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PROCESSABILITY AND DETERMINATION OF SOME MECHANICAL AND THERMAL PROSPERITIES OF FILLED AND UNFILLED POLYPROPYLENE / POLYAMIDE 6 BLEND

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ABSTRACT: - A new type of Bentonite filled PP/PA6 and red Kaolin filled PP/PA6 blends has been developed. It is Polypropylene and Polyamide 6 at constant ratio (80/20) and different weight fraction (0, 5, 10, and 15) % of both local Bentonite and Red Kaolin fillers were added respectively. Filled polymer blends were developed on a single screw extruder. Hardness, compression impact strength, and thermal stability of BN/PP/PA6 and RK /PP/PA6 blend system were determined at different temperatures, and different weight fraction of filler. The results shown hardness and compression increase while impact strength decrease with increase in weight fraction content. Also the results shown that thermal stability increases with increased weight fraction of filler. Bentonite filler produces better mechanical properties, than Red Kaolin fillers. Empirical equations are proposed and show a best fit with experimental data. Relevant contour diagrams, based on the proposed equations, for optimization of properties were also presented.

Keywords: - PP, PA6, Blend, Bentonite, Red Kaolin, Mechanical, Thermal properties.

INTRODUCTION

Polymer blends are generally considered by mixing two or more polymers that are not bonded to each other. Two or more polymers are mixed to produce blends is a well-established route to achieve a certain amount of physical properties, without need to synthesize specialized polymer systems (1).

By adjusting the composition of the polymer blend, it is possible to obtain a material possessing the desired mechanical and thermal properties. As well as a relative easy way to improve the mechanical properties of a polymer is the addition of filler materials. In general, inorganic fillers are applied to improve the stiffness of a polymer (2).

Rattana studied the reinforcement of natural rubber latex with nano size montmorillonite clay. The resulting film had greatly improved mechanical properties compared with films using micron-sized fillers. Furthermore, both modulus and toughness were improved; in many composite systems an improvement in modulus leads to a loss of toughness. X-ray diffraction results indicated that clay platelets dispersed in the rubber matrix on the nanoscale level with some macromolecules intercalated into the clay gallery. The observed considerable improvement in mechanical properties, coupled with a theoretical model of composite modulus suggest a dispersed structure of clay in the composite, while not all clay particles are exfoliated (3).

Abed-Alhakem studied the effect of Iraqi raw ceramic (Kaolin and Boxide) on epoxy resin, he showed that tensile properties (elongation at break, young modules, and stress at break) of composites filled with relatively small particles all increased with increasing filler content, with decreasing filler size, and increasing filler content(4). Hashmi studied rheological, and mechanical properties of polymer composite (red mud filled PP/PC and PP/Nylon-6 blend systems), he showed that addition of RM particles to PP/ Nylon-6 blend increases the discontinuity and restricts the growth of crystal, and therefore crystalline peak intensity reduced in the field blend composites. A new type of cheaper blend is developed by melt mixing technique and incorporation of red mud modifies the PP/ Nylon-6 structure by interlocking the chains on the surface and composite. An increase in red mud content raises the tensile strength of the PP/ PC blend (5). In this work a new type of composite has been developed composed of Polypropylene and Polyamide 6 blend at constant ratio (80/20) and adding different weight fraction (0, 5, 10, 15) % of both Bentonite (6) and Red Kaolin fillers added respectively, by using a single screw extruder. Mechanical properties: hardness, compression and impact strength were measured, thermal stability was also determined. Empirical correlation was found.

EXPERIMENT MATERIALS

Commercial polypropylene (Sabic, KSA) its melt flow index and the density of the material were 11gm/10min and 0.908 (gm/cm^3) respectively, and Commercial polyamide-6 (EXXON-MOBIL KSA) the melt flow index and the density of the material were

2.0gm/10min and 1.14 (gm/cm^3) respectively has been used without further treatment.

Red kaolin clay (RK) with particle size of 45 μm and density (2.64) gm/cm^3 and has high melting point (1755) °C. Its chemical composition is shown in table (1). Bentonite clay (BN) with particle size of 45 μm and density (2.55) gm/cm^3. Its chemical composition is shown in table (2). All the materials were used without any treatment.

COMPOUNDING

Polypropylene (PP), (PA6), (RK), and (BN) samples were kept in an air circulation oven at 70°C for 4 hr. to dry the material. Weighted amounts of polymers were mechanically mixed and fed to 25 mm single screw extruder machine (Betol BM 1820 extruder). The barrel as well as the die temperature was monitored and controlled by a thermostat, and was adjusted, to yield uniform output, of the polymer blend .Feed compression and metering zone temperature are shown in table (3).Polymer blend of PP and PA6 (80/20) were mixed with various compositions of filler (0, 5, 10, 15) % .and re-extruder again on the same machine table (3). The extrudes produced in the form of monofilaments of about 2 mm in diameter were cooled in water and cut in 3-4 mm length.

MOLDING PROCEDURE

The CININATI hydraulic press was used with maximum load (15ton) and working area (0.4*0.3) m, for compression molding. The mold used for pressing the composite material has dimensions of (30*30*4) mm. It is made of steel. The sheet was prepared by hot pressing at 150°c for PP/PA6/RK and PP/PA6/BN.A pressure of 20 kg/cm^2 was applied for 5 min to allow the composite to melt and spread out between plates. Pressure was then increased to 100 kg /cm^2 for further 5min.The compression molding parameters are listed in table (4). The pressure was removed and the mold sheet was quenched in water at room temperature.

MECHANICAL PROPERTIES

Charpy impact instrument was used in this test. Samples were prepared according to ASTM (D256-87) (7), and tests at different temperatures (25, 50, 75, and 100) °C.

Brinell method was used to determine the hardness of the composite materials using Le Bold Harris Hydraulic Press type 36110 .The measurements were carried out according to the ASTM (D638) (7). Hydraulic piston type Leybold Harris No. 36110 was used to measure the compressive strength of prepared samples at different temperatures (25, 50, 75, and 100) °c.

The test was carried out according to ASTM (D695) (7). The test specimens had the following dimensions: the length of specimen was double its width in the ratio of 2:1; the width of the sample was equal to its thickness.

Thermal analysis of samples was done on (T.G.A), samples of 6-8 mg were heated from room temperature to 600^0C. Weight loss was measured by using TG 760 SERIES STANTON Redcroft (1983) in which the weight loss was measured continuously while the temperature was increased from 25°C to 600°C at constant rate of 10°C/min (8-9).

RESULTS AND DISCUSSION

From Figs (1) and (2) one can see that the hardness of PP/PA6 increases after reinforcement with both BN and RK filler, since the test measures a plastic deformation of material under effect of external stress. The additions of BN and RK filler increase the hardness of composite material due to increase in the resistance strength of polymer to plastic deformation. These results agree well with results obtained by Abdl-hakem (4). Under the effect of temperature the hardness decreases with temperatures increase and this may be due to the matrix, which starts to change from the solid state to the plastic state. Our results agree well with those results obtained by Mohammed, and Allaf (10, 11). From the Fig (3) it is clear that the hardness of composite material made of PP/PA6/BN is higher than that of the composite material made of PP/PA6/RK, this is due to the hardness which depends on some factors such as type of the binding force between the molecules, type of surface, temperature and other effective conditions (12,13). From Figs (4) and (5) it is clearly seen that the compression strength increases with increasing weight fraction of BN and RK filler. This behavior may be due to high resistance to compression of BN and RK filler materials. The latter has a good ability to absorb the load applied by matrix PP/PA6 and increase the resistance of matrix composed of PP/PA6 and also that will increase the binding force between the molecules of matrix with filler. Also it is seen that with the increase in temperature, compression strength of composite will decrease and this may be due to the fact that temperature will soften the matrix and consequently decrease elastic modulus and compression strength. Fig (6) shows that the compression strength of composite material made of PP/PA6/BN is higher than that of the composite material made of PP/PA6/RK because BN is harder and more rigid than RK filler and also BN share the external stress with matrix better than RK(5).

From the experimental work it was observed that no failure occurs, increasing the compression stress leads to flattening the specimens, therefore, there is no compression strength at failure to be observed. Figs (7) and (8) show the relation between fracture energy of PP/PA6 and weight fraction of filler both for BN and RK respectively at different temperatures (25, 50, 75, and 100) °C.

Impact strength decreases with increase in weight fraction of filler. This decrease in impact strength is related to increase in the fracture energy of composite which is caused by the great number of parameters affecting its mechanical behavior such as the nature of reinforcing particles, the weight fraction of filler and the adhesion between the particles filler and matrix (14-15). Also From these Figs, one can see that fracture energy increases with increasing temperature cause the polymer to reach the glass transition temperature at which the polymer changes from solid material with low toughness to plastic material that has high toughness (16). These results agree well with results obtained by Allaf, and Al-Neamee (10, 17), they show in their research on composite material that the fracture energy increases with increasing temperature. When the temperature increases the impact energy is increased and this is related to the decrease in softening of matrix chain between the molecules of the material which causes movement of the matrix molecules and that cause the material to absorb some of the energy leading, to increase in the energy required for fracture.

THE ANALYSIS OF EMPIRICAL CORRELATION

The experimental results of this work are used to develop an empirical correlation. The Statistical program was used on high – speed personal computer (Pentium 4). The method of developing the present model is by introducing equations of different forms in the computer program. The calculated values of the dependent variables are compared with the actual values and the procedure is repeated until excellent agreement is obtained.

The dependent and independent variables were introduced into the computer program in such a way that the developed models could be used for the composite material to find the change in mechanical properties, The independent variables, which affect the mechanical properties of the PP/PA6/RK and PP/PA6/BN blends composite, are weight percent of filler content and the temperature. Equations (1) and (2) as shown in table (5) represent the developed models for the Brinell hardness and impact strength with weight percent of filler content and temperature respectively. A three-dimensional plot of mechanical properties (the Brinell hardness and impact strength) vs. (T) and (W) using Eqs. (1), and (2) was made for the data which are shown in Fig (11) for PP/PA6/BN and for PP/PA6/RK and these figs approximate the behavior of the system with reasonable accuracy for the values of weight present of filler (W) and temperature (T) in this studied range. The square term in the equation plots indicates a curvature in the surface obtained in three – dimensions. Also the results can be presented in terms of contour plots as shown in Figs (12) for PP/PA6/BN and (13) for PP/PA6/RK. The practical use of such contour plots is that, given the value of the property (which needs not be any experimental data points itself), the temperature and weight percent of filler content needed to attain that value of properties can be directly obtained from the plots. This procedure optimizes the value of the variables needed to get any desired value of the properties.

THERMAL STABILITY

Figs (16) and (17) show the effect of addition of both types of particle filler BN and RK respectively on thermal stability of PP/PA6 blend. An increase in filler content leads to increase in thermal stability of the composite. This may be due to the structure of the clay that makes it more thermally stable than red mud. On the other hand Fig (16) shows that PP/PA6 blend starts to decompose at a temperature of 280 °C and the residual weight is (4%) at temperature of 410 °C after the addition BN filler with different content (5, 10, 15) wf% to PP/PA6 blend composite. The initial degradation temperatures were (290,300,310) °C respectively and the residual weights were (10, 17, 23) % at a temperatures of (470,480,490) °C respectively. Fig (17) shows that the addition of RK filler at different contents (5, 10, 15) wt% to PP/PA6 blend, the initial degradation temperatures were (285, 290, 290) °C respectively and the residual weights were (8, 14, 20) % respectively at temperatures of (450, 460, 470) °c respectively. These results agree well with results obtained by Hashmi (5), he obtained that increase in Red mud content raises the thermal stability of PP/Nylon6 blend. From the above-mentioned results, it is clear that material composite PP/PA6/ BN has higher thermal stability than that of material composite PP/PA6/ RK witch may be due to the differences in behavior between BN and clay that stem from their composition and structure (18).

CONCLUSIONS

After reinforcement of PP/PA6 blend with BN or RK as filler, hardness, compression and thermal stability increase with increase the filler fraction while impact strength was observed to decrease with increase in the filler fraction. The thermal stability of PP/PA-6/BN is higher than that of PP/PA6/RK. Mechanical property can be represented as simulation function of two variables (weight fraction of filler content and temperature) by a second -

degree polynomial equations, and these equations show that the best fit with experimental data. Relevant show contour diagrams based on the proposed equation, of optimization of properties are also presented.

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Table (1): Chemical composition of Red kaolin clay (6).

SIO_2 %	AL_2O_3 %	Fe_2O_3 %	TiO_2 %	CIO %	MgO %	Na_2O %	K_2O_3 %	L.O.I %
52.48	31.31	2.94	1.43	0.462	0.33	0.28	0.23	10.93

Table (2): Chemical composition of Bentonite clay (6).

SIO_2 %	AL_2O 3%	Fe_2O_3 %	CaO %	MgO %	Na_2O %	K_2O %	P_2O_5 %	SO_3 %	CL %	L.O.I • %
56.77	15.67	5.12	4.48	3.42	1.11	0.60	0.65	0.59	0.57	9.49

Table (3): Extrusion parameter.

Polymer	**Temperature °C**				**Screw speed (RPM)**
	Zone1	**Zone2**	**Zone3**	**Zone4**	
PP/PA6	150	170	220	200	30
PP/PA6/RK	155	190	250	220	30
PP/PA6/BN	155	190	260	230	30

Table (4): Compression molding parameters

Temperature ºC		**Pressure**	**Time**
Upper plate	**lower plate**	**(Kg/Cm^2)**	**(min)**
150	150	20	5
200	200	100	5

Table (5): Equations of the mechanical properties as a function of temperature and weight percent of filler content both for BN and RK.

Equations	**Correlation Coefficient BN**	**Correlation Coefficient RK**
Eq1 : $H = a_o + a_1 T + a_2 W + a_3 T W + a_4 W^2 + a_5 T^2$	0.998	0.992
Eq2 : $I = a_o + a_1 T + a_2 W + a_3 W^2 + a_4 T W + a_5 T^2$	0.999	0.997

H: Hardness I: Impact T: Temperature W: Weight Percent of Filler

Table (6): Coefficient for the equation (1) and (2) both for BN and RK.

a	**Equations BN.**		**Equations RK.**	
	(1)	**(2)**	**(1)**	**(2)**
a_o	13.483	2.456	13.571	1.693
a_1	0.588	2.75e-4	0.336	-0.192
a_2	-0.008	0.063	-0.012	0.089
a_3	-5.104e-4	-0.001	-0.001	0.005
a_4	-0.004	7.75e-4	0.004	3.799e-4
a_5	-4e-5	-8.9e-5	-8e-6	-2.743e-4

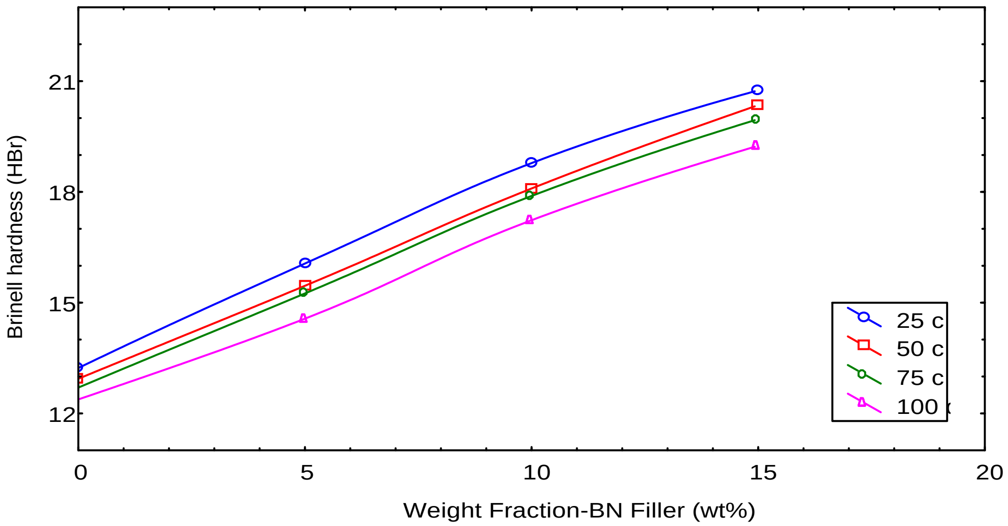

Fig. (1): The relation between Brinell Hardness and Weight fraction of PP/PA6/ BN at different temperature (25, 50, 75,100) °C.

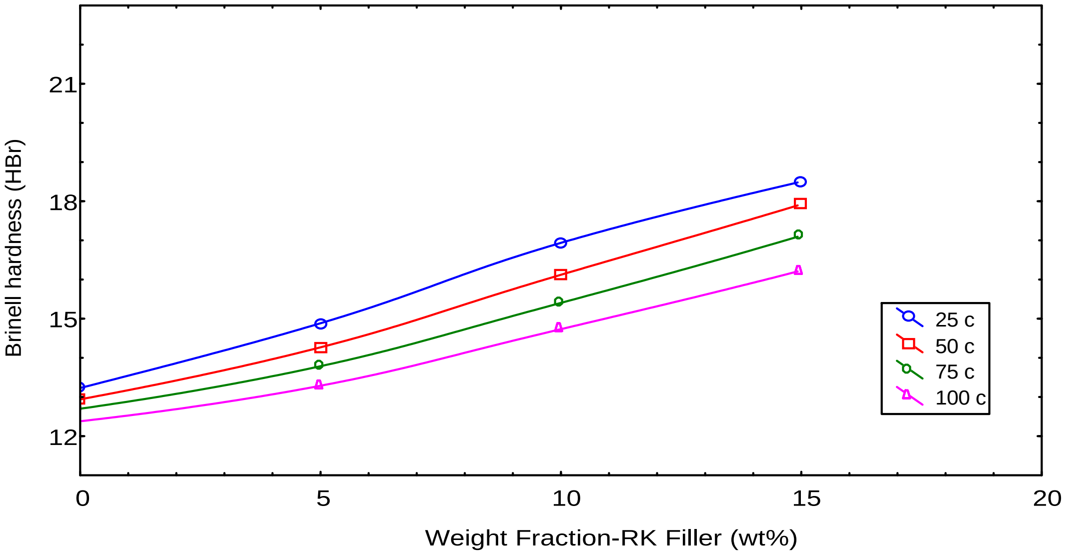

Fig. (2): The relation between Brinell Hardness and Weight fraction of PP/PA6/RK at different temperature (25, 50, 75,100) °C.

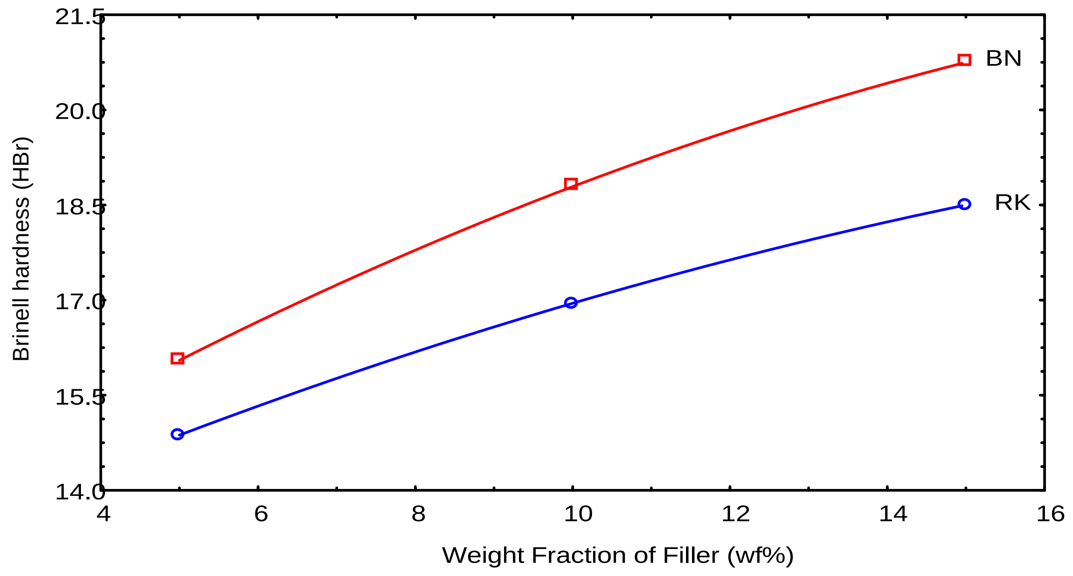

Fig. (3): The relation between Brinell Hardness of PP/PA6 with weight fraction of both RK and BN filler at temperature (25) °C

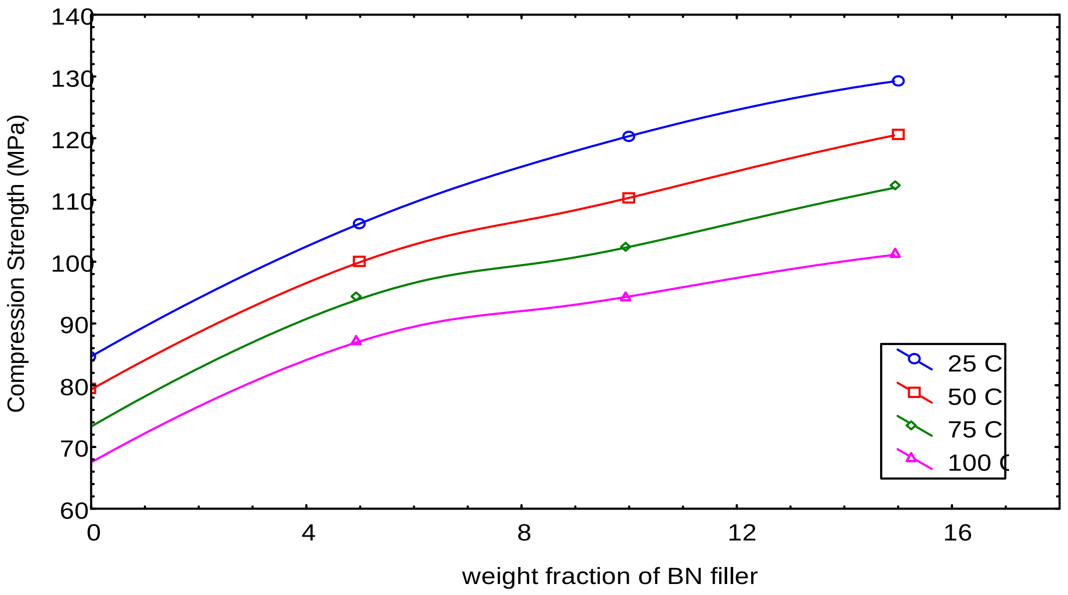

Fig. (4): The relation between compression strength and Weight fraction of PP/PA6/ BN at different temperature (25, 50, 75, 100) °C.

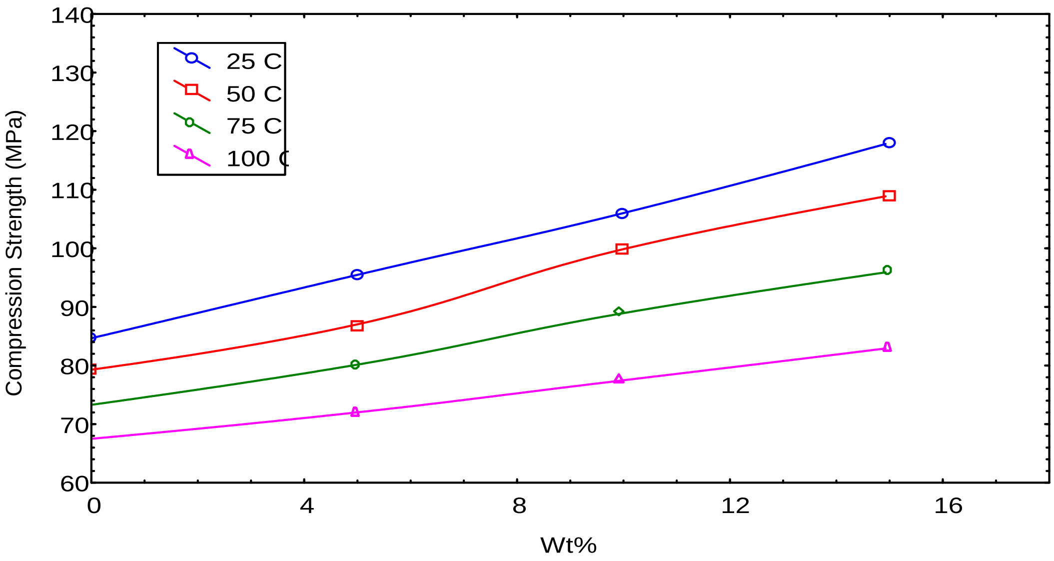

Fig. (5): The relation between compression strength and Weight fraction of PP/PA6/ RK at different temperature (25, 50, 75,100) °C.

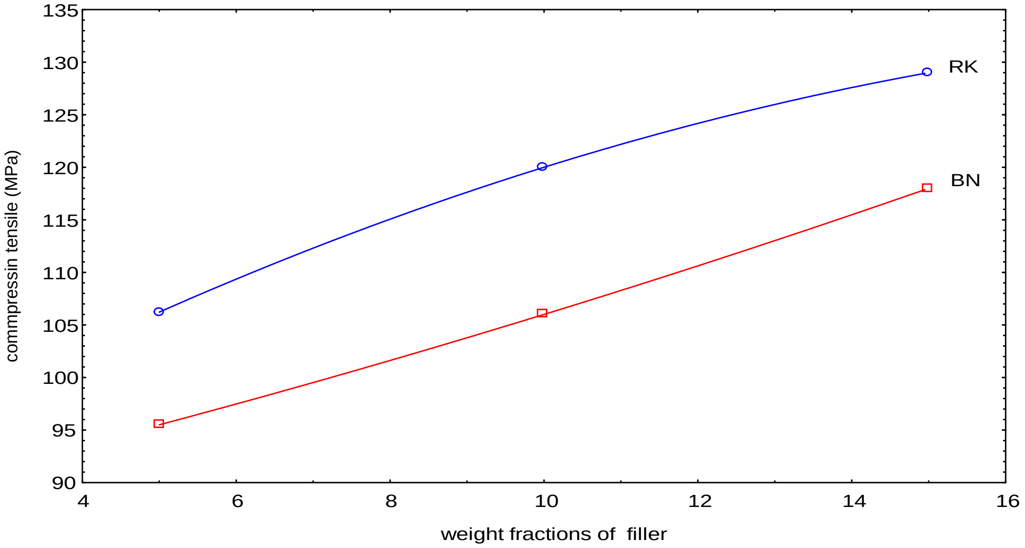

Fig. (6): The relation between compression strength of PP/PA6 with weight fraction of both RK and BN filler at temperature (25) °C

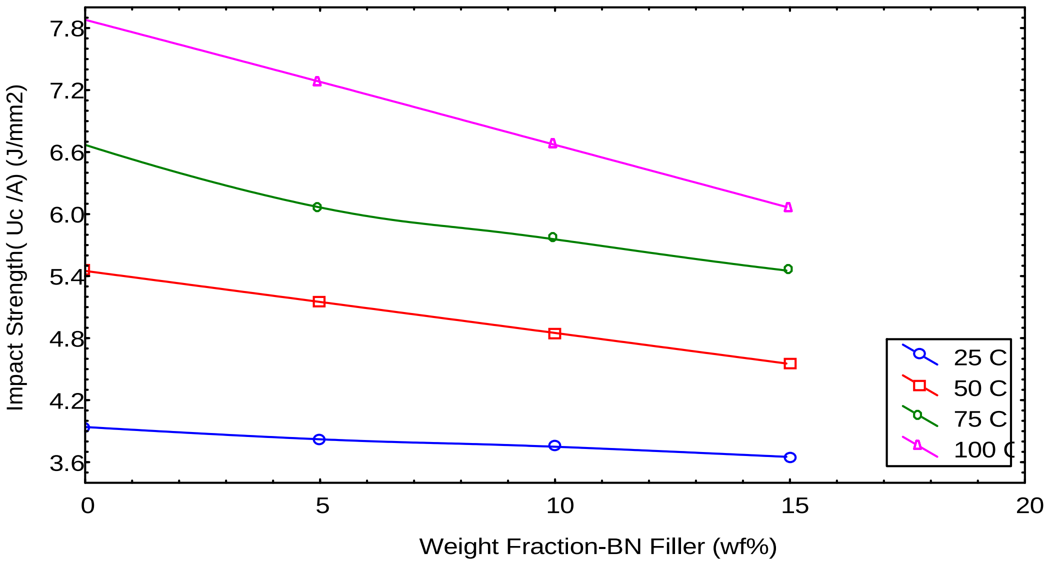

Fig. (7): The relation between impact strength and Weight fraction of PP/PA6/ BN at different temperature (25, 50, 75,100) ºC.

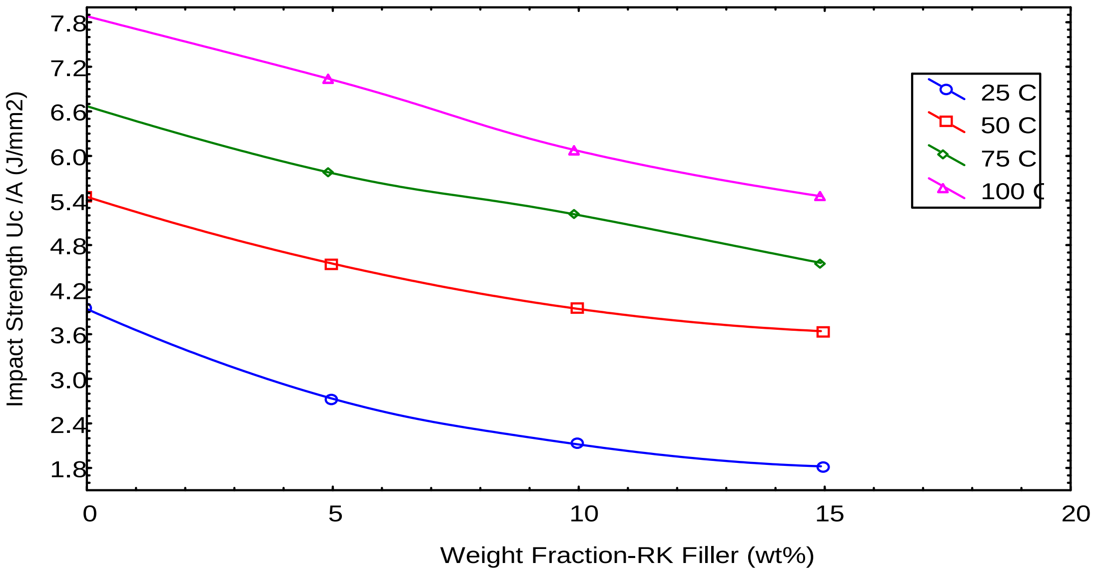

Fig. (8): The relation between impact strength and Weight fraction of PP/PA6/ RK at different temperature (25, 50, 75, 100) ºC.

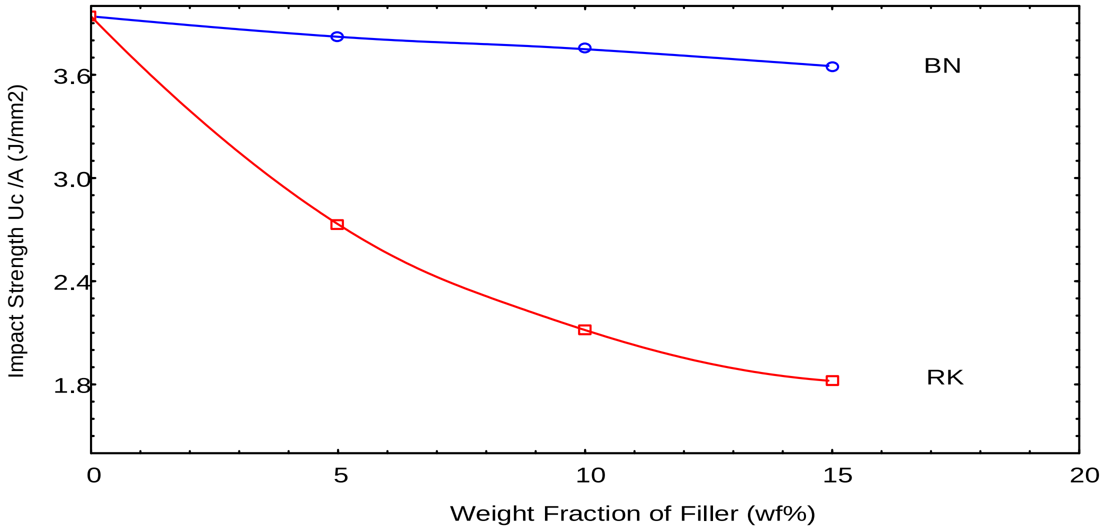

Fig. (9): The relation between impact strength of PP/PA6 with weight fraction of both RK and BN filler at temperature (25) °C.

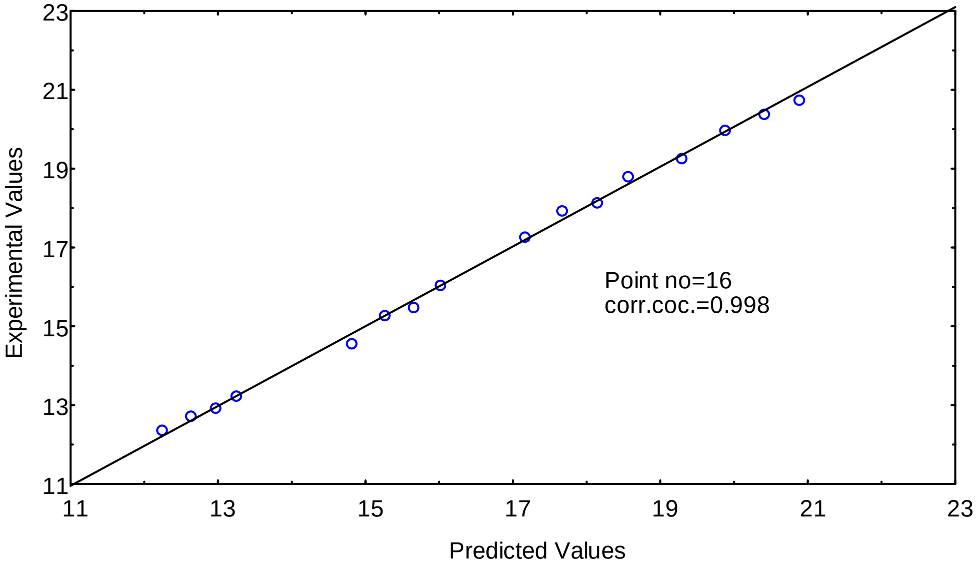

Fig. (10): Experimental versus predicted values for Eq. (1)

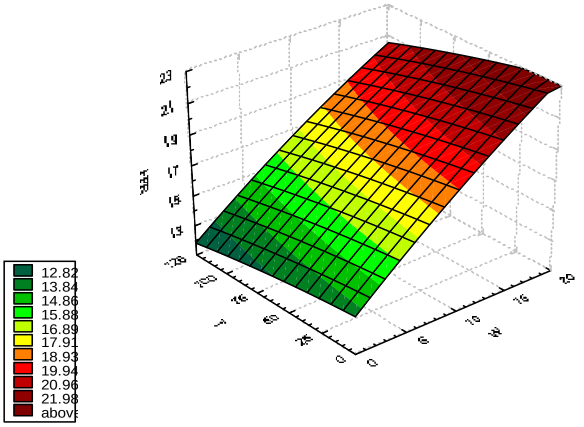

Fig. (11): Three-dimensional representation of Eq. (1) describing the variation of Brinell Hardness, with weight % of filler content BN and temp. for PP/PA6/BN.

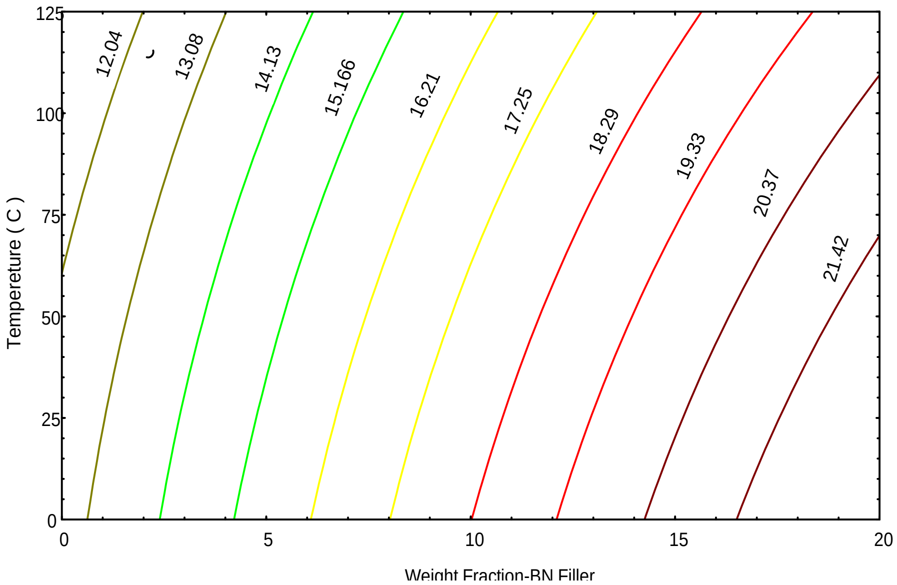

Fig. (12): Contour plot obtained from the Three-dimensional plot Fig. (11).

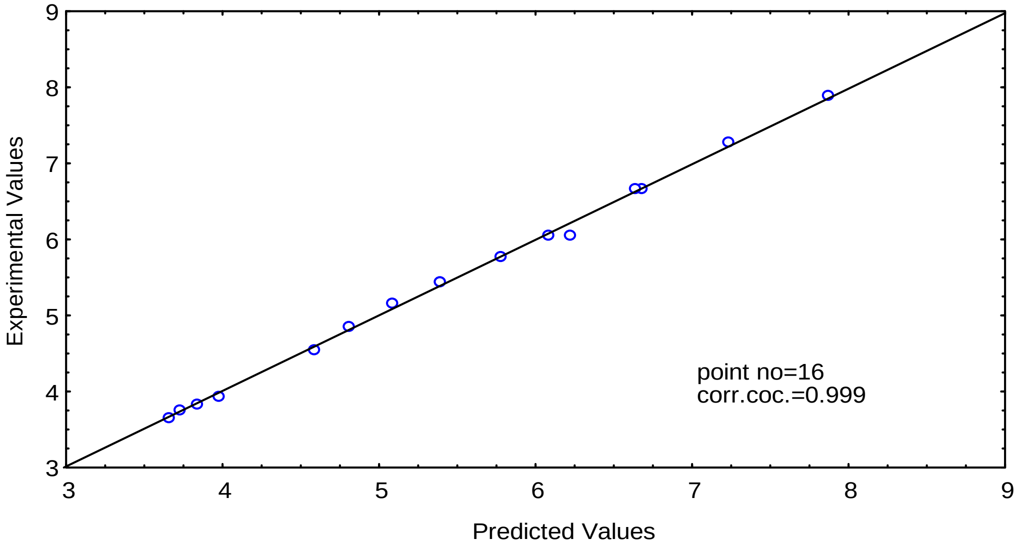

Fig. (13): Experimental versus predicted values for Eq. (2)

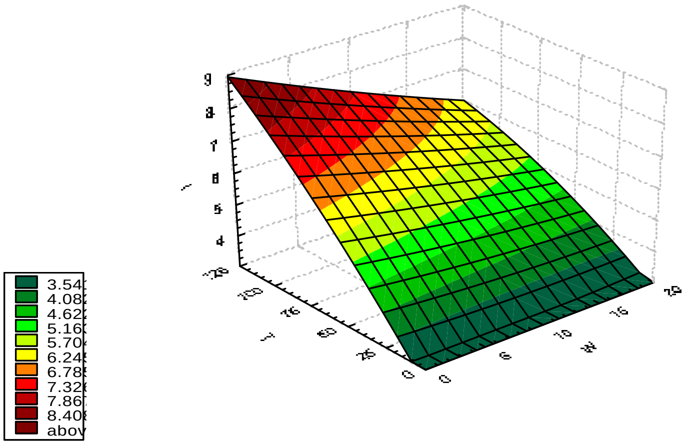

Fig. (14): Three-dimensional representation of Eq. (2) describing the variation of impact, with weight percent of filler content RK and the temperature for PP/ PA6/ RK.

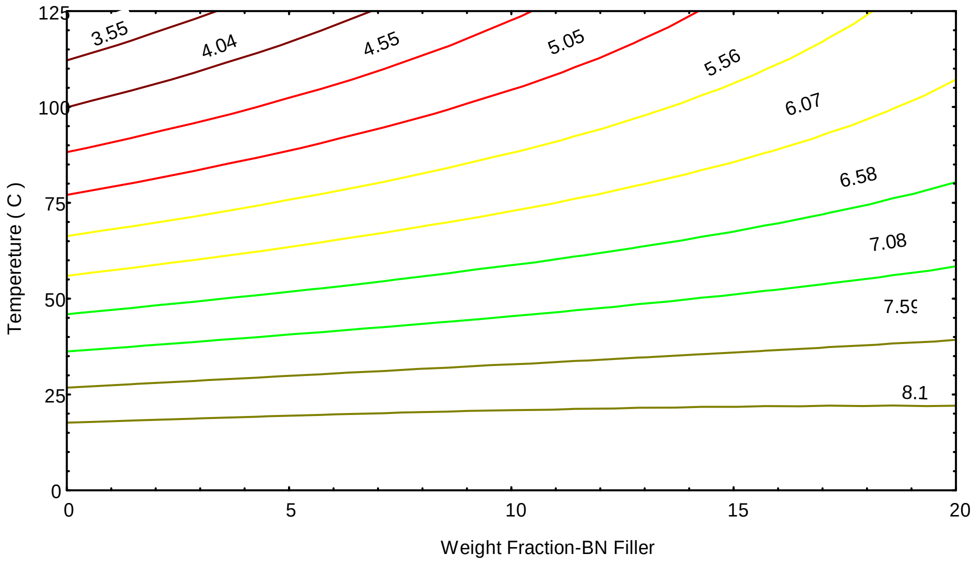

Fig. (15): Contour plot obtained from the Three-dimensional plot Fig. (14).

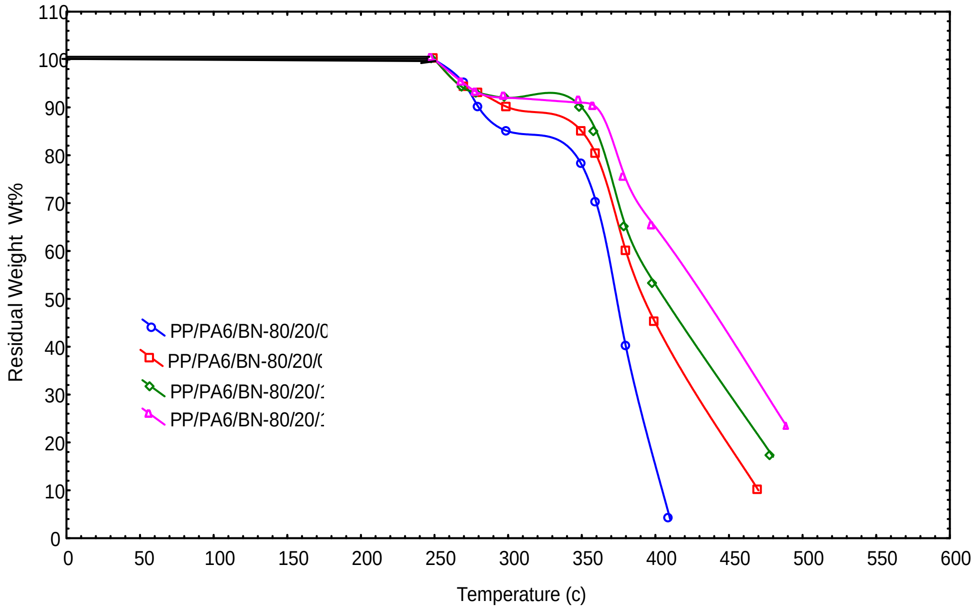

Fig. (16): Effect of addition of RK filler on thermal stability of PP/PA6 blend.

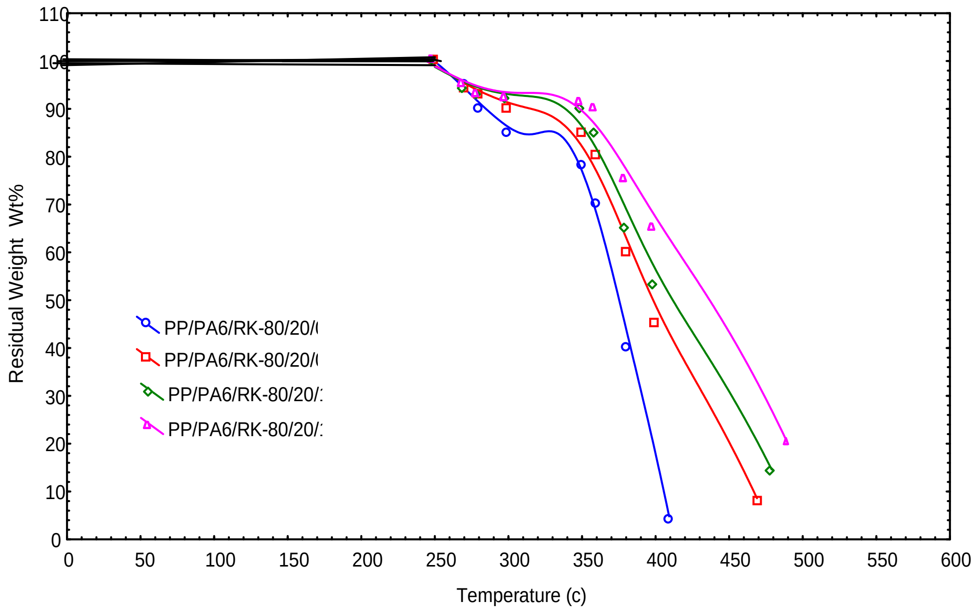

Fig. (17): Effect of addition of BN filler on thermal stability of PP/PA6 blend.

الامكانية التصنيعية وتحديد بعض المواصفات الميكانيكية والحراريه للمخلوط البوليمري بولي بروبلين/ بولي امايد6 المحشو وغير المحشو

نجاه جمعه صالح
استاذ مساعد
قسم الهندسه الكيمياوية
الجامعة التكنولوجية

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المعهد الطبي التقني/ بغداد

الخلاصة

في البحث الحالي تم تحضير مواد مركبة جديده التي هي عبارة عن مخلوط بوليمري يتكون من مزج بولي بروبلين مع بولي أميد 6-بنسبة وزنية ثابتة (20/80) مع اضافة نسب الوزنية مختلفه من مسحوقي البنتونايت والكاؤولين الأحمر المحلي على التوالي وذلك باستخدام جهاز الباثقة المنفرده.جرت دراسة الصلادة ، الانضغاطية، وقوه الصدمة والاستقراريه الحرارية لجميع المتراكبات المحضرة وقورنت النتائج المستحصلة مع تلك التي تعود لمادة الأساس بمفرده وجرت الفحوصات بدرجات حرارية مختلفة وكسور وزنية مختلفة. بينت النتائج التي تم الحصول عليها إلى زيادة كل من الصلادة، الانضغاطية والنقصان في مقاومة الصدمة بزيادة نسبة الخلط الوزني ومن ناحية أخرى تم اجراء فحص الثبات الحراري ووجد ان ازدياد الكسر الوزني للحشوة أدى إلى زيادة الثبات الحراري للمواد المتراكبة، توصلت الدراسة ان البنتونايت أعطت نتائج أفضل من الكاؤولين الأحمر، وتم استنباط المعادلات التجريبية التى تظهر العلاقة المتطابقة مع النتائج النظرية والعملية وتم رسم منحنى عدادي معتمدة على المعادلات المفترضة.