Investigating the self-healing behaviour of under-aged and 60Sn-40Pb alloy reinforced aluminium hybrid composites

O.P. Oladijo[a*], M.O. Bodunrin[b] , K. Sobiyi[c], N.B. Maledi[d], and KK. Alaneme[b]

[a]Department of Chemical, Materials and Metallurgical Engineering, Botswana International University of Science and Technology, Palapye, Botswana
[b]Department of Metallurgical and Materials Engineering, Federal University of Technology Akure, Nigeria
[c]Department of Mechanical Engineering Science, University of Johannesburg, South Africa
[d]School of Chemical and Metallurgical Engineering, University of the Witwatersrand, South Africa

*Corresponding email: seyiphilip@gmail.com

Abstract

An experimental investigation was carried out to assess the self-healing characteristics of aluminium hybrid composites reinforced primarily with mixed proportion of silica sand and bamboo leaf ash. Charpy impact test samples with 45° notch were machined from the aluminium hybrid composites and a ϕ1mm hole was drilled at 1mm away from the notch. Two different self-healing treatments were given to the test samples prepared from the composites. In the first treatment, a 3mm diameter hole was drilled along the sample and a low melting point alloy (60Sn-40Pb) which served as secondary reinforcing material was pierced into the hole and then heat treated at a temperature of 250°C. The second treatment that was adopted involved subjecting the test samples to two-steps under-ageing treatment. The first step involved ageing at 160°C for 15 minutes and quenched in water. Thereafter, the second ageing treatment was carried out at 50°C for 24h. The samples were then subjected to Charpy impact testing. The results show that the composites had lower absorbed energy compared with Al-Mg-Si alloy. Self-healing treatment improved the energy absorbed in healed samples when compared to the damaged samples. Although, the under-aged samples had a slightly higher absorbed energy in comparison with the samples containing 60Sn-40Pb, the presence of silica sand and bamboo leaf ash did not have significant influence on the absorbed energy. The highest healing efficiency obtained using the low melting point alloy approach was 61%.

Keywords: Aluminium hybrid, self –healing, bamboo leaf, Charpy impact test

1. Introduction

The possibility of achieving varieties of property combinations that are not possessed by conventional monolithic materials has been the drive for the increasing interest in the design and development of composites materials [1]. Aluminium matrix composites (AMCs) remain the most studied and widely utilised MMCs due to the ease of production, attractive properties and ease of post fabrication heat treatment [1, 2]. Some of the properties possessed by AMCs include high specific strength, high specific stiffness, and corrosion resistance. AMCs are produced using two major techniques; stir casting and powder metallurgy, for a wide range of applications which include automotive, aerospace, recreation, military and marine industries [3, 4].

Stir casting remain the most utilised method for producing AMCs commercially [4]. This technique involves reinforcing the matrix material in its liquid state with continuous ceramic fibres or discontinuous particulates [5]. The technique is well established but porosity observed in these materials is of serious concern [6, 7]. Porosity develops in AMCs due to poor interfacial bonding between the matrix and reinforcing material thus serving as potential nucleation site for mechanical and corrosion failures.

In previous studies, interfacial bonding between the metallic matrix and ceramic reinforcements has been improved by using magnesium as a wetting agent [7] and by manipulating the stirring speed and time [8] in stir cast composites. Furthermore, post fabrication deformation, careful selection of reinforcing materials and combination of two or more reinforcing materials have also been reported as a viable means of reducing porosity in composites[5], [9]. It is worthy of note that, all these methods resulted in appreciable reduction in porosity. However, these efforts still fall within the paradigm of the traditional prevention management philosophy. One major drawback of the traditional prevention management philosophy in material processing is that the materials eventually fail due to damage development [10].

Recently, self-healing approach are being developed to ensure that damage that occurs in materials systems due to propagation of defects like porosity, micro cracks formation or flaws during in-service operations, are managed through a self-repair process [11]. This self-repair process can be likened to the sealing of open wounds by blood clotting in biological systems [11]. Self-healing materials are generally classified into three groups: capsule based, vascular, and intrinsic self-healing materials [12]. The healing mechanism of the capsule based and vascular self-healing composites are similar while for intrinsic, the repair is based though inherent reversibility of bonding of the matrix polymer. For capsule and vascular self-healing materials, the structure similar to a tunnel network, in which various functional liquids are embedded in the matrix polymer. The gaps are filled with these functional liquids when a crack occurs [12]. The target objective of the healing agents (material contained in a capsule or vascular network) is to ensure that materials mechanical properties are restored substantially up to a level of >70% of its original value by a self-repair process which is triggered when damage occurs [10]. Some of the benefits of incorporating self-healing concepts into commercial processing of materials includes: prolonged service life of materials, reduction in down time of industrial process and adaptability to low cost processing in development of self-healing materials [10 – 13].

These benefits have allowed significant progress to be made in the synthesis of self-healing polymer and polymer based composites [14]. Likewise, a number of articles have reported self-healing capabilities in ceramics and ceramic based composites [15]. However, the science of self-healing is yet to be thoroughly understood, and as a result, commercial application is still very limited. Specifically, self-healing characteristics in metallic systems and metal matrix composites have not been widely reported due to strong metallic bonds and lower diffusion rates in metallic systems. This is quite unlike polymers where diffusion rate is high enough to make healing agents fill up damage nucleation sites such as micro voids and cracks [10].

The introduction of ceramic materials as reinforcements in metallic alloys to develop composites introduces more porosity into the composites. Since porosity serves as nucleation points for failures in materials systems used for structural applications, the introduction of self-healing design concept as a damage management strategy becomes appealing if catastrophic failures are to be prevented.

This research aims to investigate the adaptability of self healing phenomena to the development of aluminium based composite. The existing approaches used in achieving self-repair characteristics in metallic system include: formation of under aged precipitates [16]; use of shape memory alloys as reinforcements [17] and use of low melting point alloys as reinforcements (healing agent) in high melting temperature metallic systems [18]. Since the use of shape memory alloys as reinforcing materials in metallic systems is expensive, this experimental study focussed on the use of under-aged precipitates and low melting point alloy as low cost approach of achieving self-healing in Aluminium based composites.

2. Materials and Method

2.1 Materials

The materials utilised in this research are Al-Mg-Si alloy as the matrix, bamboo leaf ash and Igbokoda silica sand as the primary reinforcing materials and low melting point 60Sn-40Pb alloy as the secondary reinforcing material (self healing agent). The compositions of the matrix alloys and primary reinforcement are presented in Tables 1-3.

2.2 Production of aluminium hybrid composites

Detailed description on the production process of the aluminium hybrid composites used in this study is presented in [18]. In summary, the process started with determining the quantities of bamboo lead ash (BLA) and silica sand (SS) required for producing 10wt. % reinforcement in the aluminium matrix composites. The reinforcement consisted mixed proportions of BLA and SS in ratios 0:1, 1:3, 1:1 and 3:1 respectively. Prior to casting, the BLA and SS particles were initially preheated separately at a temperature of 250 °C to eliminate dampness which helps reduce particle clotting and improves wettability and dispersion of the particles in the molten Al-Mg-Si alloy. The Al-Mg-Si alloy matrix was melted in a gas-fired crucible furnace at about 750 °C ± 30 °C (above the liquidus temperature of the alloy).The molten alloy was then cooled in the furnace to a semi solid state at a temperature of about 600 °C. The pre-heated BLA and SS particles along with 1wt. % magnesium were then charged into the semi-solid melt at this temperature (600^{o}C) and stirring of the slurry was performed manually for 5 to 10 minutes. Magnesium was added to further improve wettability of the reinforcing particles by the composites. The composite slurry was then superheated to 800^{o}C ± 50^{o}C and a second stirring performed using a mechanical stirrer. The stirring operation was performed at a speed of 400rpm for 10minutes before casting into prepared sand molds fitted with metallic chills. The designation of the aluminium hybrid composites produced is presented in Table 4.

2.3 Self-healing experimental studies

2.3.1 Under-aged precipitates approach

This approach was carried out in accordance with Hautakangas et al. [16]. The parameters that yielded the optimum self-healing efficiency using this approach in the study carried out by Alaneme and Omosule [10] was adapted. Charpy impact testing was used in place of uniaxial tensile testing to evaluate the self-healing characteristics of the composite samples. The composites samples were machined to standard specifications (55x10x10mm) as shown in Figure 1. The samples were notched at an angle of 45° and a radius of 0.3mm, ϕ1mm hole was drilled at distance of 1mm from the notch; both serving as damaged sites . The samples were subjected to solution heat-treatment at a temperature of 500˚C for 1 hour and quenched in water. The samples were then thermally aged at 160˚C for 10 minutes followed by rapid cooling in a water bath. A second ageing treatment was then performed on the test samples at temperature of 50˚Cfor a period of 12 hours before quenching in water. Thereafter, the samples were subjected to impact loading on a Charpy impact tester. The absorbed energy was then recorded. The Charpy impact test sample preparation, test procedures and analysis of results were in accordance with ASTM E23-16b [20].

2.3.2 Use of 60Sn-40Pb Low Melting Point Reinforcement

For studies on healing induced by incorporation of 60Sn-40Pb low melting alloy as secondary reinforcement, the Charpy test samples (Figure 2) were incorporated with the 60Sn-40Pb secondary reinforcements fitted within the ϕ3 mm holes drilled longitudinally along the samples. Again, one set of the samples had ϕ1mm pre-cracked hole drilled 1mm away from the 45° notch and a radius of 0.3mm to pierce the low melting point 60Sn-40Pb alloy embedded within the aluminium matrix. This was done to simulate crack formation on the aluminium hybrid composite. The samples were afterwards subjected to impact testing. These set of pre-cracked samples were referred to as damaged samples. Another set of pre-cracked 60Sn-40Pb reinforced Al-Mg-Si alloy based composite samples were prepared and referred to as healed samples. The healed samples were sealed with kaolin at both ends and heated at a temperature of 250°C for 10 minutes before air cooling. The sealing of the samples at both ends with kaolin was to prevent draining of 60Sn-40Pb alloy during heating. Finally, virgin samples which were not pre-cracked were also prepared and referred to as virgin samples. The samples were all subjected to impact testing and the absorbed energy was recorded and was used to compute the healing efficiency. The test was repeated twice for each composition for reproducibility and reliability of results.

Self-healing efficiency achieved using both treatments was determined using the absorbed energy values according to equation 1 [10].

$$U\ eff. = \frac{U\ healed}{U\ virgin} \times 100\% \qquad (1)$$

Where U_{healed} is the energy absorbed by the healed specimen, U_{virgin} is the energy absorbed by the virgin specimen and $U_{eff.}$ is the self-healing efficiency.

2.4 Microstructural examination

The full composite microstructure has been discussed elsewhere [19]. The fracture surfaces of the test samples after testing were examined visually and on the scanning electron microscope to determine the fracture features.

3. Results and discussion

3.1 Self healing induced by secondary ageing approach

The results of the impact tests carried out on the unreinforced Al-Mg-Si alloy and primary reinforced Al-Mg-Si/BLA-SS hybrid composites is presented in Figure 3. It is observed that the unreinforced alloy (Sample A0) absorbed higher energy than the aluminium composites. Alaneme and Omosule [10] reported an improvement in the mechanical properties of Al-Mg-Si when subjected to secondary ageing treatment at under-aged temperature of 50°C. The improvement was ascribed to the secondary precipatitation of Mg_2Si phase from the under-aged Al-Mg-Si alloy containing solute atoms in excess of equilibrum concentration. These phases are formed at precipitate free zones which were nucleation sites within the matrix where precipitation did not occur during the under ageing primary preciptation treatment.

The lower absorbed energy observed in the composites could be as a result of the introduction of the primary reinforcing materials (BLA and SS) which are basically ceramics and known to be inherently brittle. The presence of the ceramic particles and the ceramic particle/matrix interface can accentuate a state of stress triaxiality within its vicinity which increases susceptibility to brittle mode fracture characteristics (reflected by a low impact energy) [21]. The presence of porosities (also regarded as crack precursors) as reported previously for these composite grades [19] may also have contributed to the relatively lower impact energies. Also, it was reported that, reinforcing materials when introduced into some aluminium alloy matrix, impede ageing response due to depletion of magnesium [22]. This could lead to the reduction in the amount of precipitates formed, making it difficult for self healing to effectively take place. There is need for futher studies to provide a better understanding.

Although the difference between the energy absorbed by the composites is marginal, it can be oberseved that sample A2 absorbed higher impact energy than the other aluminium hybrid counterparts. This can be attributed to the lower porosity levels in the composite as reported in our previous studies on this composite grade [19].

3.2 Self-healing induced by using 60Sn-40Pb low melting point alloys

Table 5 show the impact energy absorbed by the 60Sn-40Pb secondary reinforced aluminium hybrid composite samples and the healing efficiencies. It is observed that the impact energy absorbed by the samples subjected to self-healing treatment was higher than the damaged samples that were not treated. However, the difference in the self-healing efficiency of the samples was not significant. The highest healing efficiency of 61% was observed in sample A2.

It is worth mentioning that the self-healing efficiency of the composites is low when compared to values reported in [10]. There are many factors and parameters which can affect the performance or properties of a composite. Factors like method of manufacturing, reinforcement type and mix ratios of the reinforcement make it increasingly difficult to directly compare results of composite from other sources. Figure 4 which compares the impact energy absorbed in the virgin composites (Table 5) and the composites subjected to secondary ageing at under-aged temperature (Figure 4); reveal that more energy is absorbed by the composites that were under-aged than the virgin composites. Perhaps it could be the interaction between the primary and secondary reinforcements which is not considered in this study that is responsible for the low energy absorbed. For instance, the interaction could affect interfacial bonding between the 60Sn-40Pb low melting point alloys and the Al-Mg-Si/ BLA-SS composites which may have contributed to the low healing efficiencies observed.

3.3 Scanning Electron Microscopy

The secondary electron images of the fracture surface of sample A2 subjected to secondary ageing and self-healing treatment is presented in Figure 5(a-b). The features on the images are similar, dimples and micro-voids are clearly evident on the fracture surface. Also observed in fractographs taken at higher magnifications in Figure 6(a-b) are cleavages indicating a mixed mode fracture in the materials, as expected in composites materials that has ductile aluminium matrix and brittle reinforcements.

4. Conclusion

The self healing characteristics in aluminium hybrid composites was invesitgated using two different approaches: secondary aging at under-aged temperature and secondary reinforcement with low melting point alloy. The unreinfirced alloy had a higher self-healing efficiency as compared to aluminum hybrid composites (composites with secondary aging) resulting from the presence of ceramic particle/matrix interface in the composites, thus increases susceptibility to brittle mode fracture characteristics. The composites subjected to secondary ageing treatment absorbed a slightly higher impact energy when compared to samples that were reinforced with 60Sn-40Pb low melting point alloy. The highest self-healing efficiency that was obtained in samples reinforced with 60Sn-40Pb low melting point alloy was 61%. Poor interfacial bonding and presence of porosity in the composites are the likely reasons for the low self-healing efficiency observed in the composites. SEM images confirmed the presence of dimples, micro voids and cleavages which are features of mixed mode fracture.

5.0 Acknowledgement

This work was supported by the Botswana International University of Science and Technology, Palapye and the the University of the Witwatersrand, South Africa.

References

[1] D. K. Das, P. C. Mishra, S. Singh, and R. K. Thakur, Properties of ceramic-reinforced aluminium matrix composites - a review, *Int. J. Mech. Mater. Eng.* (9) (1) (2014) 1–16.

[2] K. K. Alaneme and M. O. Bodunrin, Mechanical behaviour of alumina reinforced AA6063 metal matrix composites developed by two step-stir casting process, *Acta Tech. Corviniensis* 4 (3) (2013) 105–110.

[3] J. M. Torralba, C. E. da Costa, and F. Velasco, P/M aluminium matrix composites: an overview, *J. Mater. Process. Technol.* 133 (1–2) (2003) 203–206.

[4] J. Hashim, L. Looney, and M. S. J. Hashmi, Metal matrix composites: production by the stir casting method, *J. Mater. Process. Technol.* 92–93 (1999) 1–7.

[5] M. O. Bodunrin, K. K. Alaneme, and L. H. Chown, Aluminium matrix hybrid composites: a review of reinforcement philosophies; mechanical, corrosion and tribological characteristics, *J. Mater. Res. Technol.* 4(4) (2015) 434–445.

[6] J. Hashim, L. Looney, and M. S. J. Hashmi, Particle distribution in cast metal matrix composites—Part I, *J. Mater. Process. Technol.* 123(2) (2002) 251–257.

[7] J. Hashim, L. Looney, and M. S. J. Hashmi, The enhancement of wettability of SiC particles in cast aluminium matrix composites, *J. Mater. Process. Technol.* 119 (1–3) (2001) 329–335.

[8] S. B. Prabu, L. Karunamoorthy, S. Kathiresan, and B. Mohan, Influence of stirring speed and stirring time on distribution of particles in cast metal matrix composite, *J. Mater. Process. Technol.* 171 (2) (2006) 268–273.

[9] K. K. Alaneme, Mechanical behaviour of cold deformed and solution-heat treted alumina reinforced AA 6063 metal matrix composites, *West Indian J. Eng.* 35 (2) (2013) 31–35.

[10] K. K. Alaneme and O. Omosule, Experimental studies of self-healing behaviour of under aged Al-Mg-Si alloys and 60Sn-40Pb alloy reinforced aluminium metal matrix composites, *J. Miner. Mater. Charact. Eng.* 3 (2015) 1–8.

[11] M. D. Hager, P. Greil, C. Leyens, S. van der Zwaag, and U. S. Schubert, Self-healing materials, *Adv. Mater.* 22 (2010) 5424–5430.

[12] B. J. Blaiszik, S. L. B. Kramer, S. C. Olugebefola, J. S. Moore, N. R. Sottos, and S.R. White, Self-healing polymers and composites. *Ann. Rev. Mater. Res., 40*, (2010) 179–211.

[13] S. Van der Zwaag, Self-Healing materials. An alternative approach to 20th centuries of materials science. Berlin: Springer, 2007.

[14] S. K. Ghosh, Self-healing Materials: Fundamentals, Design Strategies, and Applications. John Wiley & Sons, 2009.

[15] M. Madhan and G. Prabhakaran, Self-healing Ability of Structural Ceramics – A Review, in Trends in Intelligent Robotics, Automation, and Manufacturing, S. G. Ponnambalam, J. Parkkinen, and K. C. Ramanathan, Eds. Springer Berlin Heidelberg (2012) 466–474.

[16] S. Hautakangas, H. Schut, S. van der Zwaag, P. E. J. Rivera Diaz del Castillo, and N. H. van Dijk, The role of the ageing temperature on the self-healing kinectics in an underaged AA2024 aluminium alloy, in First international conference on self-healing materials, The Netherlands, 2007.

[17] P. K. Rohatgi, Al-shape memory alloy self-healing metal matrix composite, *Mater. Sci. Eng. A*, 619 (2014) 73–76.

[18] J. B. Ferguson, B. F. Schultz, and P. K. Rohatgi, Self-Healing Metals and Metal Matrix Composites, *JOM* 66 (6) (2014) 866–871.

[19] M. O. Bodunrin, O. P. Oladijo, O. O. Daramola, K. K. Alaneme, and N. B. Maledi, Porosity measurement and wear performance of Aluminium hybrid composites reinforced with silica sand and bamboo leaf ash, *Ann. Fac. Engeneering Hundeoara-Int. J. Eng.* 16 (1) (2016) 232–238.

[20] ASTM E23-16b, Standard Test Methods for Notched Bar Impact Testing of Metallic Materials, ASTM International, West Conshohocken, PA, 2016, www.astm.org

[21] Alaneme, K. K., Sanusi, K. O. Microstructural characteristics, mechanical and wear behaviour of aluminium matrix hybrid composites reinforced with alumina, rice husk ash and graphite, *Engr. Sci. Technol.,* 18(3) (2015) 416–422.

[22] A. Daoud and W. Reif, Influence of Al_2O_3 particulate on the aging response of A356 Al-based composites, *J. Mater. Process. Technol.*, 123 (2) (2002) 313–318.

List of Tables

List of Figures

Table 1: Elemental composition of Al-Mg-Si alloy [18]

Element	Wt.%
Si	0.45
Fe	0.22
Cu	0.02
Mn	0.03
Mg	0.50
Cr	0.03
Zn	0.02
Ti	0.02
Al	98.71

Table 2: Composition of Bamboo leaf ash [18]

Compound	SiO_2	Fe_2O_3	Al_2O_3	TiO_2	CaO	K_2O	Na_2O	MgO
Wt. %	94.24	0.87	1.47	0.48	0.90	0.82	0.75	0.40

Table 3: Composition of Igbokoda silica sand [18]

Compound	SiO_2	Fe_2O_3	Al_2O_3	TiO_2	CaO	K_2O	MgO
Wt. %	75.9	1.22	4.13	0.20	7.47	5.62	1.85

Table 4: Designation of aluminium hybrid composites produced via stir casting

Sample Designation	Composite Composition	Reinforcement mix ratio
A0	Al-Mg-Si	0:0
A1	Al-Mg-Si/10 wt.% SS	0:1
A2	Al-Mg-Si/2.5 wt.% BLA- 7.5 wt.% SS	1:3
A3	Al-Mg-Si/5.0wt.% BLA -5.0 wt.% SS	1:1

A4	Al-Mg-Si/7.5wt.% BLA –2.5wt.% SS	3:1

Table 5: Energy absorbed during impact testing and self-healing efficiency of secondary reinforced aluminium hybrid composites

Samples	**U virgin (joules)**	**U damaged (joules)**	**U healed (joules)**	**U. eff. (%)**
A1	12 ±1,4	5 ±0	7.0 ±0	58
A2	14 ±1,4	4.5 ±0.7	8.5 ±0.7	61
A3	11 ±0	5.5 ±2.1	6.0 ±0	55
A4	8,5 ±0.7	4 ±1.4	4.5 ±0.7	53

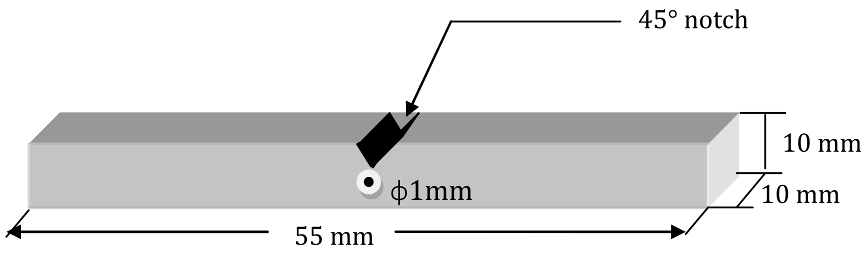

Figure 1: composite samples for Charpy impact testing (radius of notch tip =0.3mm)

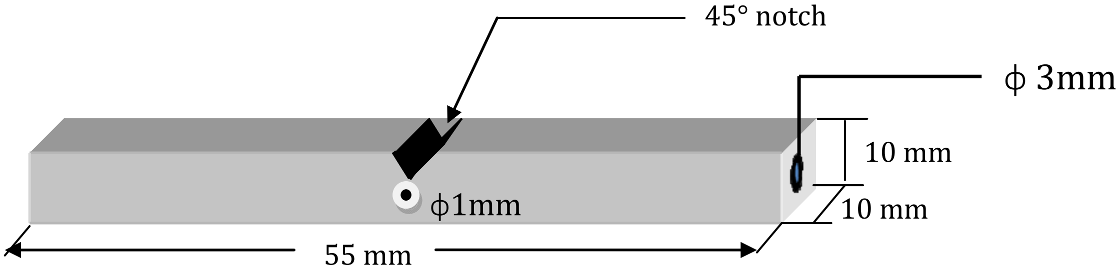

Figure 2: composite samples for Charpy impact testing (radius of notch tip = 0.3mm)

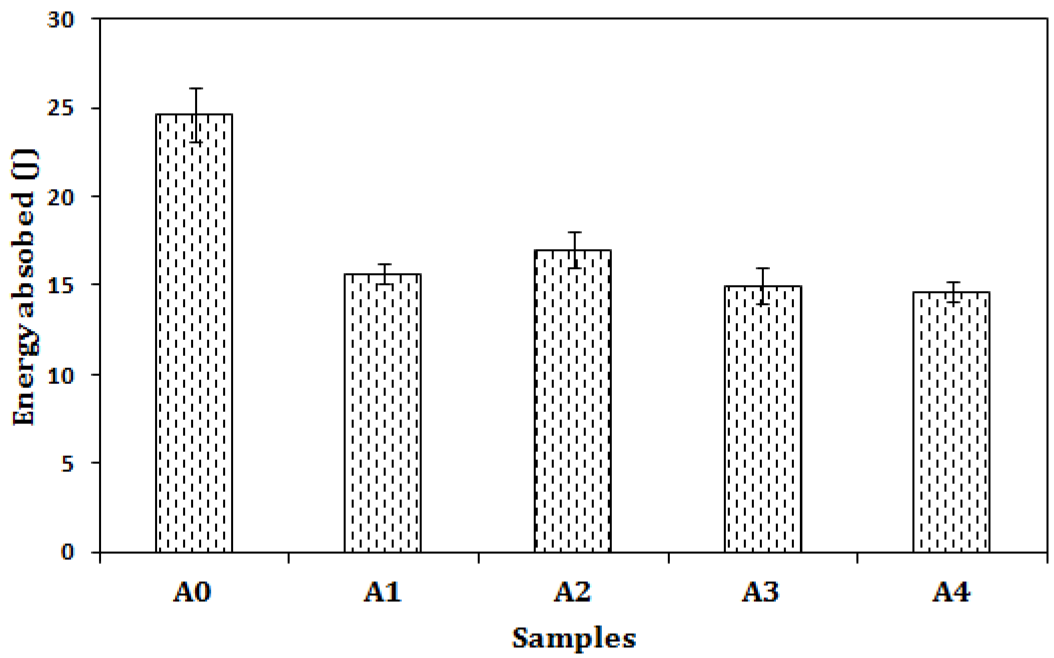

Figure 3: Energy absorbed by Al-Mg-Si alloy and aluminium hybrid composites during impact testing

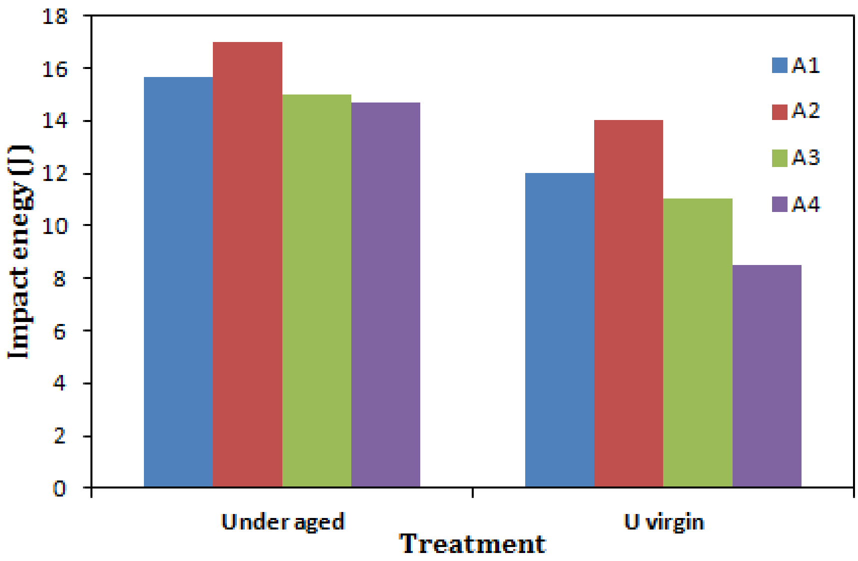

Figure 4: Impact energy absorbed by Al-Mg-Si/SS-BLA hybrid composites under different self-healing treatments

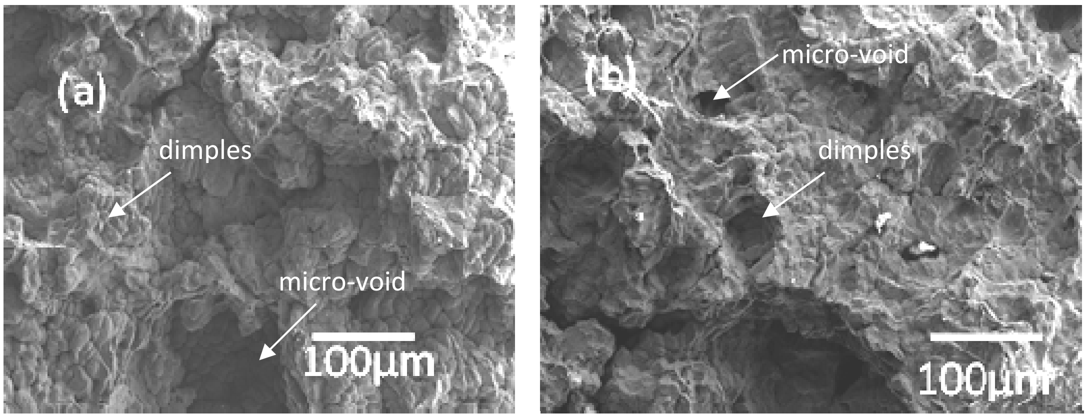

Figure 5: secondary electron images of the fracture surface of Al-Mg-Si/7.5% SS-25% BLA composites (a) secondary aging (b) 60Sn-40Pblow melting point alloy.

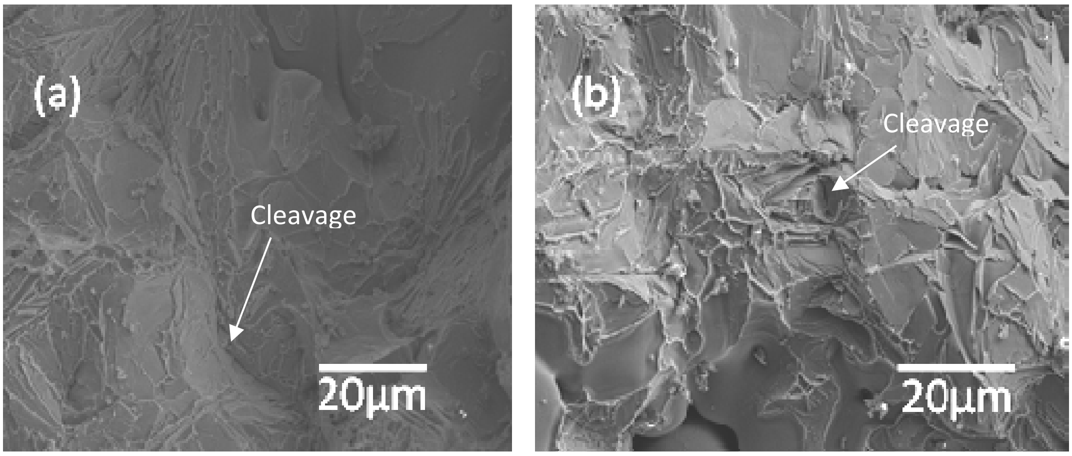

Figure 6: secondary electron images of the fracture surface of Al-Mg-Si/7.5% SS-25% BLA composites taken at higher magnification (a) secondary aging (b) 60Sn-40Pblow melting point alloy