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## Studying the effect of local mixed Libyan grasses, *Halfa* and *Esbat* on the mechanical properties of styrene-butadiene rubber composites

\*Anour Shebani<sup>1,2</sup>, Hussein Etmimi<sup>1,2</sup>, Tarik Yerro<sup>3</sup>, Osama Ahtewish<sup>3</sup>

<sup>1</sup> Libyan Polymer Research Center, Tripoli, Libya

<sup>2</sup> Research and Consultancy Center, Sirte University

<sup>3</sup> Mechanical Department, Higher Institute of Science and Technology, Nalut, Libya

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### ABSTRACT

In this study, the effect of using local mixed Libyan grasses (*Halfa* and *Esbat*) as a filler on the mechanical properties of SBR composites was investigated. This was performed by studying the effect of filler content (10, 20, 30 and 40 wt%) on mechanical properties of the composites including stress at break, elongation at break, impact strength and micro hardness. Also, a comparison study between using mixed and individual grasses has been attained. The stress at break was decreased with the addition of 10% mixed grasses, then increased as the mixed grasses content increased up to 30% and then decreased again. The incorporation of mixed grasses caused a significant decrease in the elongation at break. This decrease in the elongation at break was increased with increasing the grasses content. Impact strength was slightly increased with the addition of mixed grasses. Increasing the grasses content had no profound effect on the impact strength values of SBR composites. Hardness properties were slightly increased with the addition of mixed grasses. The optical image of SBR composite made 10% of mixed grasses content showed bad filler distribution, while composites with 20 and 30% grasses content had relatively better filler distribution. On the other hand, the image of composite with 40% mixed grasses content clearly showed the formation of filler aggregation. Mixed grasses appears to be relatively better than individual grasses (*Halaf* and *Esbat*) in order to attain composites with good strength properties. On the side, individual grasses seems to be better than mixed grasses in producing composites with relatively better impact strength and hardness properties. In both cases (individual or mixed grasses), the best content of grasses in order to provide good mechanical properties should be in the range of 20-30%. In conclusion, composites obtained from these grasses (individual or mixed) could possess acceptable mechanical properties.

## دراسة تأثير خليط من أعشاب محلية من الحلفاء والسبط على الخواص الميكانيكية لبوليمر مركب من مطاط الستايرين البيوتاديين

\*أنور الشيباني<sup>1,2</sup> و حسين التميمي<sup>1,2</sup> و طارق يروو<sup>3</sup> و اسامة احتيوش<sup>3</sup>

<sup>1</sup> المركز الليبي لبحوث اللدائن، طرابلس، ليبيا

<sup>2</sup> مكتب البحوث والاستشارات، جامعة سرت، سرت، ليبيا

<sup>3</sup> المعهد العالي للعلوم الهندسية، نالوت، ليبيا

### الكلمات المفتاحية:

الألياف النباتية  
الأعشاب الليبية  
البوليمرات المركبة  
مادة تقوية  
الخواص الميكانيكية

### الملخص

تم في هذه الدراسة دراسة تأثير استخدام خليط من الأعشاب الليبية (الحلفاء والسبط) كحشوه أو مادة تقوية على الخواص الميكانيكية لبوليمر مركب من مطاط الستايرين بيوتاديين. وذلك من خلال دراسة تأثير محتوى الأعشاب (10، 20، 30 و 40 نسبة وزنيه %) على الخواص الميكانيكية مثل الإجهاد عند الكسر، الاستطالة عند الكسر، مقاومة الصدم و الصلادة. كما تم أيضا مقارنة نتائج هذه الدراسة بنتائج دراسة سابقة والتي تم فيها استعمال هذه الأعشاب كلاً على حدا. أوضحت هذه الدراسة أن الإجهاد عند الكسر انخفض عند إضافة 10% من خليط الأعشاب ثم زاد بزيادة محتوى الأعشاب حتى 30% ثم انخفض بعد ذلك. كما تسبب إضافة

\*Corresponding author:

E-mail addresses: [anour@prc.ly](mailto:anour@prc.ly), (H. Etmimi) [hmetmimi@gmail.com](mailto:hmetmimi@gmail.com), (T. Yerro) [tarikyerro@yahoo.com](mailto:tarikyerro@yahoo.com)

, (O. Ahtewish) [osamaahtewish@gmail.com](mailto:osamaahtewish@gmail.com)

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خليط من الأعشاب في حدوث انخفاض كبير في الاستطالة عند الكسر. أن هذا الانخفاض زاد بزيادة محتوى الأعشاب. مقاومة الصدم أيضاً انخفضت بشكل بسيط جداً مع زيادة محتوى الأعشاب. الصلادة من جهة أخرى زادت بزيادة محتوى الأعشاب. نتائج المجهر الضوئي أوضحت وجود توزيع غير جيد للأعشاب في حالة العينة المحتوية على 10% من خليط الأعشاب، بينما تبين وجود تكتل للأعشاب في العينة التي احتوت على 40%. هذا التكتل لم يتم ملاحظته في العينات التي تحتوي على 20 و 30%. بالإضافة لذلك أظهرت هذه العينات توزيع جيد للأعشاب داخل البوليمر. من خلال النتائج يبدو أن استعمال الأعشاب المختلطة يعطي خصائص شد أفضل من استعمال هذه الأعشاب كلاً على حدا. من جهة أخرى استعمال هذه الأعشاب منفردة يعطي أفضل خصائص مقاومة الصدم والصلادة من استعمال الخليط. أفضل محتوى من هذه الأعشاب (سواء كانت مختلطة أو منفردة) هو من 20-30%، وفي الخلاصة، أن هذه الأعشاب يمكن أن تستخدم كحشو أو مادة تقوية لإنتاج بوليمرات مركبة بخصائص متنوعة وتكلفة منخفضة.

## 1. Introduction

Polymer composites (PCs) based on natural fibers (NFs) have attracted huge interest in recent years. They known as natural fiber (NF) reinforced polymer composites (NFRPCs) or reinforced polymer matrix (PMCs). It is important to declare that the utilization of NFs as a filler in PCs field become well known technique for different applications, especially in view of sustainable materials [1-2]. NFs have been successfully employed to replace synthetic fibers (SFs) in many applications. This is because NFs reduce waste disposal issues, reduce environmental pollution and improve the polymer properties and performance. Particularly, NFs have been competed the SFs in certain properties such as mechanical and thermal properties [3]. In fact, PMCs are considered as eco-friendlier substitutes over composites with SFs. This could be due to their inherent biodegradability, low cost, renewability, low environmental impact, non-hazardous nature, manufacturing flexibility and lightweight properties [2-4]. However, PMCs have been commercially used in many applications include; furniture, automotive, electronic industries, and building construction.

NFs are typically obtained from plants or animals or mineral. Plant fibers (PFs) considered to have a vast source of NFs. PFs are normally sourced from the leaves, stems, and seeds plants. NFs from plants like assisal, bamboo, coconut, jute, flax, hemp, kenaf, coir, kapok, palm, banana, etc., have been used in field of PMCs [5-6]. These NFs have been used as to reinforce different types of polymers such as polyethylene, polypropylene, poly vinyl chloride, nylon, acrylics, epoxies, polyesters, phenolic and others [7-8]. Moreover, these NFs have been used as a filler to reinforce polymer blends and copolymers [9-11]. Among these copolymers is styrene-butadiene rubber (SBR), which used in this study as a matrix. SBR is a rubbery material can be used instead natural rubber in many applications. SBR used in several industrial applications such automotive, footwear, and belt industries. Most importantly, adding fillers (e. g., NFs) to SBR could improve its properties and wide its range of applications. Practically, adding filler normally result increase in the modulus and significantly improves the abrasion and tear resistance [12].

In this study, mixed local Libyan grasses (Halfa and Esbat) were used as a filler to reinforce SBR. Although there are various fillers have been used for the reinforcement of SBR, no much work has been made so far to use certain grasses as reinforcing filler for SBR matrix. In a previous study [13] the SBR was reinforced using Halfa and Esbat, individually. These grasses were effectively used as a filler in SBR. Halfa showed to be better than Esbat as a filler for SBR. These grasses are known as non-woody materials and composed of cellulose, hemicelluloses, protein, lignin, and minerals, similar to woody materials. They have the same composition as woody materials, but they differ in the proportion of each component in the composition. For instance, non-woody materials contain low lignin content in comparison to woody ones. Therefore, through this study, the effect of using mixed grasses on the mechanical properties of SBR is considered, especially after successfully using them individually. According to Alessandro et al. [14] these composites could be termed as hybrid PMCs because they contain more than one type of fiber as reinforcement in a single polymer matrix.

## 2. Experimental work

### Materials

Halfa (local name of *Stipa tenacissima* grass) and Esbat (local name of *Stipagrostis pungens* grass) were involved in this study to be used as fillers. Generally, these grasses are distributed throughout Libya in dry and hot regions. The used grasses were collected from dry region close to Nalut city in Libya. Before using them, these grasses were washed with distilled water, and dried. Then they were chopped and sieved to get particle size of 212 µm. SBR from Parc Scientific (UK) was used as a matrix.

### Methods

#### Preparation of SBR composites

The used grasses were mixed and dried in an oven at 60 °C for almost 4 h. After that they were mixed with SBR using twin screw extruder from Brabender, Germany with speed of 70 r.p.m. at 180 °C. The composites were prepared with SBR different grasses content as shown in Table 1.

Table 1 Composites prepared in this study.

Sample	SBR, %	Halfa, %	Esbat, %
SBR	100	0	0
Composites with individual grasses (pervious study [13])			
SBRH1	90	10	0
SBRH2	80	20	0
SBRH3	70	30	0
SBRH4	60	40	0
SBRS1	90	0	10
SBRS2	80	0	20
SBRS3	70	0	30
SBRS4	60	0	40
Composites with mixed grasses			
SBRM1	90	5	5
SBRM2	80	10	10
SBRM3	70	15	15
SBRM4	60	20	20

### Characterization

#### Preparation of the testing samples

Injection molding apparatus from Xplore 12 ml, Netherlands was used to prepare samples for tensile strength, impact strength and microhardness tests.

#### Tensile strength test

Tensile test machine (QC-506M1, Cometheck) was used to attain the stress and elongation at break properties. The measurements were performed at room temperature. About four samples were tested from SBR and each composite prepared in this study under speed test of 100 mm/min. Samples dimension was 73 mm-4 mm-2 mm.

#### Impact strength test

Type of impact strength test performed in this study was Charpy impact test at room temperature. This test was carried out at impact energy of 15 J, using, CEAST Resil Impactor tester. This test was performed according to ASTM (D256-10) using five specimens for SBR and each composite prepared in this study.

### Micro hardness test

Vickers hardness tester MVT-1000Z was used to measure the hardness of SBR and each composite prepared in this study at room temperature. The measurements were carried out in conditions of 100 gf load and 10 s dwell time. Ten measurements were randomly taken for SBR and each composite prepared in this study.

### Morphological properties

Morphological properties of SBR and each composite prepared in this study were studied using an optical polarizing microscope (XP-501) from Turkey. This microscope is equipped with a color digital camera (Moticam 2) and software (Motic Images Plus 2).

## 3. Results and discussion

### Mechanical properties of composites with individual grasses

The results of this part have been discussed in detail in a previous study [13]. In this study, the addition of 10% and 40% Halfa and Esbat individually caused decrease in the stress at break of SBR. On the other hand, the stress at break of SBR was increased with the addition of 20% and 30% of Halfa and Esbat.

The addition of these grasses into SBR caused a noteworthy decrease in the elongation at break. This decrease in the elongation at break properties was decreased significantly with increasing the content of these grasses.

The impact strength of these composites (both with Halfa and Esbat) was increased with increasing the grasses content up to 30%. After that the impact strength was decreased, although it is still relatively higher than of neat SBR.

Increasing the content of both types of grasses resulted an increase in the hardness properties of SBR. This increase in the hardness properties was increased significantly with increasing the content of these grasses.

However, Halfa was relatively better than Esbat in reinforcing SBR, as mentioned previously. Overall, the best content of Halfa and Esbat provided good mechanical properties was in the range of 20-30%.

The mechanical properties of the SBR and composites prepared in this study is shown in Table (2). Standard deviation is given in parentheses in Table (2).

Table 2 Mechanical properties of SBR and the prepared composites with individual Halfa and Esbat.

Sample	Stress at break, N/mm <sup>2</sup>	Stress at break, %	Impact strength, KJ/m <sup>2</sup>	Micro - hardness
SBR	29.17 (0.8)	25.63 (2.9)	3.50 (0.07)	7.20 (0.3)
SBRH1	27.49 (1.3)	12.17 (1.1)	3.53 (0.06)	7.41 (0.2)
SBRH2	34.12 (1.4)	7.04 (1.2)	4.28 (0.07)	8.40 (0.4)
SBRH3	30.66 (1.1)	5.75 (0.9)	4.89 (0.07)	8.83 (0.4)
SBRH4	29.01 (1.3)	3.53 (0.9)	4.84 (0.07)	9.92 (0.7)
SBRs1	23.78 (1.5)	11.80 (1.2)	4.34 (0.06)	7.30 (0.2)
SBRs2	32.96 (0.9)	6.71 (0.8)	5.19 (0.05)	7.90 (0.2)
SBRs3	30.69 (0.3)	4.70 (0.9)	5.80 (0.08)	8.51 (0.3)
SBRs4	28.41 (0.7)	3.44 (0.8)	5.50 (0.06)	9.53 (0.5)

Standard deviation is given in parentheses.

### Mechanical properties of composites with mixed grasses

The mechanical properties of composites with mixed grasses are presented in Table (3). Standard deviation is given in parentheses, as shown in Table (3). The results in Table (3) illustrated that the stress at break, impact strength and micro hardness was increased with the addition of the mixed grasses content up to 30%. These properties were decreased with the addition of 40% of mixed grasses. On the other hand, the elongation at break decreased significantly with the addition of the mixed grasses content.

Table 3 Mechanical properties of SBR and the prepared composites with mixed grasses.

Sample	Stress at break, N/mm <sup>2</sup>	Elongation at break, %	Impact strength, KJ/m <sup>2</sup>	Micro - hardness
SBR	29.17 (0.8)	25.63 (2.9)	3.50 (0.07)	7.20 (0.3)
SBRM1	25.08 (0.2)	12.57 (0.6)	4.34 (0.19)	7.35 (0.3)
SBRM2	30.08 (0.6)	6.93 (1.2)	4.53 (0.08)	7.51 (0.2)
SBRM3	36.38 (0.5)	4.25 (0.3)	4.91 (0.09)	7.60 (0.4)
SBRM4	29.83 (0.8)	2.83 (0.3)	4.89 (0.16)	8.10 (0.4)

Standard deviation is given in parentheses.

As shown in Fig. 1, the stress at break was decreased with the addition of 10% mixed grasses, then increased as the mixed grasses content increased up to 30% and then decreased again. The highest stress at break value was obtained by composites with 30% mixed grasses content. Many studies [15-17] claimed that increasing the fiber content could resulted an increase in the tensile strength properties. Practically, the increase in the strength of PMCs could be due to certain factors such as higher cellulose, lignin contents, better dispersion of filler, and good adhesion between the filler and the matrix [18-20].

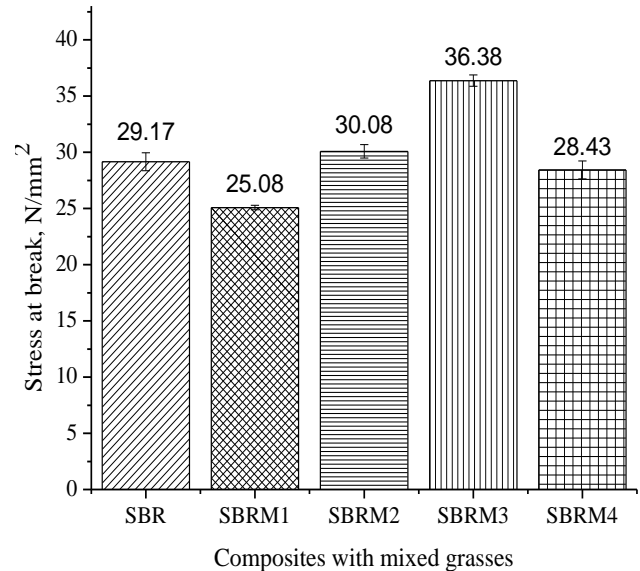
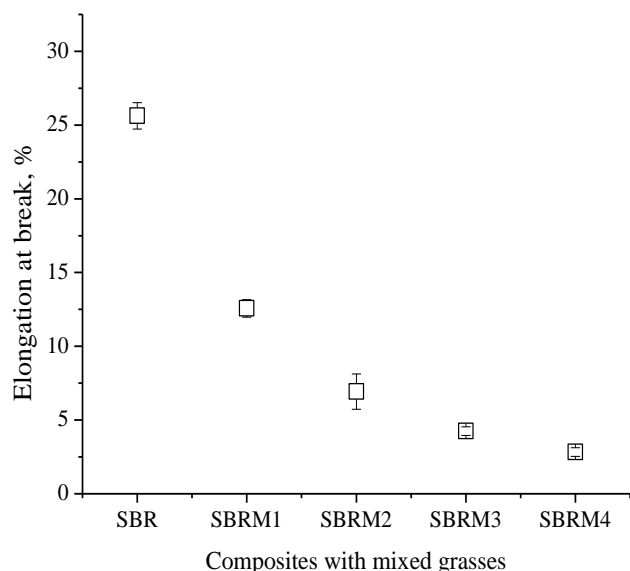


Fig. 1: Stress at break of SBR and the composites prepared with mixed grasses.

As found in the previous study [13], the effect of one grass alone on the stress at break was similar to the effect of mixed grasses. The stress at break of SBR was decreased with addition of 10% and 40% of the individual grasses. Conversely, it was increased with the addition of 20% and 30% of these grasses individually. It is important to emphasize here that the composites prepared with mixed grasses had higher values of stress at break compared to SBR and composites with individual grasses. This may indicate that the effect of mixed grasses on stress at break is more pronounced than using each grass alone.

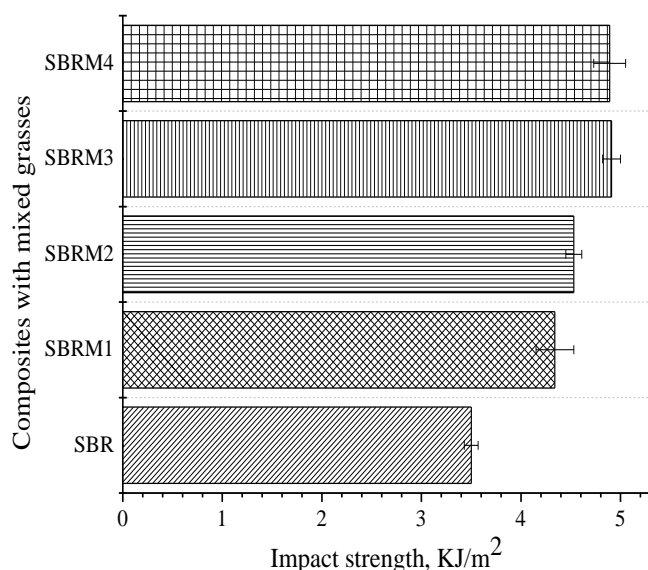
As shown in Table (3) and Fig. 2, the addition of mixed grasses caused dramatic decrease in the elongation at break properties of SBR. The decrease in the elongation at break was considerably increased with increasing the mixed grasses content. The same trend was observed when each grass was used alone, as shown in Table (2). This is a common observation, which pointed out by many researchers [21-24]. According to Ismail et al. [22], increasing the filler loading may cause an increase in the stiffness and brittleness of the PMCs, resulting a parallel decrease in the elongation at break. In other words, the increase in the stiffness of PMCs lead to decrease its ductility, which in the end the lower its elongation at break [23]. Furthermore, the addition of high filler content tends to increase the resistance to flow and lower the resistance to break [24]. This means that as filler loading increases, a higher restriction to molecular motion of the macromolecules is expected [25]. Meissner and Rzymiski [26] stated that the increase filler loading in the rubber matrix resulted decrease in the elongation at break which caused by the increase in the composite's stiffness.





**Fig. 2:** Elongation at break of SBR and composites prepared with mixed grasses.

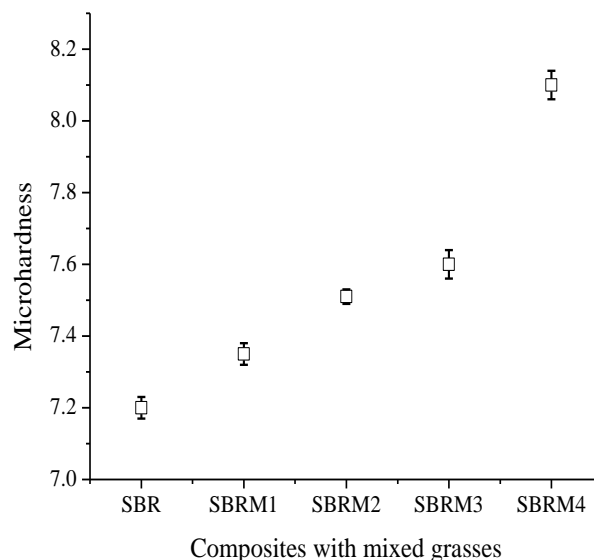
Impact strength was slightly increased with the addition of mixed grasses as illustrated in Fig. 3. Increasing the mixed grasses content had no profound effect on the impact strength values of SBR composites. As in the case of using each type of grass alone, the impact strength was increased with increasing the filler content up to 30%. Afterwards, the impact strength was increased when 40% of mixed grasses was added, then decreased. Properties like impact strength of PMCs are influenced by filler type, filler properties, filler content, matrix properties, and filler and matrix interface properties [27-29].



**Fig. 3:** Impact strength of SBR and the composites prepared with mixed grasses.

The addition of mixed grasses to SBR matrix caused slight increase in the hardness properties, as illustrated in Fig. 4. The increase in the mixed grasses content exhibited slight effect on the hardness properties of the prepared composites. Similar to the effect of the addition of individual grasses, the hardness increased with increasing the mixed grasses content. This indicates that the hardness values of all the prepared composites were higher than neat SBR. Moreover, the addition of individual grasses resulted hardness properties higher than the addition of mixed grasses. The hardness properties of composites with individual grasses were in the range of 7.30 to 9.9, while hardness properties of composites with mixed grasses were in the range of 7.35 to 8.10. This was expected because the addition of fillers into a polymer increases the stiffness and hardness of the composite [30-31]. The higher the filler incorporated in the PMCs, the harder the material would be, and the more rigid it becomes. Many studies [30-34] were reported similar increase of hardness with increasing the filler content. Essentially, the hardness of PMCs is associated to the hardness

polymer matrix, hardness of filler and filler-polymer interactions [35].

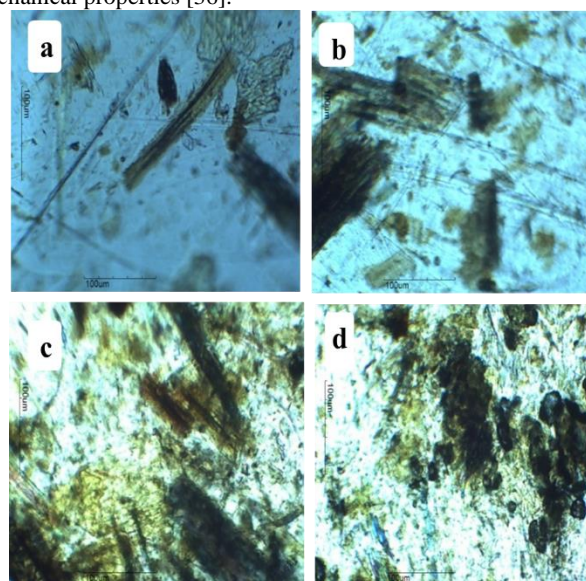


**Fig. 4:** Micro hardness of SBR and SBR composites with mixed grasses.

### Morphological properties

Fig. 5 illustrated the optical microscope images of all the composites prepared in the study with mixed grasses. There was no holes and filler pull outs from the SBR matrix were observed in these images.

Bad filler distribution was observed by composite with 10% mixed grasses content had, as shown in Fig. 5a. In contrast, composites with 20 and 30% mixed grasses content had relatively better filler distribution, as illustrated in Fig. 5b and c. Whereas, composite with 40% mixed grasses (Fig. 5d) clearly showed the formation of filler aggregation. Commonly, the filler aggregation effects poorly the filler efficiency as reinforcing material, resulting PMCs with poor mechanical properties [36].



**Fig. 5:** Optical microscope images of the prepared composites loaded with a) 10% mixed grasses, b) 20% mixed grasses, c) 30% mixed grasses and d) 40% mixed grasses.

This could may be the reason for the poor mechanical properties of composite with 40% mixed grasses content. According to the above presented measured results and previous published results [13], mixed grasses appears to be relatively better than individual grasses in producing PMCs with good strength properties. On the other hand, individual grasses were appearing to be better than mixed grasses in producing PMCs with relatively better impact strength and hardness properties. In both cases (individual or mixed grasses), the best content of grasses deliver acceptable mechanical properties is in the range of 20-30%. Future work should be directed to evaluate; thermal and morphological properties of these composites.

#### 4. Conclusion

In this present study, the effect of using local mixed Libyan grasses (Halaf and Esbat) as a filler on the mechanical properties of SBR composites was investigated. This effect was investigated by preparing different composites with mixed grasses content. Also, a comparison study between using mixed and individual (from pervious study) grasses has been achieved. It should be emphasis that composites obtained from these grasses (individual or mixed) could possess acceptable mechanical properties. In this regard, the main conclusion of this study can be as a following:

- The stress at break was decreased with the addition of 10% mixed grasses, then increased as the mixed grasses content increased up to 30%. Later, it decreased again when 40% mixed grasses were added. The highest stress at break value was obtained by composites with 30% mixed grasses content.
- The elongation at break of the prepared composites was dramatically decreased with the addition of mixed grasses. This decrease in the elongation at break was increased with increasing the mixed grasses content.
- Impact strength showed to increase slightly when the mixed grasses was added. Increasing the mixed grasses content had no profound effect on the impact strength values of SBR composites.
- The prepared composites exhibited slight increase with the addition of mixed grasses. Increasing the mixed grasses content had minor effect on the hardness properties of these composites.
- The optical image of the prepared composite made 10% mixed grasses content showed bad filler distribution, while composites with 20 and 30% grasses content had relatively better filler distribution. On the other hand, composite with 40% mixed grasses content clearly showed the formation of filler aggregation.
- Mixed grasses appears to be relatively better than individual grasses in order to produce composites with decent strength properties. On the other hand, individual grasses appear to be better than mixed grasses in producing composites with relatively better impact strength and hardness properties.
- In both cases (individual or mixed grasses), the best content of grasses that might provide acceptable mechanical properties is in the range of 20-30%.

The use of these grasses (individual or mixed grasses), as reinforcements gives interesting alternatives for production of low cost and ecologically friendly PMCs and will add value to these local grasses.

#### 5. Acknowledgement

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