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Enhanced adhesion of epoxy-bonded steel surfaces using O_2/Ar microwave plasma treatment

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ABSTRACT

Steel surfaces have been modified using low pressure microwave plasma to enhance its adhesion with an epoxy adhesive. Optimization of the wettability of the surface was done using contact angle measurements for varying plasma parameters. Maximum wettability (19.9°) was obtained at 1000 W microwave power with 20 min of treatment time, −50 V sample bias and 1.67% O_2/Ar gas flow rate ratio. Enhanced wettability of the steel surface was attributed to increased surface roughness and oxide deposition. Using atomic force microscopy, surface roughness was observed to increase from 64.4 nm for the untreated surface to 76.7 nm for the O_2/Ar plasma treated surface. Deposition of oxides on the steel surface was also confirmed by the energy dispersive x-ray spectroscopy. Moreover, the increase in the total surface energy to 53.2 mN/m for the O_2 plasma treated steel surface supported the enhancement of its wettability, and hence, the adhesion with epoxy. Based on tensile test results, the adhesion strength of epoxy-bonded O_2/Ar plasma treated surfaces at optimum settings was increased to 3816.0 N, which is significantly higher compared to 3038.3 N for the epoxy-bonded untreated surfaces.

1. Introduction

Metals are generally easy to bond. However, various problems are associated to bonding of metallic substrates. One of these is the durability of bonded metals at the interface. To promote long term durability, high bonding strength is required between the metal and its adhesive [1]. Previous researches suggested that different adhesion processes including mechanical interlocking, chemical bonding, and electrostatic attractions can be responsible for formation of strong bonds [2,3]. However, the adhesion process of an adhesive is difficult to generalize as it can vary from one application to the other.

Surface modification of a metal prior to application of an adhesive can be used to enhance the bonding strength. Plasma treatment offers an effective and non-solvent based processing of different metals, providing strong and durable bonds with many adhesives through modification of the surface functionalities [4,5]. Surface modification of steel to enhance its bonding strength with different materials has been done using various plasma techniques such as atmospheric pressure plasma [2,3,6], dc magnetron sputtering [7], radio frequency plasma [7,8] and ion implantation [8]. In 2003, Kim et al. modified stainless steel using N_2/O_2 atmospheric pressure plasma jet at room temperature. It was confirmed that reactive etching and oxidation of ions and activated species in the plasma result to a change in the hydrophilicity of the material [9]. Another study in 2006 by Tang et al. has also shown improvement of the adhesion properties of AISI 316 L stainless steel plates, using Ar/O_2 plasma at atmospheric pressure. They confirmed that new oxygen-containing groups formed through plasma treatment improved the adhesion strength of the metal [2].

In this study, a microwave plasma device from the Plasma Physics Laboratory of the University of the Philippines is used to enhance the adhesion strength of two steel surfaces with an epoxy adhesive. Various plasma parameters such as treatment time, microwave power, sample bias and O_2/Ar gas flow rate ratio are optimized based on contact angle measurements to achieve enhanced wettability, and hence, improved adhesion. Changes in the stainless steel morphology, elemental composition and interaction with its epoxy adhesive after plasma treatment are also investigated.

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2. Methodology

2.1. Sample preparation

Un-annealed steel was cut into $5\times1\times0.3$ cm^3 (Type 1), and $5\times2\times1$ cm^3 (Type 2) sample sizes. Type 1 samples were used for wettability experiments, whereas Type 2 samples were used for adhesion experiments. Surfaces of both samples were initially polished using sandpaper with grades 350 and 1000, then wiped with methanol.

2.2. Microwave plasma device

The device (Fig. 1) is composed of a 2.45 GHz magnetron with 2.0 kW continuous wave output power supply. A quartz glass allows microwaves from the waveguide to enter the vacuum chamber where the plasma is generated. Samarium–Cobalt (Sm–Co) magnets are placed outside the circumference of the plasma chamber. For the plasma treatment, argon (Ar) and oxygen (O_2) were used as working gases. These gases were controlled by a Mass Flow Controller Type 1479 A.

The vacuum system consists of a rotary pump and a diffusion pump. A PCGE 61.02-NEE Refrigerated Circulator serves as the cooling system. Other details on the microwave device can be found in the paper of Ting et al. [10], Tumlos et al. [11] and Arciaga et al. [12].

2.3. Plasma treatment

Steel samples were placed horizontally along the top of an $8\times2\times2$ cm^3 substrate holder inside the vacuum chamber. The rotary pump lowered the pressure to less than 0.01 Torr. Using the diffusion pump, the samples were exposed to O_2/Ar plasma discharge at a fixed base pressure of 9.0×10^{-5} Torr.

Plasma treatment of the samples for varying treatment time, microwave power, sample bias and O_2/Ar gas flow rate ratio were performed successively to obtain the optimum settings for enhanced wettability based on contact angle measurements. Table 1 shows the list of parameter values used during the treatment.

In this study, initial plasma settings were fixed at 20 min of treatment time using 1000 W of microwave power, −30 V sample bias and 3.33% O_2/Ar gas flow rate ratio.

2.4. Contact angle measurement

Contact angle measurement was performed using Dino-Lite Digital Microscope and DinoCapture software. Deionized water (DI) and ethylene glycol (EG) were used as polar components, whereas, diiodomethane (DM) was used as the non-polar component [13]. Each test liquid was dropped onto the surface of the samples using a 0.5 mL syringe. In this experiment, 10-point measurement on each sample was performed at random positions, and the average value was considered as the contact angle of the sample.

2.5. Adhesion and tensile tests

Two pieces of Type 2 samples (Fig. 2) were bonded in pairs using a bisphenol A-epichlorohydrin-based epoxy. The area of the bonding surface is 2×1 cm^2. Four sample combinations were done. Sample 1 includes two untreated sides (U–U), Samples 2 and 3 include one untreated and one treated sides (U–T) using Ar plasma and O_2/Ar plasma, respectively, and Sample 4 included two treated sides (T–T) using O_2/Ar plasma. Samples were prepared to determine the effect of Ar and O_2/Ar plasma treatments on the adhesion of two steel surfaces. For treated samples, plasma treatment was done at optimum plasma parameters.

Table 1
Variations of plasma parameters.

Parameter	Values
Treatment time (mins):	5, 10, 20, 30, 40
Microwave power (W):	200, 400, 600, 800, 1000
Bias (V):	0, −10, −20, −30, −50
O_2/Ar gas flow rate ratio (%):	0, 1.67, 3.33, 5.00, 6.67
Oxygen (SCCM)	0, 0.5, 1.0, 1.5, 2.0
Argon (SCCM)	30

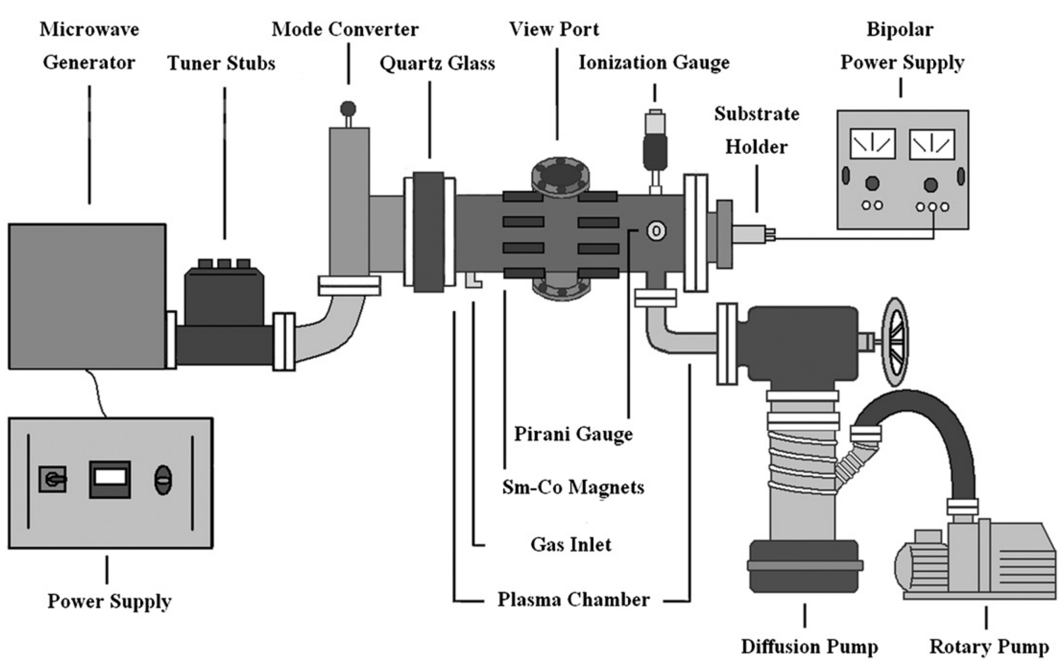

Fig. 1. Schematics of the microwave plasma device.