Comparative Study of the Mechanical Properties of Two Groups Ternary Polymer Blends Composites

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ABSTRACT

Thermoplastic polymer blends composites were prepared as strips by double screw extruder. The prepared composites consists of two groups, the first group was prepared as ternary polymer blend according to the formula [84%LLDPE:15%((100-X)%PP:X%PVP):1%B.P] with X values (0,4,8,12,16%).The second group was prepared with replacing the weight ratios for each of the LLDPE and PP according to formula [84%PP:15%((100-X)%LLDPE:X%PVP):1%B.P] with same X values. Mechanical properties for all prepared samples (tensile, flexural, compression, impact toughness, hardness and creep tests) were studied. The results of this research showed that the fracture strength, young modulus, compressive strength, flexural modulus and creep modulus for the two groups increase with increasing in PVP percentage and the highest values were at 8%PVP, while elongation decreases with addition PVP to the composites. Moreover flexural strength, shear stress and hardness values increase slightly with increasing in PVP percentage for both groups. In addition to that, the impact strength and fracture toughness for the second group samples were high and reach the maximum values (0.45 kJ/m²) at 4%PVP and (4.58 Pa√m) at 8%PVP respectively, as well as all results show that the second groups samples have better mechanical properties.

Keyword: polymer composites, PVP, PP, LLDPE

دراسة مقارنة للخواص الميكانيكيه لمجموعتين من الخلائط البوليمرية الثلاثية المتراكبة

الخلاصة

الخلائط البوليمرية اللدائنية المتراكبة تم تحضيرها بشكل شرائط بواسطة باثقة ثنائية اللولب المتراكبات المحضرة تتكون من مجموعتين,المجموعة الاولى حضرتكمزيج بوليمري لدائني ثلاثيوطبقا الى الصيغة التالية :(PVP%X :PP%(X-100)) %15 :LLDPE %84] B.P%1]وعندقيم X (16,12,8,4,0). والمجموعة الثانية حضرت باستبدال النسب الوزنية لكل من LLDPE و PP فيما بينهما وحسب الصيغة التالية -100((15:PP%84] LLDPE:X%PVP):1%B.P]%(X وبنفس قيم X . تم دراسة الفحوصات الميكانيكية للعينات

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المحضرة والمتمثلة (فحص الشد والانحناء والانضغاطومتانة الصدمة والصلادة والزحف)وبينت النتائج البحث ان كل من مقاومة الكسر ومعامل المرونة ومقاومة الانضغاط ومعامل مرونة الانحناء ومعامل الزحف لكلا المجموعتين تزداد بزيادة نسبة PVP في الخليط وسجلت أعلى قيم عند النسبة PVP(8%) بينما تناقصت قيم الاستطالة لكلا المجموعتين عند أضافة PVP للمتراكبات المحضرة ,أما قيم مقاومة الانحناء واجهاد القص والصلادةازدادت قليلاً مع زيادةنسبة PVP في المجموعتين الاولى والثانية.ومن خلال فحص مقاومة الصدمة لوحظ ان عينات المجموعة الثانية تمتلك اعلى قيم لمقاومة الصدمة ومتانة الكسر والبالغة (0.45 kJ/m²)عند نسبة PVP 4%و(4.58 Pa√m) عند نسبة PVP8% على الترتيب. كما وبينت النتائج أن عينات المجموعة الثانية تمتلك خواص ميكانيكية افضل مما تمتلكها عينات المجموعة الأولى.

INTRODUCTION

There is currently a strong interest for polymer blend composites. The basic aim of polymer blend research was to develop new materials with optimum properties. The increasing use of thermoplastics in many applications can be attributed to their cost of effectiveness, corrosion resistance and ease of fabrication [1]. The blending of two or more thermoplastic polymers is not only for development of polymers with new properties, but also for recycling of green materials [2-4].Among the polymer blend attracting commercial interest are polypropylene (PP-based) ones, indeed, there are extensive literatures on the blends of polypropylene (PP)/polyethylene (PE) [5-9].

Three different types of polyethylene (PE), namely low density polyethylene (LDPE), high density polyethylene (HDPE), and linear low density polyethylene (LLDPE) were used to modify the physical and mechanical behavior of PP by forming physical blends [5]. One of the reasons for addition of (PE) to (PP) is improvement of its low temperature impact strength and young's modulus, and environmental stress cracking properties. The PP drawbacks can be considerably improved by blending PP with other polymers [10-11].

Blending of PP and different PE largely depends on the miscibility or immiscibility of the two polymers. PP and three types of PE (LDPE, LLDPE and HDPE) are generally considered immiscible in the whole composition range and shows a remarkable phase separation during cooling /crystallization [10,12]. However PP/LLDPE miscibility is restricted by the processing conditions, compositions and high temperatures. If a blend of PP and LLDPE is cooled from a miscible melt it may separate into two phases resulting in an immiscible blend [11,13]. Studies elsewhere have shown that the uncompatibililzed immiscible polymer blends provide synergy of mechanical properties when the processing and compositional parameters are near optimum values [14].

Two ways are used to improve polymer properties, polymer blending and the use of filler to form polymer composites. These two methods were extensively, but separately used. Studies dealing with ternary polymer blends reinforced with rigid fillers to give multiphase polymer composites are still fairly new. Very few studies of ternary systems involving natural fibers as reinforcement were reported [3, 4, and 11].

In this study two groups of composites were prepared from ternary polymer blend (PP/LLDPE/PVP) and reinforced with basalt particle. The two groups were prepared according to the formulas [84%LLDPE:15%((100-X)%PP:X%PVP):1%B.P] and

[84%PP:15%((100-X)%LLDPE:X%PVP):1%B.P] respectively. Mechanical properties were studied for both groups.

Experiential Part
Materials

In this research three types of polymer materials were used, two of them were provided from National Company for Plastic and Chemical Industries, Zaffrania-Baghdad. These polymer materials are linear low density polyethylene (LLDPE) which produced from (Dow Company); with trade name "Dowlex 2042/2045". Isotactic polypropylene (PP): This was supplied from Sabic Company, Saudi Arabia, with trade name "520LPP", melt flow rate [10(g/10min)] and E-modulus [1600(N/mm²)].Polyvinylopyrrolidone (PVP) type (K-30) produced from "Sinopharm chemical reagent company", in china with trade name "Luviskol or Kollidon" and polymer structures $[C_6H_9NO]_n$.

Basalt particles (B.P) were used as a filler material for the polymer blend composite. They were produced from basalt stones which were obtained from "That Al-Sawary State Company for Chemical Industries".

Sample Preparation

Basalt stones were washed thoroughly with detergent powder and then with hot distilled water in order to remove dust and mud. After that these stones were dried in a hot air oven at 60 °C for 3 hours, and then crushed by cracking machine to small grains. Finally the basalt grains were ground by porcelain mill for several times, each time was done for three hours, until grains become fine particles, then the grinded particles were sieved garbled by sieving process to get very fine particles. The particles size distribution of basalt particles was carried out using laser diffraction particle size analyzer type (SHIMADZU SALD2101) located in the Ministry of science and Technology. The result of particle size distribution is shown in Figure (1). The mean particle diameter was (4.848 μm) and the median value was (5.872μm).
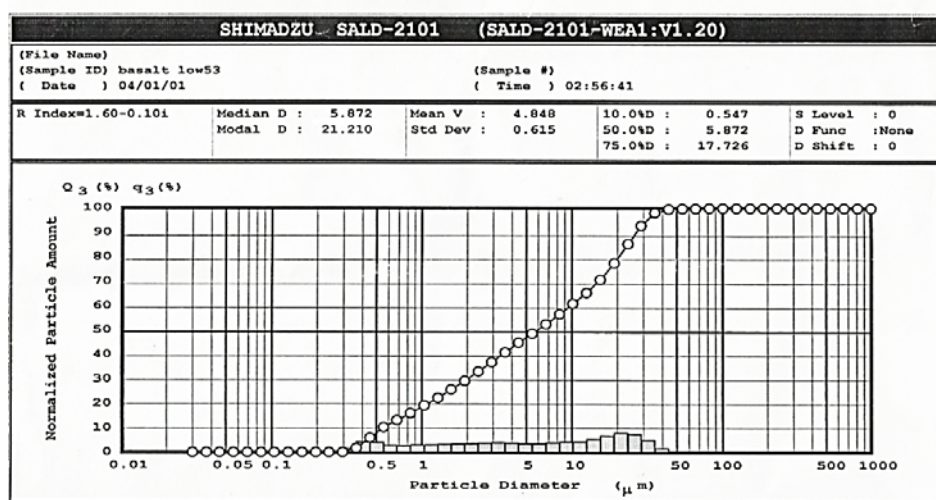

Figure. (1) Shows particle size analysis of basalt.

Pellets of polymer materials and particles of reinforced material were weighed according to selected percentages to prepare the batches for extrusion. For good distribution, the polymer blends and reinforced materials were mixed together in dry condition at room temperature for 20 minutes by mill made of porcelain material. The first group of Polymer blends composites were prepared according to the formula [84%LLDPE:15%((100-X)%PP:X%PVP):1%B.P] and the second group were prepared by replacing the weight percentages for each LLDPE and PP according to the formula [84%PP:15%((100-X)%LLDPE:X%PVP):1%B.P] with X values (0, 4, 8, 12, 16%), for both groups poymer blends composites were melted in double screw extruder machine with a screw L/D of 30:1.The extrusion temperatures for three zones (feed zone, compression zone and melting zone) were 170, 170, and 160°C respectively with screw speed 50 r.p.m.At the end of the extruder, the melt passes through a die to produce an extrudate as a strip with 1.5mm thickness. In order to get a suitable thickness for samples test, three strips of extrudate polymer blend located in mold made of steel (115* 50*5mm) which is previously heated at 150 °C for one hour, compression technique carried out at pressure 350kg/cm² for 10 minutes.

Mechanical test

Prepared samples for the tensile test were cut according to ASTM D638 [15, 16].The machine used for testing is micro computer controlled electronic universal testing machine (model WDW 200 E) made in china. The test was conducted at constant cross head speed of the order of (1 mm/min) at ambient temperature. Tensile stress was applied till the failure of the sample and stress-strain curve was obtained. Each sample was tested three times and the average values of the results have been reported.

Bending behavior of the prepared sample was tested using a three point test instrument, (model WDW 200 E) according to ASTM D-790-78 [15, 16], at room temperature with velocity (5mm/min) until the failure of the specimen occurs. Each sample was tested three times and averagevalues of the result have been reported, flexural strength (σ_{bend}), maximum shear stress ($\tilde{\iota}$) and flexural modulus are valued from the relationships (1-3) respectively.

$$\text{Flexural strength for three - point bend test} = \sigma_{bend} = \frac{3\,FL}{2wh^2} \qquad \cdots\cdots\cdots\cdots(1)$$

Where F is the fracture load, L is the distance between the two outer supporting points, w is the width of the specimen, and h is the thickness of the specimen.

$$\text{Maximum shear stress } \tilde{\iota} = \frac{3F}{4wh} \qquad \cdots\cdots\cdots\cdots (2)$$

$$\text{Flexural modulus} = E_f = \frac{FL^3}{4\,\partial wh^3} = \frac{mL^3}{4wh^2} \qquad \cdots\cdots\cdots\cdots (3)$$

Where∂ is the deflection of the beam when a force F is applied, m is the slope of the load (F) /deflection curve.

Compressive test is performed at room temperature according to ASTM-D695 [15] with dimensions (8x4x4) mm3. A computerized universal testing machine made in china, model (WDW 200E) were used for compressive test. The compression stress was applied till the failure of sample. Each sample was tested three times and average values of the results have been reported.

Impact test is performed at room temperature according to ASTM ISO 179 [16].
Izodcharpy tension impact (measurement test machines Inc, Amityville-New York).
Hardness test carried out on a Durometer shore D scale a according to ASTM D-2240
[15].
A creep test is performed under a constant applied load (1500gm) at room tempera-
ture according to BS 1178 [16].Creep modulus has been calculated by applying the
formula (4). Each sample was tested two times and average values of the results have
been reported.

$$E = \sigma / \epsilon_{(t)} \qquad \cdots\cdots\cdots\cdots (4)$$

Where σ: is the initial applied stress, and∈: creep strain after a particular time
and constant.

Results and Discussions

Tensile inspection was carried out mainly to investigate the effect of addition PVP on
stress-strain curves behavior for polymer blends composites. Figure.(2) shows the
stress-strain curves of the first group composite which contains
[84%LLDPE:15%((100-X)%PP:X%PVP)%:1%B.P] with different ratios of PVP
where X has the values (0, 4, 8, 12, 16). As shown from these curves, all the prepared
samples exhibit plastic behavior with tensile load, and the behavior changes from
weak and soft for [84%LLDPE:15%PP:1%B.P] composite to strong and tough with
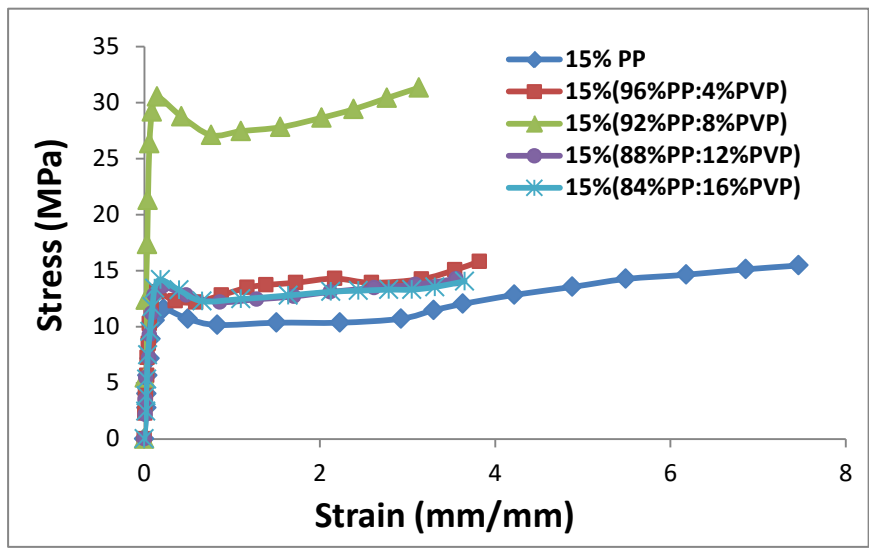the addition of PVP in 8%wt ratio as shown in Figure.(2).

**Figure. (2) The stress-strain curve of [84%LLDPE:15%((100-
X)%PP:X%PVP):1%B.P] as a function of PVP content in the composites.**

Figure.(3) illustrates the effect of PVP content on the stress-strain curves of second
group composite which contains [84%PP:15%((100-X)%LLDPE:X%PVP):1%B.P]
with different ratios of LLDPE and PVP by increasing weigh percentage of PVP
(0,4%,8%,12%,16%) and decreasing LLDPE content in the polymer blend at the

same ratio. As shown from those curves, all the prepared composites exhibit plastic behavior with tensile stress and significant decrease in the elongation at break with the increasing of PVP content in the polymer blend.
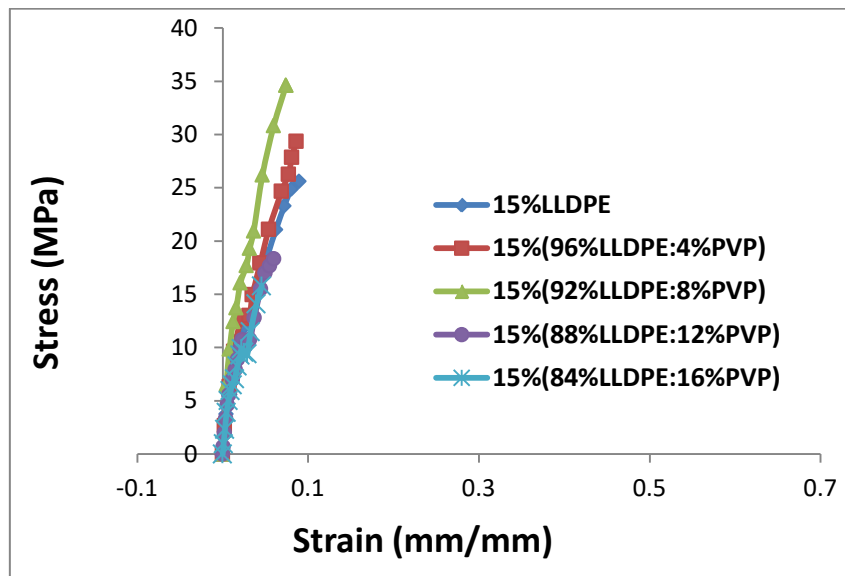

Figure.(3) Stress-Strain curves of [84%PP: 15 %((100-X)%LLDPE:X% PVP):1%B.P] as a function of PVP content in the composites.

Figures (4) and (5) illustrate the effect of PVP content on the fracture strength and Young modulus values respectively of the prepared composites. These figures indicate that the addition of PVP to the basic polymer blend composites (without PVP) increases the fracture strength and young modulus values of these composites, with maximum values at PVP 8%wt (34.6MPa) and (0.48GPa) respectively for the second group composite and (31.32MPa) and (0.140GPa) respectively for the first group at the same ratio of PVP (8%wt). The increase in PVP content larger than 8% ratio produces sharp decrease in these values to less than the values of the basic composite (without PVP). These behaviours related to the characteristics of PVP which may act as agent to improve the compatibility between constituents of polymer blend composite specially at slight ratio of PVP content, as well as PVP may promote optimum dispersion of particle phase in the composites [1, 15]. However, amounts of PVP to larger than 8% ratio may produce accumulation in the dispersed phases and this makes passive behavior in mechanical properties of prepared composites. Also it can be noticed from Figures (4) and (5) that the fracture strength and Young's modulus values of the second group composite are higher than that values for the first group composites having the same ratios of PVP, and that is related to the nature of is otactic molecular structure of PP which has a methyl group, the substitution of a methyl group with every second carbon atom of the polymer main chain restricts rotation of the chains, so the polymer blend composite is prepared with higher ratio of PP to produce strong composite compared with the other one having low ratio of PP.
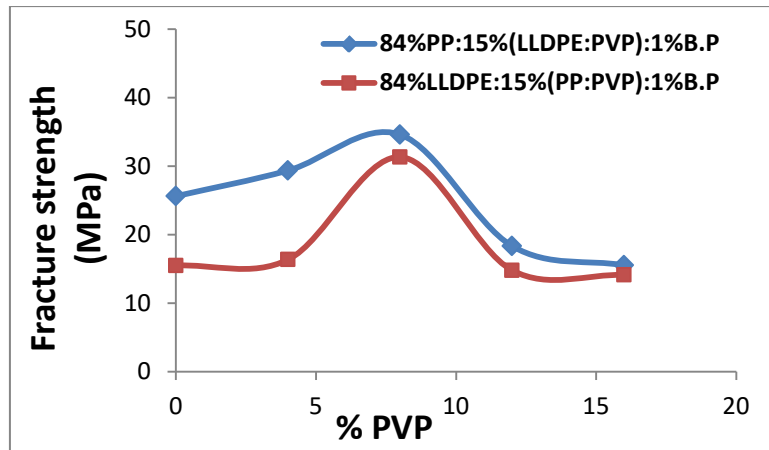
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**Figure.(4) Fracture strength of [84%LLDPE:15%((100-
X)%PP:X%PVP:1%B.P] and [84%PP:15%((100-
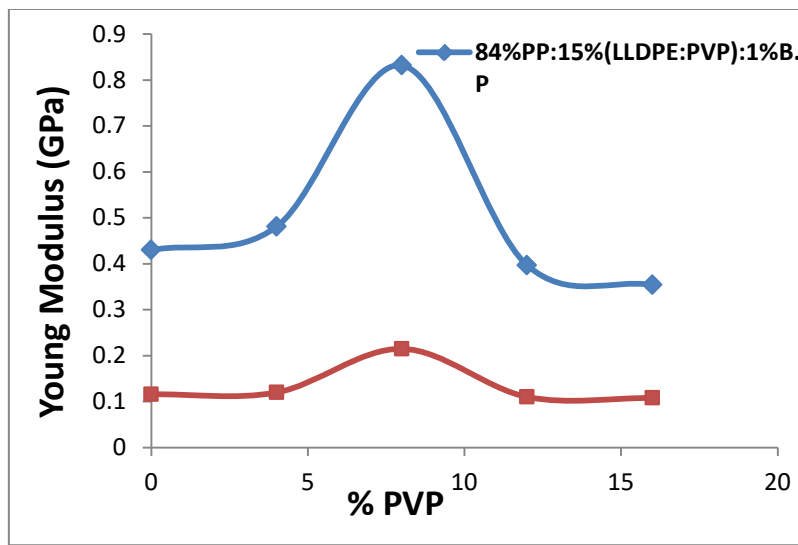X)%LLDPE:X%PVP):1%B.P] as a function of PVP content in the composites.**

**Figure.(5) Young modulus of [84%LLDPE:15%((100-X)%PP:X%PVP:1%B.P]
and [84%PP:15%((100-X)%LLDPE:X%PVP:1%B.P] as a function of PVP con-
tent in the composites.**

Figure. (6a, b) show that the % elongation values of both group polymer blend
composites decrease with increasing PVP content in the polymer blend and the %
elongation values of all samples of the first groups composites (having high ratio of
LLDPE (Fig6a)) are much higher than the values of the second group composites

(having high ratios of PP(fig6b)) which are at the same ratios. That is can be attributed to the molecular structure of is otactic PP which has a methyl group on every second carbon atom of the PP main chain and this restricts motion of the chain under the tensile stress compared with the LLDPE which has a simple linear chain structure with short side branches, the branched chain structure reduces intermolecular bonding force [15,16] so the polymer blend composite prepared with higher ratio of LLDPE produces composites having very high % elongation compared with the one that having low ratio of LLDPE and high ratio of PP.
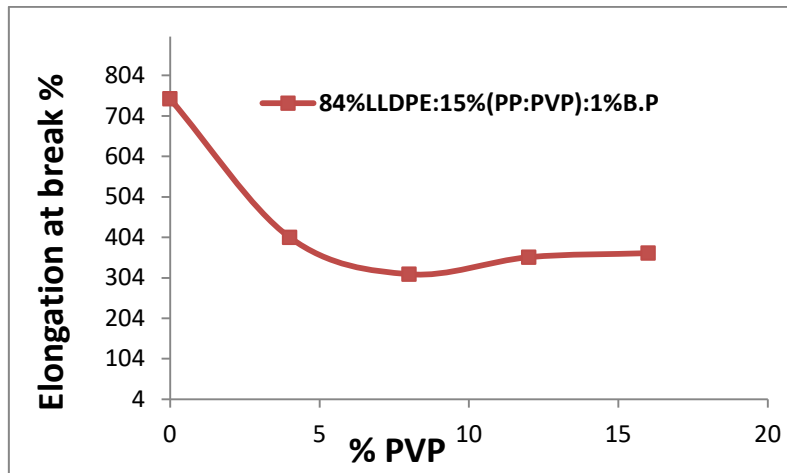

Figure.(6 a) Elongation of [84%LLDPE:15%((100-X)%PP:X%PVP):1%B.P] as a function of PVP content in the composites.
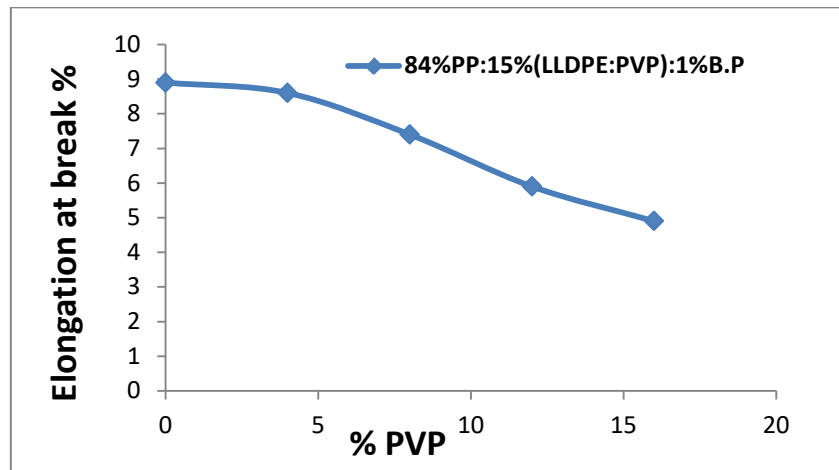

Figure.(6 b) Elongation of [84%PP:15%((100-X)%LLDPE:X%PVP):1%B.P] as a function of PVP content in the composites.

From Figure. (7), it is found that the compressive strength increases with addition of PVP to the basic polymer blend composites (without PVP), until reaches its maximum values at PVP 8%wt (27.4MPa); (61.4MPa) for the two groups of composites respectively. The increase in PVP content in polymer blends composites larger

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than 8% ratio decreases the compressive strength values, and becomes nearly stable at PVP content larger than 12%wt in the two groups polymer blends composites, as well as these values remain larger than the values at the basic composites (without PVP). As mentioned before, it is related to the characteristics of PVP which may act as a good adhesive agent to improve the compatibility between constituents of prepared composites [15]. Also from Figure. (7) it is found that the compressive strength values of the second group composites (having high ratio of PP) are larger than the values of the first group composites (having high ratio of LLDPE) for the same ratio of PVP and that is due to is otactic molecular structure of PP which has a methyl group and the chain structure of LLDPE which has short branch structure and that lowers the strength and stiffness of the first prepared composites [1].
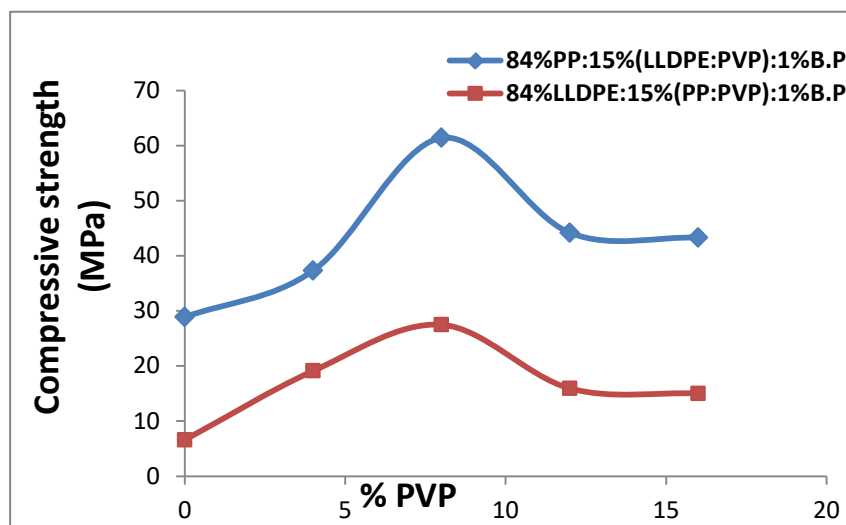

Figure.(7) The compressive strength of [84%LLDPE:15%((100-X)%PP:X%PVP):1%B.P] and [84%PP:15%((100-X)%LLDPE:X%PVP):1%B.P] as a function of PVP content in the composites.

The flexural strength and maximum shear stress values for two groups composites as a function of PVP content are shown in Figs. (8) and (9) respectively. From these figures it is found that slightly increase in flexural stress and maximum shear stress values with the increasing in PVP content in the polymer blend composites. Moreover it can be noticed that there is asymmetry in these values for two groups composites except at 8% PVP ratio. Furthermore the flexural strength and maximum shear stress for the second groups composites specimens have higher values than first group composites, and this may be related to the difference in the molecular chain structure of LLDPE and PP.
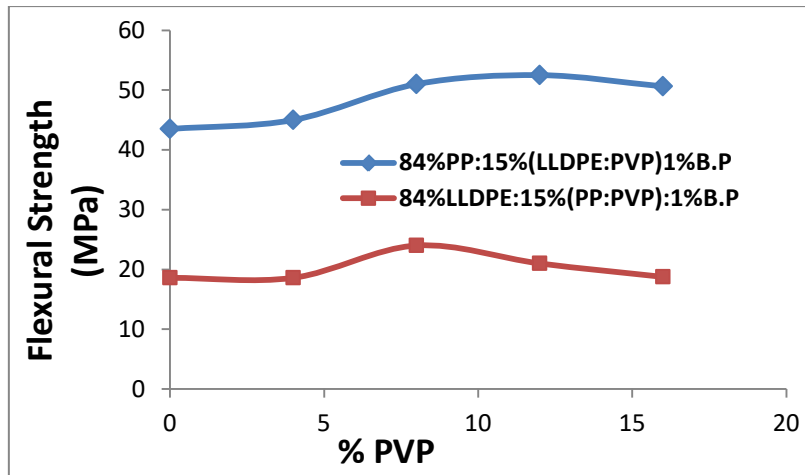
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Figure.(8) Flexural strength of [84%LLDPE:15%((100-X)%PP:X%PVP):1%B.P] and [84%PP:15%((100-X)%LLDPE:X%PVP:1%B.P] as a function of PVP content in the composites.
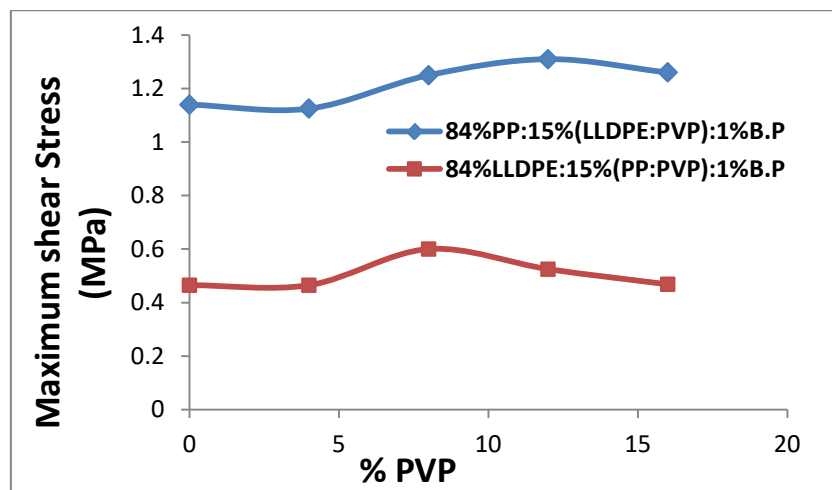

Figure. (9) Maximum shear stress of [84%LLDPE:15%((100-X)%PP:X%PVP):1%B.P] and [84%PP:15%((100-X)%LLDPE:X%PVP):1%B.P] as a function of PVP in the composites.

Fig.(10) shows the effect of PVP ratio on the flexural modulus of both groups of polymer blend composites, which reveals that the addition of PVP in the basic polymer blend composites (without PVP) increases the flexural modulus values of both groups prepared composites, and the rate of increment of flexural modulus increases with increased PVP ratio until reaches its maximum values (1.725GPa) and (5.07GPa) at PVP ratio of 8%wt for both composite groups. Also the rate of increment in flexural modulus values of the second group specimens is larger than the values of the first group at the same ratio of PVP. Increasing PVP ratio to larger than 8%wt decreases the flexural modulus values for the two groups composites. As well as from Fig. (10) it is found that flexural modulus values for the second group speci-

mens [PP:(LLDPE:PVP):B.P] are higher than the values of the first group [LLDPE:(PP:PVP):B.P] at the same ratio of PVP and that is due to the nature of molecular chain structure of PP and LLDPE.
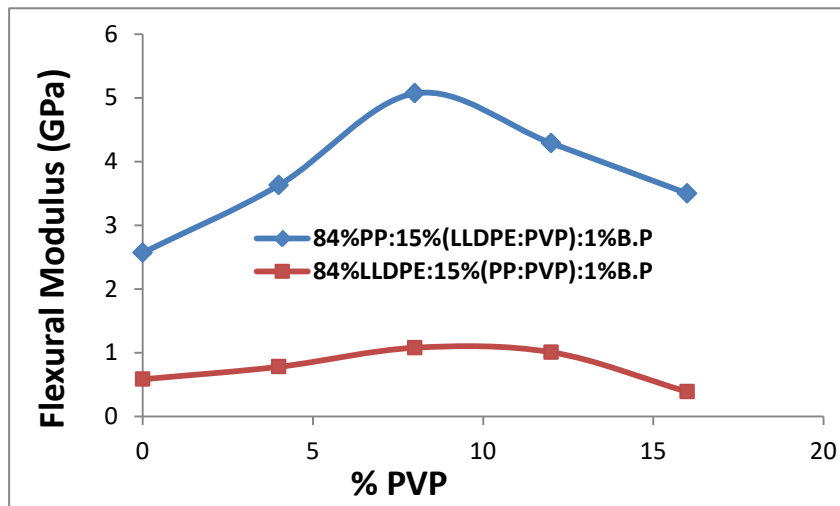

Figure.(10) Flexural modulus of [84%LLDPE:15%((100-X)%PP:X%PVP):1%B.P] and [84%PP:15%((100-X)%LLDPE:X%PVP:1%B.P] as a function of PVP content in the composites.

Figure.(11) and (12) show the effect of PVP ratio on the impact strength and fracture toughness of both groups of composites. The impact strength and fracture toughness for the second group composite [PP:(LLDPE: PVP):B.P] increase with addition of PVP to the basic polymer blend composites (without PVP) and the rate increases with increase in PVP content in the composite and the impact strength value reaches maximum value of (0.45 KJ/m²) at (4%wt PVP), while fracture toughness reach to maximum value of (4.58MPa√m) at (8%wt PVP).Then these values decrease with increase in PVP content to 12%wt after that these values become nearly stable with increased PVP ratio in the composite, whereas the impact strength of the first group in the polymer blend is nearly stable with the addition of PVP to the basic polymer blend composite until reaching 8% PVP after that it slightly increases with increased PVP content in the polymer blend, whereas the fracture toughness values of the first group composite (no break) slightly increase with increased PVP ratio till to 12% PVP ratio then the fracture toughness decreases with increased PVP ratio. Also from Figs. (11) (12) it is found that the impact strength and fracture toughness values for the second group are larger than the first group values at the same PVP ratio, and that is related to the nature of chain structure of LLDPE and PP and the PVP which acts as a good adhesive between the constituents of composite [16,17,18].
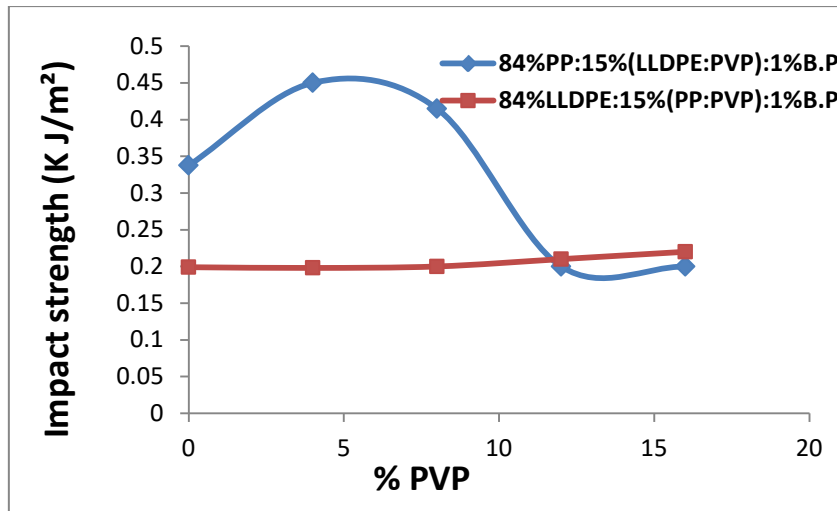
Figure.(11) Impact strength of [84%LLDPE:15%((100-X)%PP:%XPVP):1%B.P] and   [84%PP:15%((100-X)%LLDPE:X%PVP):1%B.P] as a function PVP content in the composites.
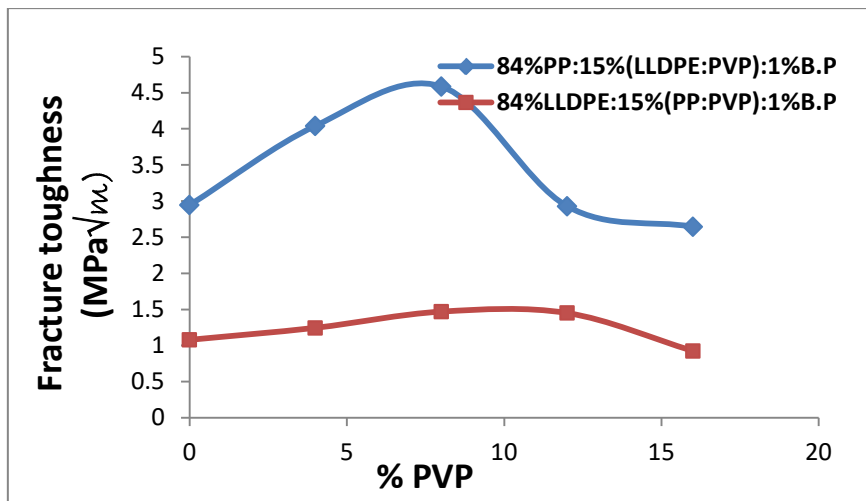

Figure.(12) Fracture toughness of [84%LLDPE:15%((100-X)%PP:X%PVP:1%B.P] and [84%PP:15%((100-X)%LLDPE:X%PVP:1%B.P] as a function of PVP content in the composites.

The effect of addition of PVP on hardness values of both groups of composites is shown in Figure (13). From this figure it is found that the hardness values for both groups of composite slightly increase with the addition of PVP ratio into polymer blend composite, as well as these values lightly increase with increasing in PVP content in both groups composites. Furthermore, it is noticed that hardness values of the second group of composite [84%PP:15%(LLDPE:PVP):1%B.P] are higher than hardness values of [84%LLDPE:15%(PP:PVP):1%B.P] composites due to the difference in the chain structure of PP and LLDPE as has been mentioned previously, so

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PP is stronger and more rigid than LLDPE, therefore the polymer blend composite prepared with higher ratio of PP produces composites having high hardness values compared with the other one having high ratio of LLDPE and lower ratio of PP[1,16].
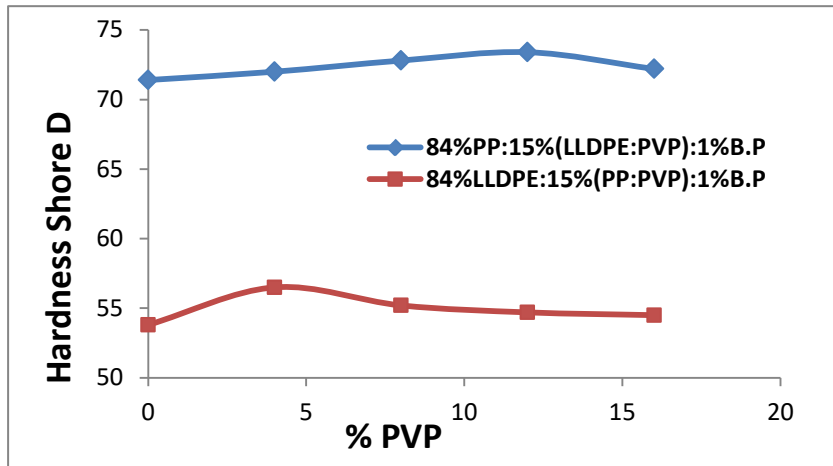

Figure.(13) Hardness Shore D of [84%LLDPE:15%((100-X)%PP:X%PVP):1%B.P] and [84%PP:15%((100-X)%LLDPE:X%PVP:1%B.P] as a function of PVP content in the composites.

The major difference between plastics material and the more traditional materials is the time dependent behavior of polymers. Figs. (14) and (15) show (strain-time) curves of both groups of polymer blend composites respectively. These figures indicate that the creep rates at room temperature and at constant tensile stress increase rapidly with the time for all prepared samples, but after six hours the creep rates slightly increase with time, and it can also be observed from these figures that the creep rates depend on the PVP content as well as on the constituent of composites.
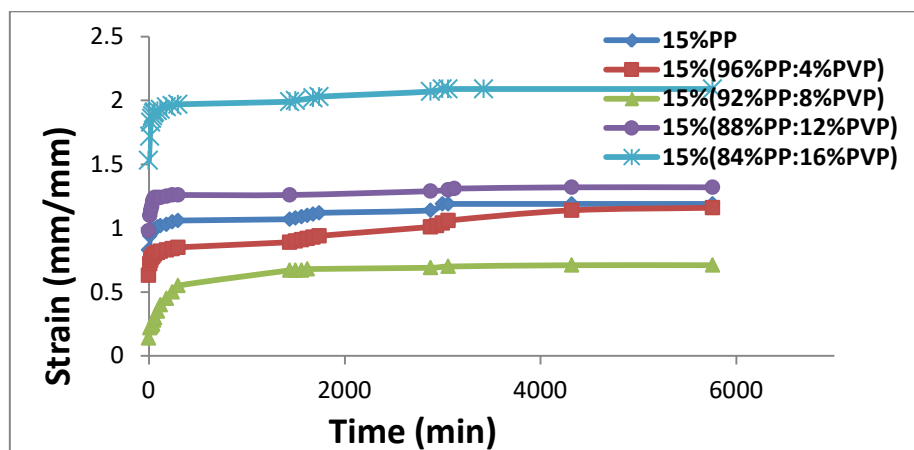

Figure.(14) Creep behavior of [84%LLDPE:15%((100-X)%PP:X%PVP):1%B.P] as a function of PVP content in the composites.
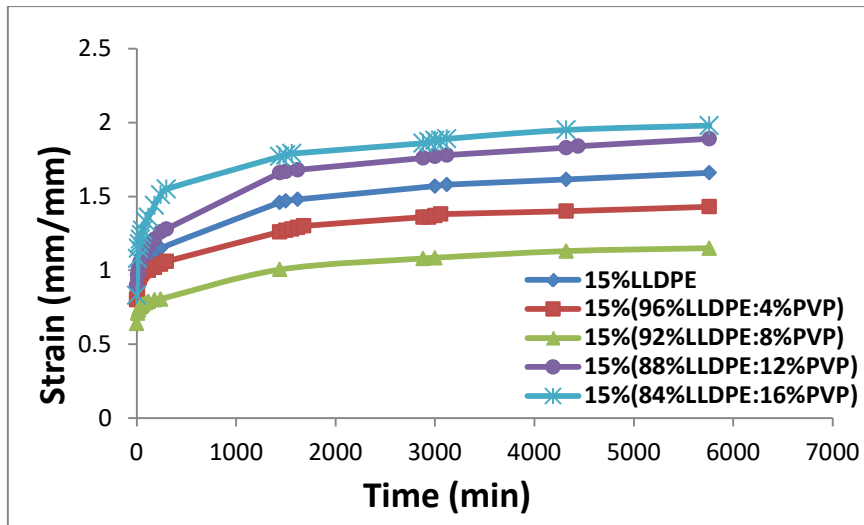
Figure.(15) Creep behavior of [84%PP:15%((100-X)%LLDPE:X%PVP):1%B.P] as a function of PVP content in composites.

The effects of PVP content on the creep modulus (the ratio of the initial applied stress to the creep strain ϵ (t) after a particular time and at constant temperature testing [16]) of two composites groups for different creep times of (240min, 2880 min and 5760 min) and at room temperature are shown in Figs. (16 and 17) respectively.
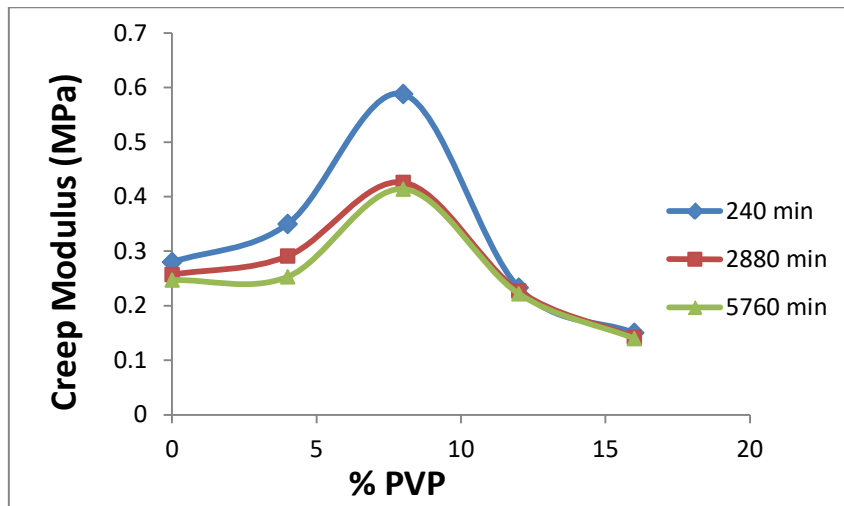

Figure.(16) Creep modulus of [84%LLDPE:15%((100-X)%PP:X%PVP:1%B.P]
as a function of PVP content in the composites.

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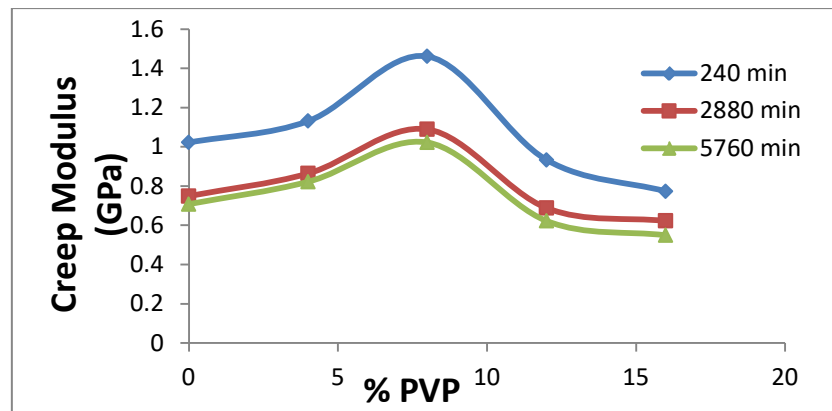

Figure.(17) Creep modulus of [84%PP:15%((100-X)%LLDPE:X%PVP):1%B.P] as a function of PVP content in the composites.

These figures illustrate that the creep modulus increases with addition of PVP to the composites for all creep times and for both groups and reach maximum values at 8%wt PVP and at creep time 240 min for two groups of composites. The maximum values of creep modulus for first and second group composites were (0.588GPa) and (1.46GPa) respectively. The high creep modulus values of polymer blend composite imply low creep rate as shown in Figures (14) and (15).These values decrease rapidly with the increased PVP ratio in the polymer blend to reach less than its values in the basic composites (without PVP) for the same creep time. And that is related as mentioned before to the characteristics of PVP which may causes (when it is in small ratio) good compatibility between polymer blend composite constituents [16].

From Fig. (18) it is found that the creep modulus values of the second group composites have larger values as compared to the first group of composite samples at the same ratio of PVP, this is related to the difference in molecular chain of LLDPE and PP as mentioned before.
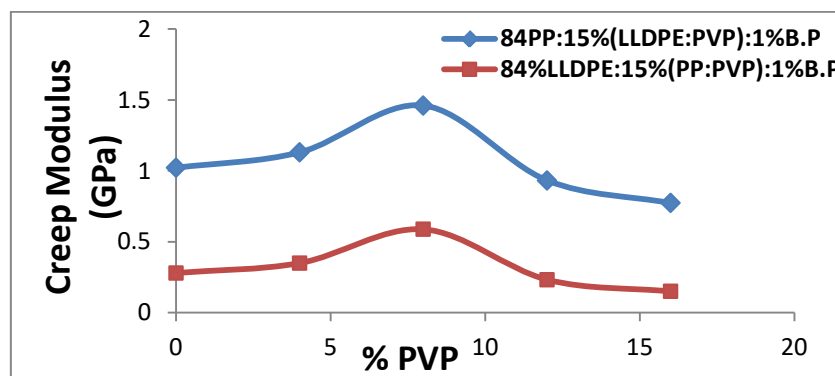

Figure.(18) Creep modulus at (240 min) of [84%LLDPE:15%(PP:PVP):1%B.P] and [84%PP:15%(LLDPE:PVP):1%B.P] as a function of PVP content in the composites.

CONCLUSIONS

The mechanical properties of two groupsternary Polymer blend composites(84%LLDPE:15%(PP:PVP):1%B.P.)and (84%PP:15%(LLDPE:PVP):1%B.P.), were investigated as a function of PVP ratio. The following conclusion can drawn from those study:-

1-The mechanical properties (fracture strength, young modulus, compressive strength, flexural modulus and creep modulus for two groups composites increase with increasing the ratio of PVP to reach highest values at 8% PVP ratio, then decrease slightly with increase PVP content, Whereas the elongation rates decreases with increasing PVP ratio in the blend.

2-The characteristics of PVP may be act as a good adhesive agent to improve the compatibility between constituents of prepared compositesespecially at slight ratio of PVP content.

3-The test values of mechanical properties for the second group composites are higher than the values of the first group composites and the highest values of fracture strength, flexural strength, impact strength and fracture toughness reach to 34.6MPa, 52.5MPa, 0.45KJ/m2 and 4.5869MPa$\sqrt{m}$ respectively.

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