PLASMA-ASSISTED ADHESIVE BONDING OF ALLUMINIUM AND ITS ALLOYS

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Abstract: Aluminium and its alloys are significantly used as light weight materials in many industrial sectors including aerospace but it exhibits poor adhesion properties due to complex surface properties. Surface treatments of aluminium alloys play a very important factor from the point of view of high corrosion resistance, joint strength as well as durability of adhesive joints. The plasma deposition of thin film coatings on aluminium that exhibit strong interfacial bonding could provide an alternative to the traditional carcinogenic chromate-based treatments. The plasma deposited coatings confer protection against corrosion and provide a good interface for strong, durable adhesive bonds. Cold plasma represents an efficient, non-polluting and economical alternative to clean, activate and, then, to increase the adhesive properties of alluminium surfaces. Therefore, cold plasma reduces the amount of primer needed to be applied and the weight of whole structure. In addition to that the adhesive bonding of composite patch to repair cracks in metallic structures is an accepted technology in aerospace and automobile industries. The bond strength between composite patch and metallic structure is significantly affected by the surface preparation of the composite patch and metallic structure. The surface treatment effect of alluminium by plasma on the bond strength of alluminium/CFRP composites exhibits 33% higher shear strength and 6 times higher T-peel strength than those of untreated allluminium/CFRP composites. The optimal plasma treatment time and the ratio of acetylene gas to nitrogen gas are found to be 30 sec and 5:5 respectively. The surface treatment effect of alluminium foam and alluminium by plasma on the bond strength of alluminium foam/ alluminium composites exhibit 13% higher bending strength and 30% higher shear strength compared to no plasma treatment.
Keywords: Adhesive bonding, adhesion, Plasma treatment of Aluminium

INTRODUCTION: Presently, many industrial sectors including aerospace industry, there is a significant use of aluminium as lightweight materials, which eventually results cost savings and low fuel consumption. Aluminium and its alloys have high specific strength, good machinability, and formability and corrosion resistance but exhibits poor adhesion properties due to complex surface properties. Very often aluminium is fabricated for desired structure by adhesive bonding rather than welding, riveting or brazing. In this context, pre-treatment of aluminium alloys prior to adhesive bonding or painting is a very important factor. However, use of traditional wet chemical methods of pretreatments shows ecological challenges and therefore, there is a clear interest in ecologically cleaner vacuum-based plasma technology. The plasma deposition of thin film coatings on aluminium that exhibit strong interfacial bonding could provide an alternative to the traditional chromate-based treatments. Plasma cleaning, etching and deposition in a single plasma reactor could provide a complete cycle before painting, avoiding the use, handling and disposal of hazardous materials [1]. Further, the 2024 aluminium alloy structure of an aircraft or a helicopter is commonly protected from severe conditions and heavy stresses by a paint coating [2]. Therefore, the effect of cold plasma treatment and its various parametric aspects prior to painting or adhesive bonding for corrosion behavior and adhesive joint performance have been commented upon.

The adhesive bonding of composite patch to repair cracks in metallic structures is an accepted technology in aerospace and automobile industries [3-5]. The bonded composite patch minimizes stress concentration at the joint and offers superior fatigue resistance of the repaired section than the conventional repair method. In addition, high modulus and strength of composites makes the patch thin, thus minimizing the aerodynamic drag of protruded repair section [6]. The bond strength between composite patch and metallic structure is significantly affected by the surface preparation of the composite patch and metallic structure [7, 8]. The optimal plasma treatment conditions and the effect of plasma surface treatment on the bond strength of aluminium and CFRP composites have been discussed [9].

PLASMA TREATMENT OF ALUMINIUM FOR IMPROVED ADHESION WITH PAINT

The 2024 aluminium is commonly protected from severe conditions and heavy stresses by a paint coating. The current industrial finishing process to apply a protective coating on a 2024 aluminium alloy surface contains three steps:

(i) Cleaning by Methyl-ethyl ketone (MEK) to remove grease and dust

(ii) XPD® primer application to enhance adhesion of the paint coating to the aluminium surface and

(iii) Painting.

After cleaning, the surfaces appear uniform and a 15 to 20 μm thick XPD® primer film is applied on the aluminium surface. XPD® primer is an epoxy-polyamide resin according to MIL-P-23377. The primer remains efficient after 7-8 hr from its application. Finally, layers of an acrylic paint are applied on the activated aluminium surface. The elimination of the traditional process of MEK cleaning by cold plasma means a reduction in polluting substances whereas eliminating the primer results in a decrease in the weight of the aluminium structure.

Polini[2] W et al have studied the two cases of plasma application in processing of aluminium before painting- the substitution of MEK cleaning and XPD® primer application with cold plasma (Al 2024 + cold plasma + paint) and the combination of cold plasma treatment with the traditional XPD® primer application (Al 2024+ cold plasma + XPD® primer + paint). They have compared the industrial and cold plasma process in terms of average percentage of area involving removal of paint under tape test and squaring test as shown in Fig 1.

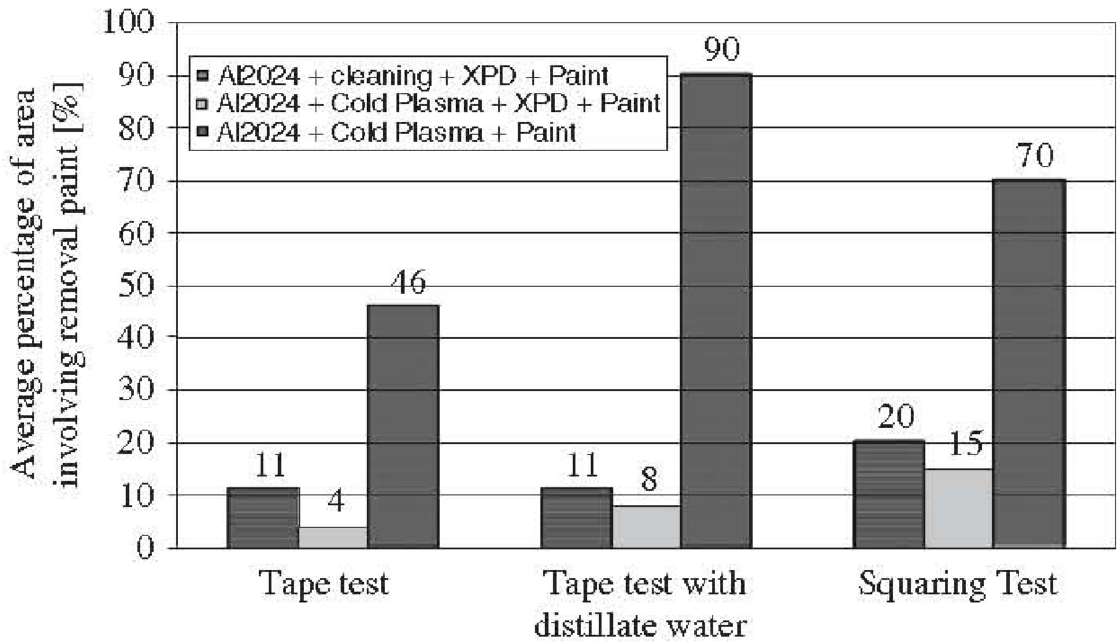

Figure1. Final comparison of samples processed by traditional process (Al2024+cleaning+ XPD® + paint) and those treated by cold plasma before XDP® primer application (Al2024 + cold plasma + XPD® + paint), in terms of Average percentage of area involving removal paint.

Under tape test, the samples where cold plasma has completely substituted primer deposition give a percentage of paint removed much higher than accepted limit of 12 %. Cold plasma added to the primer application results in a decrease of the area removed between the aluminium alloy and the paint by more than 50 %. Therefore, the cold plasma has allowed to substitute completely the MEK cleaning and to reduce the amount of XPD® primer to apply on the surface before painting. The samples under tape test with distilled water show similar results to those previously obtained by the tape test. In fact, all the process conditions give an average percentage value of the area involved by paint removal near that to the traditional process, with the exception of those where cold plasma substituted MEK cleaning and XPD® primer application. Under squaring test, the samples processed according to the traditional technology have passed the aeronautical prescription of percentage of an area of paint detachment lower than 35%. In contrast, the samples processed by cold plasma have given good results with primer application and bad results without primer application. The use of cold plasma process improves the adhesive properties when it is applied before the primer and, therefore decreases the area of paint removal by about 25% in comparison with the traditional technology (cleaning + XPD® primer). The increase in adhesive properties is attributed to the improvement in both wettability and sample surface uniformity. The wettability increase is connected to the activation and cleaning of the sample surface. The increase in sample surface uniformity is due to the cleaning effect of cold plasma treatment. The value of the contact angle of a sample without any treatment (i.e. without cleaning by MEK and without cold plasma) is equal to 850; cleaning by MEK decreases this significantly to 480. The value of the contact angle of a sample treated with cold plasma is equal to 340 lower than that due to cleaning by MEK. In addition, cold plasma allows decreasing strongly the percentage of both carbon and oxygen.

PLASMA-COATED ALUMINIUM ALLOY FOR IMPROVED CORROSION RESISTANCE AND ADHESIVE JOINT PERFORMANCE

The electrochemical impedance spectroscopy (EIS) and scanning Kelvin probe (SKP) are used to evaluate the corrosion behavior of plasma-coated aluminium alloy. The adhesion properties are assessed by wedge cleavage tests. Fernandes[1] J. C. S. et al. have studied the corrosion behavior and adhesive properties of plasma-coated Alclad AA2024-T3 when hexamethyl disiloxane (HMDSO) monomer and add-gases such as O_2, N_2, H_2 and Argon were employed. Plasma coatings from HMDSO and O_2 have proven to be effective primer coatings for adhesion promotion to Al [10]. From the literature [11], it is known that the onset of pitting corrosion is followed by a change in the shape of electrochemical impedance spectroscopy (Bode plots). From the Bode plot, the value of log[z] taken at a fixed angular frequency of ω=0.1 rad.s^{-1} is a useful measure of corrosion susceptibility. In their study, a threshold of $\log[z]_{\omega=0.1} \geq 6$ was set. When this parameter was below 6, it was taken to indicate that the specimen under study was displaying corrosion. It was taken that the samples had a good resistance to pitting corrosion if $\log[z]_{\omega=0.1}$ was ≥ 6 after 7 days immersion in 0.5 M NaCl solution.

Figure 2 represent the plot of $\log[z]_{\omega=0.1}$ vs. immersion time in 0.5 M NaCl solution for untreated and plasma coated Alclad 2024-T3. Both samples display good resistance to [pitting corrosion, with $\log[z]_{\omega=0.1} \geq 6$ after seven days of immersion. However, the untreated sample fails for an immersion time of 9 days, whereas the plasma-coated specimen resists pitting corrosion for more than 60 days.

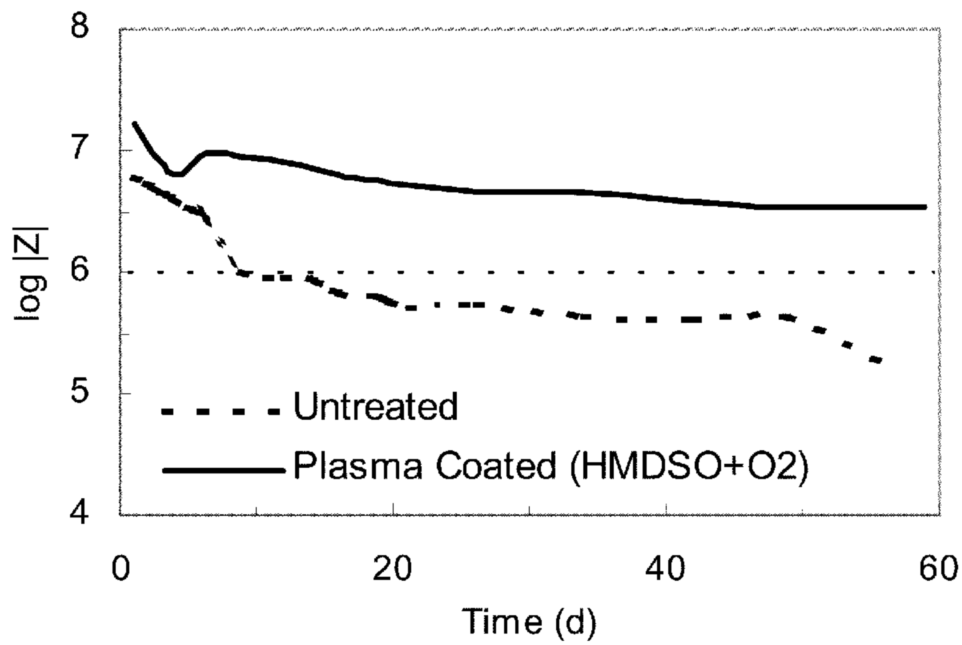

Figure 2. Plot of log[z]ω=0.1 vs. immersion time for untreated and Plasma coated Alclad 2024-T3 [1].

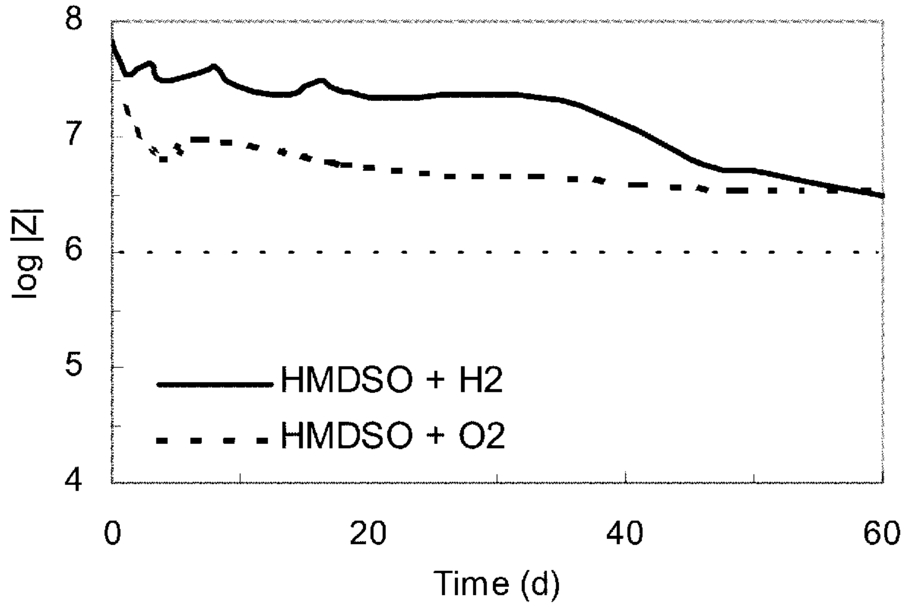

Figure3. Effect of the add-gas - Plot of log [Z] vs. time [1].

Figure3 represent the importance of add-gas used in the deposition by comparing samples coated using the gas mixtures of HMDSO+H_2 and HMDSO+O_2 in the same proportions. Although the best results from the adhesion tests are obtained with samples prepared from mixtures of HMDSO+O_2, the use of HMDSO+H_2 mixture lead to higher corrosion resistance. However, this enhanced resistance disappears for longer immersion times.

PLASMA TREATMENT OF ALUMINIUM FOR IMPROVED BONDING STRENGTH

Rhee[9] Kyong. Y. et al have investigated the surface treatment effect of aluminium by plasma on the bond strength of aluminium/ Ar^+ irradiated CFRP composites. The optimum plasma treatments conditions (treatment time, ratio of acetylene gas to nitrogen gas) and the optimal Ar^+ ion dose were determined. The peel strength and shear strength of surface-treated aluminium/ CFRP specimens were compared with those of untreated aluminium/ CFRP specimens. The optimal Ar^+ ion dose in the treatment of composite panels was determined based on water contact angle and surface energy. It was shown in a previous study [12] that IAR (ion-assisted reaction) method in oxygen environment forms more stable hydrophilic groups than those of the conventional ion beam method. It was determined based on the results of contact angle and surface energy that the optimal ion dose in the treatment of composite panel is in the range of 10^{15}-10^{17} ions/cm^2. Therefore, the surface of composite panel was treated with 10^{16} ions/cm^2 under oxygen environment.

In order to determine the optimal plasma treatment condition of aluminium panels, the optimal volume ratio of acetylene gas/nitrogen gas was determined first by measuring the water contact angle and then the scotch tape peel strength for the treatment times of 30 and 60 s. The contact angle was 820 before plasma treatment and changed between 400 and 1350 after plasma treatment. The contact angle was minimum at a volume ratio of 3:7 for the treatment time of 60 s, and at a volume ratio of 5:5 for the treatment time of 30 s. The scotch tape peel strength of untreated case was 1.8 N. The T-peel strength increased to a maximum value of 13.5 N at a volume ratio of 5:5 for the treatment time of 30 s while it increased to a maximum value 9.1 N at a volume ratio of 3:7 for the treatment time of 60 s. Therefore, the optimal volume ratio of acetylene gas/ nitrogen gas was determined to 5:5.In order to determine the optimal plasma treatment time, the water contact angle and scotch tape peel strength were measured as a function of treatment time at a 5:5 volume ratio of acetylene gas/ nitrogen gas. The contact angle was minimum (~ 400) when the treatment time was 30 s and the peel strength was maximum, when the treatment time was 30 s. therefore, the optimal plasma treatment time was determined to 30 s.

Figure 4 shows a comparison of shear strength for four types of specimens. It can be seen from e figure that the shear strength of plasma-treated aluminium/ irradiated CFRP specimen is largest and that of untreated aluminium/untreated CFRP is smallest. The T-peel strength has similar trend as with Shear strength.

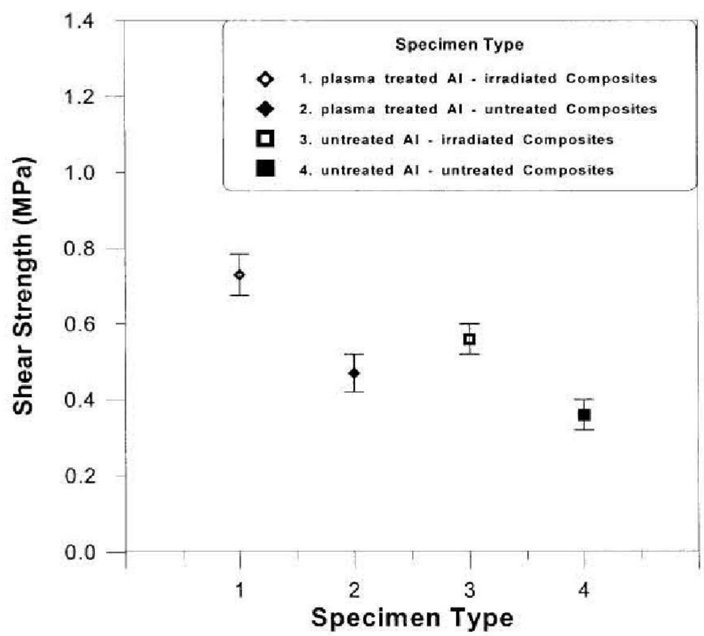

Figure 4. Comparison of shear strength for four types of specimens [9].

In another study, Sperandio[13] et al reported the influence of atmospheric plasma treatment of aluminium 1024 surface on its surface energy and bond strength with different proportions of plasma gases of nitrogen and oxygen. It was found in their study as shown in figure 6 that the optimum requirement of oxygen is 20 % which increases the hydroxide and/or oxyhydroxil groups and so the polar surface energy. These polar contributions enhance the wetting phenomena and so a high bond strength.

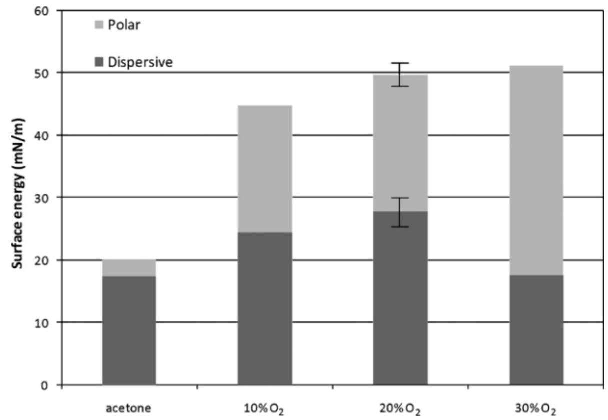

Figure 5. Surface energy measured on aluminium surface [13]

CONCLUSIONS

(1) Cold plasma substitute MEK cleaning step prior to painting, thus avoiding the use of polluting substances.

(2) Cold plasma improves the adhesion between paint coating and the 2024 aluminium alloy surface.

(3) Cold plasma may reduce the primer needed before painting and the weight of the whole structure.

(4) Plasma coatings obtained by the plasma deposition of HMDSO improve the corrosion resistance of the Alclad 2024-T3 alloy.

(5) In plasma treated aluminium/CFRP composite joint, it was found that the water contact angle was minimum and the peel strength was maximized when the aluminium was plasma treated for 30 s at a volume ratio of acetylene gas to nitrogen gas of 5:5.

(6) The bond strength (T-peel strength and shear strength) of plasma treated aluminium/irradiated CFRP specimen was largest and that of untreated aluminium/CFRP specimen was smallest.

(7) The shear strength and T-peel strength of plasma treated aluminium/untreated CFRP composite joints under the optimal treatment conditions was 33% and six times higher respectively than that of untreated Aluminium/untreated CFRP composite joints.

(8) The untreated Aluminium/irradiated CFRP specimen produced better bond strength than the plasma treated aluminium/untreated CFRP.

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