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Full Length Research Article

ALUMINIUM-COPPER-CULLET METAL COMPOSITE: A SECONDARY SOURCE OF ALUMINIUM FROM WASTE MATERIALS

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ABSTRACT
In this work, Aluminium composites were fabricated from recycled materials, using recycled Aluminium (Al) obtained from scrap door and window frames, copper (Cu) wire scrap and cullet powder (CP). The Al-Cu-CP metal composite was fabricated via stir casting method due to its simplicity and economic importance. The composite and the unreinforced Al were analysed for physical, mechanical and morphological properties. The analyses indicated that the chemical composition of the products covered all the initial components. The results of hardness tests were found to have improved significantly due to the impact of the reinforcement materials. The hardness increased from 40.625 HV of the unreinforced Al to 124.704 HV for Al-Cu-CP metal composite. The microstructural analysis of the composite revealed appreciable distribution of the reinforcement materials within the Al matrix. Evolution of new phases was also revealed which furthermore contributed immensely towards enhancing the strength of the composite. It is obvious that the properties of the Al-Cu-CP composite were relatively enhanced significantly. Thus, it could be used where high strength with less density composite is required such as automobile brake shoes and brake disc as a result of the upgraded mechanical properties.

Keywords: Aluminium, Composite, Copper, Cullet powder, Percentage composition, Stir casting.

INTRODUCTION

The importance of metal matrix composites (MMCs) cannot be separated from their structures in addition to their superior properties compared to conventional engineering materials. These composites combine the greater strength in reinforcements and the ductility from the metallic matrices. The inclusion of the reinforcement usually makes composites fit for tribological practises. MMC is an environmental friendly technology which embraced advancement in the tribological industry owing to its various benefits. A number of factors are closely studied when considering the choice of materials to be used in order to perform a particular engineering function (Joseph and Babaremu, 2019; Magibalan *et al*., 2018; Ajibola *et al*., 2015). The major factors which determine the structure and properties of the composites are interfacial relations between matrix, its reinforcement and nature of bonding between the two components (Behera *et al*., 2020). Modern composite materials form a significant percentage of the engineering materials' market, varying from everyday goods to sophisticated niche uses. MMCs proffer greater combination of properties in such a way that today no existing monolithic material can show and for this reason are increasingly being used in the aerospace and automobile industries. The major advantage MMCs possess over other materials lies in the enhanced strength and hardness on a unit weight basis (Ramesha *et al*., 2018; Rao and Ramanaiah, 2017; Laxminarayana and Reddy 2014). Aluminium is one of the leading elements in fabrication of metal matrix composites (MMC) in spite of its better strength, excellent wear resistance and good dimensional stability (Sakthivelu *et al*., 2020). Al-based MMCs, reinforced with ceramics or metallic particles were developed as a substitute to materials with superior strength–weight and strength–cost ratios and high stiffness, which have great effects on improving the mechanical properties of the composites (Tash and Mahmoud 2016). Al-based composites are gaining more consideration in both structural and automotive industries. These Al-MMCs have advantages over conventional engineering materials like ferrous materials especially in the reduction of weight which leads to lower moment inertia, less consumption of fuel and also better corrosion and wear resistance (Roshan *et al*., 2013). When observed from both scientific as well as technological perspective, their admirable mechanical properties with reasonable cost effectiveness in terms of production which makes the Al-MMCs very suitable candidates for a range of applications (Saravanan *et al*., 2015).

The major setback in the general production and applications of composites is the cost of the raw materials. Though, the problem is immediately being attended by scientists and researchers toward the development of composites using reinforcement materials which are comparatively low-cost. One of the ways is through the use of recycled materials for the fabrication of composite materials. Using recycled materials for composite fabrication gives relief to the non-renewable resource and getting rid of the scrap materials off the environment, this is another advantage apart from producing relatively low-cost materials (Agbeleye *et al*., 2017; Oladele and Okoro, 2016; Abd Rahim *et al*., 2015; Kumar and Shankar, 2012). Thus, the recycling process is more economical as it saves a lot of energy. Al has confirmed to be reclaimed, refined and recycled for further use at a relatively lower energy cost of 5% of the required amount to produce the same quantity of the metal from its ore. Al recycling has provided major economic and environmental benefits (Al-Imari, 2014).By using complete recycled components, the cost of Al production through secondary sources will be reduced further. This material complements Al supply from primary resources.

Hence, this research can be considered as a secondary source of Al. The research could be an additional way of removing solid wastes from the environment. It is also eco-friendly research. The process could also be viewed as a way of promoting technological sustainability and it will also support the zero-waste campaign for promoting green technology.

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MATERIALS AND METHOD

Fabrication

Al scraps from door and window frames were cut into smaller sizes with the average size of 30 × 19 mm dimension. Vertical band-saw machine was used in cutting the Al scrap. The Al scrap cuts were placed in a steel crucible which was put into an electric furnace. When the Al scraps become melted, it was then poured into a steel mould to produce an ingot from the Al scraps. The Al ingot was removed from the cylindrical mould after it was allowed to cool to room temperature. The unreinforced Al ingot produced was used to fabricate the composite. After the unreinforced Al got melted, a variable percentages of reinforcement materials were separately added, that is, 5 - 20 wt. % of preheated Cu pieces and CP in equal proportion and stirred thoroughly.

Microhardness Test

Hardness measurement was performed via Vickers hardness tester. The test was carried out by loading of 980.7 mN for a period of 10 seconds in each and every indentation. The indenter was removed and the indentation shape was measured. The smaller the indentation appeared on the surface of the sample, the more resistant or the harder the material. The hardness value is determined by measuring the depth or the area of the indentation. The automated microhardness tester is connected to a computer which displays the already computed hardness values. Samples from both the matrix and the composite fabricated were cut to the desired size, ground with abrasive papers of different grit sizes ranging from 240 to 1000.

Microstructural Analysis and XRD

The microstructural analysis of the samples was conducted with the guidance of the American Society for Testing and Materials (ASTM E-1382) standards for metallurgical samples' preparation. The etching process of the samples was done as described under the section of "Chemical Compositions analyses". The specimens of Al-Cu-CP composites. The ground specimens were polished with alumina suspension and etched using Kroll's reagent. Each of the specimens was immersed in the etchant for 10 seconds with mild agitation. Table 1 presents the components of the Kroll's which was used as the etchant. The etched specimens of the unreinforced Al as well as the Al-Cu-CP composite were subjected to metallographic analysis with aid of SEM and XRD. The etched specimens were then subjected to Scanning electron microscopy and optical microscopy to examine their microstructures.

Table 1: Chemical Composition of Kroll's reagent (ASTM E3 11)

Component	Volume (cm^3)
Distilled water	92
Trioxonitrate (V) acid	6
Hydrofluoric acid	2

Energy Dispersive X-ray (EDX) spectroscopy was carried out on the etched surfaces of the samples to uncover the phases present in the composite. This analysis was carried out in accordance with the concept of Bragg's law to determine the intensity of peaks using the relationships of 2-theta (2θ). The phase patterns present were identified by comparing the pattern of all the samples with those in the database of the EVA software.

RESULTS AND DISCUSSION

Hardness Analysis

Hardness values of materials give an impression on their strength and durability (Kadir*et al.*, 2017). The values of the hardness testing ranged from 49.0 HV at 5% to 124.7 HV at 20% reinforcement. That is, it increases as the wt. % of the reinforcement materials increased. However, the hardness value of 25 wt. % was found as 74.2 HV which shows that the hardness value has declined when the reinforcements were increased from 20 wt. % to 25 wt. %, as illustrated in Figure 2. The hardness value of the composite has decreased by 40.53 % when the reinforcements' percentage composition was increased from 20 wt. % to 25 wt. %. Agbeleye *et al*. (2017) also observed a similar trend that is an increase in hardness of Al composite as the reinforcement material was increased until the optimum composition was attained afterwards the hardness value declined.

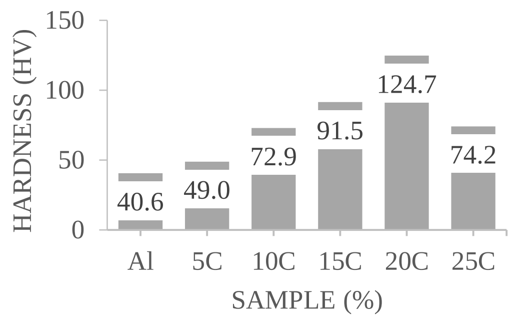

Figure 2: Hardness values of Al-Cu-CP composites of different wt. %

This finding indicates that the composite at 20 % reinforcement has the highest hardness value among all the Al-Cu-CP composite compositions. The improvement in the hardness value at 20 wt. % is up to 200 % rise when compared with the hardness of the unreinforced Al, 40.6 HV. The reason for the improvement in the context of highest value of hardness of Al-Cu-CP metal composite is due to the homogeneous dispersion of the reinforcement phases owing to the thorough stirring as well as the addition of the wetting agent which resulted in uniform dispersion of the reinforcement materials in the course of the fabrication. A similar observation was reported by Haque *et al*. (2016). The uniform dispersion of reinforcement materials in fabricated composite improves the resulting mechanical properties remarkably (Behera *et al*., 2020). The improvement in the hardness of the Al-Cu-CP metal composite observed in this study could also be attributed to the high hardness of the reinforcement materials and strong interfacial bonds between the Al matrix and the reinforcements' phases, metallic bond in $CuAl_2$ and dative bond in $Cu_{15}Si_4$. A similar observation was made by Ramesha *et al*., 2018 and, Nuruzzaman and Kamaruzaman (2016). Another factor that is also worth considering is the existence of hard reinforcement particles acting as obstacles in the motion of dislocation thereby making it difficult for the grains of the Al matrix to slide over one another which also improves the hardness of the material as earlier observed by Jain *et al*.(2016) and, Mallireddy and Siva (2020). Mahdi (2017) reported that hardness is a fundamental property in the selection and fabrication of MMCs. Evolution of new phases due to reaction between the Al matrix and reinforcement materials contributed to the enhancement in the strength of the composite as earlier reported by Tash and Mahmoud (2016).

Microstructural Analysis

The strength and the performance of Al-MMCs rely on the homogeneity of the particulate reinforcement phase(s) in their base matrix while agglomeration and sedimentation of the particulate reinforcement phase(s) during fabrication could cause a non-homogeneous composite with inferior properties (Sharma *et al*., 2015). Figure 3(a - e) shows SEM micrographs of Al-Cu-CP metal composites containing different weight percentages of reinforcements' compositions but fabricated under the same conditions via stir casting method. It is evident that the reinforcements' particles are well dispersed within and along the grain boundaries. The essence of this SEM analysis is to reveal the level of distribution of the reinforced materials. It could be noticed that the porosity increases as the weight percentage of the reinforcements are being increased. Similarly, it was observed that there are fewer pores alongside the grain boundaries compared to the observed porosity within the grains in all the samples. This is more obvious from the composite containing 15 wt. % to 25 wt. % as shown in Figure 3(c-e). A similar observation was earlier reported by Fathy *et al*. (2015).

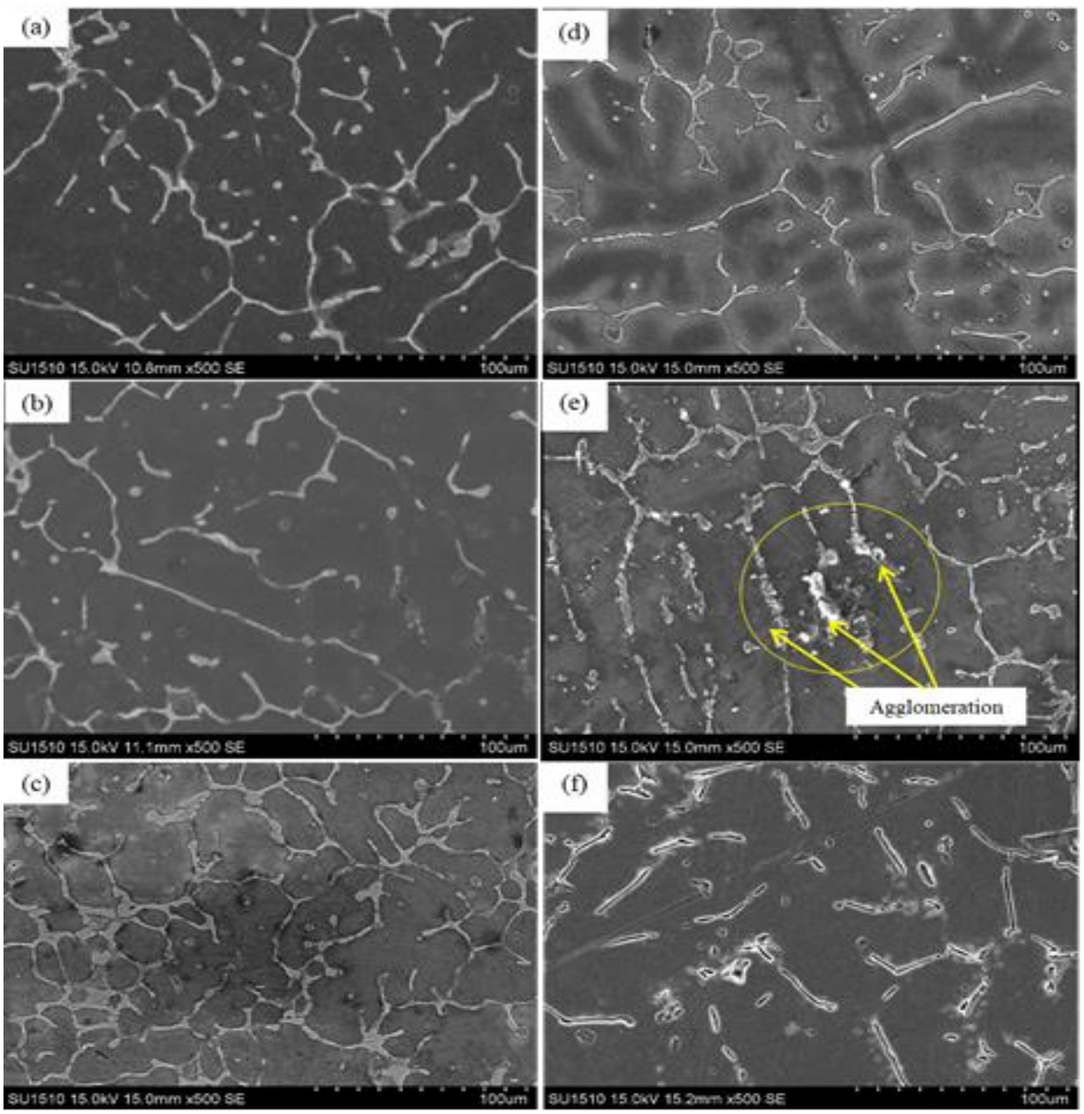

Figure 3: Surface morphology by SEM of Al-Cu-CP metal composite with variable reinforcement percentages (a) 5 wt. % (b) 10 w. t% (c) 15 wt. % (d) 20 wt. % (e) 25 wt. % and (f) unreinforced Al

As the percentage of the reinforcement materials increased (to 25 wt. %), agglomeration of the reinforcement materials sets in and becomes glaring when viewed under the microscope, as presented in Figure 3. Due to the weight percentage of the reinforcement increase, the amount of Cu content in the matrix also increases which leads to the formation of an alloy with the Al matrix. At this stage, it forms an interface between the Al metal matrix and the Cu reinforcement. Hence, the resultant strength of sample with 20 wt. % of reinforcement materials is a combination of the alloy formation, reinforcement and good interface bonds. However, when the agglomeration reached a certain level, it diminishes the strength of the material. This is obvious as can be seen in the image of 25% due to the increase in the reinforcement's contents. This results to decline in the mechanical properties of the material, as well as deterioration in its microstructure as illustrated in Figure 3(e). A related observation was reported by previous scientists, Li *et al*. (2016); Madhusudan *et al*. (2016); Al-Imari (2014), and Canakci and Varol (2014), that the agglomeration of particulates multiplies the effects of the drop in strength. Thus, suggested that 20 wt. % of reinforcement material as the optimum composition of reinforcement in Al metal composite fabricated via stir casting method.

Phase Analysis of Al-Cu-CP Composite.
The XRD analysis of the Al-Cu-CP metal composites with varying compositions of reinforcement materials was carried out and the diffractograms have shown a gradual evolution of new phases in the pattern of the composite as presented in Figure 4. As the weight percent of the reinforcement increases from 5 wt. % to 25 wt. % evolution of new phases at different 2θ values were noted. Figure 4 shows that few phases have evolved around 2θ positions of 42.23° and 42.75°, 47.40° and 47.85°. These are the most notable and common features of the XRD patterns of all the composite samples with different wt. % of the reinforcement materials. The value of the d-spacing maintained a linear relationship with the percentage weight of the reinforcement composition. That is, it increases as the reinforcement percentage increases.

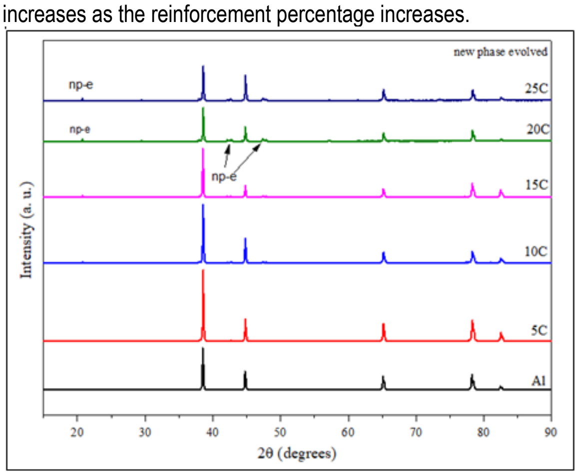

Figure 4: XRD results of Al-Cu-CP metal composite of variable Cu and CP compositions

The trend continued until 20C, which has the least d-spacing value. The decrease in d-spacing for the 20 wt. %is another factor responsible for the increase in hardness of the composite as earlier reported (Ahmad *et al*., 2017). Table 2 presents the detailed variations in the diffraction angle at (111) plane of Al-Cu-CP metal composite obtained due to changes in the reinforcement percentage compositions of the composites. The 2θ values of the first peak of (111) plane kept on changing with respect to change in the percentage composition of the reinforcements of the composites. The change in the peak intensity of the Al-Cu-CP metal composite at (111) is due to the change in orientation of its crystals which brought about change in its hardness property (Kuijpersa*et al*., 2003).

Figure 5 depicts a diffractogram of 20C which evolved new phases in contrast with the unreinforced Al. The new phases were closely studied and analysed to find out their chemical identities. The dominant peaks of the diffractogram still represent Al, indicating that the material is a composite of Al. Both Cu and Si were found present in their elemental forms and in combined forms as well. The emergence of Cu and Si peaks spotted in the XRD diffractogram reveals that there are some unreacted Cu and Si atoms within the α-Al grains, a similar observation was earlier reported by Tash and Mahmoud (2016). An intermetallic compound, Al-copper, $CuAl_2$ (also known as Khatykite) was found to be present in the Al-Cu-CP metal composite.

Table 2: Variations in (111) plane of Al-Cu-CP metal composite due to changes in reinforcement percentage composition

Sample	2θ (°)	Intensity (a. u.)	d-spacing (Å)
Al	38.510	15419	2.33588
5C	38.559	1844	2.33349
10C	38.559	1514	2.33302
15C	38.514	1258	2.33564
20C	38.551	12629	2.33276
25C	38.524	987	2.33503

This could be as a result of a reaction between the two metals in their molten forms at the eutectic region which is usually the reaction zone of composites due to the high concentration of the reinforcements at the zone. The formation of $CuAl_2$ in Al composites was earlier reported by Islak*et al*. (2016) and Kumar *et al*. (2012).

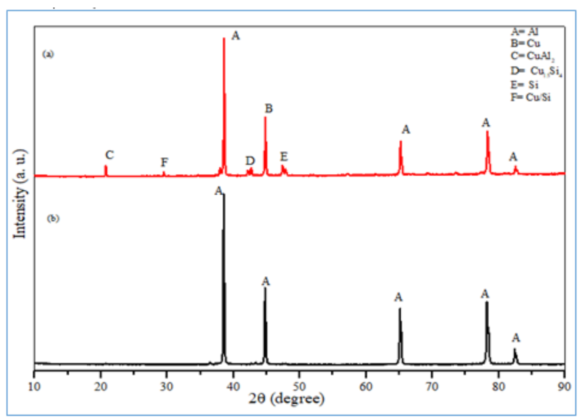

Figure 5: Diffractograms of (a) Al-Cu-CP metal composite (b) unreinforced Al

The reaction between Al and Cu could be represented in Equation (1) as:

$$2Al + Cu \rightarrow CuAl_2 \quad (1)$$

Another compound identified in the Al-Cu-CP metal composite was copper silicon with chemical formula $Cu_{15}Si_4$.

The reaction representing the combination of copper and silicon in the composite is represented by Equation (2) as:

$$15Cu + 4Si \rightarrow Cu_{15}Si_4 \qquad (2)$$

A peak representing Cu/Si phase was also found within the composite as Cu and Si, in their elemental forms (Cu, Si) and not in the combined state. This observation most probably found in the eutectic region whereby the Si atoms were embedded in the Cu rich zone. These new phases evolved are said to be responsible for the overall improved properties of the composite especially the intermetallic phase as earlier suggested by Majidian*et al*. (2016). Based on the analyses conducted so far, 20 wt. % appears to be the best possible composition by proving the best performance all through. Therefore, it could be concluded that 20C has the optimum composition of Al-Cu-CP metal composite as well as the fabrication parameters. Among the earlier researchers who reported 20 wt. % of reinforcement composition in Al-MMCs include Al-Imari (2014) and, Rahman and Al Rashed (2014).

Conclusion

The chemical analysis confirmed the presence of the starting materials, Al and Cu while the presence of Si from CP was also confirmed in the composite fabricated. The effect of varying percentage composition of the components was investigated by studying properties of the Al-Cu-CP metal composite which include hardness, microstructure and crystal phase. The investigation revealed the optimum conditions for the fabrication of the composite as well as improvement in its properties. Characterizations and analyses of the properties of the Al-Cu-CP metal composite were investigated and found to be better than the unreinforced Al at 800 °C casting temperature, 60 minutes melt holding time and with addition of 20 wt. % of the combination of Cu and CP as reinforcement materials.

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