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EFFECT OF GRAPHITE PARTICLES ADDITION AND PROCESS PARAMETERS ON THE MECHANICAL PROPERTIES OF POLYETHYLENE/GRAPHITE SURFACE COMPOSITE FABRICATED BY FRICTION STIR PROCESSING

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Abstract: Friction stir processing (FSP) is a novel solid state technique; it is employed for the improvement of the mechanical properties of a material and the production of surface layer composites instead of conventional processing technologies. This research aims to study the ability of applying Friction Stir Processing (FSP) on thermoplastic polymer in the form of high density polyethylene (HDPE) reinforcing by graphite particles with particle size of 25µm, filled in a groove of 1.5 mm depth on PE substrate. Friction stir process was carried out, using tool with scrolled shoulder to produce surface layer composite. The effect of processing parameters including rotational and transverse speeds on the mechanical properties of composite layer was studied. Wear test results showed a pronounced improvement in wear resistance of HDPE surface through reinforcement additions of graphite particles, where wear rate decreased by 70%, as compared with as received PE. The hardness of composite layer increased 18% higher than the as received material. OM revealed that high tool rotational speed resulted in homogeneous distribution of graphite particles and vice versa.

Keyword: *Friction, Friction stir process, Composite Materials, Wear Resistance.*

تأثيرات اضافة حبيبات الكرافايت وظروف العملية على الخواص الميكانيكية لسطح الكرافيت/البولي اثيلين المصنع بواسطة عملية الخلط الاحتكاكي

الخلاصة: ان عملية الخلط بالأحتكاك هي تقنيه جديده لتصنيع المواد في الحاله الصلبه تستخدم لتحسين الخواص الميكانيكيه للمواد وكذلك في انتاج طبقات سطحيه من المواد المركبه بدلا من تقنيات التصنيع التقليديه.يهدف البحث الى دراسة امكانية تطبيق تقنية الخلط بالأحتكاك لانتاج طبقه من ماده مركبه على سطح مادة البولي اثيلين عالي الكثافه بأضافة حبيبات الكرافايت بحجم حبيبي 25 مايكرون, وضعت في شق عمقه 1.5 ملم على سطح البولي اثيلين.ولغرض الحصول على توزيع متجانس للحبيبات على سطح الماده الأساس تم اختيار اداة ذات لفائف دائريه التي انتجت أقل تشوه على سطح الماده البوليمريه.تم دراسة تأثير عملية الخلط الاحتكاكي كالسرعة الدورانية والسرعة الخطية على الخواص الميكانيكية للطبقه السطحيه ذات الماده المركبه الناتجه بالمقارنة مع الماده الأساس.أظهرت نتائج اختبار البليان تحسناً ملحوظاً لسطح الماده البوليمريه حيث انخفض معدل البليان بمقدار 70 % بوجود حبيبات الكرافايت , اما أختبار الصلاده فقد اظهر أن صلادة الماده المركبه السطحيه المدعمه بحبيبات الكرافايت ازدادت بمقدار 18% عن الماده الأساس. بينت نتائج الفحص المجهري ان لسرع عملية الخلط الأحتكاكي تأثير على توزيع حبيبات مادة التدعيم خلال الماده المركبه الناتجه, حيث ان زيادة السرعه الدورانيه أدت الى توزيع حبيبي متجانس اضافة الى زيادة المسافات مابين الحبيبات وبالتالي التقليل من تكتل أو تجمع هذه الحبيبات.

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1. Introduction

FSP is a solid-state processing technique depended on friction stir welding basics, which was contained a special design of rotating tool with moves down the surfaces of material plates. This is achieved by generating a highly plastically deformed region by the associated stirring action. The resultant structure of stirred zone (SZ) contains equiaxed and fine grains, which formed as a result of dynamic recrystallization [1].

FSP can be modified and changes the local properties of the samples, without influencing the properties in the residual part of the samples. However, it varies in that FSW treaties with joining of two similar or dissimilar materials, whereas FSP was used to strengthen (not necessarily join) some regions of a material [2]. There are many parameters affect the overall process, such as traverse speed, rotational speed and pass number. Any varying in these parameters results in a significant influence on the mechanical properties, for example, the hardness and strength of the final product [1,2].

Newly, there have been numerous investigations on the using of the FSP to fabricate a composite layer directly on the surface of a sheet in situ by making a channel or groove on the sheet surface which is packed with reinforcement particles. However, it remains a challenge, during the FSP, how to prevent the reinforcement particles from being ejected out of the channel or groove, as well as the particles have typically non homogeneous distribution unless multiple FSP passes are applied to produce homogeneous microstructure. The resultant composite layer generally exhibits better properties and microstructure due to clean particle-matrix interface, improved wet ability of particles which form strong bonds between the reinforcement and the material matrix [2].

Thermo plastics, such as Polypropylene (PP) and Polyethylene (PE) have been treated by friction stir processing [3,4]. It is noted that FSP of polymers and polymeric composites (PMCs) are still much less studied than FSP metals and metallic composites. There are some concerns related to applying of friction stir process on polymers due to the rotation tool pin and the severe plastic flow in the nugget zone could break macromolecular chains and change the properties of the materials [3].

Y. Morisada et al. (2006) studied the uniformly dispersed of SiC particles into an AZ31 matrix using FSP. The micro hardness of the processed zone that reinforced with SiC particles was increased, due to the distribution of the SiC particles and to the grain refinement [5]. P. Asadi et al. (2010) employed FSP to form a composite layer on the surface of magnesium alloy (as-cast AZ91) using SiC powder. The grain size of magnesium matrix (AZ91) decreases severely as well as micro hardness increased, by incorporation of SiCnano particles. Second pass of FSP decreases the grain size and increases the micro hardness of the samples FSPed with SiC particles and enhances SiC particles distribution [1]. Adem Kurt et al. (2011) incorporated of SiC particles into the commercially pure aluminum to form particulate surface layers by using Friction Stir Processing (FSP). Specimens were subjected to several transverse and rotational speeds with and without SiC particles. Results showed that increasing the tool rotational speed causes a more homogeneous of SiC particles distribution. Bending strength of the resultant surfaces composite was significantly higher than processed specimen and untreated base Al. The hardness of the resultant surface composite was improved by three times as compared to that of base metal [6]. Thangrasu et al. (2012) studied the fabrication of aluminum matrix surface composite (AMC) reinforced with TiC powders using FSP. (SEM) showed a homogeneous distribution of TiC particles which formed a strong bond with the matrix alloy. Hardness of the Al/TiC surface composite was increased about 45% higher than that of the base metal [7].R. Sathiskumar et al. (2013) utilized Friction stir process technique (FSP) to fabricate copper surface composites reinforced by boron carbide (B_4C) particles. Increasing tool rotational speed with decreasing transverse speed produced higher area of surface composite with uniform distribution of B_4C particles due to high frictional heat generated which increased stirring and transportation of material. The microstructure evaluation showed that the grains of copper matrix were refined due to the pinning effect of B_4C particles [8].E.T.akinlabi et al. (2014) studied the effect of process parameters on the wear resistance behavior of Al/TiC surface composites fabricated by friction stir process. The wear rate property was found to decrease as a result of the TiC powder addition. The right combination of the process parameters was optimized to improve the wear resistance property of the resultant surface composites produced. The micro hardness profiling of the FSP samples showed an increase in hardness value when compared to the base metal, which can be attributed to the dispersed TiC particles via FSP [2].

From the above, it can be seen that all of the FSPs were done on metallic material, such as (Al, Cu, Mg…etc.) and there are very rare researches deal with FSP on polymer. In this study, FSP was applied in fabricating Polymer Matrix Composite (PMC) by the reinforcing of high density polyethylene (HDPE) with graphite particles. The effect of FSP parameters (rotational and transverse speeds) on the mechanical properties (wear resistance and hardness) and microstructure was studied as compared with as received one.

2. *Experimental work*

Clamping assembly consisting of aluminum flat plate (backing plate) with dimensions of 80x30x2 cm was used in this study. Vertical drilling of adequate number of screw holes was used; those holes were distributed on the fixture to facilitate the installation of samples sheet and to work out on more than variable through the section to save time and effort. The high number of screws was used to clamp the work piece tightly to avoid any movement, and to ensure that the blocks are positioned horizontally all the time. HDPE pieces of 600x200x5 mm were cut and used to fabricate polymer matrix surface composite by FSP, Fig. 1.

The FSP procedure is schematically shown in Fig. 2. A groove or channel was made in the middle of the plate along the line of FSP; with 1.5x2 mm dimensions. The groove was fully filled with the graphite particles, Average particle size and purity of graphite particles were< 25 μm and 99%, respectively.Then, FSP tool rotating clockwise is inserted into the prepared sample to carry out FSP and mixing the reinforcement particles with matrix plate to produce surface composite, as shown in Fig 3.

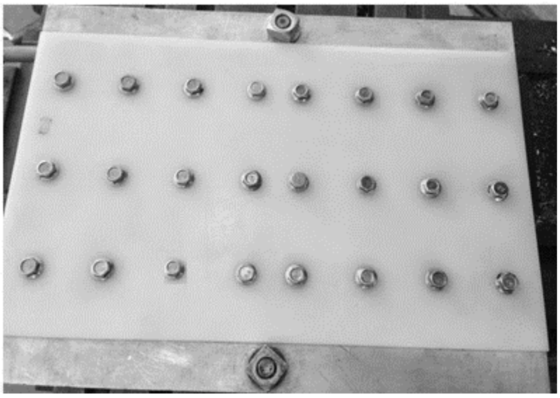


Figure 1. Polyethylene substrate set on fixture

DIN 100 MnCrW 4 tool steel, with an average of 56 HRC hardness, was used in this processing. This tool consists of scrolls on shoulder involved of a flat end surface with a spiral channel cut from the edge towards the center. The channels serve to make the material flow from the edge of the shoulder to the center, thus eliminating the need to tilt the tool [9].

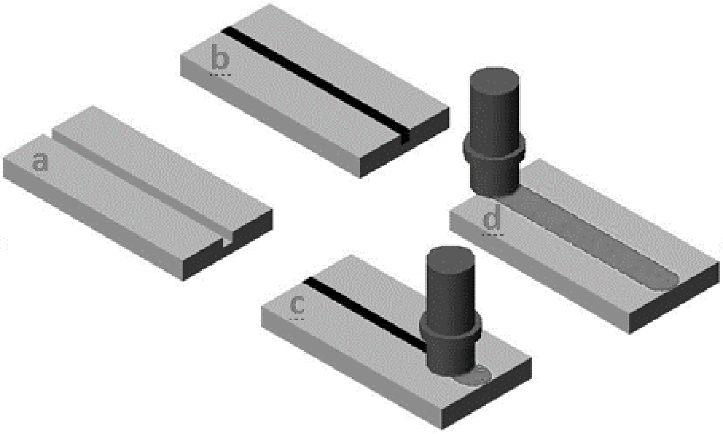


Figure 2. FSP procedure to fabricate surface composite: (a) making a groove, (b) compacting the groove with particles, (c) processing using a pinless tool and (d) processing using a FSP tool

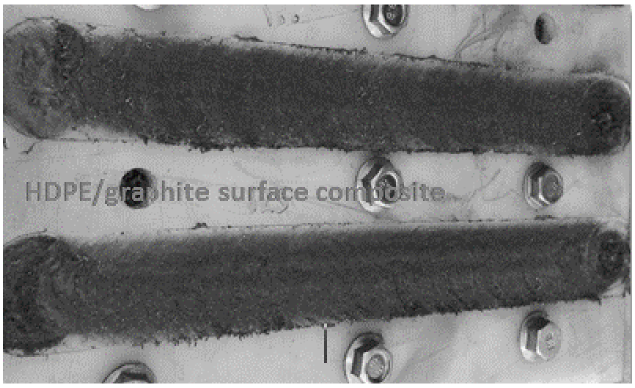
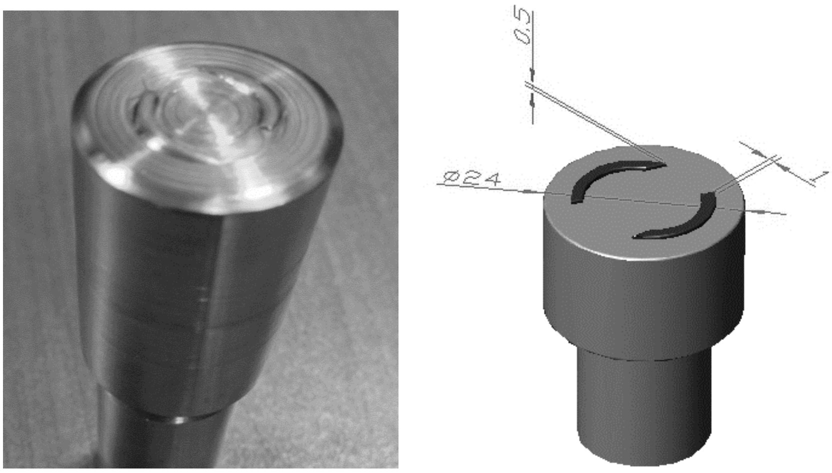


Figure 3.FSPed zones resulted by using Scrolled shoulder

This shoulder has 24 mm diameter, 0.5 mm scrolls thickness, with 1mm width [10], the schematic and photographic profiles of this tool are presented in Fig. 4(a) and 4(b).

The advantages of using scrolls shoulder to produce PMCs are: (a) Very high degree of homogenous distribution of reinforcement particles in HDPE matrix, (b) the scroll shoulder design prevents material displacement away from center and takes advantage of the greater flexibility in the contact area between the substrate and shoulder [11].



(a) (b)

Figure (4): (a) Scrolled shoulder, (b) Sketch of the scrolled tool

In order to determine an optimum range for the processing parameters, many tests were performed to find the appropriate range for these variables. Table 1 contains FSP parameters used in fabricating a composite layer on HDPE surface.

Table (1) FSP parameters for fabricating of HDPE surface composite

Rotational speed (rpm)	Transverse speed (mm/min**)**
1600	74
820	74
650	74
1600	98
820	98
650	98
1600	132
820	132
650	132

Wear test specimens were fabricated according to ASTM G 99 – 04 standardization, where they were of (10 mm) diameter, inserted in a cylindrical collet of 10mm diameter and 3mm length, under varying of applied load (2.5, 5, 7.5 and 10) N, constant speed of 250 rpm, and the duration of test was 15 minutes. Hardness test was performed on samples which had dimensions of (20x20x5) mm according to ASTM D2240 using the Durometer tester. Macro and micro surface structures were carried out by using of optical microscope to study the homogeneity of the mixture.

3. Results and Discussions

3.1. *Wear Resistance test results*

The wear resistance test was evaluated on both the as-received HDPE and processed samples. The weight loss of each specimen after wear testing was determined in order to calculate the wear rate (gm/cm) of the HDPE surface composite. The formula used to convert the weight loss into wear rate (gm/cm) is [12]:

$$Wr = (\Delta W / S) \qquad (1)$$

Wr= Wear rate (gm/cm)
$\Delta W = W_1 - W_2$= Weight difference of sample before and after each test (gm).
S = Total sliding distance (cm)

$$S = V \times t \times 100 \qquad (2)$$

V = Linear velocity (m/min)
t = Time of running (min)

$$V = 2\pi \times r \times n \qquad (3)$$

r = Distance from the center of pin to the center of disc, (m).
n= Disc rotational speed (rpm).

HDPE was reinforced with graphite particles under different processing parameters (Table1); these parameters affected clearly the wear resistance of HDPE/surface composite. Fig.5 shows that the wear resistance of samples increases (decrease of wear rate) by reinforcing with graphite particles under all processing parameters.

Too high rotational speed for a polymeric material such as pure HDPE tends to produce high quantities of frictional heat that caused high mixing between polymeric materials with the powder. Dilution requires to be minimized so as to enhance the wear resistance of the work piece surface, melting does not occur during the process; this is one of the main advantage of using FSP [2].

High rotational speed results in the increasing of the process temperature due to the frictional heat generation, therefore, HDPE sheet begins to burn, melt and degradation. Furthermore, excessive turbulence of mix will occur, that’s lead to voids content [13].

At (1600 rpm), the wear rate was found to decrease with the increase in the transverse speed, it became (5.26272×10^{-8} gm/cm) at 10 N load (decreased approximately 63%) at 132 mm/min, as shown in Fig.6. This can be attributed to proper melting of the material; hence dilution of graphite powder can occur at high feed rate which would increase the wear action [2] (i.e., at high rpm, adequate softening of matrix can be achieved when the feed rate increased).

As the transverse speed increases, the shoulder also moves faster, which would not provide enough time to polymer sheet to be melted. On the other hand at low transverse speeds, excessive heat input per unit area and material processing will result in void effect. The increasing in the process time and consequently decrease in the process efficiency is why that small amount of travel speed was avoided [13, 14].

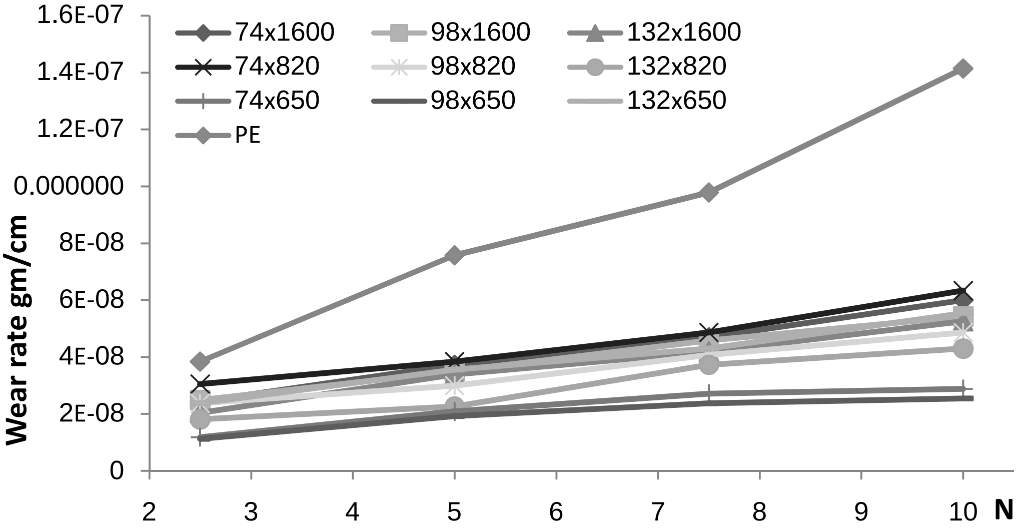


Figure 5. Wear rate of HDPE reinforced with graphite by different rotational and transverse speeds

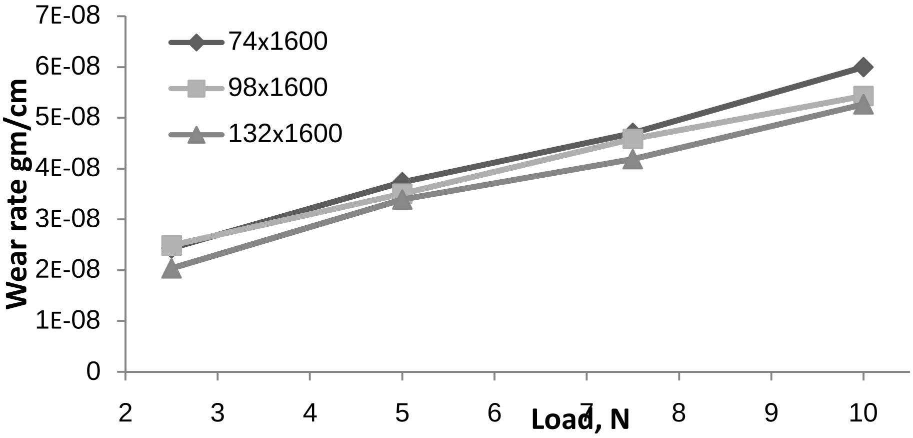


Figure 6. Wear rate of HDPE reinforced with graphite by 1600 rpm and (74, 98, 132) mm/min

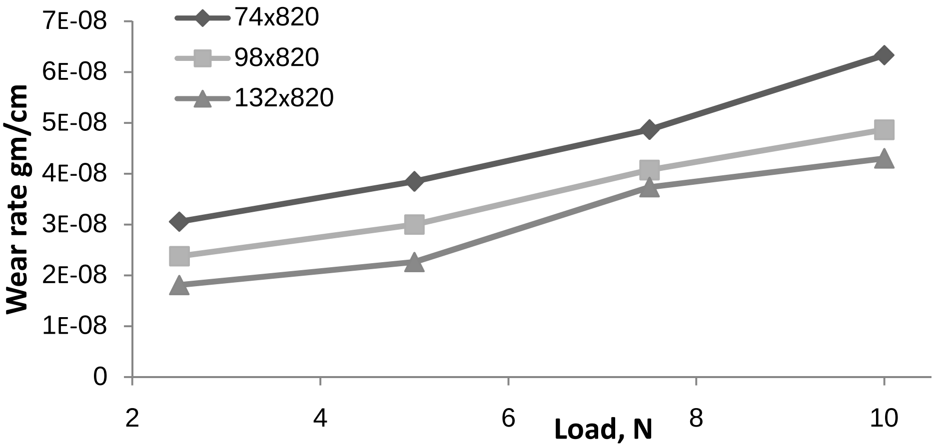


Figure 7. Wear rate of HDPE reinforced with graphite by 820 rpm and (74, 98, 132) mm/min

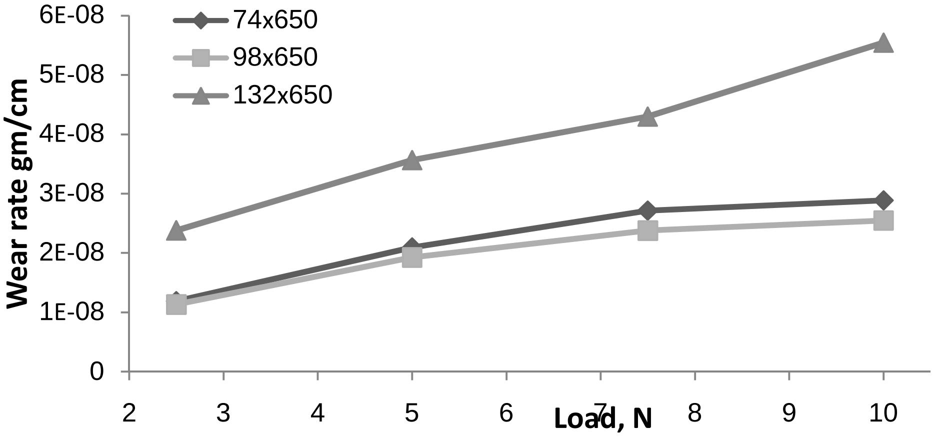


Figure8. Wear rate of HDPE reinforced with graphite by 650 rpm and (74, 98, 132) mm/min

At (820 rpm), the wear rate was also found to be decreased as the transverse speed increased from 74 mm/min to 132 mm/min to reach ($4.300*10^{-8}$ gm/cm) at 10 N load, as shown in Fig.7. A good wetting and proper melting can occur at 132mm/min, and wear rate decreased approximately 70% at this condition due to the suitable amount of heat generation that caused the substrate to be softened which enhanced mixing quality.

At lower rotational speed of (650 rpm) and transverse speed of98 mm/min, the minimum loss of mass was observed, wear rate reached to ($2.5464*10^{-8}$ gm/cm) at 10 N load as shown in Fig.8, which was decreased approximately by 82%.This represents the optimum conditions of wear resistance of HDPE reinforced with graphite particles, due to the proper amount of heat generated at these conditions providing high quality of mixing between matrix and filler at the surface as well as good environment which form strong bond between matrix and filler. Also, slippage and wettability of particles increase, leading to decreasing wear rate at this condition [2,15,16].

3.2. *Durometer surface Hardness Test Results*

Results of Shore-D Hardness tests showed that all HDPE/surface composites have higher hardness than that of the as received one. The increase in this property is due to the high hardness property of the graphite particles in addition to the strengthening resulted from carrying of the load by these particles. Fig.9refers to the average of five values for the specimens manufactured by FSP at three zones (retreating (RZ), center (CZ), advanced (AZ)).From this figure, it can be seen that the hardness increases as the rotational speed (rpm) increases from 650 rpm to 820 rpm. When tool rotational speed was increased to 1600 rpm, hardness of composite layer decreased. The hardness was found to be less than that of the as received HDPE, especially in the retreating zone at low transverse speed (74 mm/min).

The explanation for that behavior is the presence and amount of agglomerations which cause higher difference in hardness across the composite layer. The possibility of indenter resting directly on the particles increased, due to agglomerations when the measurement of hardness was carried out at even spacing, which lead to higher hardness at 650 rpm and 820 rpm [8].

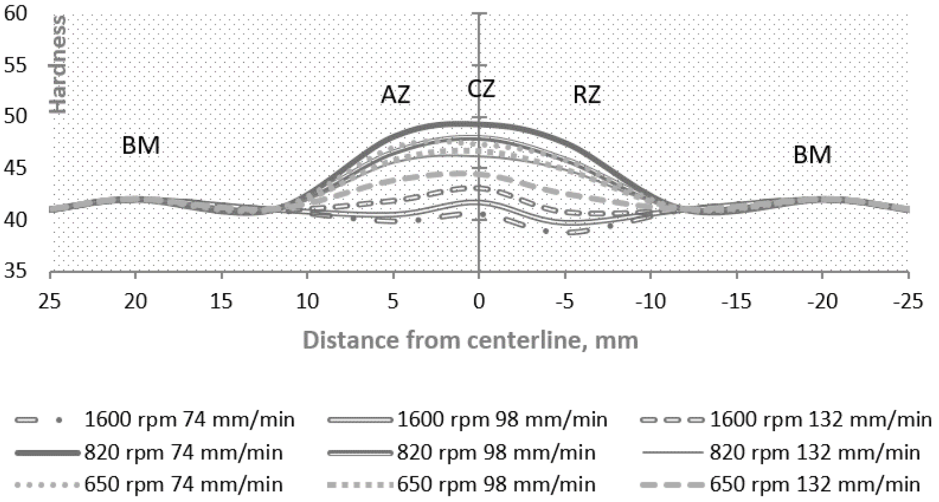


Figure 9. Hardness profile along stir zone of HDPE reinforced with graphite after FSP applied with all parameters

When tool rotational speed was increased to 1600 rpm, the hardness decreased which can be attributed to the breaking up of particles agglomerations and then their uniform distribution. Since the area (dimensions) of surface composite layer simultaneously increases, it resulted in a general drop in particles volume fraction, where the same amount of particles packed in the groove was distributed into larger area of softened HDPE. Therefore, the hardness of the surface composite becomes lower at 1600 rpm for the three types of reinforcements [8].

The effect of transverse speed on the hardness of HDPE/surface composite is also shown in Fig.9. At 1600 rpm, the hardness increased when transverse speed was increased. While, it decreased at 74 mm/min and that can be attributed to the drop in the particles volume fraction, despite having homogeneous distribution. The presence of agglomerations resulted in higher hardness values when transverse speed was increased. On the other hand, at 650 and 820 rpm, the hardness decreased with the increase of transverse speed. That is attributed to the increase in the volume fraction of graphite in the processed zone, as a result of the reduction in the amount of the reinforcement particles mixed with the softened matrix, due to the decrease in the amount of heat generated at these conditions.

Polymeric composite material (especially which was reinforced by particulate using FSP technique) does not depend only on the process parameters or the properties of components, but also depends on the nature of the interface between the components, the weight fraction, and sometimes on the geometry of the particulates [17].

It is obviously clear that the graphite-reinforced composite exhibits higher hardness values compared to that as received one, where they were, 49.3 and 41.5 HD, respectively. This is mainly due to the interfacial bond as well as good inherent hardness of the graphite particles.

3.3 *Microstructure Evaluation*

Optical microscope was used to reveal the distributions of reinforcement particles (graphite) in the HDPE matrix. Fig.10, 11 and 12 shows the effect of rotational speeds (650, 820 and 1600 rpm) respectively on the particles distribution at constant transverse speed (74 mm/min).

At 650 rpm the distribution was not uniform due to the presence of agglomeration of graphite particles at several places. The agglomerations were gradually disappeared when tool rotational speed was increased. Excellent distribution was shown when rotational speed increased to 1600 rpm, where the average spacing between graphite particles increased. Tool rotational speed has two more functions in addition to heat generating due to friction, stirring the softened materials as well as influencing matrix sample flow behavior across the processed zone [8].

The presence of particles agglomerations at low rotational speed (650 rpm) can be attributed to inadequate stirring and insufficient material flow from the advancing side to retreating side. The graphite particles, which were filled the groove, did not mix with the softened HDPE substrate appropriately, therefore, particles agglomeration were formed. When rotational speed increased, the quantity of stirring and material flow increased too. The friction stir processing zone (surface composite layer) was submitted to high plastic strain. This plastic strain enhanced the stirring shatter of the agglomerations into fine dispersion in the HDPE matrix.

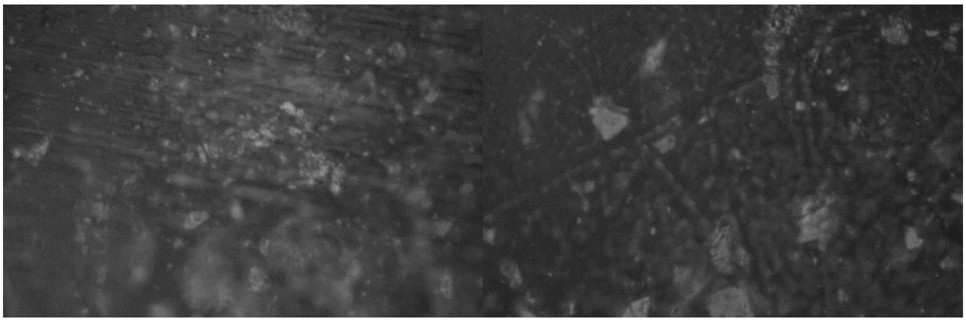


Figure 10. Optical photomicrograph (800 X) of HDPE/graphite surface composite at 650 rpm

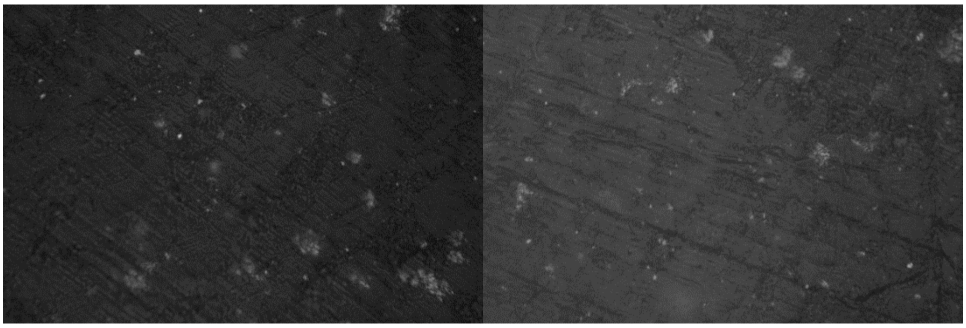


Figure 11. Optical photomicrograph (800 X) of HDPE/graphite surface composite at 820 rpm

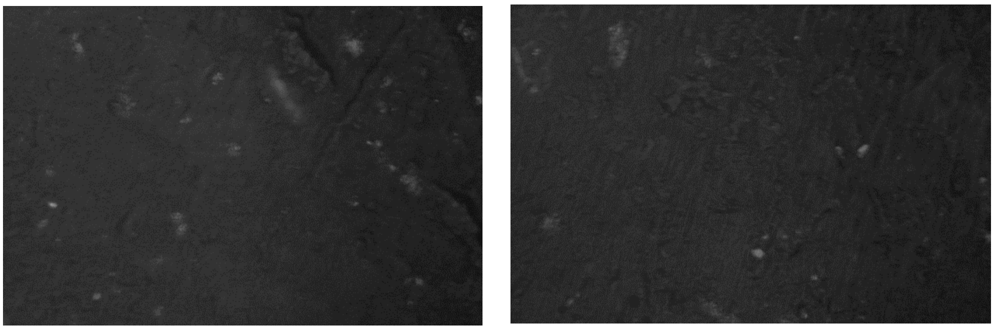


Figure 12. Optical photomicrograph (800 X) of HDPE/graphite surface composite at 1600 rpm

5. Conclusions

The conclusions derived from this experimental work can be summarized as follows:

1- HDPE/graphite surface composite with homogenous particles distribution can be achieved by the scrolls shoulder, in which the frictional heat generated and flow of softened HDPE were adequate to yield defect free surface composites.

2- Right combination of processing parameters (rotational and transverse speed) was necessary to really improve the wear resistance and hardness of the composite layer produced.

3- There was insufficient melting of matrix at low rotational speed. As well as, too high heat generated at too high rotational speed that caused high dilution of the filler which reduces the wear resistance and hardness.

4- A moderately rotational speed of (650, 820) rpm formed the surface composited layer with the optimum wear resistance and hardness properties depending on FSP parameters optimize in this work and can be considered as the best parameter.

5- High tool rotational speed resulted in good distribution of particles and vice versa.

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6. References

1. P. Asadi, M. BesharatiGivi, K. Abrinia, M. Taherishargh and R. Salekrostam. (2011)."*Effects of SiC Particle Size and Process Parameters on the Microstructure and Hardness of AZ91/SiC Composite Layer Fabricated by FSP*". Journal of Materials Engineering and Performance JMEPEG, Vol. 20, pp. 1554–1562.
2. E. T. Akinlabi, M. Mahamood, A. Akinlabi and E. Ogunmuyiwa. (2014). "*Processing Parameters Influence on Wear Resistance Behavior of Friction Stir Processed Al-TiC Composites*". Hindawi Publishing Corporation, Advances in Materials Science and Engineering Article ID 724590,12 pages.

3. Yong X. Gan, Daniel Solomon and Michael Reinbolt. (2010)."*Friction Stir Processing of Particle Reinforced Composite Materials"*. Materials, Vol. 3, pp. 329-350.
4. R. Prasad and P.madhuRaghava. (2012). "*FSW of Polypropylene Reinforced with Al_2O_3 Nano Composites, Effect on Mechanical and Micro structural Properties"*. International Journal of Engineering Research and Applications (IJERA), Vol. 2, pp.288-296.
5. Y. Morisada , H. Fujii , T. Nagaoka and M. Fukusumi. (2006). "*Effect of friction stir processing with SiC particles on microstructure and hardness of AZ31"*. Materials Science and Engineering, Part A, Vol. 433, pp. 50–54.
6. Adem Kurt, Ilyas Uygur, ErenCete. (2011). "*Surface modification of aluminium by friction stir processing"*. Journal of Materials Processing Technology Vol. 211, pp. 313–317.
7. A. Thangarasu, N. Murugan, I. Dinaharan and S. Jvijay. (2012). "*Microstructure and microhardness of AA1050/TiC surface composite fabricated using friction stir processing"*. Indian Academy of Sciences, Vol. 37, Part 5, pp. 579–586.
8. R. Sathiskumar, N Murugan, Dinaharan and S J Vijay. (2013). "*Role of friction stir processing parameters on microstructure and microhardness of boron carbide particulate reinforced copper surface composites"*. Indian Academy of Sciences Vol. 38, Part 6, pp. 1433–1450.
9. Y. N. Zhang, X. Cao, S. Larose and P. Wanjara. (2012). "*Review of tools for friction stir welding and processing"*. Canadian Metallurgical Quarterly Vol. 51, No 3.
10. Rajiv S. Mishra. (2007). "*Friction Stir Welding and Processing"*, Center for Friction Stir Processing, University of Missouri-Rolla Murray W. Mahoney, Rockwell Scientific Company.
11. R. Mishra and Z. Ma. (2005). "*Friction stir welding and processing"*. Materials Science and Engineering, Vol. 50, pp. 1–78.
12. ASTM Designation D 2240 – 02a, Standard Test Method for Wear Testing with a Pin-on-Disk Apparatus.
13. Ehsan Azarsa and Amir Mostafapour. (2014). "*Experimental investigation on flexural behavior of friction stir welded high density polyethylene sheets"*. Journal of Manufacturing Processes, Vol.16, pp. 149–155.
14. Erica Anna Squeo, Giuseppe Bruno, Alessandro Guglielmotti and FabrizioQuadrini. (2009). "*Friction Stir Welding of polyethylene sheets"*. FASCICLE V, Technologies in Machine Building, pp. 241-246.
15. Omar O. Salman. (2006).*"Investigation of mechanical properties of epoxy with ceramic material"*, M.Sc. Thesis, Mechanical Engineering/College of Engineering of Nahrain University, Iraq.
16. Mohamed K. H. (2011).*" Ceramics (Al_2O_3+SiC+graphite) and fiber (aramide) reinforcement effect on the mechanical properties of composite material"*. Higher Diploma Thesis, Material Engineering Technology, Engineering Technical College / Baghdad, Iraq.

17. Ali Kifah Ghazi. (2012). *"Mechanical properties and fatigue study for different polymer composites using micro- nano reinforcement".* Higher Diploma Thesis, Material Engineering Technology, Engineering Technical College / Baghdad, Iraq.