



## Uniform and pitting corrosion events induced by $\text{SCN}^-$ anions on Al alloys surfaces and the effect of UV light

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### ABSTRACT

The influence of the alloying elements on the uniform and pitting corrosion processes of Al-6061, Al-4.5%Cu, Al-7.5%Cu, Al-6%Si and Al-12%Si alloys was studied in 0.50 M KSCN solution at 25 °C. Open-circuit potential, Tafel polarization, linear polarization resistance (LPR) and ICP-AES measurements were used to study the uniform corrosion process on the surfaces of the tested alloys. Cyclic polarization, potentiostatic current-time transients and impedance techniques were employed for pitting corrosion studies. Obtained results were compared with pure Al. Passivation kinetics of the tested Al samples were also studied as a function of applied potential,  $[\text{SCN}^-]$  and sample composition by means of potentiostatic current transients. The induction time, after which the growth of stable pits occurs, decreased with increasing applied potential and  $[\text{SCN}^-]$ . Regarding to uniform corrosion, alloyed Cu was found to enhance the corrosion rate, while alloyed Si suppressed it. Alloying elements of the tested samples diminished pitting attack to an extent depending on the percentage of the alloying element in the sample. Among the investigated materials, Al-Si alloys exhibited the highest corrosion resistance towards uniform and pitting corrosion processes in KSCN solutions. The passive and dissolution behaviour of Al was also studied under the conditions of continuous illumination (300–450 nm) based on cyclic polarization and potentiostatic techniques. The incident photons had a little influence on pit initiation and a marked effect on pit growth. These explained in terms of a photo-induced modification of the passive film formed on the anode surface, which render it more resistant to pitting. The effects of UV photons energy and period of illumination on the morphology of the pitted surfaces were also studied.

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### 1. Introduction

The electrochemical behaviour of aluminium and aluminium alloys is a subject of a large number of studies due to increased industrial applications of these materials (construction, metallurgy, mechanical engineering and transport vehicle industry, from automobiles to aircrafts, as well as food processing industry in the production of packaging material, etc.). These applications are often possible because aluminium spontaneously forms stable oxide films in air and in aqueous solutions.

Although pure Al is too soft to be used as a heavy duty material for large structures, high-strength Al alloys can be produced by addition of appropriate alloying elements, such as Cu, Mg and Zn and by suitable heat treatment procedures [1–3]. For these reasons, studies of the corrosion and stability of aluminium and its alloys are still of considerable interest. Aggressive anions, such as the halides,

induce breakdown of the protective passive film [4–7] and can result in catastrophic failure of the material. There have been extensive investigations on the role of chloride ions in the breakdown of the passive film, repassivation and initiation of localized corrosion of Al and high-strength Al-based alloys. Furthermore, the effect of alloy content [4,8–10], electrolyte composition [11], heat treatment [12] and mechanical stress [13] on pitting and repassivation of Al-based alloys in halide media have been reported.

A program is in progress in our lab to study the pitting corrosion, corrosion inhibition and electrochemical behaviour of Al and some of its alloys in some aqueous media [14–17]. It was shown recently in our lab that Al and Al-Zn alloys pit in KSCN solutions [18,19]. The purpose of this work is to study the effect of the alloying elements on the uniform and pitting corrosion processes of the following alloys: Al-6061, Al-4.5%Cu, Al-7.5%Cu, Al-6%Si and Al-12%Si in KSCN solutions. Pure Al was also used in this work for comparison. Various electrochemical techniques including Tafel polarization, linear polarization resistance (LPR), cyclic polarization, potentiostatic and impedance were applied in this study. An independent method of chemical analysis, namely inductively coupled plasma atomic emission spectroscopy (ICP-AES) was also used to clarify the

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**Table 1**  
Composition (wt%) of the tested Al–Cu and Al–Si alloys.

Alloy	Al	Cu	Mn	Si	Mg	Zn	Sn	Cr	Ti
Al–4.5%Cu	Balance	4.5	0.02	0.03	0.02	0.022	0.005	0.001	0.01
Al–7.5%Cu	Balance	7.5	0.02	0.03	0.02	0.022	0.005	0.001	0.01
Al–6%Si	Balance	0.02	0.02	6.0	0.03	0.01	–	–	–
Al–12%Si	Balance	0.02	0.02	12.0	0.03	0.01	–	–	–

role of the alloying elements in the uniform corrosion process of the tested Al samples. The kinetics of the passivation and pit growth on the surfaces of Al and its tested alloys has been studied as a function of applied anodic potential and  $[\text{SCN}^-]$ . It was also the purpose of the present work to study the photo-inhibition of localized corrosion of Al in KSCN solutions and the factors affecting the degree of photo-inhibition based on cyclic polarization and potentiostatic techniques. Some SEM examinations were performed to clarify the effect of the incident photons on the morphologies of the pitted surfaces.

## 2. Experimental

The working electrodes employed here were made of pure Al wire (Sigma–Aldrich, 1.0 mm diam., 99.999), Al–Mg rod (Al-6061 alloy): 6.35 mm diam., Stock #42052, Lot #G14K32 (97.93% Al, 1.0% Mg, 0.60% Si, 0.27% Cu, 0.20% Cr), Al–4.5%Cu, Al–7.5%Cu, Al–6%Si and Al–12%Si alloys. The composition of the tested Al–Cu and Al–Si alloys is presented in Table 1. The pure Al wire was mounted into Teflon leaving a constant surface area of  $0.00785 \text{ cm}^2$  to contact the solution. The tested Al–Cu and Al–Si alloys were also in the form of cylindrical rods of base diameters 0.50 and 0.4 cm, respectively. The rods of Al-6061, Al–Cu and Al–Si alloys were welded with Cu-wire for electrical connection and mounted into glass tubes of appropriate diameter using Araldite to offer an active flat disc shaped surface for the working electrode, to contact the test solution. The surface pre-treatment of each electrode was carried out by grinding with different grades of emery papers down to 1200 grit. The electrode was then, rinsed with acetone, distilled water and finally dipped in the electrolytic cell.

Experiments were performed in a three-electrode PTFE cell equipped with a quartz window to allow irradiation of the test electrode. A saturated calomel electrode (SCE) was used as the reference electrode and a platinum wire, coiled inside the PTFE cell, was used as the auxiliary electrode. The working electrode was irradiated at wavelengths between 300 and 450 nm using a 150 W UV-enhanced Xe lamp (Oriol model 6254) and a 1/8 monochromator (Oriol model 77250). The incident power density at 300 nm was  $400 \text{ mW cm}^{-2}$ , giving a photon flux of  $6.0 \times 10^{14} \text{ s}^{-1} \text{ cm}^{-2}$ . Electrochemical experiments were carried out under the conditions of non-illumination and continuous illumination of the working electrode in KSCN solutions at  $25^\circ\text{C}$ .

The experiments were carried out in 0.50 M KSCN solution freshly prepared from analytical grade chemical reagents using water purified by a Millipore Milli-Q system, and was used without further purification. For each run, a freshly prepared solution as well as a cleaned set of electrodes was used. Each run was carried out in aerated stagnant solutions at the required temperature ( $\pm 1^\circ\text{C}$ ), using water thermostat.

Tafel extrapolation and linear polarization resistance (LPR) methods were used to evaluate the effect of the alloying elements on the rate of the uniform corrosion process. Cathodic and anodic polarization curves were determined by the potentiostatic method using 10 mV increments of potential, and steady state currents were observed within 30 min at each applied potential.

ICP–AES (inductively coupled plasma-atomic emission spectroscopy) has been also used as an independent chemical

(non-electrochemical) method to monitor the rate of corrosion and to confirm validation of corrosion rates measured by Tafel extrapolation method. In the ICP method, the  $\text{Al}^{3+}$  ions concentration was determined as a function of sample composition and immersion time (from 1.0 day up to 30 days) using PerkinElmer Optima 2100 Dual View inductively coupled plasma atomic emission spectrometry (ICP–AES) instrument connected with AS 93 Plus auto sampler.

Cyclic polarization measurements were carried out by sweeping linearly the potential from the starting potential into the positive direction at a scan rate of  $0.50 \text{ mV s}^{-1}$  to required potential value and then reversing the scan direction at the same scan rate until forming a well-defined hysteresis loop. These measurements were performed to evaluate the influence of the alloyed elements on the pit growth of the tested Al samples. Potentiostatic technique was also employed to study the kinetics of the passivation, and pit initiation and growth processes. The potentiostatic current-time measurements were carried out after holding the working electrode at a constant anodic potential ( $E_a$ ), where the anodic current was recorded against time. The stabilization period prior to collecting data was 12 h. The open circuit potential of the working electrode was measured as a function of time during this stabilization period. This period of 12 h was quite sufficient to reach a quasistationary value for the open circuit potential.

Impedance measurements were carried out using AC signals of amplitude 5 mV peak to peak at the open circuit potential in the frequency range 100 kHz to 1.0 mHz. To determine the impedance parameters of Al and its tested alloys in these solutions, the measured impedance data were analyzed by using the computer program EQUIVCRT [20].

For surface morphology studies regarding pitting corrosion and its control using the UV light, the pure Al wire was exposed to pitting attack in 0.50 M KSCN solutions at  $25^\circ\text{C}$  under illuminated and non-illuminated conditions. The pure Al electrode was exposed to an anodic potential of 400 mV ( $>E_b$ ) as a function of duration of illumination at a given wavelength (300 nm) and energy of the incident photons at a fixed duration time of 10 min. The electrode was held at the given potential for a certain time under the conditions of illumination and non-illumination, and finally washed thoroughly and submitted to 20 min of ultrasonic cleaning in order to remove loosely adsorbed ions.

## 3. Results and discussion

### 3.1. Uniform corrosion process

#### 3.1.1. Open-circuit potential vs. time measurements

The time evolution of the open-circuit potential (OCP) for pure Al, Al-6061, Al–4.5%Cu, Al–7.5%Cu, Al–6%Si and Al–12%Si alloys in 0.50 M KSCN solution is presented in Fig. 1. It can be seen, in all cases, that in the first moments of immersion the open circuit potential moves rapidly towards less negative values due to the initial formation and growth of the passive oxide film. Then the electrode potential shifts towards the more negative values to an extent depending on sample composition. This trend may be attributed to dissolution of the protective alumina layer as the result of  $\text{SCN}^-$  adsorption and subsequent formation of Al–SCN soluble complex

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