Improved Corrosion Resistance of Carbon Reinforced Aluminium Laminates in Atmospheric Environment: Role of Environment Friendly Jute Fibre/ Alumina nano Coating

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ABSTRACT

In the Fibre Metal Laminate (FML), Carbon Reinforced Aluminium laminate (CARALL), aluminium is placed next to carbon fibres. The potential difference between the aluminium and carbon is larger, leads to galvanic corrosion, which tries to bring down the durability of the FML. To bring down the effect of corrosion, a material layer is introduced between fibres and aluminium so as to separate them thus lowering the possibility of occurrence of corrosion. Another approach is to coat the surfaces of aluminium with different proportions of aluminium oxide nano particles prior to fabrication of the FML. For both the cases, corrosion rates and polarization resistances are determined and compared. In addition, the surface morphological studies were carried out using SEM to reveal the intensity of corrosion. The electrochemical results revealed that the jute reinforced CAJRAL FML and CARAL FML coated with 10% alumina has better corrosion resistance compared to other FMLs..

KEYWORDS: *Carbon Reinforced Aluminium Laminate, Carbon-Jute Reinforced Aluminium Laminate, Polarization resistance, Alumina nano particles.*

1. INTRODUCTION

Today's world is in need of light weight materials in order to consider the economy and energy consumption, since the availability of power producing fuels becomes less. It is essential to ascertain not to compromise on the strength aspect of such materials. Hence a new variety of composite material emerged at the end of seventies[1,2]. Fibre metal laminates (FML) are

J. Polym. Mater. Vol. **36,** No. 1, 2019, 1-11

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DOI : https://doi.org/10.32381/JPM.2019.36.01.1

hybrid composites, which are made as a laminated material of alternately thin aluminium alloy sheets and fibre composite layers and were used in wing structures, fuselage and for ballistic protection[1-4]. This material possess low weight, high specific strength and is easily tailorable to any shape. Such a fibre metal laminate is being commercially produced in threes types viz., Aramid Reinforced Aluminium Laminate(ARALL), in which aluminium is reinforced with aramid fibres, Glass Reinforced Epoxy Laminate (GLARE) in which aluminium is reinforced with glass fibres[5,6] and Carbon reinforced aluminium Laminate(CARALL) in which aluminium is reinforced with carbon fibres. ARALL has good specific strength, specific modulus, high impact resistance but poor compression strength[5]. Whereas CARALL and GLARE show high specific modulus, low specific strength, strain to failure and impact resistance.

CARALL is used as a structural material in commercial Airbus A380. CARAL FML is formed by reinforcing carbon fibres with aluminium in a polymer matrix. Carbon, an efficient cathode and aluminium, an efficient anode in CARALL[5], induces galvanic[7-13] corrosion when it is used in electrolytic solutions of sea/rain/acidic waters. This is extremely undesirable as this may corrode the metal thus depreciating the shelf life of the FML[14-16]. Tavakkolizadeh et al and Fovet et al.[17-18] found that pitting was initiated as a result of high intensities of galvanic corrosion, thus a reduction in service life of the FML was noticed. Xiaoqiang Zhan[19] et al found that smaller corrosion current densities of magnesium alloy were obtained when coated with different composite coatings. Gavrilo Sekularac[20] et al studied the corrosion behaviour of aluminium alloy in artificial sea water consisting of different concentrations of sodium sulphide and evidenced that low concentration is beneficial with regard to corrosion protection of the alloy. The intensity of corrosion is high during the friction stir welding of dissimilar aluminium alloy shown by Sori Won[21] due to galvanic effect arising in one of the welds. Donatus[22] et al have observed two pathways of corrosion in aluminium alloys, one in the corrosion front and the other along the crystallographic channels where the corrosion is more dependent on the second phase particles and the grain boundary character.

Surfaces coated with nano materials have great benefits in improving the strength, wear resistance, bonding ability, corrosion resistance[23,24] and durability of materials. Alumina nano particles[25] which are cost effective, find wide utility in improving the surface morphology of materials that can work well in corrosive environments[26,27], as an agent that improves the bond strength between layers of a composite and also acts as a thermal barrier[28,29]. Ianina Santana[30] et al have developed a new anti-corrosive sol-gel coating which consists of cerium and clay nano particles. ZHANG Shenglin[31] et al have improved the corrosion resistance of AA6061 alloy by coating with zinc phosphate with Rare Earth Nitrate (REN) additive.

It is reported that the jute fibre[32-36] is used as reinforcement in composite material, as it is a natural fibre[37-41] possessing good properties like low density, low cost and less processing compared to the other natural fibres[42].

In this work, the carbon fibres are separated from aluminium by inducing a layer of jute fibre in CARALL to bring down the intensity of corrosion. This new FML is named as CAJRALL (Carbon-Jute reinforced Aluminium laminate). Another approach to curtail the severity of corrosion is to coat with a layer of alumina nano particles on the surfaces of aluminium before fabricating the FML. Two different proportions of alumina nano particles are used: 5% and 10% by weight[43]. The polarization resistances and the corrosion rates of FMLs are evaluated experimentally for both the cases in the atmospheric environment. This condition is simulated by exposing the samples to 0.5M sodium chloride solution. The same procedure is used for FMLs fabricated with two other fibres, glass and Kevlar (an aramid fibre).

MATERIALS AND METHODS

Fabrication of FMLs

The materials used for the preparation of FML samples are carbon (240 gsm), jute (240 gsm), glass (200 gsm) and kevlar (200 gsm) fibres, alumina nano particles, aluminium 2024 T3 sheet with thickness of 0.17 mm, epoxy resin of grade LY 556 and Araldite hardener of grade X3543. Carbon and glass fibres, epoxy resin and hardener are purchased from M/s S.M.Traders Ltd, Chennai. Aluminium and jute are purchased from M/s JNS Steels, Chennai and M/s Small industries Production Promotion Organisation Ltd., Madurai, respectively. Kevlar (grade 49) was supplied from Nickunj Eximp Enterprises Private Limited, Mumbai. Alumina powder is purchased from Lab Chemicals, Chennai.

The FML specimens CARAL, GLARE and ARALL prepared with and without jute considered for analysis are fabricated by combination of hand lay-up and compression moulding techniques. The epoxy is mixed with the hardener in the ratio of 10:1. Initially the FMLs are prepared by hand lay-up technique using the above mentioned materials. Then it is cured at room temperature and is pressed for ten minutes in compression moulding machine at 70 kg cm^{-2} and at 70°C[44] to maintain uniformity in distribution of the matrix and to avoid voids that form during fabrication.

The FML samples are coated with two different proportions of alumina nano particles of 5% and 10% by weight. The coating is done by blending the nano particles with epoxy in the above quoted proportions. The FML samples are fabricated with this mixture of resin as described above.

Electro Chemical Studies
Potentiodynamic polarization studies

The FMLs fabricated as above are cut with an area of 1 cm^2 for exposure to electro chemical analysis for determination of polarization resistance and corrosion rate as per ASTM G102 standard. The test is carried out in atmospheric condition using a electrochemical cell consisting of FML sample as the working electrode, platinum foil as the counter electrode and a saturated calomel electrode (SCE) as the reference electrode. The working electrode is immersed in an electrolyte of 0.5 M solution of NaCl and is initially allowed to stabilize for 30 minutes. A current of 1.5mA/cm^2 is applied for 15 min duration to reduce the surface oxides. Polarization studies were conducted using a potentiostat (Model PGSTAT 12) and data analysis was done using a customized GPES software version 4.9. The polarisation curves for the FML samples were recorded by sweeping the voltage at the rate of 1 mV/s. The polarization resistance is measured from Tafel Exploration plot obtained at the end of the test. The polarization resistance assessed is given by eq (1)[45].

$$RP = \frac{1}{2.303}\left[\frac{(\beta_a \times \beta_c)}{(\beta_a + \beta_c)}\right] \times \frac{1}{I_{corr}} \qquad (1)$$

Weight loss technique

The weights of the FML coupons without jute of size 2 cm x 2 cm x 0.4 cm and FML coupons with jute of size 2 cm x 2 cm x 0.7 cm are measured before and after polarization test and thus the weight loss due to corrosion attack is measured. The coupons are immersed in 300 ml of 0.5M NaCl solution at room temperature for 3 hours. Moreover, corrosion rates of the FML samples are determined using weight loss method through eq. (2)[46].

$$CR = \frac{w \times K}{\rho \times A \times t} \text{ miles per year} \quad (2)$$

where *CR* is the corrosion rate, *K* = 3.45 x 10^6 is a factor and *t* is the time of exposure in hours.

RESULTS AND DISCUSSION

Polarization resistances of FML samples

The cathodic and anodic polarization plots for CARALL and CAJRALL are shown in Figure 1(a) and (b). As the potential increases, the current density also increases, showing the active region of corrosion. With further increase

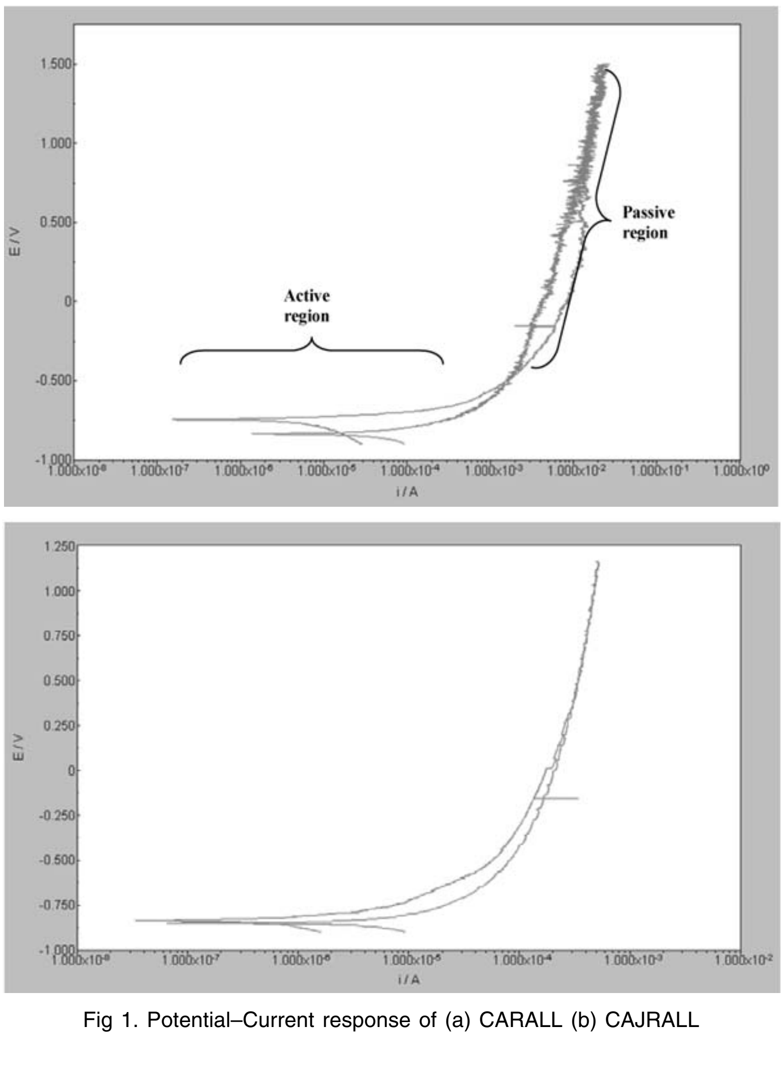

Fig 1. Potential–Current response of (a) CARALL (b) CAJRALL

in potential, the current density of the sample remains almost constant which indicates the passive region of the polarization curve. Here the specimen starts passivating and the corrosion is controlled considerably due to loss of chemical reactivity under this environmental condition. This may be as a result of the formation of film on the metal surface.

The polarization resistances of various FML samples prepared, without and with nano coating are tabulated in Table 1 and Table 2 respectively.

It is observed from the Tables 1 and 2 that the polarization resistances of the CARALL and GLARE with jute fibres are found to be higher than those without jute fibres but lower than those with nano coatings. However ARALL with jute fibres show similar results but that with nano coatings provides differing results. This is due to the hygroscopic nature of kevlar fibre[47]

TABLE 1. Electrochemical parameters of FML samples without nano coating

Sequence	β_a v/decay)	β_c (v/decay)	Current density (amps)	E_{corr} (V)	Polarization resistance(R_p)
Bare Al	0.146	0.327	1.499E-6	-0.842	6.612E+2
A/J/Epoxy	0.125	0.143	1.12E-7	-0.790	4.212E+4
A/C/Epoxy	0.034	0.153	5.100E-7	-0.0823	3.212E+3
A/J/C/Epoxy	0.138	0.138	2.422E-10	-0.841	4.294E+3
A/G/Epoxy	0.033	0.044	1.24E-7	-0.748	4.35E+3
A/J/G/Epoxy	0. 023	0.113	1.23E-7	-0.686	4.52E+3
A/K/Epoxy	0.420	0.092	1.199E-6	-0.802	3.112E+3
A/J/K/Epoxy	0.363	0.121	1.312E-6	-0.728	3.442E+3

TABLE 2. Electrochemical parameters of FML samples with nano coating

Sequence	β_a (V/decay)	β_c (V/decay)	E_{corr} (V) 5% Alumina	E_{corr} (V) 10% Alumina	Current density (amps) for 5% alumina	Current density (amps) for 10% alumina	Polarization resistance (R_p) for 5% alumina	Polarization resistance (R_p) for 10% alumina
A	0.143	0.354	-0.832	-0.873	1.41E-6	1.38E-6	1.647E+4	1.87E+4
A/C/Epoxy	0.136	0.340	-0.868	-0.840	1.373E-6	1.232E-6	1.754E+4	1.98E+4
A/G/Epoxy	0.021	0.167	-0.750	-0.690	8.2072E-8	8.002E-8	2.854E+4	2.91E+4
A/K/Epoxy	0.036	0.250	-0.895	-0.81	1.858E-6	1.653E-6	2.299E+3	2.312E+3

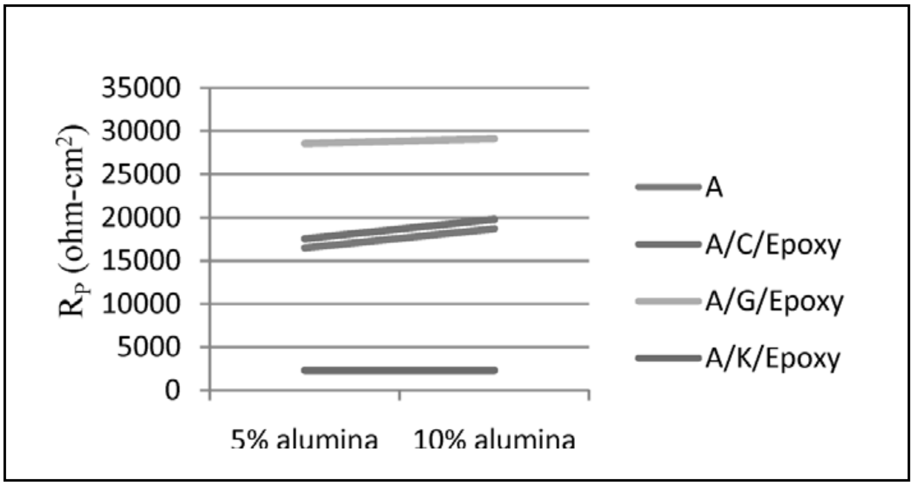

Fig. 2. Percent variation of polarization resistances of FMLs with nano coating

which absorbs more amount of moisture and retains the same amount, when it contacts alumina nano powder which is coated on the aluminium sheet. Hence the resistance to corrosion of the A/K/Epoxy laminate decreases after coating which is evident from Figure 2.

Thus the polarization resistance of CARALL with jute i.e., CAJRALL is 34% more than that of CARALL, that of GLARE with jute reinforced is 3.9% more than that of GLARE without jute and the polarization resistance of ARALL with jute is 11% more than that of ARALL without jute.

The effect of proportion of alumina nano particles on the polarization resistances of the FML samples are shown in the Figure 2.

It is observed from Figure 2 that, for all cases, FMLs with 10% alumina coating yields high resistance to corrosion than the FMLs with 5% coating. Moreover the polarization resistance takes a decreasing trend in the order of coated FMLs GLARE, CARALL, bare aluminium metal and ARALL which is apparent from Figure 2.

Metal loss Rate and Corrosion Rate of FML samples

The corrosion rates obtained for various FML samples without and with coating (with 5% alumina and 10% alumina) are shown in the Table 3 and 4 respectively.

TABLE 3. Metal loss and corrosion rates of FML samples without nano coating

Sequence	Weight loss	Corrosion Rate (CR)
Bare Al	0.022	237.90
A/Epoxy	0.011	118.96
A/J/Epoxy	0.004	43.16
A/C/Epoxy	0.014	151.41
A/J/C/Epoxy	0.005	53.95
A/G/Epoxy	0.015	161.85
A/J/G/Epoxy	0.029	312.9
A/K/Epoxy	0.026	280.54
A/J/K/Epoxy	0.016	172.60

TABLE 4.Corrosion rates of FML samples with nano coating

Sequence	Weight loss		Corrosion Rate (CR)	
	5% alumina coating	10% alumina coating	5% alumina coating	10% alumina coating
A/Epoxy	0.009	0.011	97.11	118.69
A/C/Epoxy	0.012	0.007	129.48	75.53
A/G/Epoxy	0.002	0.005	21.58	53.90
A/K/Epoxy	0.19	0.08	205.01	197.90

It is observed from the Table 3, that the corrosion rate of the CARALL and ARALL with jute fibres reinforced is found to be lesser than those without jute fibres. However GLARE with jute provides adverse results. This is due to the fact that glass fibres do not corrode aluminium metal. From Figure 3 (a) and (b), it can be seen that corrosion pits are formed on aluminium surface of CARALL whereas no such pits or corrosion marks are observed in case of CAJRALL, which proves the above statement.

It is observed from Tables 3 and 4 that, the corrosion rate of CARALL coated with 5% Alumina is 15% less than that of uncoated

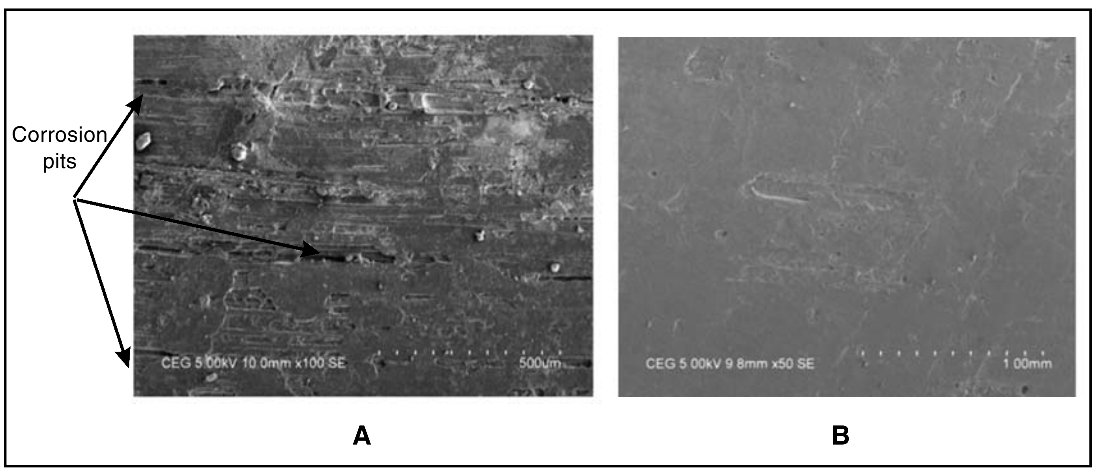

Fig. 3. SEM Images of aluminium surface in (a) CARALL (b) CAJRALL

CARALL and that coated with 10% Alumina is 50% less than that of uncoated one. Also the corrosion rate of GLARE coated with 5% Alumina is 86% less than that of uncoated GLARE and that coated with 10% Alumina is 66% less than that of uncoated one. Likewise the corrosion rate of ARALL coated with 5% Alumina is 27% less than that of uncoated FML and that coated with 10% Alumina is 29% less than that of uncoated one.

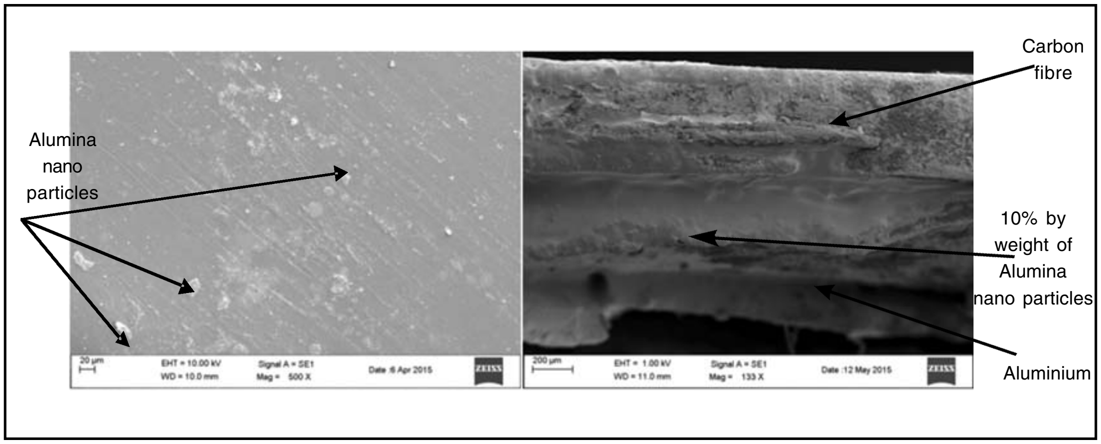

Fig. 4. SEM Images of aluminium surface in (a) CARALL (b) CARALL

Tested sample of CARALL fabricated with aluminium coated with alumina nano particles is shown in Figure 4 (a). It can be inferred that no corrosion pits are formed on the metal surface. Figure 4(b) shows the picture of the cross section of the tested sample of CARALL prepared with 10% by weight of alumina nano coating. This is also proved from EDX analysis from Figure 5 (a) and (b) and Figure. 6 (a) and (b), that CARALL with 10% nano coating has oxides of 26% by weight whereas that without nano coating has only 7% by weight.

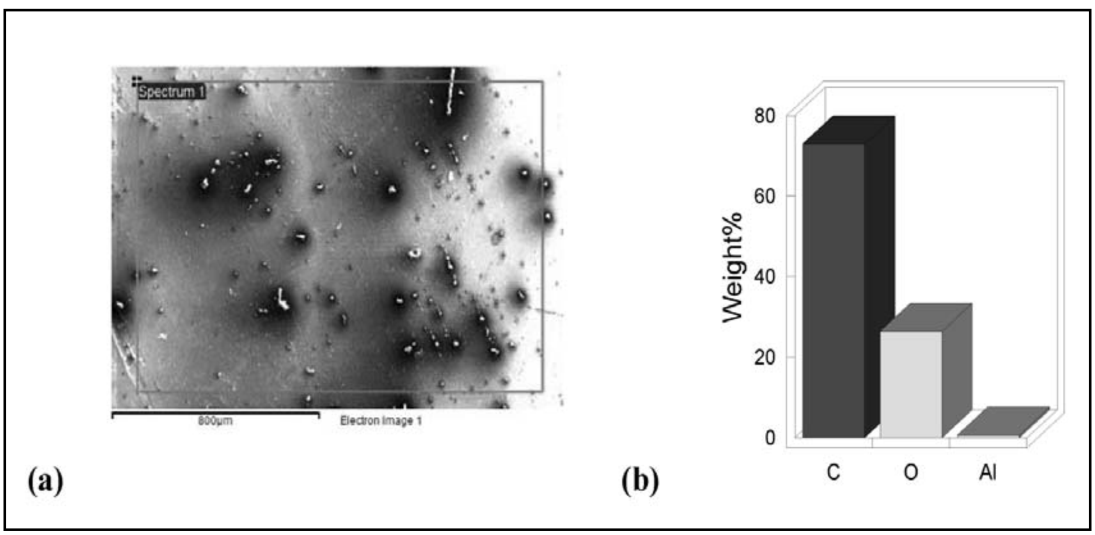

Fig. 5. (a) SEM Image of cross section (b) EDX analysis of CARALL with aluminium coated with 10% alumina by weight

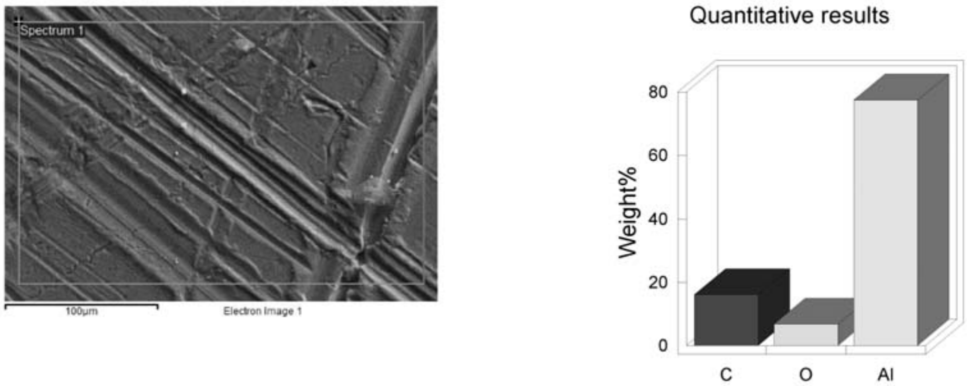

Fig. 6.(a) SEM Image (b) EDX analysis of CARALL without alumina nano coating

CONCLUSIONS

Inclusion of jute in FMLs CARALL and ARALL increases the corrosion resistances of the FMLs thus bringing down their corrosion rates. GLARE has an adverse effect and hence addition of jute does not find much significance here. Alumina nano coating of 10% by weight seems to dominate jute addition in terms of corrosion resistance for FMLs CARALL and GLARE but does not suit ARALL due to the non-conformance of alumina with Kevlar. This study, recommends the economical approach of addition of jute fibre in FMLs CARALL and ARALL and also recommends the utility of alumina nano particles as a surface coating material in all FMLs except for ARALL,to reduce the risk of galvanic corrosion in these fibre metal laminates.

ACKNOWLEDGEMENT

The authors thank the Chemistry department, College of Engineering, Guindy, Anna University, Chennai for providing technical support in performing tests.

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Received: 20-12-2018

Accepted: 27-02-2019