EFFECT OF CHLORIDE AND COPPER ION FOR REACTOR TANK MATERIAL INTEGRITY OF RSG GAS PRIMARY COOLING SYSTEM

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ABSTRACT

EFFECT OF CHLORIDE ION AND COPPER ION FOR TANK MATERIAL INTEGRITY OF RSG GAS PRIMARY COOLING SYSTEM. RSG GAS as Multipurpose Reactor designed to operate for 30 years, now RSG GAS has been operating for approximately 29 years. Al₂Mg₃ as a reactor tank material has a weakness in its use, that is not resistant for environment that contains of chloride ions and copper ion. The purpose of this research was to know the impact of Cl⁻ and Cu²⁺ for tank material integrity of the RSG GAS primary cooling system. The analysis is done by simulation on Al₂Mg₃ corrosion in Cl⁻ and Cu²⁺ solution using Potensiogalvanostate IG & G. The results showed that the concentration of chloride ion in the primary cooling water that greater than 6 ppm have potentially to degrade the integrity of reactor tank material. The existence of copper ions in the primary cooling water will accelerate the damage to the reactor tank. Material integrity of RSG GAS reactor tank can be maintained by limiting the concentration of chloride and copper ions in each of the primary cooling water in accordance with the standard requirement.

Keywords: integrity, the reactor tank material, the primary cooling water, RSG-GAS.

ABSTRAK

PENGARUH ION KLORIDA DAN ION TEMBAGA TERHADAP INTEGRITAS MATERIAL TANGKI PADA SISTEM PENDINGIN PRIMER REAKTOR RSG GAS. Reaktor Serba Guna G.A. Siwabessy (RSG GAS) dirancang untuk beroperasi selama 30 tahun, sampai saat ini RSG GAS telah beroperasi selama kurang-lebih 29 tahun. Al₂Mg₃ sebagai bahan tangki reaktor mempunyai kelemahan dalam penggunaannya yaitu tidak tahan dengan lingkungan yang mengandung ion klorida CI dan ion tembaga Cu²⁺. Penelitian ini bertujuan untuk mengetahui pengaruh CI dan Cu²⁺ terhadap integritas tangki Reaktor RSG GAS. Analisis dilakukan melalui simulasi uji korosi Al₂Mg₃ dalam lingkungan ion CI dan Cu²⁺ menggunakan Potensiogalvanostate IG&G. Hasil penelitian menunjukkan bahwa pada konsentrasi ion klorida dalam ATR lebih besar dari 6 ppm berpotensi mendegradasi integritas tangki reaktor. Keberadaan ion tembaga dalam ATR akan mempercepat kerusakan tangki reaktor. Integritas tangki reaktor RSG GAS dapat dipertahankan dengan membatasi konsentrasi ion klorida dan ion tembaga dalam air pendingin primer masing-masing tidak melebihi standar yang dipersyaratkan.

Kata Kunci: integritas, tangki reaktor, air pendingin primer, RSG-GAS.

INTRODUCTION

Multipurpose Reactor G.A. Siwabessy (RSG GAS) designed to operate for 30 years, now RSG GAS has been operating for approximately 29 years. Because of aging process, very possible that the condition of systems, structures and components (SSCs) of the reactor will degraded the function of reactor material. Degradation of system functions and components can disrupt the continuity of the operation of the reactor and interfere the reactor operation safety. Operation safety of RSG GAS should be maintained so that the operation of reactor can be extended for 10 years. Therefore, it is necessary to monitor the reactor safety, by predicting the strength of the reactor tank material [1]. Aging management is one program that must be done to get a reactor operating life longer [2]. Monitoring of the reactor material is very important to do, especially for Al₂Mg₃ as the RSG GAS reactor tank

material. Aluminium alloy has a weakness in its use which is not resistant to environments containing chloride ions, especially when there is a copper Cu ion. That will trigger the corrosion process on aluminum alloys. Therefore, there should be analysis of the influence of chloride ion and Cu ion in primary cooling water for RSG GAS material integrity. The use of water as a coolant in research reactors could pose problems for the integrity of structures, systems and components of the reactor, such as the corrosion of the reactor material, the formation of oxide layer in the cladding of fuel elements and heat exchangers pipes so that will reduce the efficiency of heat conductivity, radiation exposure increases due to the activation of particles dissolved in the water, and the emergence of aggressive species generated by radiolysis process [3]. Various mitigation needs to be done to minimize the negative effects arising from the use of water as a coolant. Water treatment is needed to be important in obtaining water quality which low aggressiveness [2] It is necessary to minimize the negative effects arising from the reactor cooling water to the reactor material. Therefore, it is necessary to periodically monitoring system against reactor water chemistry quality conditions so that if any irregularities will be immediately anticipated and reactor water quality will be maintained according to the requirements [4]. Chloride ion and copper ion is very aggressive towards aluminium (Al₂Mg₃) as reactor tank material, so that if the concentration exceeds the concentration required it will damage the reactor tank material and lead operations will shorter. The purpose of this study was to know the impact of Cl and Cu²⁺for material integrity on the RSG GAS primary cooling system. The existence of trace elements in the primary cooling water on RSG GAS especially chlorida ion and copper ion have not been monitored periodically. Monitoring on water quality conditions of the primary cooling water on RSG GAS done on the pH (degree of acidity) and electrical conductivity of water.

ALUMINIUM CORROSION ON WATER

Corrosion degraded the quality of material caused by reactions with the environment [5]. The mechanism of corrosion process in water by electrochemically occurring simultaneously at the anode and cathode regions on the electric current circuit closed [6-7].

1. Reaction on anoda (oxidation)

$$M \rightarrow M^{n+} + ne^{-}$$
 (1)

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2. Reaction on catoda (reduction)

a. acidic solution

Without
$$O_2$$
 : $2H^+ + 2e^- \rightarrow H_2$ (2)
With O_2 : $4H^+ + O_2 + 4e^- \rightarrow 2H_2O$ (3)

b. Alkali or neutral solution

$$: 2H_2O + O_2 + 4e^- \rightarrow 4OH^-$$
 (4)

c. The reduction of the dissolved metal ions

$$: M^{n+} + ne^{-} \rightarrow M$$
 (5)

The stages on corrosion process in 4 phases, as follows: [8].

- 1. Anode, oxidation reaction occurs that releases electrons, so the corrosion process will take place.
- 2. Cathode, there will be a reduction reaction that will capture electrons.
- 3. The electrolyte as a medium of contact between the anode and the cathode so as to allow the flow of electricity.
- 4. The electrical circuit between the anode and cathode which allows the flow of electrons Water is a one agrresive substance towards metal which pasted. Because water will be ionized to produce ions H + and OH- ions in 10⁻⁷ mol per liter (pH = 7). H⁺ ions will acts as an oxidant in corrosion process in the structure and components of the material reactor. So the presence of hydrogen ions in the water has a chance to damage the metal, support for the corrosion process such as the occurrence of defects on the surface[9]. Corrosion will occur in the structure and components on the material reactor if water is used in reactor coolant contains aggressive ions such as chloride ion and cupper ion. For example, the aggressiveness of ion chloride and copper ion in form of pitting on aluminum alloy such as Figure 1 [10-12].

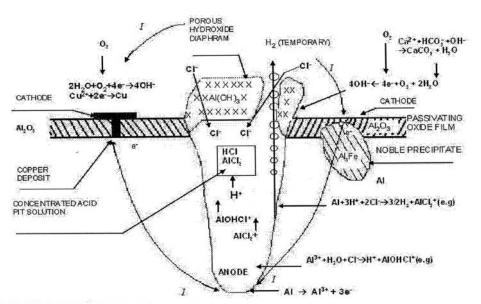


Figure 1. Formation process of pitting corrosion due to the effect of chloride ions and copper ions on aluminum alloy [10].

METHODOLOGY

If the primary cooling water in RSG GAS containing chloride ions and copper ions, the integrity of the material reactor tank needs to be analyzed to ensure the continuity and safety of operation. The analysis was performed through corrosion process using a potensiogalvanostate for Al_2Mg_3 as the reactor tank material on the solution that contains chloride and copper ion. The element content on Al_2Mg_3 as described in the Table 1.

| Table 1. Element content on Al₂Mg₃ [13]. | | | | | | | | |
|---|---------|--------|--------|--------|--|--|--|--|
| Element | Al | Fe | Mn | Cu | | | | |
| W (%) | 98.9034 | 0.4939 | 0.4189 | 0.1838 | | | | |

 Al_2Mg_3 had cutting with 1.6 cm diameter cylindrical shape as a sample corrosion test. The sample then polishing by using sandpaper with 100, 400, 800, 1200 mesh. Simulations carried out by using a natrium chloride solution with varying concentrations of 0, 2, 4, 6 ppm. Varian concentration of copper solution with 0.02 ppm, 0.1 ppm, 0.15 ppm is add on the natrium chloride solution 2 ppm. The corrosion rate is calculated using the phenomenon of Faraday's law [14]:

Corrosion process on the metal (M), occurs electrochemically by follow the equation such as

$$M \rightarrow M^{+} + e \tag{6}$$

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$$1/2M_2 \to 1/2M_2^{2+} + e \tag{7}$$

$$1/3M_3 \to 1/3M_3^{3+} + e \tag{8}$$

 M_1 = monovalent metal M_2 = bivalent metal and M_3 = trivalent metal

If in the corrosion test, has result the current (i corr) of 1 Faraday (96500 Coulomb) means there has been a corrosion process on metal as much as 1 equivalent. Faraday formulation to calculate the corrosion rate.

$$Corrosion.rate = \frac{0.13i.eq}{\rho A} mpy$$

corrosion rate = mpy (mili inchi per year)

A = area of the metal (cm^2)

eq = equivalent weight

 ρ = metal density (gr/cm³)

i = current (mili ampere)

RESULTS AND DISCUSSION

The water quality requirements (safety analysis report) on Triga Reactor-Bandung, Kartini reactor-Yogyakarta, RSG GAS-Serpong are shown in Table 2:

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Table 2. Safety analysis report of water quality requirement

| | | F | Reactor research | | | | |
|-----|-------------------------------------|--------------|------------------|-----------|--|--|--|
| No. | Parameter | Kartini | Triga | RSG GAS | | | |
| 1. | рН | 5.5 - 6.5 | 5.5 - 6.5 | 5.8 - 7.0 | | | |
| 2. | λ (μS/cm) | < 3.5 | < 3.5 | < 8.0 | | | |
| 3. | TDS (ppm) | - | - | - | | | |
| 4. | Ca (ppm) | < 1 | < 1 | - | | | |
| 5. | Mg (ppm) | < 1 | < 1 | - | | | |
| 6. | Si (ppm) | < 1 | < 1 | - | | | |
| 7. | Na (ppm) | < 1 | < 1 | 0.0232 | | | |
| 8. | Fe (ppm) | - | - | - | | | |
| 9. | Mn (ppm) | 0.004 | < 0.1 | - | | | |
| 10. | Al (ppm) | - | - | 0.0115 | | | |
| 11. | Cu (ppm) | - | - | 0.0209 | | | |
| 12. | Ni (ppm) | - | - | - | | | |
| 13. | Zn (ppm) | - | - | - | | | |
| 14. | Cd (ppm) | - | - | - | | | |
| 15. | Be (ppm) | - | - | - | | | |
| 16. | Cl (ppm) | - | - | 0.049 | | | |
| 17. | SO ₄ ²⁻ (ppm) | - | | 0.0528 | | | |

Effect of chloride ion to Al_2Mg_3 corrosion rate as RSG GAS reactor tank material is shown in Figure 2. Effect of copper ion concentration of 0.2; 0.10; 0.150 ppm to Al_2Mg_3 corrosion rate on chloride solution as shown in Table 3. We can see that there are the corrosion process on Al_2Mg_3 on concentration chloride ion solution 0 ppm with corrosion rate of 0.0121 mpy (0.000307 mm/year). It is possible there are the corrosion process on Al_2Mg_3 , supported by reaction 1,2 and 3. Water will contribute the corrosion process, in the presence of hydrogen ions from the dissociation process, it is exactly to effect for the corrosion process. If ion concentration at primary cooling water can be maintained under the standard requirement, especially ion concentration 0 ppm, a reduction in the thickness of the reactor tank material for 30 years was 0.363 milli-inch (0.009210 mm). Reduction in thickness due to corrosion of the reactor material tank in these conditions is small relatively so that the integrity of the reactor material tank is still high relatively.

The RSG GAS safety analysis reports of chloride ion concentration on the primary cooling water is 0.049 ppm. Based on the curve of Figure 2, can be determined that the corrosion rate of RSG GAS reactor material tank is 0.0144 mpy (0.000365 mm/year). So, thinning tank reactors over 30 years of age RSG will not exceed 0.432 milli-inch (0.010970 mm). This condition is still safe for thinning reactor material tank still small relatively. So that the integrity of the material tank is high relatively.

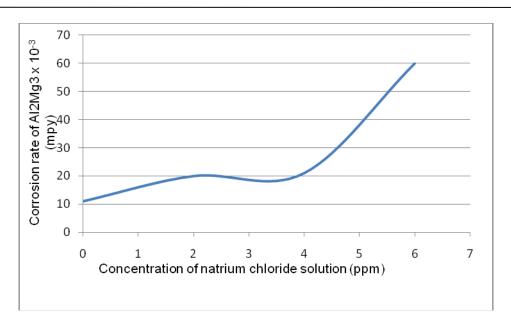


Figure 2. Effect of chloride ion to Al₂Mg₃ corrosion rate

Table 3. Effect of copper ion to Al₂Mg₃ corrosion rate

| | | | <u> </u> | | |
|--|------------|------------|------------|------------|------------|
| Konsentration Cl ⁻ / Cu ²⁺ (ppm) | 0,00/0,00 | 2,00/ 0,00 | 2,00/0,02 | 2,00/0,10 | 2,00/ 0,15 |
| Corrosion rate | 0,0121 mpy | 0,0213 mpy | 0,0639 mpy | 0,0958 mpy | 0,1478 mpy |
| (mpy) | (0,000307 | (0,000541 | (0,001623 | (0,002434 | 0,003754 |
| | mm/year) | mm/year) | mm/year) | mm/year) | mm/year) |

The corrosion rate of Al_2Mg_3 at 2, 4 and 6 ppm chloride ion concentration, is 0.0213 mpy (0.000541 mm/year), 0.0258 mpy (0.000655 mm/year) and 0.0609 mpy (0.001546 mm / year). So can be expected the possibility of thinning material for 30 years is 0.639 milli-inch (0.016230 mm), 0.774 milli-inch (0.019650 mm), and 1.827 milli-inch (0.046400 mm). On the condition of the chloride ion concentration below 6 ppm, thinning of the reactor tank is small relatively so that the integrity of the reactor material tank at these concentrations can still be maintained. It can be seen table 3, that the corrosion rate of Al_2Mg_3 in the chloride solution with concentration of 2 ppm add the copper solution with 0.150 ppm is 0.1478 mpy (0.003754 mm/year), much higher than the corrosion rate of Al_2Mg_3 in the chloride solution with concentration of 2 ppm with and without copper solution is 0.0213 mpy (0.000541 mm / year). We can see that increase of the corrosion rate is 7 times. It can be confirmed that the presence of copper ions in the primary cooling water would be able to trigger an increase in the corrosion rate of Al_2Mg_3 .

Monitoring of primary cooling water quality especially chloride ions and copper ions as periodically should be done so that the integrity of the reactor material can be maintained. If there are symptoms of increased ion is then immediately do a more intensive refining primary cooling water thus further corrosion of the tank can be avoided.

CONCLUSION

A concentration of chloride ions in primary cooling water is greater than 6 ppm potentially degrade the integrity of the material reactor tank. Presence of copper ions in the primary cooling water would accelerate damage to the material reactor tank. Monitoring of primary cooling water quality especially chloride and copper ions should be done periodically so that the integrity of the reactor material can be maintained. If there are symptoms of increased ion is then immediately do a more intensive refining primary cooling water thus further corrosion of the tank can be avoided. Monitoring of primary cooling water quality especially chloride ions and copper ions as periodically should be done so that the integrity of the reactor material can be maintained.

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REFERENCES

- 1. YEON SOO KIM, G.I HOFMAN, A.B. ROBINSON, "Oxidation of Aluminum Alloy Cladding for Research and Test Reactor Fuel", Elsevier: Journal of Nuclear Materials, vol.378,pp: 220-228 (2008).
- 2. IAEA, "Ageing Management For Research Reactors", IAEA Safety Standards Series No. SSG-10, Vienna, International Atomic Energy Agency (2010).
- 3. MAHBUBOOR R. CHOUDURY, MING-KAI HSIEH., RADISAV D VIDIC., DAVID A. DZOMBAK., "Corrosion Management In Power plant Cooling Systems Using Tertiary-Treated Munical Wastewater as Makeup Water"., Elsevier: Corrosion Science. Vol 61: 231-241.(2012).
- 4. SINDELAR ROBERT L, CHANDLER GREG T AND MICALONIS JOHN L" Water Quality and Corrosion: Considerations for Nuclear Reactor Systems, "Journal of the South Carolina Academy of Science, Vol. 9: Iss. 1, Article 13 (2013).
- 5. PETER L. ANDERSEN, Corrosion Sci of Journal., Vol. 69, No. 10, pp. 1024-1038 (2013).
- 6. IAEA,TRS No. 418, "Corrosion Of Research Reactor Aluminium Clad Spent Fuel In Water" IAEA, Vienna (2003).
- 7. LIJIN DONG and EN-HOU HAN,"Environmentally Assisted Crack Growth in 308L Stainless Steel Weld Metal in Simulated Primary Water", Corrosion Science, Vol 117, pp.1-10 (2017).
- 8. RAKHMAD INDRA PRAMANA.,"Studi Ekstrak Beluntas (Plunchea Indica Less) Sebagai Inhibitor Korosi Ramah lingkungan Terhadap Baja Karbon Rendah Di Linkungan 3,5 % NaCl"., Tesis FT-UI, Jakarta(2012)
- ADRIAN,"Perilaku Korosi Material Baja Paduan Akibat Pengaruh Kondensat Yang Berasal dari Pembangkit Listrik Tenaga Panas Bumi", Tesis, FMIPA-UI, Jakarta (2009).
- 10. P. M. NATISHAN AND W. E. O'GRADY, "Chloride Ion Interactions with Oxide-Covered Aluminum Leading to Pitting Corrosion: A Review", *J. Electrochem. Soc.* Vol. 161, issue 9, pp.421-432 (2014)
- 11. ROSA VERA AND PATRICIA VERDUGO, "Corrosion of Aluminium in Copper-Aluminium Couples Under a Marine Environment: Influence Of Polyaniline Deposited Onto Copper", Corrosion Science, Vol 52, Issue 11, pp.3803-3810 (2010).
- 12. H. NADY, M.M.EL-RABIEI, M.SAMY, "Corrosion Behavior and Electrochemical Properties Of carbon Steel, Commercial pure Titanium, Copper and Copper-Aluminum-Nickel Alloy in 3.5% Sodium Chloride Containing Sulfide Ions", Egyptian Journal of Petroleum, Vol 26, Issue 1, pp:79-94 (2017).
- 13. GENI RINA SUNARYO, SRIYONO, "Aplikasi Program Pengawasan korosi Untuk Kolam Penyimpanan Reaktor RSG-GAS", Prosiding Seminar Nasional ke-16 Teknologi dan Keselamatan PLTN Serta Fasilitas Nuklir ISSN: 0854 2910, pp: 158 -164 (2010)
- SOFYAN YUSUF, "Laju Korosi Pipa Baja Karbon A106 Sebagai Fungsi Temperatur dan Konsentrasi NaCl Pada Fluida Yang Tersaturasi Gas CO₂", Tesis, FMIPA-UI, Jakarta (2008)