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# A morphological and electrochemical comparison of the corrosion process of aluminum alloys under simulated acid rain conditions

Husnu Gerengi<sup>a,\*</sup>, Gozen Bereket<sup>b</sup>, Mine Kurtay<sup>a</sup>

<sup>a</sup> Corrosion Research Laboratory, Kaynasli Vocational College, Duzce University, Kaynasli 81900, Duzce, Turkey

<sup>b</sup> Department of Chemistry, Faculty of Arts and Science, Eskisehir Osmangazi University, Eskisehir 26480, Turkey

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## ABSTRACT

The corrosion behavior of aluminum 6060 and 6082 alloys was examined by scanning electron microscopy (SEM), atomic force microscopy (AFM) and dynamic electrochemical impedance spectroscopy (DEIS) after exposure to simulated acid rain for 1, 5, 24 and 48 h. The electrochemical parameters obtained from DEIS were compared with the AFM and SEM studies. Results showed that DEIS is a sensitive technique detecting even very moderate changes in charge-transfer resistance ( $R_{ct}$ ); it exhibits a very good correlation with surface morphological changes in relation to exposure time. This research also demonstrated the importance of performing corrosion measurements after a longer time period.

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## 1. Introduction

Aluminum and its alloys are widely applied in a variety of fields such as construction, electrical engineering and transportation due to their excellent mechanical properties and corrosion resistance. In this respect, AlMgSi (6000 series) alloys are generally regarded as corrosion-resistant [1]. These alloys are normally exposed to outdoor conditions without protection because under normal atmospheric conditions, a passive oxide film develops naturally on the alloy surface. However, the natural oxide film on aluminum does not offer sufficient protection, and thus, Al and its alloys are prone to corrosion in humid, polluted atmospheric conditions or in aqueous environments. In the case of industrial applications, it is of great importance to recognize the corrosion behavior of aluminum and its alloys under polluted atmospheric conditions. Outdoor exposure tests under different environmental conditions along with laboratory accelerating experiments have been carried out in order to study the influence of atmospheric environmental factors, including corrosive ions (e.g., Cl, SO<sub>2</sub>, NO<sub>x</sub>, and CO<sub>2</sub>), relative humidity, exposure time to wetness, etc., on the corrosion behavior of aluminum [2–14]. It is well known that pollution has been increasing rapidly as a result of industrialization, population explosion, deforestation, vehicular exhaust, and the burning of fossil fuels containing nitrogen, sulfur and carbon. Sulfur, nitrogen, and carbon do not undergo complete combustion, but evolve in the atmosphere by reacting with atmospheric oxygen to form oxides

of nitrogen, sulfur, and carbon (NO<sub>x</sub>, SO<sub>x</sub>, CO<sub>x</sub>). This results in an increase in the concentration of these gases in the atmosphere [15,16].

Consequently, the acid rain conditions developed in a polluted atmosphere lead to the deterioration of metals and alloys subjected to such environments. The corrosion behavior of 6060 and 6082 type aluminum alloys immersed in simulated acid rain was investigated using dynamic electrochemical impedance spectroscopy (DEIS) and potentiodynamic polarization techniques in a previous study [17]. It was found that the AA6082 type alloy was more susceptible to pitting than the 6060 type alloy, although there was increased Mn and Cr content in the 6082 alloy. The decreasing corrosion rate as the Al content was increased was explained by the distribution of the Al-rich phase.

The aim of this study was to examine the corrosion kinetic of 6060 and 6082 type alloys in simulated acid rain. For this purpose, DEIS was conducted over a 48-h period. In addition, two complementary techniques, scanning electron microscopy (SEM) and atomic force microscopy (AFM), were used to reveal time-dependent corrosion and the surface morphological changes during the 48 h.

## 2. Experimental approach

### 2.1. Materials

Silver/Silver chloride (Ag/AgCl) and Pt-mesh were used as reference and auxiliary electrodes, respectively. The working electrodes were first prepared using 400–2000 grade abrasive paper; following this procedure, they were rinsed with distilled water and degreased with acetone. The exposed surface area of the working electrodes in

\* Corresponding author. Tel.: +90 5053987953.

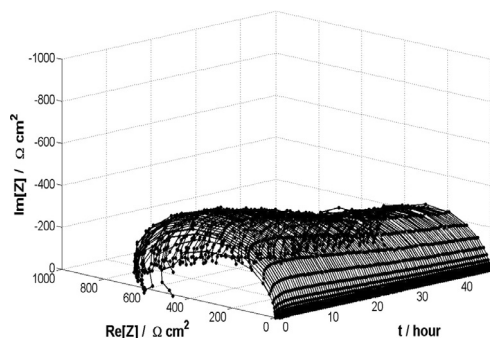
E-mail address: [husnugerengi@duzce.edu.tr](mailto:husnugerengi@duzce.edu.tr) (H. Gerengi).

**Table 1**  
Chemical compositions of working electrodes (wt.%).

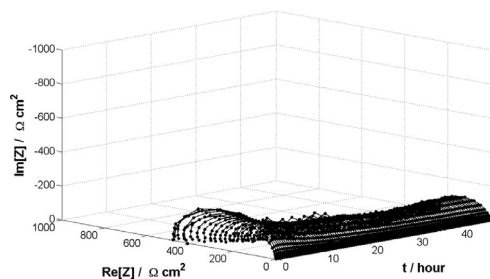
Samples	Si	Fe	Cu	Mn	Mg	Cr	Zn	Ti	Al
AA6060	0.3–0.6	0.1–0.3	0.1	0.1	0.3–0.6	0.05	0.15	0.1	98–99
AA6082	0.7–1.3	0.5	0.1	0.4–1.0	0.6–1.2	0.25	0.2	0.1	95–97

**Table 2**  
Chemical composition of the artificial acid rain.

Material	H <sub>2</sub> SO <sub>4</sub>	HNO <sub>3</sub>	Na <sub>2</sub> SO <sub>4</sub>	NaNO <sub>3</sub>	NaCl	(NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub>
(g/l water)	0.032	0.015	0.0128	0.0084	0.0336	0.0184



**Fig. 1.** Impedance changes of AA6060 alloy in artificial acid rain solution for 48 h obtained from DEIS.



**Fig. 2.** Impedance changes of AA6082 alloy in artificial acid rain solution for 48 h obtained from DEIS.

the solution was 0.785 cm<sup>2</sup> and all measurements were performed starting after immersion for 60 min in artificial acid rain solution. The chemical compositions of the electrodes are given in Table 1. The chemical composition of the artificial acid rain is given in Table 2 [18]. The DEIS measurements were performed at room temperature and during all measurements; the solution was stirred with a magnetic

stirrer (300 rot/min) in order to maintain a homogenous solution. Each experiment was repeated at least seven times to ensure that the most accurate agreement was achieved. All chemicals were obtained from Merck, with purity listed as follows: H<sub>2</sub>SO<sub>4</sub> (98%), HNO<sub>3</sub> (65%), Na<sub>2</sub>SO<sub>4</sub> (99%), NaNO<sub>3</sub> (99%), NH<sub>4</sub>SO<sub>4</sub> (99.5%). The same bottles were used throughout the experiment.

## 2.2. Method

### 2.2.1. Dynamic electrochemical impedance spectroscopy (DEIS)

The DEIS electrochemical method can be used successfully under both galvanostatic and potentiostatic conditions [19]. The generation of the current perturbation was performed with a National Instruments Ltd. PCI-4461 digital–analog card. The same card was used to measure the current and voltage signals. The sampling frequency was 12.8 kHz. The perturbation signal was a package composed of current sinusoids of the frequency range from 4.5 kHz to 700 mHz.

### 2.2.2. Surface morphology studies

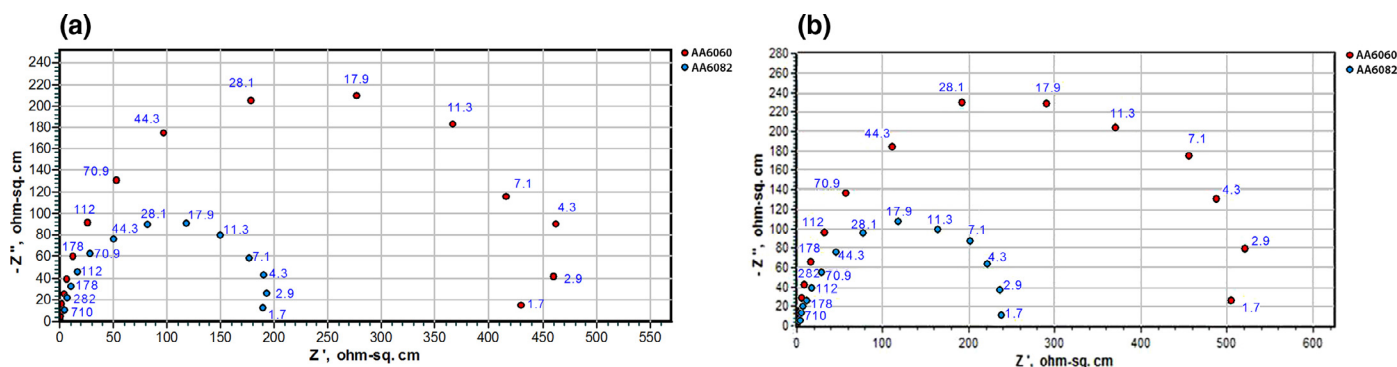
The aluminum alloys were immersed in artificial acid rain solution at 25 °C for different time periods of exposure (after 50 s, then after 1, 5, 12, 24 and 48 h) and their surface morphologies were then examined by means of SEM (FEI, Model: Quanta FEG 250) and AFM (PARK SYSTEMS, Model: XE-100E).

## 3. Results and discussion

### 3.1. DEIS measurements

Dynamic electrochemical impedance spectroscopy was applied for evaluating and tracking the changes in corrosion during the 48 h immersion in simulated acid rain solution. Figs. 1 and 2 display the DEIS results obtained from the AA6060 and AA6082 samples subjected to 48 h exposure to artificial acid rain in order to determine the change in corrosive behavior over time. As can be seen from the figures, the DEIS plots of the two alloys are very similar, and individual impedance spectra presented in the form of a Nyquist plot appear in the shape of flattened semicircles in the high frequency range, the magnitude of which decreases with time.

Fig. 3 shows a comparison of the first and last Nyquist plots of AA6060 and AA6082 obtained via DEIS after 50 s (a) and 48 h (b) exposure to artificial acid rain solution. The differences between (a)



**Fig. 3.** The first and last Nyquist plots of AA6060 and AA6082 alloys obtained via DEIS after (a) 50 s and (b) 48 h exposure to artificial acid rain solution.

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