to an amorphous form<sup>(18)</sup>. The results agree well with results obtained by Vlasova<sup>(6)</sup> and Abed-Alhakem<sup>(13)</sup>.



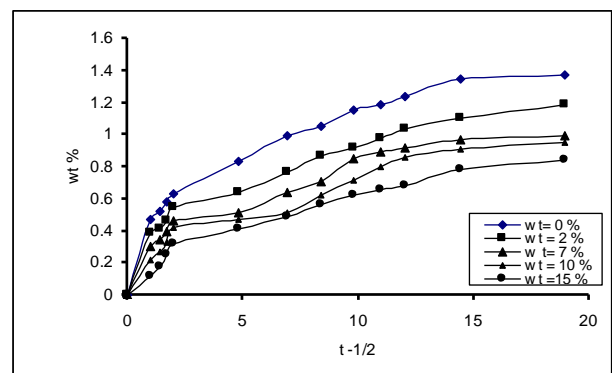
**Fig.5. The Relation Between Modulus of Elasticity of LDPE With Weight Fraction of Both Grain and Calcinations Kaolin Filler.**

Figure (6) shows the variation of impact strength of LDPE at different wt. % of grain and calcined kaolin. From this figure, it is clearly seen that the impact strength increases with increasing weight fraction of kaolin. This may be due to a good adhesion between the matrix and filler. Also, from this figure, it is found that the impact strength of composite material made of LDPE/ calcined kaolin is higher than that of composite material made of LDPE/ grain kaolin and this is attributed to high strength that LDPE/ calcined kaolin filler possesses. Because the structure of Kaolin is not symmetrical the crystal layers become highly polarized. The polarization of the layer crystals allows the kaolin to adsorb water molecules (which are also polar) between the layers.



**Fig.6. The Relation Between Impact Strength of LDPE With Weight Fraction of Both Grain and Calcined Kaolin Filler.**

Diffusion is the process by which matter is transported from one part of the system to another as a result of random molecular motion. In general, diffusion behavior and transport in glassy polymers have been classified according to the relative rates of mobility of the penetrate and of the polymer segments. Figures (7 and 8) show that adsorption water is reduced with increase in weight fraction of filler, because has a high ability to bear water compared with polymeric material. The absorption percent was reduced with increase in the weight fraction of filler. The amount of water absorbed will depend primarily upon the chemical nature of the polymer matrix and the environment to which it exposed. The results agree well with results obtained by Al-Neamee<sup>(14)</sup>.



**Fig.7. Shows the Relation Between the Weight Gains % of LDPE/ grain Kaolin With the Square Root of Time.**

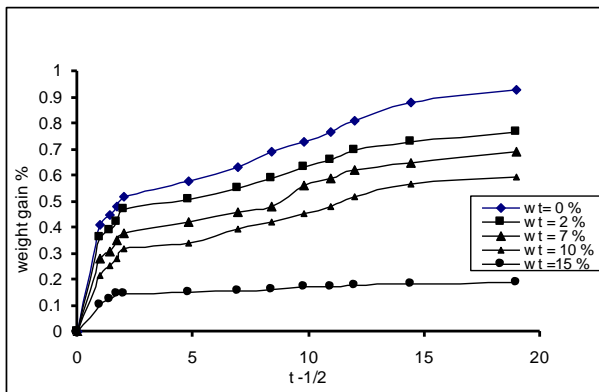


Fig.8. Shows the Relation Between the Weight Gains % of LDPE/Calcined Kaolin With the Square Root of Time.

#### 4. Conclusion:

The addition of kaolin was found to improve the modulus of elasticity, shore hardness, impact strength and elongation at break. On other hand, pure LDPE has a higher value of tensile strength. Water absorption values were observed to be the highest in pure LDPE, and these values decreases with increasing different percentage of kaolin. Kaolin calcinations filler produces better mechanical, and physical than grain kaolin fillers.

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## تأثير اضافات (الحبيبات والكلسنة) الكاؤولين على بعض الخصائص الميكانيكية والفيزيائية على متراكبات البولي اثلين الواطئ الكثاف

زينب يوسف شنين

قسم الهندسة الكيماوية/ الجامعة التكنولوجية

### الخلاصة

في البحث الحالي تم تحضير مواد مركبة والتي هي عبارة عن مخلوط بوليمري ، يتكون من مزج بولي اثلين الواطئ الكثافة مع الكاؤولين المحروق بدرجه حرارية (850 م°) والغير محروق. وذلك باستخدام جهاز الباتقة الأحادية حيث تم تشغيلها عند درجات حرارة تتراوح بين (150-200 م°). تم استخدام ماكينة البثق أحادية اللولب في تحضير المواد المتراكبة. وبعد ذلك جرت دراسة بعض الخصائص الميكانيكية مثل الشد وإجهاد الشد عند الكسر والاستطالة، قوة الصدمة وصلادة شور ، ولجميع المتراكبات المحضرة وقورنت النتائج المستحصلة مع تلك التي تعود لمادة الأساس بمفرده . و جرت الفحوصات بكسور وزنيه مختلفة. من النتائج التي تم الحصول عليها لوحظ ازدياد كل من: معامل المرونة، الاستطالة عند الكسر ،صلادة شور وقوة الصدمة ونقصان مقاومة الشد بزيادة نسبة الخلط ألوزني. تم إجراء فحص الامدصاصية لكل نموذج قبل وبعد التدعيم بمسحوق الكاؤولين وأظهرت النتائج نقصان في الامدصاصية بعد التدعيم . من النتائج التي تم الحصول عليها تبين أن الكاؤولين المحروق أعطى نتائج أفضل من الكاؤولين الغير محروق.