JMATPRO	Journal of Advanced Materials and Processing 10 (2): 47-58, Spring 2022 http://jmatpro.iaun.iau.ir 20.1001.1.2322388.2022.10.2.6.1

Research Paper

Using of Thermoplastic Polyurethane Granule (TPU) as a Reinforcing Phase and Self-Healing Agent in a Polymer Composite Resin Epoxy to Exhibit Mechanical Properties Recovery

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ARTICLE INFO

Article history:

Received 3 February 2022
Accepted 17 March 2022
Available online 1 April 2022

Keywords:
Self-healing
Polyurethane
Resin epoxy
Mechanical properties

ABSTRACT

In this study, for the first time, thermoplastic polyurethane granule (TPU) is used as a reinforcing phase and self-healing agent in a polymer composite epoxy resin (ER) to exhibit mechanical properties recovery. When the polymer composite is damaged or cracked, TPU granules are released at the site of damage and cause auto-repair of surfaces. Therefore, TPU granules with different composition percentages were mixed in silicon molds containing epoxy resin polymer composite. 4 samples with different TPU granules percentages were selected (A= 0 Wt.% TPU, B=10 Wt.% TPU, C=20 Wt.% TPU, and D=30Wt.% TPU). At first, making a deep cut in 4 polymer composite samples, the self-healing process and mechanical properties improvement are investigated by mechanical tests. In the self-repairing behavior of self-healing samples, it is observed that polymer composite samples with self-repairing agents of ER+20 Wt.% TPU granules had the highest self-healing efficiency (60.2%) compared to other specimens. A mechanical test shows that Sample C has a higher Young's modulus (4.837 MPa) and higher tensile strength (9.46 MPa). Also, the impact test illustrated Sample C has a higher impact energy of 7.1 (J/m). Therefore, sample C has the highest mechanical properties among self-healing samples.

Citation: Hosseini, M.; Ghasemi, A. (2022). Using of Thermoplastic Polyurethane Granule (TPU) as a Reinforcing Phase and Self-Healing Agent in a Polymer Composite Resin Epoxy to Exhibit Mechanical Properties Recovery, Journal of Advanced Materials and Processing, 10 (2), 47-58. Dor: 20.1001.1.2322388.2022.10.2.6.1

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1. Introduction

Recently, self-healing materials have become the most important research interest because of community awareness regarding the prolongation of the lifetime of materials, which can save natural resources. The self-healing of materials such as polymers, metals, and concrete was studied to improve the service life of these structures [1,2]. These materials can be divided into two distinct groups: intrinsic and extrinsic self-healing materials. In the first group, based on the action of an added phase or structure, intrinsic self-healing is a mechanism that is in the structure of the material [3-9]. Polymer composites are extensively used in a range of applications, and they have been noteworthy candidates to introduce the self-healing theory into up-to-date engineering materials [2,10]. Not only does this class of polymers improve the ability to heal damage autonomously, rehabilitate mechanical properties, and increase the lifetime of polymers, but also the utilization of highly sophisticated chemical and physical principles has entered the field of merchandise polymers [11, 12]. Self-healing of polymer composite materials can recover from damage without external stimuli and intervention. This sort of material is particularly suited for use in applications that are highly exposed to external impacts and fatigue loading [13,14]. Epoxy resins are kernel materials for self-healing because they can react with a diversity of curing agents or hardeners at various temperatures [15]. Furman et al. [16] introduced the synthesis and characterization of a new diamine curing agent (tetramethylated diaminodiphenylsulfone) for self-healing epoxy resins. Polyethylene-co-methacrylic acid (EMAA) [17,18] and Poly (ethylene-*co*-glycidyl methacrylate) (PEGMA) [19] are also used as healing agents in brittle epoxy polymer. Roy et al. [20] exploit mesoporous silica as an amine immobilizer to endow epoxy resin's healing functionality. Polyurethanes (PUs) are a versatile class of polymers used in both synthesis and applications, and they were extensively used in modern society [21-23] because they are widely used in fields such as aerospace, automotive industry, and architecture, self-healing Polyurethanes have recently become a major research issue [24,25]. Thermoplastic polyurethanes (TPUs), as block copolymers, are the most important class of polyurethanes [26,27] with many great properties such as prolonged mechanical strength, good flexibility in low temperature, prominent wear and abrasion resistance [28-31]. Luan et al. [32] investigated the mechanical and self-healing properties of thermoplastic polyurethane (TPU)-graphene composites. Park et al. [33] developed self-healing TPU at room temperature with robustness, stretchability, and durability surpassing those of current room-temperature self-healable materials. As seen in previous research, TPU granules have not been used as a self-healing in the Epoxy Resin. For the first time, the crack created in the Epoxy Resin (which reduces its mechanical properties) is supposed to be repaired by TPU granules.

In this work, a series of self-healing thermoplastic polyurethane and Epoxy Resin (TPU/ER) composites with different TPU contents were prepared. TPU granules with different composition percentages (0 Wt.%, 10 Wt.%, 20 Wt.%, and 30 Wt.%) were mixed in silicon molds containing epoxy resin polymer composite. When the polymer composite is damaged or cracked, TPU granules are released at the site of damage-causing auto-repair of surfaces by heating. For this purpose, a deep cut in polymer composite samples with different percentages of TPU granules was conducted. Then, all of the samples with different percentages of TPU were in the oven for heating. After that, the healing ability is understudied to study the effect on self-healing behavior. Moreover, the effect on mechanical properties is estimated by three tests: tensile test, impact test, and three-point flexural test.

2. Experimental

1.2. Materials and reagents

The materials used in Thermoplastic polyurethane/Epoxy resin (TPU/ER) composites included: Thermoplastic polyurethane granule (LARIPUR 7025, Coim Company, Italy), Epoxy resin liquid (EP411 Laminating & Casting, DSM Company, the Netherlands) and hardener Epoxy Resin EP411 from DSM Company, and silicon (RTV/2-4425) made in Canada.

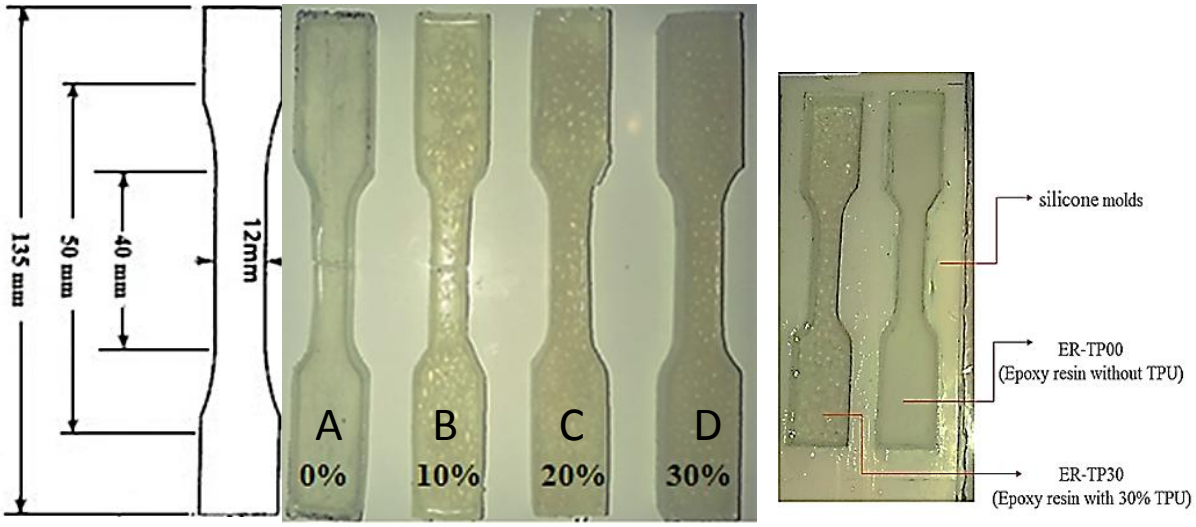

Fig. 1. Prepared samples of TPU/ER composites with different TPUs contents.

2.2. Method and fabrication of composites

At room temperature (20–25 °C), different ratios of epoxy resin (ER) and hardener (15:100) were mixed in a beaker with a mechanical stirrer for 2 minutes. TPUs were added to the mixture with different percentages. Fig. 1 shows that samples with different TPUs contents were named Sample- A, Sample B, Sample C, and Sample D, which correspond to TPU content of 0 Wt.%, 10 Wt.%, 20 Wt.%, and 30 Wt.%, respectively. The prepared liquid composites were cast into silicone molds already made for mechanical testing. Then, they were dried and rigid for two days at room temperature (20–25 °C). After two days, the specimens were taken out of the molds for mechanical tests. The test samples were also obtained by the same casting process mentioned above.

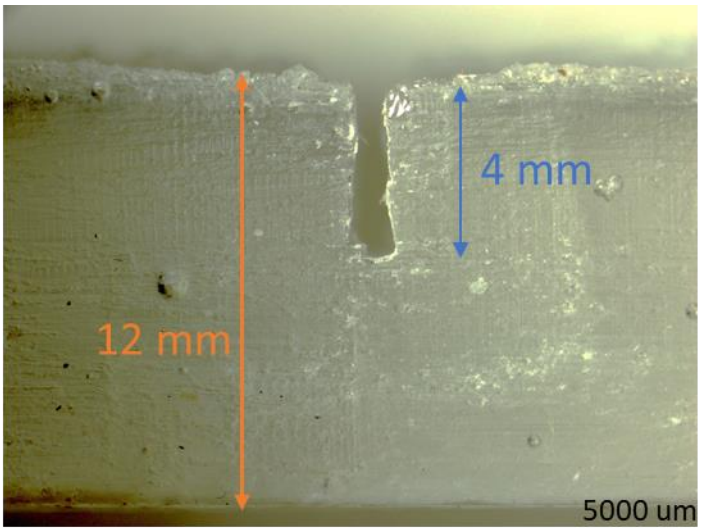

Fig. 2. Illustration of cut TPU/ER sample by the blade

2.3. Characterization of composites

For the healing test, samples of the composite were cut to a depth of 4 mm by using a razor blade in a frozen state to avoid any accidental deformation of the samples, as shown in Fig. 2. Care was taken to ensure that healing of the scratched or cut samples occurs mainly by heating. The temperature of healing was 150 °C by the oven (memmert–UNB40 made in Germany) for 80 min. After that, all the samples were extracted from the oven and froze at room temperature (20–25 °C) for 5 hours. The healed specimens were subjected to the mechanical test. The self-healing efficiencies were obtained on the recovery of tensile strength following Eq. (1),

$$\text{Healing of Efficiency (\%)} = \frac{Tensile\ Strength_{healed}}{Tensile\ Strength_{virginal}} \times 100 \quad (1)$$

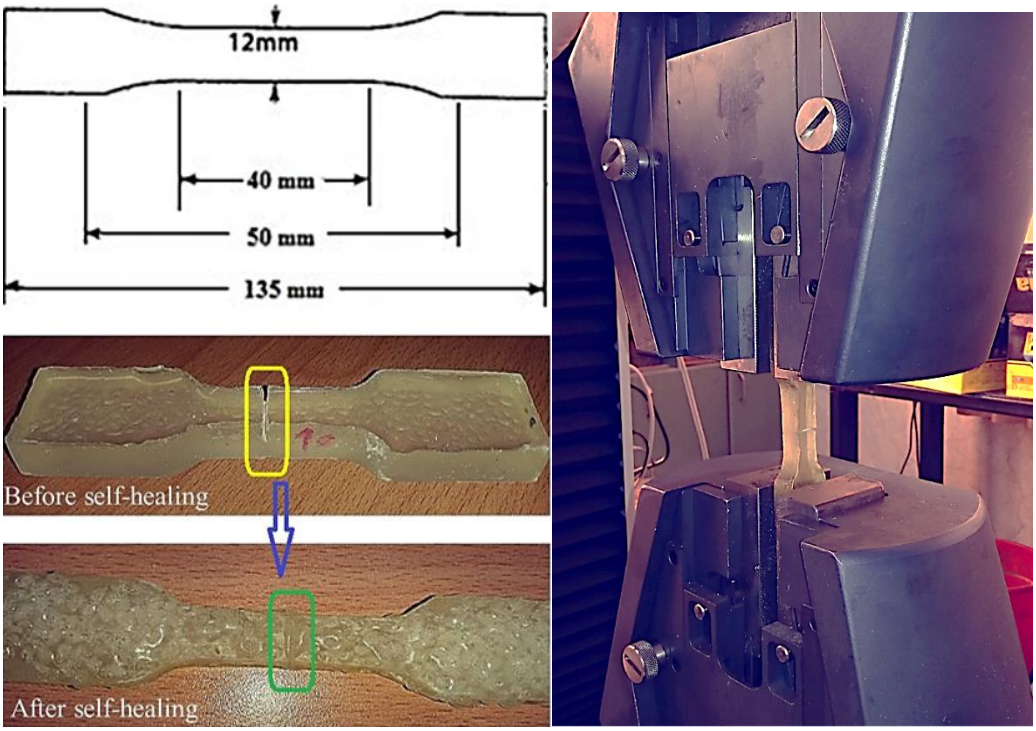

Fig. 3. Samples before and after self-healing, and their placement under tensile testing

Where tensile strength healed and tensile strength virginal are obtained in tensile test of healed and virgin materials for a given concentration of healing. The stress-strain relation, including tensile modulus of TPU/ER composites, is determined as a function of TPU content according to ASTM D3039/D3039M-17using an Instron-4486 made from Britain apparatus, as shown in Fig.3. A 2500 N load cell and an extensometer with a gauge length of 10 mm are used. A displacement control with a speed of 2.00 mm/s is applied. The tensile properties are measured for TPU/ER composites with 0, 10, 20, and 30 wt.% of TPUs.

The flexural test for self-healing is shown on the universal testing machine for three-point bending, as shown in Fig. 4. The equipped specimen is placed on rollers provided at the extreme ends. The specimen diameters are carried out according in proportion to the width of the specimen as specified by ASTM D790 on an MTS 810 Material Test System (MTS Systems Corp., Eden Prairie, MN) at a crosshead velocity of 2.00mm/min and a span of 50 mm. The load and deflection are shown on a computer. These samples are loaded until the first visual appearance of the crack.

The fracture behavior of the material at high strain rate levels was investigated by testing surfaces obtained after Charpy impact tests were performed according to ASTM D6110 standard, with digital pendulum impact indicator provided by SIT-50, the series of Santam Company, as shown in Fig. 5.

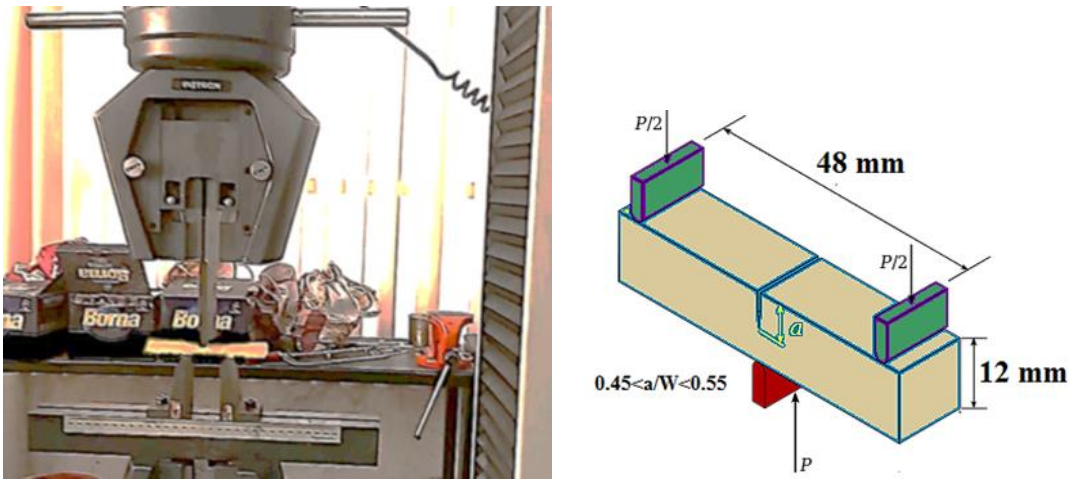

Fig. 4. Flexural test for self-healing and sample dimensional.

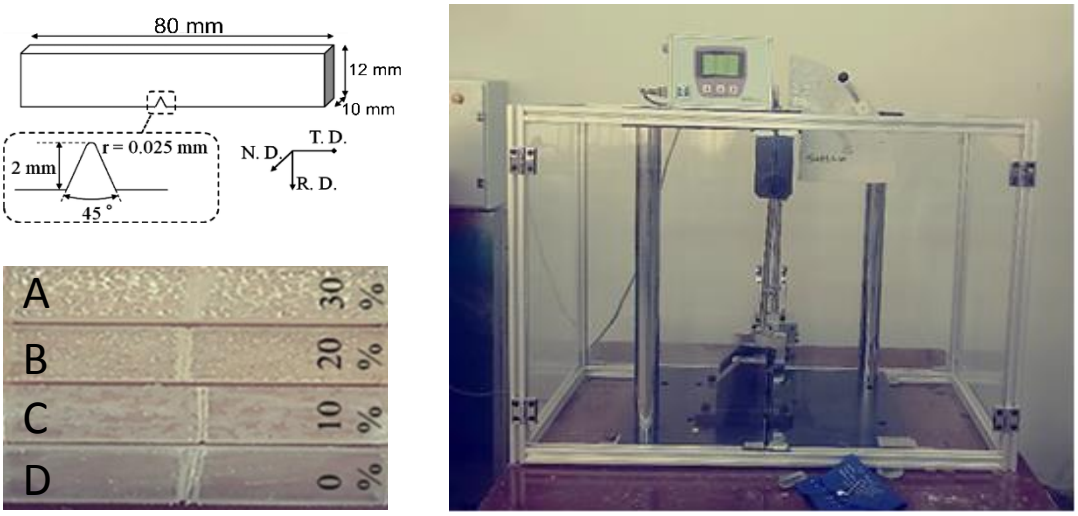

Fig. 5. Samples of Impact Test and Charpy impact test machine

The rectangular dimensions of the Charpy impact specimens with different TPU contents were 80 × 12 × 10 mm^3, as shown in Fig. 5. The speed of the impact test was 2.9 m/s with an incident energy of 5 J at room temperature and regular atmospheric conditions The XRD studies on the sample were carried out on an X-Ray Diffractometer, Phillips, Germany instrument. Fig. 6 represents the XRDs of the amorphous structure in the sample and that shows there is no crystal structure in it.

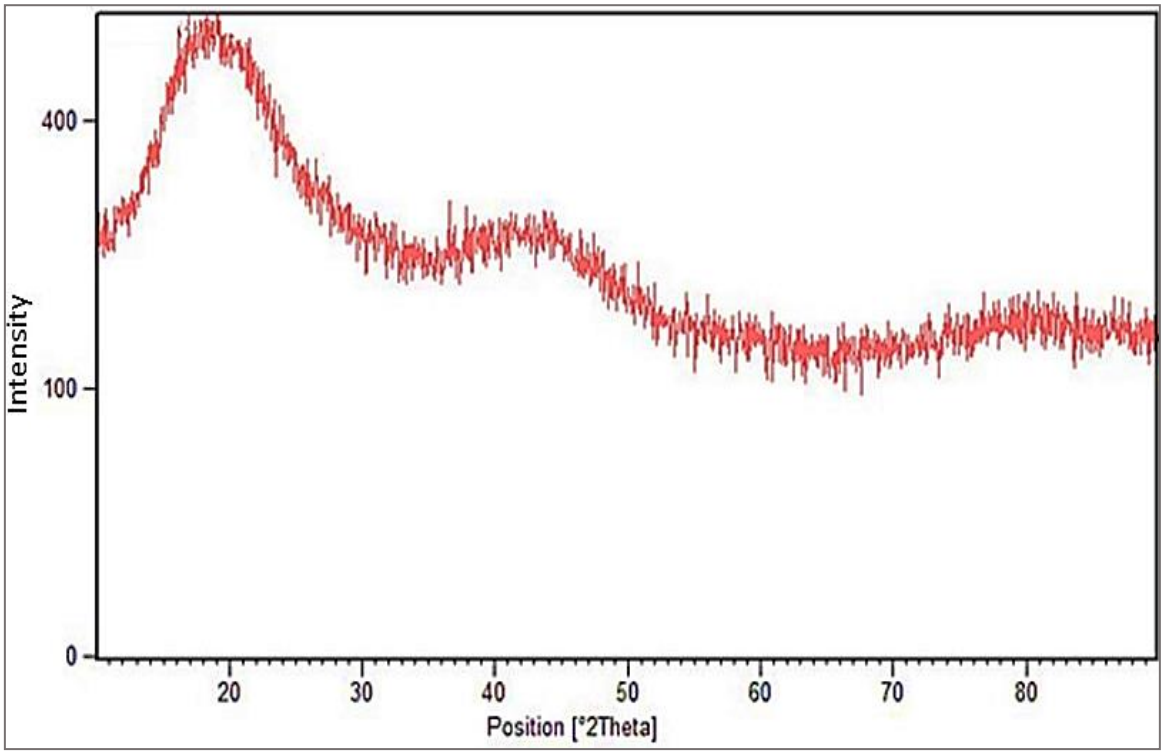

Fig. 6. XRD of TPU/ER

3. Results and discussion

3.1. Self-healing properties

The self-healing behavior of TPU/ER composites under thermal treatment was first investigated by observing the improvement of the cracks with the contents of different TPU granules. The healing test was conducted by heating the cut specimens to 150°C for 80 min while no external force was applied. Figs. 7-10 show the results of the surface analysis conducted by stereoscope (Olympus SZX16) before and after the healing process; then, qualitative observations were performed using image processing software. According to Figs. 7-10, with the removal of external force, gaps were sealed for all samples, and only trace lines were observed, indicating that the influence of introducing TPU on the self-healing of epoxy resin has been successful. Figs. 7-10 shows the evolution process of cracks in TPU/ER samples from various TPU values during heat treatment at 150°C.

It can be seen that the widths of all cracks are reduced with the extension of heat treatment time Fig. 7 shows that all TPU/ER samples except for Sample A have self-healing properties. The reason for this is that sample A was not used with TPU granules, as shown in Fig. 7. However, the crack in TPU/ER sample from Sample B does not disappear completely; rather, it only becomes part of the restored cracks, manifesting that the self-healing process has not finished, as illustrated in Fig. 8. The reason is that in Sample B, a little bit of granule has been used in comparison to the other two samples. By contrast, the healing degree is different for three specimens at the same heat treatment time. Cracks in TPU/ER samples from Sample C and Sample D completely disappeared when the percentage of the granules increased, as shown in Fig. 9 and Fig. 10. However, in the Sample C and Sample D specimens, recovery of a gap was better than that in Sample B and Sample A specimens. Although the cuts were closed, further tests were employed to evaluate the recovery of mechanical properties, which was mainly due to the different contents of polyurethane.

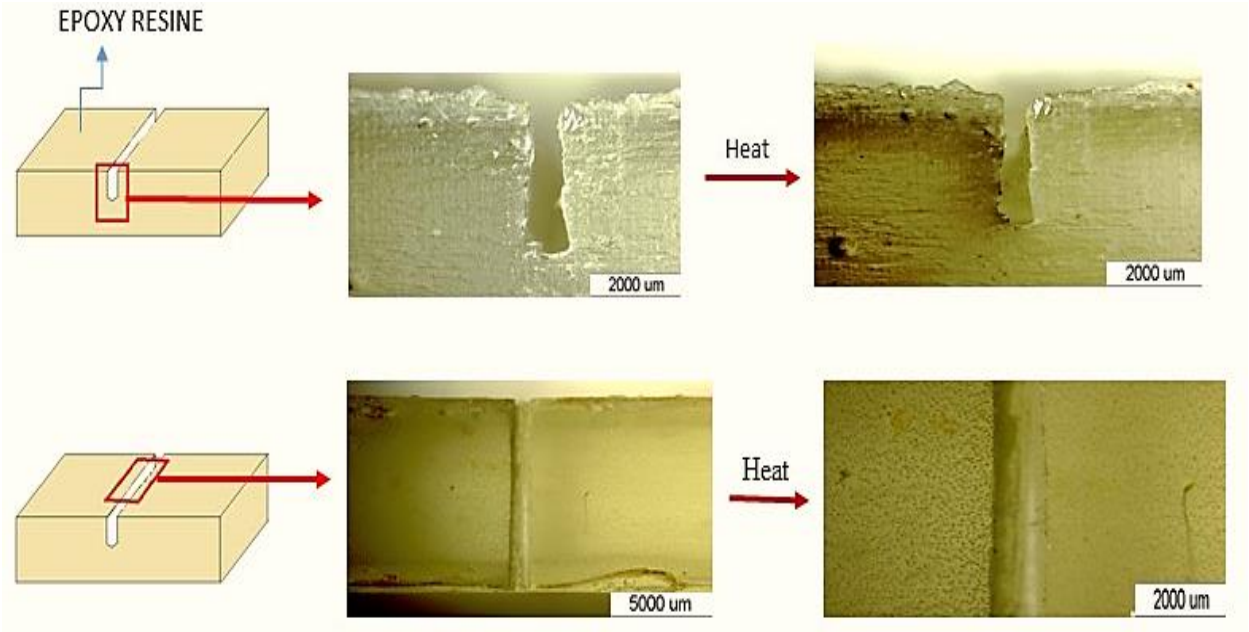

Fig. 7. Sample A shows no self-healing after a certain temperature and heating.

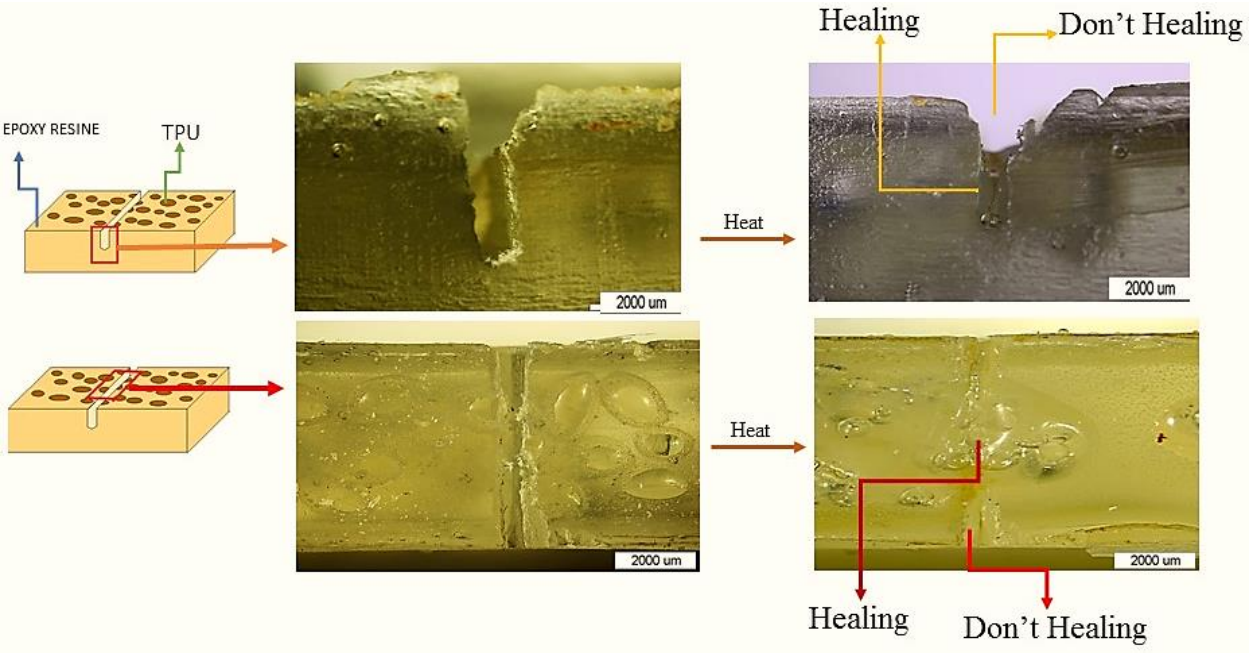

Fig. 8. Sample B shows healing after heating for a certain time, but healing is not complete

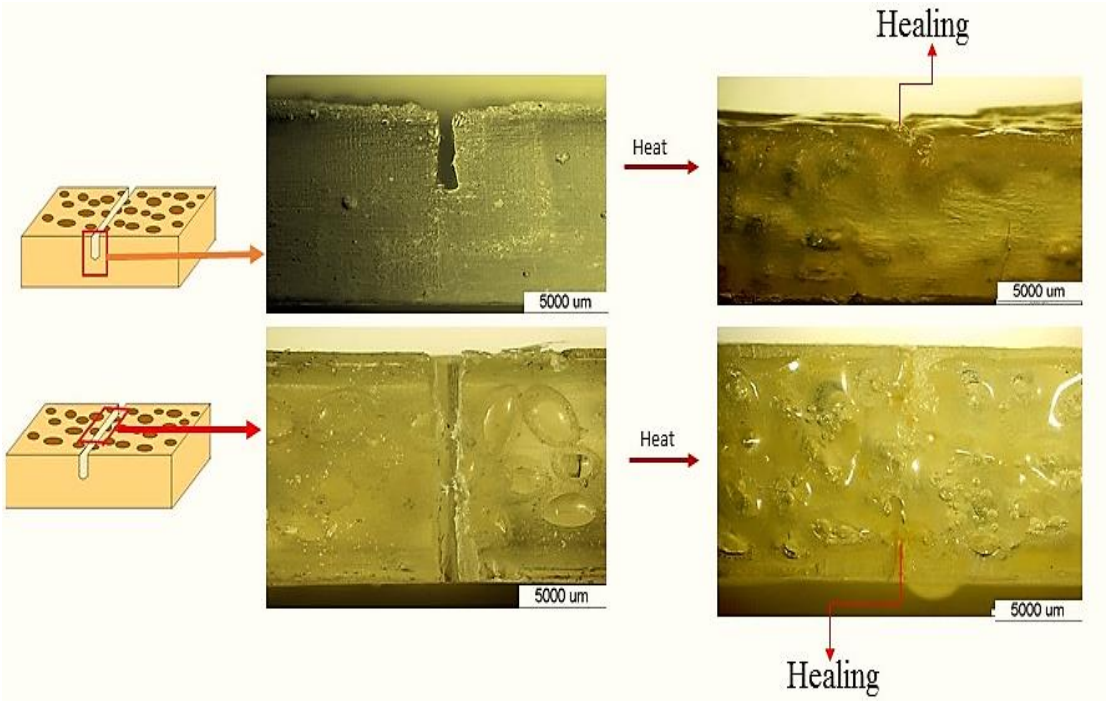

Fig. 9. Sample C illustrated that TPU was melted and self-healing after heating at a certain time was completed.

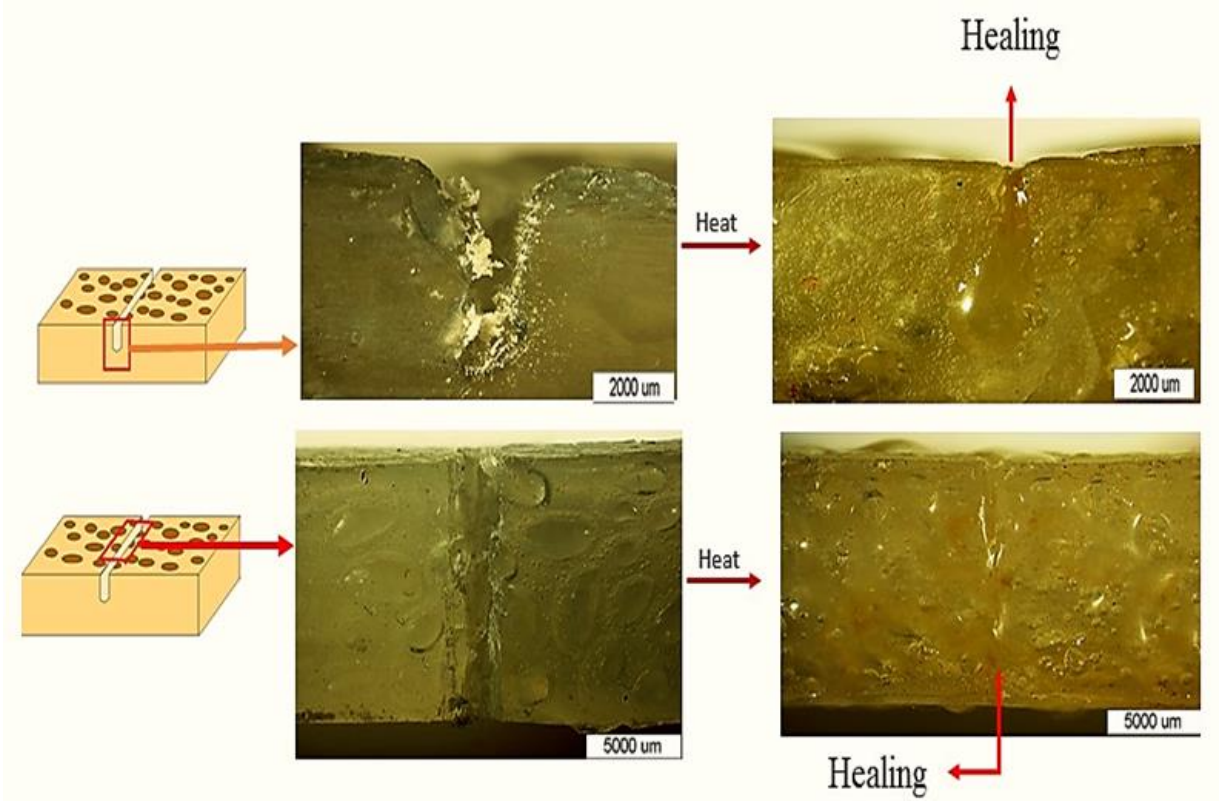

Fig. 10. Sample D shows that this sample like sample C was self-healed completely after heating in the same time (80 min, in150°C)

3.2. Flexural properties

The flexural properties of different self-healing groups (Sample A, Sample B, Sample C, and Sample D) are shown in Fig. 11. These data show that both the modulus and the strength of composites decrease with an increase in TPU content. The reason is that epoxy resin, used as a matrix in polymer composite specimens, has a brittle property. Furthermore, composite specimens have become more fragile by adding TPU granules to the epoxy resin matrix. Therefore, it should select the most suitable percentage of TPU granular composition by tensile test and impact test to obtain the highest mechanical properties.

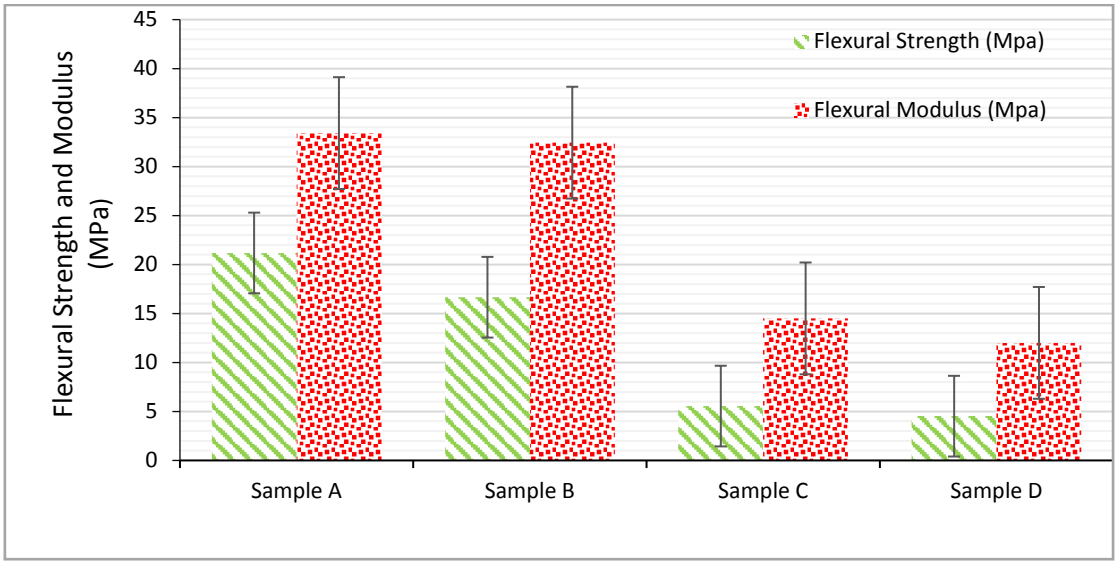

Fig. 11. Effect of processing on the flexural properties of TPU/ER composites

3.3. Healing efficiency properties

In this study, the stress at the first inflection point was used to calculate the healing efficiency of the epoxy resin. The experimental results for obtaining the ultimate tensile strengths of the healed specimens are illustrated in Fig. 12.

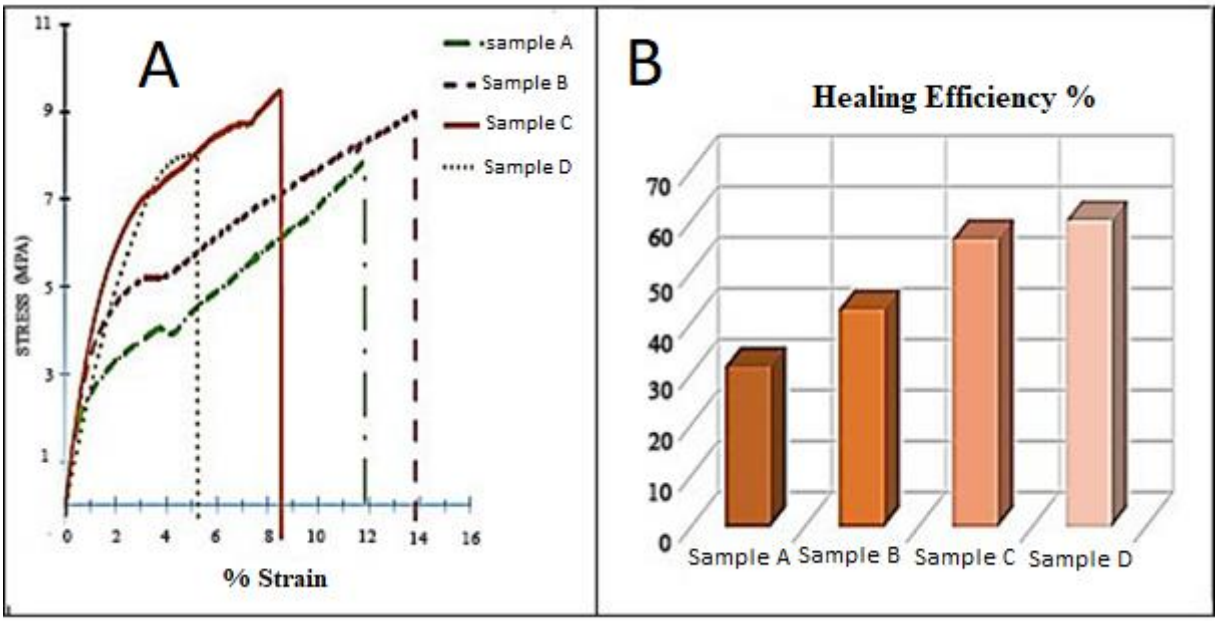

Fig. 12. A: Tensile strength of different TPU/ER structurally reinforced TPU composites of the healing specimens
B: The self-healing efficiencies were obtained on the recovery of tensile strength Eq. (1)

The experimental healing parameters for the four kinds of samples are listed in Table 1. As shown in Table 1, there is a clear difference in the ultimate tensile strength among the healed samples with variable healing conditions. By comparing Sample C and Sample A, and Sample B, we can see the higher ultimate tensile strength and Young's modulus. When comparing Sample C and Sample D, we can conclude that Sample C has the higher tensile strength, elongation at break, and Young's modulus. Moreover, the self-healing efficiency of the three samples is also different. Sample A exhibits quite a low healing efficiency of 31.6% because there are no TPU granules for sample repair. In contrast, Sample B from a lower TPU value exhibits excellent mobility because of the lower content of hard segments, which reduces the tensile strength and lead to sample shape deformation upon heat treatment. Thus, a relatively low healing efficiency of 42.7% is obtained. It is worth noting that a much higher self-healing efficiency than 56.5% and 60.2% is obtained for Sample C and Sample D. However, after the healing process, Sample C has a higher Young's modulus, tensile strength, and healing efficiency. Compared to other specimens, it indicates that a suitable TPU value has been selected for Sample C.

Table 1. Experimental Parameters for Four Specimens of Self-Healing Experiments

Sample	Healing Temperature (°C)	Healing Time (Min)	Ultimate Tensile strength (MPa)	Elongation at break (%)	Young's Modulus (MPa)	Healing Efficiency (%)
Sample A	150	80	7.84422	11.83687	3.948	31.5
Sample B	150	80	8.99474	13.83687	4.165	42.7
Sample C	150	80	9.465389	8.529687	4.837	56.5
Sample D	150	80	8.007692	5.22375	2.784	60.2

3.4. Tensile properties

The stress-strain relation for TPU/ER composites, obtained by tensile testing, as a function of the TPU content, is shown in Fig. 13. This figure represents stress-strain curves of virginal (as-prepared), cut (with a crack of 4 mm in depth), and healed (heat-treated at 150°C for 80 min) TPU/ER samples from different TPU values. Tensile test data and Young's Modulus of TPU/ER from different TPU values are listed in Table 2. It can be found from Fig. 13 and Table 2 that the tensile strength, elongation at break, and Young's modulus of each cut TPU/ER sample with a crack, will decrease as compared to those of the virginal specimen. On the contrary, the tensile strength and elongation at break of each healing TPU/ER sample with healing increase compared to those of cut specimen Young's modulus healing of Sample A and Sample D decrease compared to cut TPU/ER samples. However, all three mechanical properties of each healed TPU/ER samples are lower than those of virginal ones, indicating that all TPU/ER samples exhibit certain self-healing performances while cracks cannot be repaired completely.

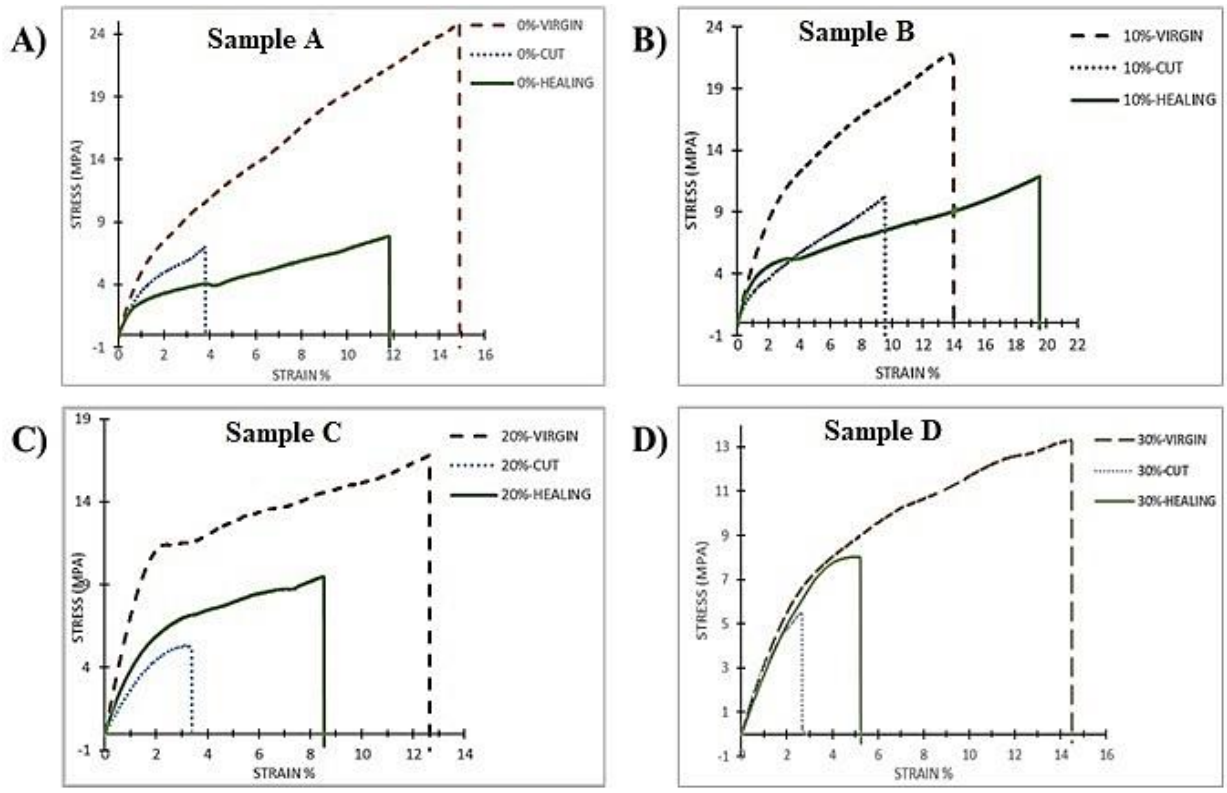

Fig. 13. Representative stress-strain curves of TPUER composites as a function of different TPU contents. A= stress-strain curves Sample A (with out TPU granules in epoxy resin), B = stress-strain curves Sample B (ER+10 Wt.% TPU granules), C= stress-strain curves Sample C (ER+20 Wt.%TPU granules), D= stress-strain curves Sample D (ER+30 Wt.% TPU granules)

Moreover, a comparison of mechanical properties of TPU/ER samples from different TPU values indicates that the tensile strength and Young's modulus of Sample B and Sample C are better than the other two samples in healing.

Table 2. Tensile test data and Young's Modulus of TPU/ER from different TPU values.

Sample from different TPU values	Condition	Tensile strength(MPa)	Elongation at break (%)	Young's Modulus (MPa)
	Virgin	24.89247	14.89375	5.583
TPU/ER (Sample A)	Cut	7.007532	3.805312	4.054
	Healing	7.84422	11.83687	3.948
	Virgin	21.0226	13.97219	5.693
TPU/ER (Sample B)	Cut	10.19753	9.544375	3.554
	Healing	8.99474	13.83687	4.165
	Virgin	16.78831	12.63875	7.311
TPU/ER (Sample C)	Cut	5.229481	3.377812	2.758
	Healing	9.465389	8.529687	4.837
	Virgin	13.28292	14.4875	3.373
TPU/ER (Sample D)	Cut	5.504717	2.66475	3.135
	Healing	8.007692	5.22375	2.784

3.5. Impact test

The experimental results concerning the impact properties of the tested samples for four self-healing TPU/ER samples are presented in Fig. 14. The Charpy impact test was carried out to evaluate the impact energy of the healing TPU/ER composites. The obtained results revealed that Sample C has higher impact energy than the other sample because not only is Sample C completely self-healing, but also the TPU content is appropriate in the Epoxy resin.

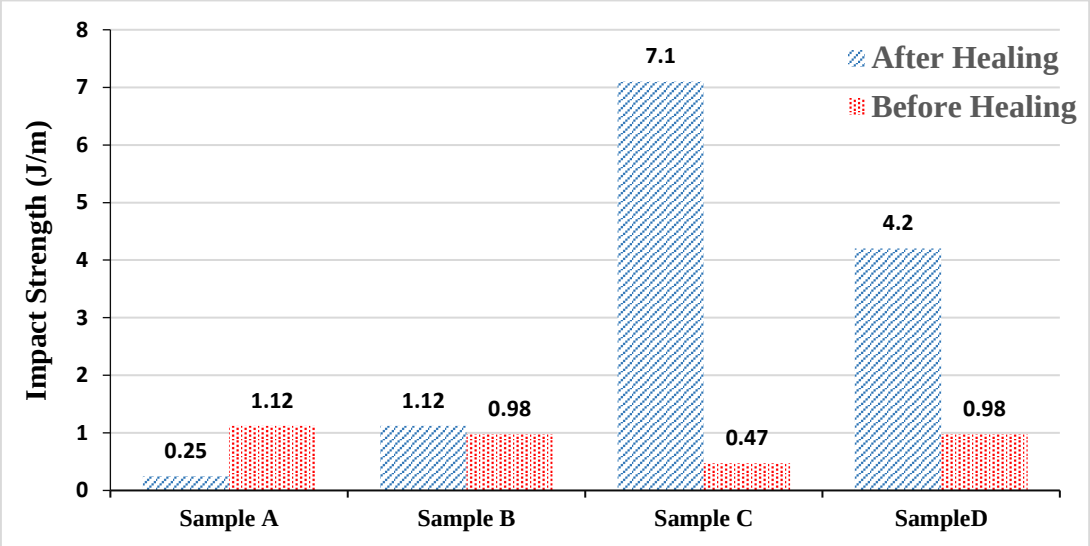

Fig. 14. Representative impact test of TPU/ER composites

4. Conclusions

The study of self-healing is of great significance for epoxy polymers. Epoxy polymers are applied in all aspects of life, including aerospace, automotive, and electronic fields. Epoxy polymers are also widely used in construction and transportation fields, such as, building materials and epoxy asphalt [34].

In the present paper, a series of self-healing TPU/ER composites were prepared. TPU/ER composites with a certain amount of TPU were successfully able to heal cracks automatically and improve structural function via a granule embedded in the matrix. Mechanical tests show the incorporation of an appropriate content of TPU in the matrix to reinforce and recover the mechanical properties of these samples after damage. Results of the tensile test and impact test revealed that incorporating 20Wt.% of TPU in Sample C would promote mechanical

properties in comparison with other samples. In the before research has shown to be capable of recovering between 50% and 70% of its pre-fracture strength after healing [35]. Stereoscope observation shows that cracks with 4 mm in depth in TPU/ER samples can be healed upon heat treatment of 150°C for 80 min, while the self-healing efficiency can reach as high as 60%. They further confirm that the excellent self-healing performance of TPU/ER is due to the combined actions of thermo-reversible TPU reaction and thermal movement of molecular chains. Consequently, our present understanding shows that the TPU bond and its thermal reversibility contribute much to the improvement of the mechanical property while the thermal movement efficacy of molecular chains acts as an auxiliary recovering force that speeds up the whole healing process.

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