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Development And Characterization Of Copper-Coated Basalt Fiber Reinforced Aluminium Alloy Composites

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Abstract:
The aim of the work was to investigate the effect of copper coated basalt short fiber reinforced Al alloy composites and was compared with uncoated basalt short fiber Al metal matrix composites (MMCs). Five different wt. % of basalt short fiber reinforced Al MMCs were prepared by squeeze casting technique 2.5, 5, 7.5 and 10% basalt short fiber MMCs. Both type of MMCs (coated and uncoated basalt fiber reinforced Al MMCs) were tested for elastic modulus, ultimate tensile strength and ductility along with microstructural change as per ASTM standard. The result shows the coating of Cu on basalt short fiber increases the Young's modulus, due to the homogenous distribution of basalt short fiber and the alignment of these fibers parallel to the axis with minimum segregation in the alloys, the ultimate tensile strength also increased due to their matrix strengthening and reduction in the alloy grain size, but the ductility significantly decreases due to the voids. The microstructure and fracture surfaces of both MMCs were examined using optical and SEM micrographs respectively. The lack of observed fiber pull-out on fracture and improved mechanical properties resulted due to the good wetting of the fibers by the liquid alloy.

Keywords: Metal matrix composite (MMCs), Basalt fibers, Short fiber composites, Electroless coating, squeeze casting technique.

1. INTRODUCTION

Metal matrix composites (MMCs) properties are strongly depending on the interfacial phenomena between the metal matrix and fiber surfaces [1]. The interface plays a most vital role in the overall performance of a composite material. The wettability of reinforcement by liquid metal is the key factor to achieve high interface bonding strength. There are several methods to improve the interfacial bonding including the modification of the matrix composition [2], coating of the reinforcement [3] and control of process parameters [4]. Among these methods, modification of fiber surface or metal coating on fiber to improve the wetability between matrix and reinforcement is prominent [5]. Although many technologies available for metal coating on the fiber surface but electroless copper coating is high preferable in research community [6] due to simple, low-cost and an easy to use process. And also it has been successfully applied to prevent undesired interfacial reactions and promote the wettability through increasing the overall surface energy of the reinforcement [7-8].

Wettability is one of the major criteria during the fabrication of fiber reinforced MMCs due to the repulsion of the negative electron of the reinforcement present in the fiber by negative oxygen anion monolayer present at the aluminium surface [9]. However, the reaction between the fiber and matrix takes place above 500°C [10]. It has also been reported [11] that the reaction is possible even below this temperature. Thus, by applying a suitable coating on the fiber and/or modifying the matrix composition to avoid repulsion of the negative electron. Some researcher the copper coating on the glass fibers facilitated their wetting with the molten aluminium [12]. During coating, the gas film is chemically displaced by metal film which easily interacts with the molten metal during composite processing. $CuAl_2$ phase formation during Cu-coated fibers dispersion, being endothermic in nature lowers the fluidity around the interface. Some undissolved copper remained at the interface protects the fibre from coming in contact with molten aluminium and prevents the formation of $A1_4C_3$. The magnesium in melt improves wettability because of the lower surface tension of magnesium [13]. In the present study, an attempt has been made to investigate the effect of copper coating on basalt fiber reinforced aluminium alloy composites properties prepared by squeeze casting for microstructure and mechanical properties.

2. Experimental work

2.1 Materials and methods

The Al and basalt short fiber used as the MMCs in the present study are obtained from commercial ingots with correct chemical composition as shown in Table 1, the presence of these elements have been confirmed by SEM / EDS spectra. The aluminium alloy 7075 was selected as matrix and basalt fibers (mineralogical research company) in the form of form of continuous basalt fibers (CBF) were used as reinforcements for making the composites. In the present investigation the deposition of copper coating on short basalt fibers by an electroless route has been optimized.

Table 1 Chemical composition of Al alloy (Weight percentage) & short basalt fiber

Element	Si	Fe	Cu	Mn	Mg	Cr	Zn	Ti	Al
%	0.4	0.5	1.6	0.3	2.5	0.15	5.5	0.2	Bal

Element	SiO_2	Al_2O3	Fe_2O3	MgO	CaO	Na_2O	K_2O	TiO_2	MnO
%	69.51	14.18	3.92	2.41	5.62	2.74	1.01	0.55	0.04

2.2 Pre-procedure

The continuous basalt fibers of average diameter 6 µm were chopped down to short fibers of about 1 to 2 mm length. The complete process of coating starts with the treatment of fibers in a muffle furnace for 10 min. at 500 °C to eliminate the pyrolytic coatings around as received fibers. The electroless process used to deposit the copper coatings on the basalt fiber relies on a sequence of sensitizing, activation and metallization, with important cleaning, rinsing, washing and drying stages also being included.

2.3 Electroless coating

The electroless process used to deposit the copper coatings onto the basalt fiber relies on a sequence of sensitizing, activation and metallization, with important cleaning, rinsing, washing and drying stages also being included. The conditions used are detailed in Table

2. Subsequently they were coated with copper using the electroless process, as described elsewhere [14-15]. A two-Step Process Chemical Concentration Temperature (°C) Time (min)

2.4 Experimental procedure

The short basalt fiber was cleaned in distilled water and dried at 90°C. The sizing and finishing treatment from the surface of the fibers, prior to coating were removed by heating them to about 970 K for 10 min in air. Fibers have elastic modulus of 90 GPa, and a yield stress of 4500 MPa. The coating procedure consist of three well defined stages namely sensitization, activation and metallization. The heat cleaned fibers are first treated with glacial acetic acid to activate the surface, and then again activated using stannous chloride ($SnCl_2$) and they were sensitized for different times (5, 10 & 15 min.) under continuous stirring. Fibers are then filtered and cleaned with distilled water. In order to have catalytic surfaces, the sensitized fibers were exposed to an aqueous solution containing palladium chloride ($PdCl_2$) and HCl under ultrasonic agitation. This process, called activation, produces the formation of Pd sites on the fiber surface which allow the subsequent metallization with copper. The complete process of metallization starts with the treatment of fibers in an open oven for 10 min. at 500°C to eliminate the pyrolytic coatings around as received fibers.

Table 2: Chemical compositions

Stage and conditions	Concentration of chemicals
Sensitisation	12 g/l $SnCl_2 \cdot 2H_2O$
5min, 10min & 15 min at room temperature	40 ml/ HCl
Activation	0.2g/l $PdCl_2$
5min, 10min & 15 min at room temperature	2.5 m/l HCl
Metallization	10g/l $CuSO_4 - 5H_2O$
Multiple conditions tested	45g/l EDTA
40°C and 50°C	20 g/l NaCOOH
pH 12 and pH 13	16 ml/l HCHO 36%
2 min - 20 min	NaOH for adjusting Ph

Metallization is produced by immersion of activated fibers into a solution containing $CuSO_4$-$5H_2O$ as metal ion sources also held under agitation. Different metallization conditions have been tested, pH (12 &13), time (2- 20 min) and temperature (40 & 50°C), and continuous and crystalline coatings with homogeneous thickness have been obtained. The reactive volume used assures that the concentration of the diluted copper can be considered constant during the deposition. The coatings obtained at different metallization temperature, times and pH values were studied by SEM and the thickness of the copper layer was determined in transversal cross section.

2.5 Preparation of composites by squeeze casting technique

In squeeze casting liquid metal is injected into the interstices of short fibre usually called as a preform. In this process liquid metal is pressurized while they solidify and hence near net shapes can be produced with sound and dense quality. The apparatus was designed to make a cylindrical billet of 100 mm diameter and 40 mm thick. A die of 100 mm diameter was manufactured and was installed on the 150 ton hydraulic press. It consists of a simple die and punch set with an ejector rod to facilitate removal of the solidified composite. The die is heated by a band heater to the required temperature. The casting operation consists of preheating the die, with the preform inside it, typically to 300–400 ^{0}C, and melting a weighed charge of the alloy to be cast, heating it to a temperature typically 150–250 ^{0}C above its liquidus temperature. The molten metal is then poured into the die and then the punch is driven into the cavity to compress the charge. The pressure applied is varied from 20 to 30 MPa. The constant ramspeed of 10mm/swas used. The pressure is maintained during solidification period and then after a further cooling period of 5–10min the ram is withdrawn and the composite extracted. The process was repeated without the preform in it to get the squeeze cast metal. The composites were solutionised at 530 ^{0}C for 2 h, quenched and the specimens were artificially aged at 180°C for 12hrs followed by furnace cooling.

2.6. Characterization of composites

All mechanical tests were conducted in accordance with ASTM standard. Tensile tests were conducted at room temperature using a Shimuzu universal testing machine (of 10 tons capacity) equipped with a pair of extensometers in accordance with ASTM E8-82. The tensile specimens of diameter 8.9 mm and gauge length 76 mm were machined from the cast composites with the gauge length of the specimen parallel to the longitudinal axis of the castings. The modulus measurements were carried out on the specimens in tension, using 'loading unloading method' with a crosshead velocity of 5 mm/min. Young's modulus values were calculated from the slope of the ruler fit straight lines joining the two ends of the loading-unloading curves. Four different specimens were tested and average values of the UTS and ductility were measured.

Microstructure specimens of the Al/basalt fiber MMC were prepared as per ASTM E3 standards and the structure was examined under conventional optical microscopy to understand the variation in microstructure and fiber distribution. To increase the visibility of the embedded fibers in the matrix, the composites were etched deeply with Palmerton etching reagent (Chromic oxide-200g, Sodium sulfate-15g, water-1000ml) polishing with 9 and 1 lm diamond paste. The choice of a sample for microscopic study is very important for analyzing composites. Scanning Electron Microscopy (SEM) was performed on the fractured surface to understand the failure mechanisms. The fractured surfaces of the squeeze infiltrated AMC and the squeeze cast metal were examined under scanning electron microscope (SEM to determine fiber/matrix bonding).

3. RESULTS AND DISCUSSION

3.1 Characterization of the copper coating

Fig. 1 shows the photographs of un-coated and coated basalt fibers. It can be seen that, almost all (more than 90%) chopped fiber are coated uniformly and the copper coating is continuous over the fibers. Fig. 1 shows the surface characteristics of uncoated fibers (Fig.1a.), of copper coated ones (Fig. 1 b-f) under different metallization conditions, and Fig. 1d shows the transversal section of copper coated fibres as observed with light microscopy. As it can seen in the images, the thickness and morphology of the copper layers is highly dependant on the metallization conditions, mainly sentisization time, activation time, metallization time, temperature and pH. Bath temperature must be kept below 45°C because higher temperatures unstabilize the metallization solution. At this or higher temperatures dentritic growth of the coating (Fig.1b) is promoted. Using lower bath temperatures under the same pH conditions (pH 13) homogeneous thickness can be achieved on the coatings. To obtain any copper deposition, the pH of the metallization solution must be higher than 12. For pH 12 or higher the deposition rate increases as the pH increases so shorter deposition times are needed to get the same thickness. For pH 12, the minimum time needed to get continuous copper coating is 3 min. After this first stage, the thin copper coating grows homogeneously in thickness without evidence of dentritic growth (Fig. 1c and d)

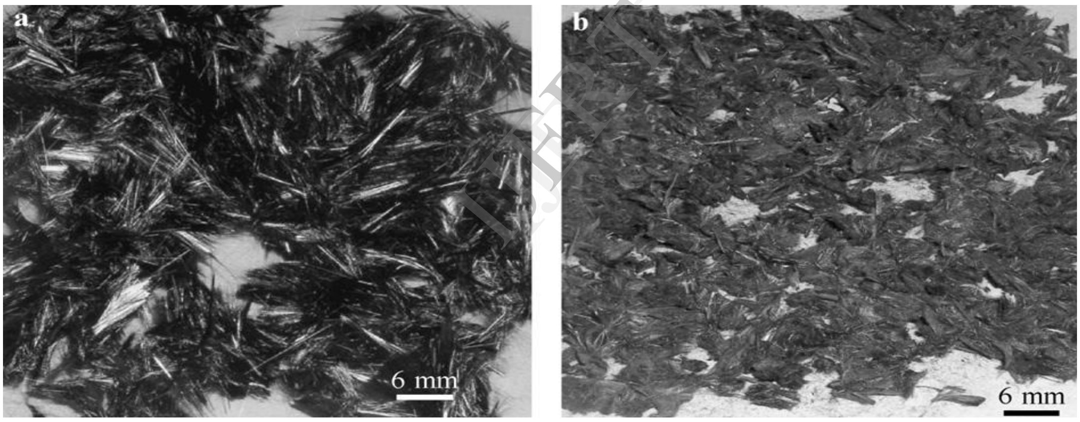

Fig. 1 – Photographs of (a) un-coated basalt fibers and (b) coated basalt fibers.

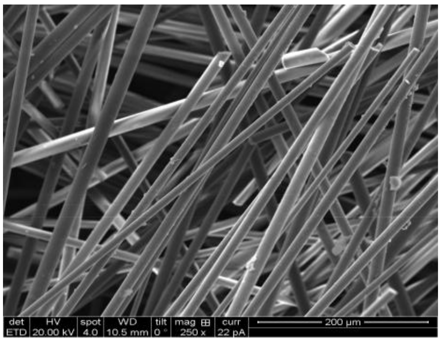

Fig. 1 (c) SEM Micrographs of coated basalt fibers

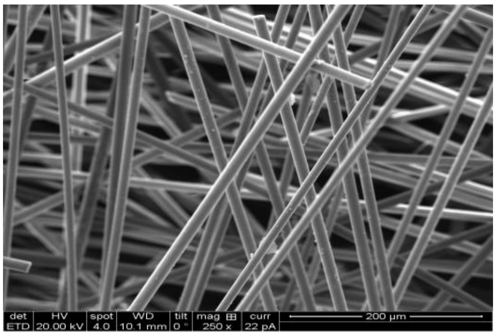

Fig. 1 (d) SEM Micrographs of coated basalt fibers

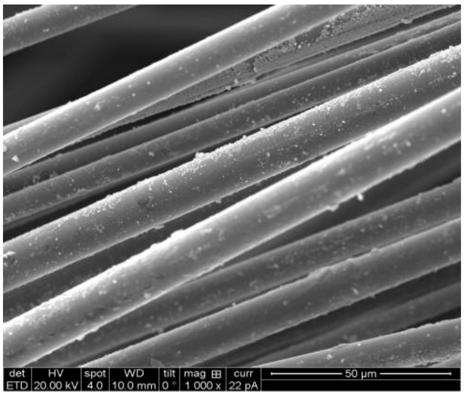

Fig. 1 (e) SEM Micrographs of coated basalt fibers

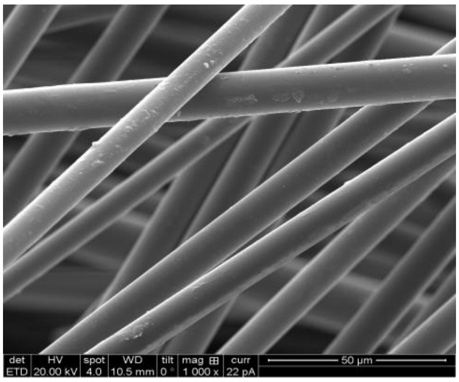

Fig. 1 (f) SEM Micrographs of coated basalt fibers

3.3. Microstructural studies of composites

Effort was made to observe distribution of fibres in the cast composites produced. After casting, ingots were cut into four pieces along the longitudinal direction and the porosity and distribution of fibres were observed. The surface of the composite castings were machined first to remove the surface layer of the metal and the microscopic examination of this surface generally gives the over all trend in the distribution of the reinforcements.

To study wettability and reaction processes that takes place in the matrix/fiber interface regions. Typical microsections of the composite fabricated with uncoated basalt fibers dispersed in Al matrix showed fiber matrix interactions. Reaction products were observed almost at the entire interface. Uncoated fibers are pushed away by the solidification front and appear concentrated in the outer zones of the specimens and locally, in the interdendritical spaces as shown in Fig. 3(a)

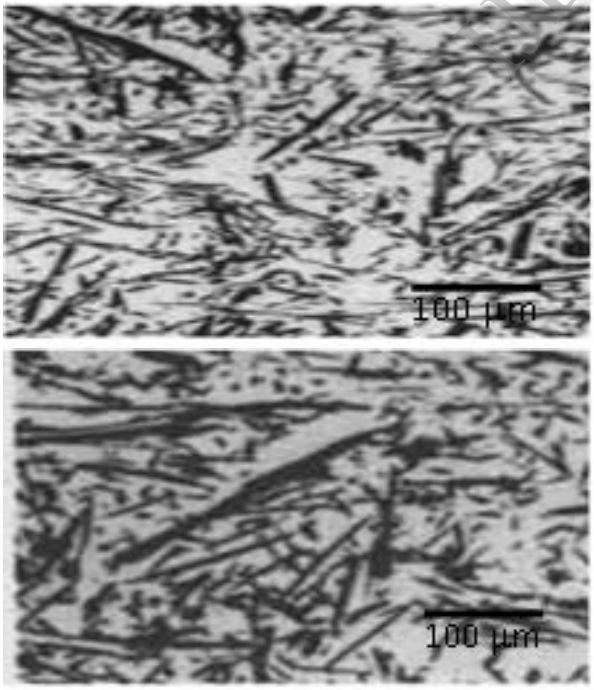

Fig. 3 Microstructure of the Al/10 % basalt short fiber MMCs uncoated (a) and coated (b) conditions.

Microstructures of aluminium composites containing 2.5, 5, 7.5 and 10 wt % copper-coated short basalt fiber are shown in Fig. 4. It is seen that basalt fibers are embedded in the aluminium matrix in the form of flattened bundle. In the composites, the matrix metal has thoroughly penetrated into the bundle of fibers and is well bonded to every single fiber. The optical micrographs of composites containing 2.5, 5, 7.5 and 10 wt % short basalt fiber show that the fibers are more or less uniformly distributed and also observed that some pores are present in the composites. As the fiber content increases, the uniformity of distribution of the fibers in the composite increases up to 7.5 % fiber content and beyond this uniformity of distribution of fibers decreases because of the agglomeration of fibers in the composites. The uniformity of fiber distribution in the composite containing 10 wt% basalt fiber is slightly decreased and voids and inclusions are present in the composite.

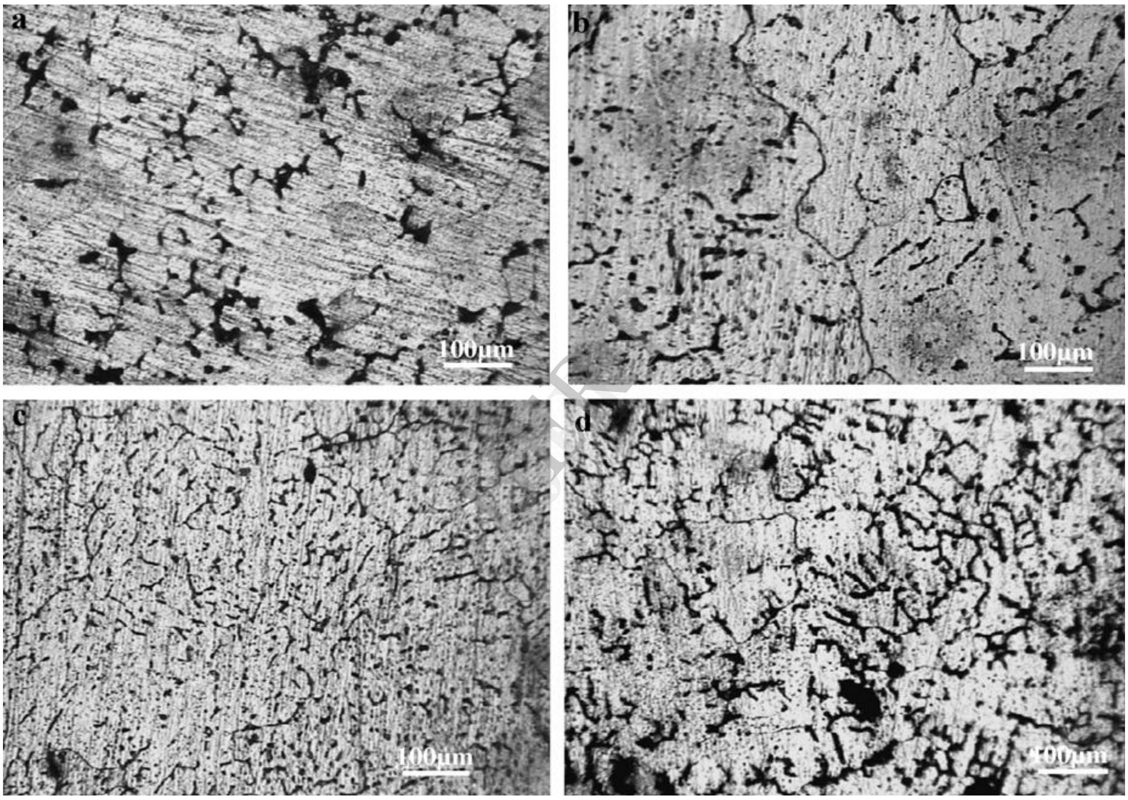

Fig. 4 – Optical micrographs of copper-coated basalt fiber reinforced composites. (a) 2.5 wt%, (b) 5 wt%, (c) 7.5 wt% and (d) 10 wt% fiber reinforcements.

3.4. Mechanical properties

3.4.1 Elastic modulus

The measured mean values of elastic moduli were plotted as a function of weight percentage of basalt short fiber as shown in the fig.5 (a). The sharp increase in Young's modulus has been observed, in young's modulus of Cu coated basalt short fiber when compared with uncoated Al\basalt short fiber MMCs, this is probably due to the homogenous distribution of copper coated basalt short fiber and the alignment of these coated fiber parallel to the axis with minimum segregation in the alloys. McDanels [16] is of the opinion that the elastic modulus increases with increase in reinforcement content. However, elastic modulus has been found to be independent of the type of reinforcement.

As short basalt short fiber content was increased from 2.5 to 10 % by wt., an improvement in Young's modulus of 13.26% has been observed.

3.4.2 Tensile properties

Fig.5 (b) shows the effect of Cu coated basalt short fiber on UTS in Al MMCs, It can be seen that as the Cu coated basalt short fiber content is increased, the UTS of the alloy material also increases when compared with uncoated Al/basalt short fiber MMCs. There is a marked increase in the UTS of the alloy from 15.5 to 28.7% as the Cu coated basalt short fiber content is increased from 2.5 to 10 wt.%. The increase in UTS is attributed to the presence of hard basalt short fiber, which imparts strength to the matrix alloy, thereby providing enhanced tensile strength. Vogelsangs' et al. [17] believes that the improvement in UTS may be due to the matrix strengthening. The reasons assigned are reduction in the alloy grain size and generation of a high dislocation density in the matrix, which is a result of the difference in thermal expansion between the metal matrix and the basalt short fiber reinforcement.

3.4.3 Ductility

Fig. 5 (c) shows the effect uncoated and Cu coated basalt short fiber content on the ductility (% of elongation) of the MMCs. It can be seen from the graph that the ductility of the alloy decreases with the increase in Cu coated basalt short fiber content from 2.5 to 10 wt. % (the ductility decrease by about 35.9%) when compared uncoated Al\basalt short fiber MMCs. This decrease in ductility in the MMCs is the commonly encountered disadvantage in MMCs when compared with the base alloys. Mummery et.al [18] is of the opinion that this behaviour is probably due to the voids, which nucleate during the plastic strains of the reinforcement or by reinforcement interface.

3.4.4 Optical and scanning electron microstructure analysis

The general features of the microstructure, e.g. fiber distribution, wt. % of reinforcement, and orientation, were studied using an optical microscope and fractured surfaces were studied using a scanning electron microscope. Typical fractured surfaces of Al/basalt short fiber composite, obtained from tensile tests, are shown Figure 6 (a). According to Withers [19], the experimental observation also suggests that short fibers rarely fracture. Only very few fibers can be seen as fractured and there is also an evidence of ductile failure in the matrix. The failure is few fibers split longitudinally and transversally as shown in the Figure 6 (b). The failure of fibers in the composite may be attributed to the increase in stress on the specimen. As the load on the fiber increases, it induces strain in the fibers, and the most heavily loaded fiber fractures. Some fiber "pull-out" has occurs in the samples, but the failure appears to be at the matrix end and not at the interfacial regions, as indicated by the conical cavities with rippled surface. Apparently more "pull-out" occurred in the matrix composite as expected due to the lower strength of the matrix. In some places where the fiber end was exposed to SEM, it appears that the matrix sheared away from the fiber.

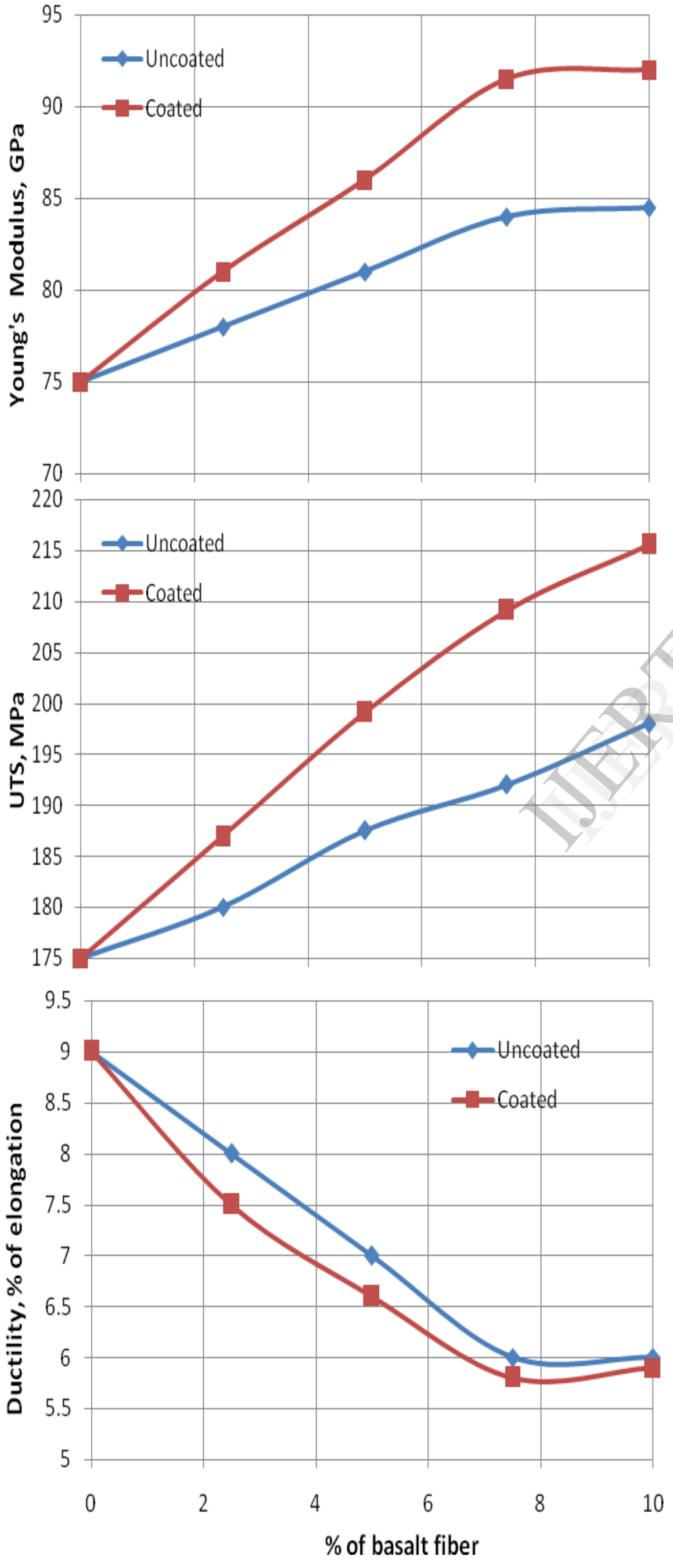

Fig. 5 Effect of Cu coating on basalt short fiber reinforced Al MMCs a. Young's modulus, b) UTS and c) Ductility

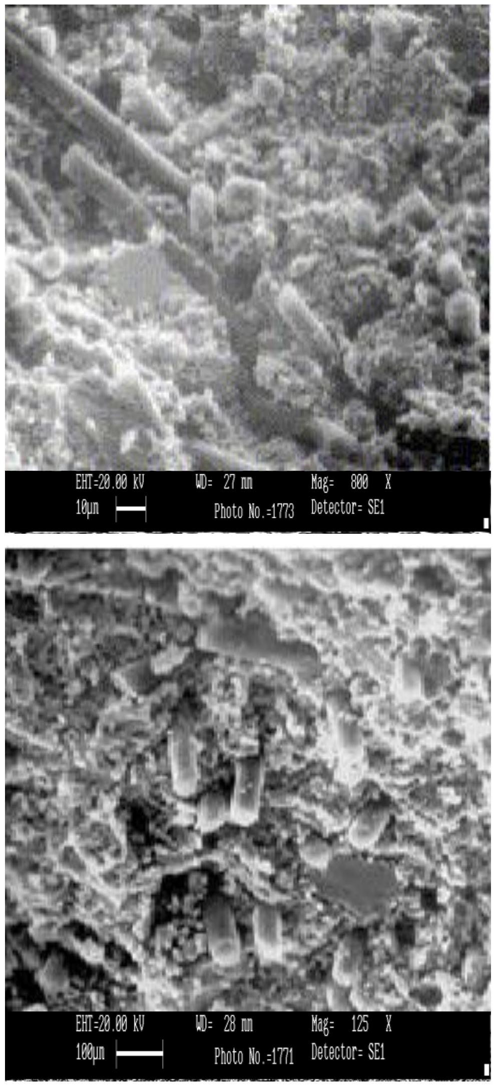

Figure 6 (a) Fractrographs of the tensile specimen shows (a) fractured short Basalt fibers and (b) fiber puIlout from the

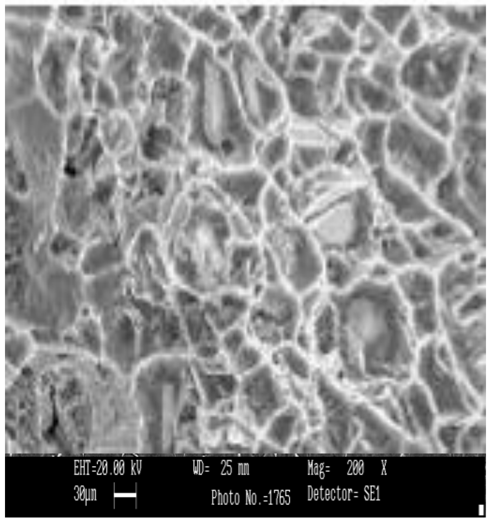

Figure 6 (b) Fractographs of the tensile specimen unreinforced shows uneven distribution of the large dimples

4. Conclusions

The following conclusions are made from the study:

1. A fairly uniform and continuous coating of copper on short basalt fibers can be prepared successfully by electroless technique. Around 95% of the fiber had a continuous coating with a fine crystallite-type deposition of copper was found over continuously coated copper
2. The low wettability of high modulus basalt fibers by molten aluminium has a marked effect on the subsequent final solidification of the composite, determining not only heterogeneous distribution of the short basalt fibers in the aluminium matrix but also favouring the microsegregation of alloying elements (i.e. magnesium) in the matrix/fiber interfaces.
3. The copper-coated fibers, up to 7.5 wt%, are uniformly distributed in the matrix with little agglomeration.
4. The tensile strength increases with increase in Cu coated reinforcement content, an increase in UTS from 15.5 to 28.7% was observed.
5. The Young's modulus and UTS of the composite material increases with increase in fiber wt %, an improvement in Young's modulus of 13.26% has been observed.
6. The ductility of the MMC decreases gradually with increase in fiber wt %.

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