ANALYSIS ON AlMg$_2$ AS RSG-GAS CLADDING MATERIAL CORROSION IN CHLORIDE CONTAINING WATER

ANALISIS KOROSI TERHADAP MATERIAL AlMg$_2$ SEBAGAI KELONGSONG BAHAN BAKAR RSG-GAS DALAM AIR MENGANDUNG KLORIDA

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Diterima 17 April 2018, diterima dalam bentuk perbaikan 6 September 2018, disetujui 14 September 2018

ABSTRACT

CORROSION ANALYSIS ON AlMg$_2$ AS RSG-GAS CLADDING MATERIAL IN CHLORIDE CONTAINING WATER. *The AlMg$_2$ is one of an alluminium alloy that used as cladding material for the RSG GA. Siwabessy (RSG-GAS) research reactor in Serpong, Indonesia. The reactor uses demineralized water as primary coolant with 6.5 to 7.5 of pH. A poor treatment of water in primary coolant can lead to the problem of AlMg$_2$ integrity. The primary coolant concentration of chloride must be lower than 0.0094 ppm to protect cladding corrosion. The purpose of this study is to determine the effect of temperature and chloride ion concentration to AlMg$_2$. The method in this research is to observe the corrosion rate for AlMg$_2$ material by using Potentiostat. The laboratory experiments were conducted in various temperatures (28, 35, 40 and 45°C) and concentrations of sodium chloride were 0.005, 0.010, 0.015, 0.020, 0.025, 0.030 and 0.035 ppm. The results show the corrosion rates were very small, and the highest corrosion rate occured was 1.23 x 10^{-3} mpy in 0.035 ppm of NaCl at 45˚C.*

Keywords*: AlMg$_2$, corrosion, cladding material, chloride ion, temperature, Potensiotat.*

ABSTRAK

ANALISIS KOROSI TERHADAP MATERIAL ALMg$_2$ SEBAGAI KELONGSONG BAHAN BAKAR RSG-GAS DALAM AIR MENGANDUNG KLORIDA. *AlMg$_2$ adalah salah satu paduan aluminium yang digunakan sebagai bahan kelongsong bahan bakar Reaktor Serbaguna GA. Siwabessy (RSG-GAS) di Serpong, Indonesia. Reaktor ini menggunakan air bebas mineral sebagai pendingin primer dengan pH 6,5-7,5. Buruknya kondisi air pendingin primer dapat menyebabkan masalah integritas AlMg$_2$. Konsentrasi klorida pada pendingin primer harus lebih rendah dari 0.0094 ppm untuk menghindari terjadinya korosi kelongsong bahan bakar. Tujuan dari penelitian ini adalah untuk menentukan pengaruh suhu dan konsentrasi ion klorida terhadap AlMg$_2$. Metode penelitian ini adalah mengamati laju korosi AlMg$_2$ dengan menggunakan Potentiostat. Penelitian dilakukan pada berbagai suhu (28, 35, 40 dan 45 °C) dan dalam 0.005, 0.010, 0.015, 0.020, 0,025, 0.030 dan 0.035 ppm natrium klorida. Data hasil penelitian menunjukkan laju korosi AlMg$_2$ sangat kecil, dan laju korosi tertinggi terjadi pada suhu 45˚C dalam 0.035 ppm NaCl yakni 1,23 x 10^{-3} mpy*

Kata kunci*: korosi AlMg$_2$, material kelongsong, ion klorida, suhu, Potensiotat.*

INTRODUCTION

Batan has 3 research reactors. The biggest one is Reaktor Serbaguna GA Siwabessy (RSG-GAS). It has been operated for almost 30 years. The reactor has been operated safely. It is possible the degradation and corrosion process occurred at structure materials and reactor components. The AlMg$_2$ is used as fuel cladding for this reactor. It has its own excellent corrosion resistance and its usage as one of the primary metals of research reactor. It can form itself the oxide layer film that is bonded strongly on its surface. Aluminum has high resistance to corrosion under the majority of service conditions, and no colored salts are formed to stain adjacent surfaces or discolor products with which it comes into contact [1,2].

Corrosion involves the reaction of a metallic material with its environment and it is a natural process in the sense that the metal is attempting to revert to the chemically combined state in which it is almost invariably found in the earth's crust. Corrosion is a natural process that can cause the degradation on material, component and other structures [3].

The corrosion process can not be able to be stopped. Corrosion can be controlled through many ways; such as, using the material resist to corrosion or controlling the environment. The environment of material has an influence to the integrity of material toward corrosion.

Many type of material are used in nuclear reactor depend on function and environment. Aluminum and its alloy is one of the widely used corrosion-resistant metals. The corrosion rate of aluminum depends mainly on the thickness of the oxide layer and its compactness or crystallographic structure. The corrosion process is also influenced by the electric change of the surface. It is known that the charge on the surface of the oxidized aluminum is the consequence of the interaction of the oxide and the surroundings. The results of the former investigations of the anodic oxidized aluminum have shown that the charge of the surface depends on the oxidation process.

$AlMg_2$ as material cladding has to keep its integrity in order to the reactor can be operated safely. When aluminum surfaces are exposed to the atmosphere, a thin invisible oxide skin forms immediately, this protects the metal from further oxidation. This self-protecting characteristic gives aluminum its high resistance to corrosion. Unless exposed to some substance or condition that destroyed this protective oxide coating, the metal remains fully protected against corrosion [4]. Aluminum is highly resistant to weathering, even in industrial atmospheres that often corrode other metals. It is also corrosion resistant to many acids. Alkalis are among the few substances that attack the oxide skin and therefore are corrosive to aluminum although the metal can safely be used in the presence of certain mild alkalis with the aid of inhibitors, in general, direct contact with alkaline substances should be avoided.

$AlMg_2$ is used in RSG–GAS as cladding material for containing fuel and fission products. Aluminum and its alloys have a good resistance toward corrosion at low temperature and will form a thin oxide film at its surface which separate the metal from its environment and protects from further oxidation. This self-protecting characteristic gives aluminum its high resistance to corrosion. Unless exposed to some substance, such as chloride or condition that destroys this protective oxide coating [5,6]. This oxide film can inhibit further corrosion process, but some of them vulnerable to localized corrosion in solution containing chloride. Most of metal and alloys, including aluminum have capability to build passive film from metal oxide. This film will separate the metal from its environments. For aluminum, pit and crevice corrosion are most commonly produced by halide ions, of which chloride (Cl^-) is the most frequently encountered in service [7]. The passive film will be destroyed in environment containing the aggressive ion such as; chloride ion. Temperature also has an effect to corrosion of materials. Increasing temperature is usually followed by increasing corrosion rate [4,8].

$AlMg_2$ degradation can cause an effect to reactor safety. The cladding failure will release radioactive to environment. In research reactors, corrosion processes are strongly affected by operational measured variables such as environment medium, pH, temperature, conductivity and chloride ion content. The protection mechanism to be outlined focuses mainly on maintaining high quality of coolant water.

The RSG-GAS has two coolant systems, i.e. the primary and secondary cooling systems. Both of these systems ensure that the reactor coolant temperature is controlled. During normal operation, heat generated in the core is taken by primary cooling system and transferred to the secondary cooling system via heat exchangers. The heat is discharged into the environment through cooling towers by forced circulation.

Table1. RSG-GAS demineralized water specification [3,9]

No	Parameter	Value
1	pH	6,5 – 7,5
2	Conductivity (max)	2 μS/cm
3	Chloride ion (max)	0,0094 ppm
4	Copper ion (max)	0,0056 ppm

Demineralized water is used for primary coolant and it is produced by demineralized production system that has requirement as noted in Table 1 [9]. The primary cooling system plays an important role in ensuring the integrity and reliability of the fuel cladding that made of $AlMg_2$. The plant's demineralized water system, also known as the GCA01, produces that water. The reactor pool and all subsystems in the primary system are filled by demineralized water, but the secondary system uses freshwater supplied by the utility supplying Puspiptek (the Research Center for Science and Technology, namely the agency which manages the research area where the RSG-GAS is located). Prior to use, however, the water from Puspiptek is given mechanical treatment which includes flow through precipitation pools and sand filter pools.

The primary water coolant specification of the RSG GAS is shown in Table 1. The conductivity, chloride ion and copper ion in reactor coolant are very small. Chloride ion as an aggressive ion in small concentration can cause the pitting corrosion. Pitting is a localized form of corrosive attack. Pitting corrosion is typified by the formation of holes or pits on the metal surface. Sometimes pitting corrosion can be quite small on the surface and very large below the surface. Passive metals, such as stainless steel, aluminum and its alloy resist in corrosive media and can perform well over long periods of time [10]. However, if corrosion does occur, it forms randomly in pits. Pitting is most likely to occur in the presence of chloride ions, combined with such depolarizers as oxygen or oxidizing salts [11,12]. Aggressive ions such as chloride tend to prevent the formation of protective oxide films on the metal surface and thus increase corrosion [13,14].

Leckie [15] states that the presence of neutral and acid salts in water tends to increase the corrosion rate of iron, steel and low steel alloy by influencing anodic polarization, the conductivity of the solution, the diffusion and solubility of oxygen, and the solubility of the corrosion products.

The purpose of this study is to observe temperature and chloride ion effect to $AlMg_2$ RSG–GAS cladding material. The method that used in this research is to observe the corrosion rates of $AlMg_2$ material in NaCl concentration at several temperature conditions using Potentiostat. Tests were conducted in 28, 30, 35, 40 and 45 ˚C. The RSG – GAS cooling water temperature in normal operation is around 40 ˚C. The $AlMg_2$ corrodes in solution containing NaCl solution. The chloride ion content of the water should be maintained as low as achievable and at less than 0.0094 ppm for optimum corrosion protection. The chloride ion in cooling water is possible come from the water make - up system. Chloride ions break down the passive film on aluminum and promote metal dissolution. The chloride ions in these chemicals in small amount will destroy the passive film on aluminum and cause aggressive pitting corrosion. The corrosion process of aluminum increases with increasing temperature. Corrosion resistance is used as criterion in determining the lifetime of components and structures. The aim of this research is to determine the corrosion of $AlMg_2$, based on corrosion rate in various NaCl cocentrations and temperatures. The experiment used electrochemical method by using Potentiostat.

METHODOLOGY

Specimen Preparation

$AlMg_2$ was used as specimen in this experiment. The specimen dimension is 10 mm of length, 10 mm of width and 5 mm of thickness. The specimen surface was treated with polished paper from grade 400, 600, 800 and 1000 and then polished with Metadi II diamond paste

Corrosion Test Using Potentiostat

Anodic polarization was obtained by a potential scanning from –500 mV at an open circuit potential (OCP) to 600 mV OCP at a rate of 0.2 mV/sec. A silver/silver chloride (Ag/AgCl) and Pt wire were used as reference electrode and a counter electrode, respectively. For each condition, temperature was changed to simulate the corrosion testing. Corrosion testing was conducted in various temperatures (28, 35, 40 and 45 °C)and in 0.005, 0.010, 0.015, 0.020, 0.025, 0.030 and 0.035 ppm of sodium chloride concentrations. The demineralized water with pH of 6.7 and conductivity 0.4 μS/cm was used in the experiment.

Data Analysis

Corrosion current that find from the experiment was analyzed by using Tafel slope analysis to get corrosion rate. Tafel analysis is performed by extrapolating the linear portions of a logarithmic current versus potential plot back to their intersection. The value of either the anodic or cathodic current at the intersection is I_{corr}. Corrosion rate is calculated by using equation (1).

$$C_r = \frac{I_{corr}.K.E_w}{d.A} \quad \text{.....(1)}$$

Where I_{corr} is a corrosion current in Ampere, K is a contant that defines by the units in corrosion rate, E_w is equivalent weight in gram/equivalent, d is density in gram/cm^3, and A is sample area in cm^2 [15].

RESULT AND DISCUSSION

Temperature Effect to $AlMg_2$ Corrossion

Almost all of chemistry reaction rate will increase linearly with temperature. At high temperature increasing of the corrosion rate is accelerating by oxygen diffusion to the system. In open vessel, where the oxygen is free to escape, the corrosion rate increases with temperature up to 80 ˚C. Then, as the temperature is increased further, the corrosion rate decrease. This is due to a drop in the oxygen solubility of water above 80 ^{o}C. In closed system, oxygen cannot escape, and the corrosion rate increase with temperature until all of the oxygen is consumed. The rate of diffusion of oxygen increases with rise in temperature. However, owing to the reduction in the solubility of oxygen with rise in temperature, the curves expressing the relation of the rates of corrosion of the above metals to temperature show maxima. The limiting diffusion current varies with temperature in a similar way. It would be expected that in a "closed system" both the limiting diffusion current and the rate of corrosion processes controlled by diffusion would rise continuously with increasing temperature.

This would explain that the temperature dependence of the corrosion rate shown in Fig. 1 as well as its dependence on NaCl concentration. For a thermally activated process, corrosion rates are expected to increase with increasing temperature. Increasing the corrosion rate also occur at temperature 28 ˚C, 40 ˚C and 45 ˚C with increasing NaCl concentration.

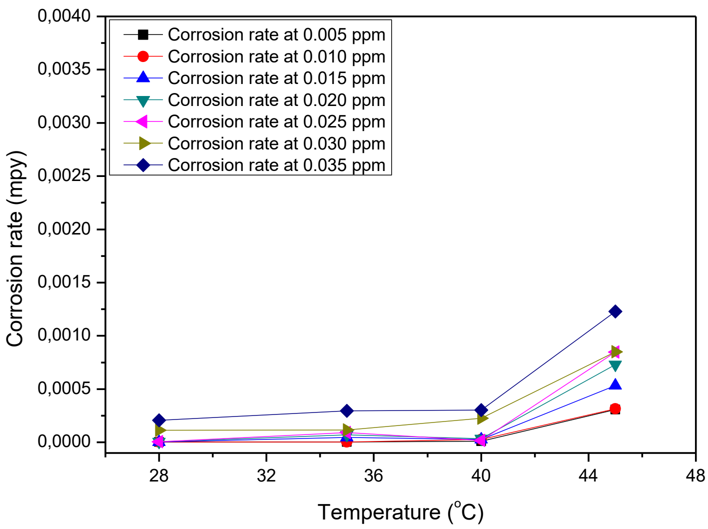

Figure 1. $AlMg_2$ corrosion rate at various temperatures in several NaCl concentrations in demineralized water with pH 6.7 and conductivity 0.4 μS/cm

Effect of NaCl Concentration on $AlMg_2$ alloy Corrosion Rate

The Table 2 shows experiment results that NaCl addition tend to increase $AlMg_2$ corrosion rate. At temperature 35˚C in 0.005 ppm NaCl, corrosion rate is 2.63×10^{-6} and increases to 3.21×10^{-6} mpy in 0.010 ppm NaCl, at the same temperature. Then at the same (35˚C), $AlMg_2$ corrosion rate in 0.015 ppm NaCl is 4.59×10^{-6} mpy and increase become 7.12×10^{-6} mpy in 0.020 ppm NaCl. At the same condition (35˚C), corrosion rate continue to increase with increasing NaCl concentration, for example; is 2.96×10^{-4} mpy in 0.035 ppm NaCl. Chloride as an aggressive ion can destroy the passive film on the alloys and metal surface. Many metal and alloys have an ability to form the passive film from metal oxide which separated metal from its media. Herefore ion chloride concentrations determine the incubation time of pitting corrosion. Ion chloride and oxygen will compete to be adsorbed at material surface. The higher the ion chloride concentration is the higher possibility chloride ion to be absorbed at material surface. It can be seen at Figure 2, since 0.030 ppm NaCl concentration, $AlMg_2$ corrosion rate increase sharply.

Table 2. $AlMg_2$ corrosion rate at several temperatures and NaCl Addition in *outlet Mix Bed water* (pH 6.7) and conductivity 0.4 μS/cm

Temp (°C)	Corrosion rate (mpy) at various NaCl Concentration (ppm)						
	0.005	0.010	0.015	0.020	0.025	0.030	0.035
28	2.08×10^{-6}	2.61×10^{-6}	2.70×10^{-6}	4.59×10^{-6}	6.63×10^{-6}	1.14×10^{-4}	2.06×10^{-4}
35	2.63×10^{-6}	3.21×10^{-6}	4.59×10^{-5}	7.12×10^{-5}	9.08×10^{-5}	1.15×10^{-4}	2.96×10^{-4}
40	9.56×10^{-6}	2.63×10^{-5}	2.70×10^{-5}	3.36×10^{-5}	1.77×10^{-5}	2.26×10^{-4}	3.03×10^{-4}
45	3.09×10^{-4}	3.15×10^{-4}	5.34×10^{-4}	7.30×10^{-4}	8.47×10^{-4}	8.51×10^{-4}	1.23×10^{-3}

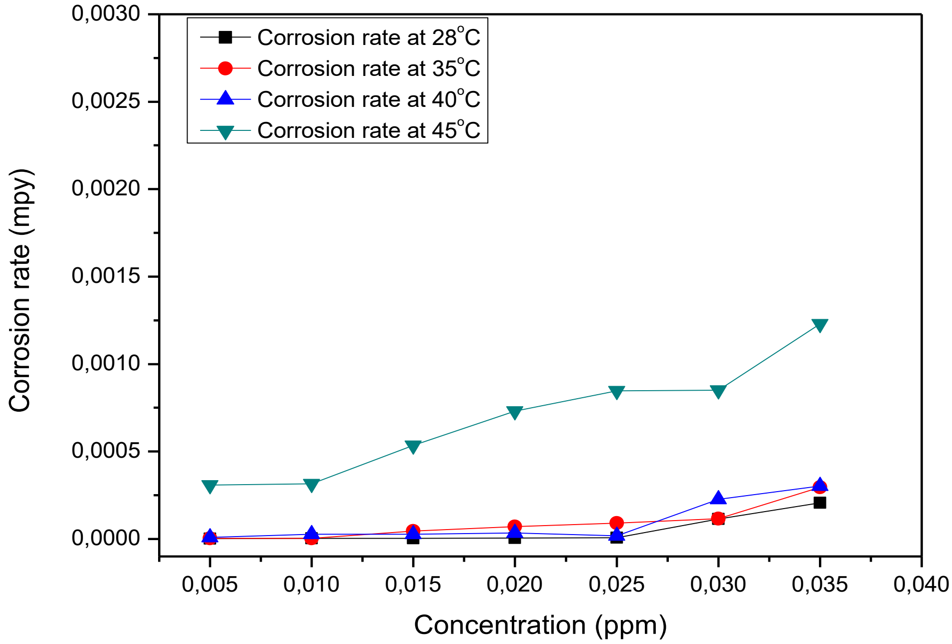

Figure 2. $AlMg_2$ corrosion rate in various NaCl concentrations at various temperatures in demineralized water with pH 6.7 and conductivity 0.4 μS/cm

CONCLUSION

The laboratory experiment result showed the relation between increasing temperature and corrosion rate. From the Tafel slope analysis, it can be seen that NaCl addition into solution affected to $AlMg_2$ corrosion rate. At higher NaCl concentration that added in the solution will affect the higher corrosion rate of $AlMg_2$. The result of experiment shows that the highest corrosion rate occur in NaCl 0.035 ppm solution at temperatur 45 °C, 1.23×10^{-3} mpy. From the $AlMg_2$ corrosion rate data, it was found that NaCl can cause corrosion to $AlMg_2$ as fuel cladding. The corrosion rate of $AlMg_2$ as fuel cladding material is very small in all of experiment conditions.The corrosion resistance material is important requirement for material that used in nuclear reactor. Ion chloride consentration has been controlled in tightly to ensure the reactor safety.

ACKNOWLEDGEMENT

This work has been carried out under Center for Nuclear Reactor Technology and Safety (PTKRN) in the year of 2017 budget program, National Nuclear Energy Agency of Indonesia (BATAN). The authors appreciate thank you very much to the research coordinator of “Ageing of RSG GAS” for her support.

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