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MODEL SIMULATION OF THE EFFECT OF ZINC PHOSPHATE INHIBITOR ON CORROSION RATE OF SECONDARY COOLING PIPE FOR PRESSURIZED WATER REACTOR USING PREDICTOR-CORRECTOR METHOD

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ABSTRACT
MODEL SIMULATION OF THE EFFECT OF ZINC PHOSPHATE INHIBITOR ON CORROSION RATE OF SECONDARY COOLING PIPE FOR PRESSURIZED WATER REACTOR USING PREDICTOR-CORRECTOR METHOD. Corrosion is one of the unavoidable problems in nuclear reactor cooling systems. Secondary Cooling Pipe Multi-Purpose Reactor G.A Siwabessy (MPR-GAS) based on low carbon steel is very easy to oxidize because it is an open circulation cooling system. Zinc-phosphate inhibitor ($ZnPO_4$) is one of the inhibitors used to inhibit the corrosion reaction that occurs between the low carbon steel cooling pipe and the cooling water environment. The research has been carried out on numerical calculations to find a solution for the corrosion rate model of steel with and without an inhibitor by the Predictor-corrector method. The simulation results show that the effect of $ZnPO_4$ inhibitor on the corrosion rate of steel is seen as a relationship between inhibitor concentration and stable time, if the inhibitor concentration increases, the stable time will also increase. The rate of corrosion that occurs (reducing the concentration of corroded product). Therefore, the recommended inhibitor concentration value is 0.9% for models in 0.1 M HCL medium. This concentration is the most appropriate concentration to slow down the corrosion rate most minimal.

Keywords: model, simulation, corrosion, carbon steel, inhibitor, $ZnPO_4$.

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ABSTRAK

SIMULASI MODEL PENGARUH INHIBITOR ZINC PHOSPHATE TERHADAP LAJU KOROSI PIPA PENDINGINAN SEKUNDER REAKTOR AIR BERTEKAN MENGGUNAKAN METODE PREDIKTOR-KOREKTOR.Korosi merupakan salah satu permasalahan yang tidak dapat dihindari pada sistem pendingin reaktor nuklir. Reaktor Serba Guna Pipa Pendingin Sekunder G.A Siwabessy (MPR-GAS) berbahan dasar baja karbon rendah sangat mudah teroksidasi karena merupakan sistem pendingin tipe sirkulasi terbuka. Zinc-phosphate inhibitor ($ZnPO_4$) merupakan salah satu inhibitor yang digunakan untuk menghambat reaksi korosi yang terjadi antara pipa pendingin low carbon steel dengan lingkungan air pendingin. Telah dilakukan penelitian perhitungan numerik untuk mencari solusi model laju korosi baja dengan dan tanpa inhibitor dengan metode Predictor-Corrector. Hasil simulasi menunjukkan bahwa pengaruh inhibitor $ZnPO_4$ terhadap laju korosi baja dilihat dari hubungan antara konsentrasi inhibitor dengan waktu stabil, jika konsentrasi inhibitor meningkat maka waktu stabil juga akan meningkat. Laju korosi yang terjadi (mengurangi konsentrasi produk yang terkorosi). Oleh karena itu, nilai konsentrasi inhibitor yang direkomendasikan adalah 0,9% untuk model dalam medium HCL 0,1 M. Konsentrasi ini merupakan konsentrasi yang paling tepat untuk memperlambat laju korosi paling minimal.

Kata kunci: model, simulasi, korosi, baja karbon, inhibitor, $ZnPO_4$.
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INTRODUCTION

Corrosion is the degradation of a material caused by chemical reactions with other materials and the environment. In metallurgy, the corrosion process can be viewed as an event or reaction of chemical compounds back to their original form or can be referred to as the reverse of the extraction metallurgical process. Corrosion occurs because of chemical and electrochemical reactions. The main elements that can cause corrosion are materials, environment, metallic contacts, and electrolytes[1]–[3].

Corrosion is one of the unavoidable problems in the nuclear reactor cooling system that uses water as a cooling water medium for heat transfer, especially for an open circulation type cooling system. The secondary cooling system of the Pressurized Water Reactor (PWR) is an open recirculation cooling system so it is very easy for the components in the system to interact with oxygen in the surrounding environment or open air[4], [5]. The interaction of metals with oxygen causes oxidation or corrosion processes. Corrosion control in the secondary cooling system of the PWR is carried out by adding chemicals as corrosion inhibitors using an injection system, which is operated continuously if the secondary cooling system operates. The addition of this inhibitor material causes the corrosion rate of the PWR secondary cooling pipe to be around <3 mpy (miles per year)[6]. The basic material used as a corrosion inhibitor is a synthetic chemical compound called zinc phosphate ($ZnPO_4$)[7]–[10].

In previous studies, research was conducted on the effect of variations in zinc phosphate concentration on the corrosion rate of carbon steel in water solution medium[11]. In this experiment, the data is about the corrosion process in carbon steel on an HCl solution medium. The data obtained from the results of corrosion experiments carried out in the laboratory are insufficient to show that zinc compounds ($ZnPO_4$) are corrosion inhibitors suitable for use in the field. This is due to differences in data in the field and in the laboratory due to the large number of parameters not considered in laboratory experiments. Another study used a balanced state model to model the effect of inhibitors on the reaction rate [12]. The model is derived with an emphasis on the corrosion mechanism from the point of view of the Transition State Theory, meaning that the metal first turns into metal ions (transition state) before becoming the reaction product (corroded product).

Based on the things mentioned above, this research will study the simulation model of the effect of Zinc Phosphate inhibitor on the corrosion rate of steel. inhibitor with the Predictor-Corrector method. The numerical solution of each case will be compared to determine how much influence the addition of inhibitor has on the corrosion rate. So, from the simulation, the inhibitor profile in controlling the corrosion rate of carbon steel is useful for minimizing damage to the secondary cooling pipe of the PWR, which is made of carbon steel.

METHODOLOGY

Before simulation, the experiment was conducted at the Radiometallurgy Installation Laboratory of the Research Center for Nuclear Material and Radioactive Waste Technology. The material used for corrosion analysis comes from carbon steel, the PWR secondary pipe material. The pipe material is cut and turned into a rectangular shape with an area of 10 mm and 10 mm. The test sample/specimen is then mounted using an epoxy resin. The sample's surface is left open so that it is in direct contact with the test solution. When mounted, the specimen relates to a wire (copper) as a current connector. The specimen's exposed surface was gradually sanded with SiC sandpaper with mesh sizes of 500, 800, and 1200. After that, the test sample was washed with acetone solution and rinsed in demineralized water and then dried. The corrosion test specimen is shown in Figure 1.
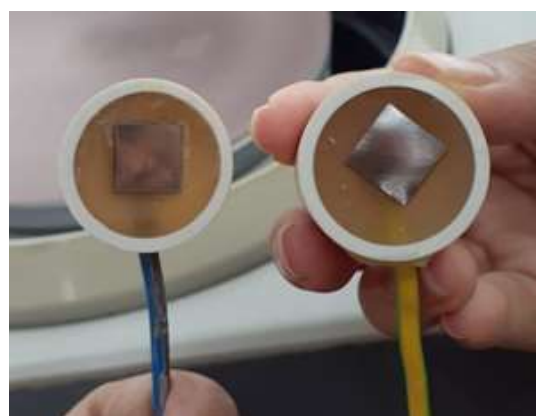

Figure 1. Sample preparation results for corrosion test.

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The corrosion medium used is demin water, secondary cooling water for the PWR and several solutions with various concentrations for corrosion testing. The inhibitor used in this study was a scale inhibitor. The basic ingredient of scale inhibitors is the synthetic chemical compound zinc phosphate ($ZnPO_4$). Corrosion rate measurements were also carried out in water and 0.1 M HCl media with the additional variation of 0 - 0.9% of zinc phosphate inhibitor. Determination of the rate of corrosion quantitatively through the tafel method. The Tafel corrosion test was conducted at a potential range from -250 mV to +250 mV concerning open circuit potential (Eocp). The test begins with the measurement of Eocp until the condition is stable for about 60 minutes [13]. The corrosion cell used for measuring the corrosion rate of the tafel method consists of three electrodes, as shown in Figure 2.


Figure 2. Cell circuit for corrosion rate measurement

Two 20th-century scientists, Leonor Michaelis and Maud Leonora Menten, proposed the model known as Michaelis-Menten Kinetics to account for enzymatic dynamics. The model explains how an enzyme can cause kinetic rate enhancement of a reaction and how reaction rates depend on the concentration of enzyme and substrate. The chemical reaction that occurs is as follows:

$$x \underset{b}{\overset{a}{\rightleftharpoons}} y \overset{c}{\to} z$$
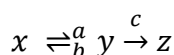
According to the concept of reaction equilibrium, chemical reaction is a reversible reaction (back and forth reaction). Reactions that indicate a reversible reaction are as follows:

$$x \underset{b}{\overset{a}{\rightleftharpoons}} y$$
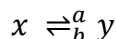
Metals will change into metal ions with a reaction rate of a, while metal ions can change back into metal with a rate of a but this happens very slowly. A reversible reaction will continue until reaction equilibrium is reached. Meanwhile, the reaction that occurs when metal ions become corroded products only takes place in one direction, as mentioned:

$$y \overset{c}{\to} z$$

with a reaction rate of c. It used to obtain the value of the Michaelis Menten through the equation:

$$c = \frac{px}{r + x} \tag{1}$$

r is the Michaelis Menten constant, and p is the maximum corrosion rate which has a value of 1 (according to assumptions).

The addition of inhibitors does not influence the rate of change in metal concentration per unit of time or the differential equation for equation (1) because metal ions function as primary catalyst centers. After the presence of new metal ions, the inhibitor that is added will influence it. So:

$$\frac{dx}{dt} = by - ax \tag{2}$$

Equation (1) is equal to the rate of change in metal concentration per unit time in the absence of inhibitors. In the reaction, the initial inhibitor works after the presence of metal ions and then the inhibitor becomes a corrosion product. This is the same as the reaction product described in Michaelis Menten's Kinetics, only the substrate concentration is replaced by the inhibitor concentration, namely m. Then the rate of reduction of inhibitor per unit time is:

$$\frac{dm}{dt} = -\frac{pm}{r + m} \tag{3}$$

The (-) sign means that as time passes the inhibitor concentration will decrease. The rate

of reduction of metal ions to metal using an inhibitor will be the same as the rate without the inhibitor, only the rate will be slowed down by the presence of the inhibitor, so that:

$$\frac{dy}{dt} = ax - by - \frac{px}{r+x} + \frac{pm}{r+m} \qquad (4)$$

The sign (+) in the equation above explains that the function of the inhibitor is to inhibit the formation of metal ions instead of accelerating it. The rate of change in the concentration of corrosion products per unit time is influenced by the reaction of the metal with the environment and the reaction of the inhibitor with the environment. The inhibitor factor is (-) because the concentration of corrosion products decreases as the inhibitor is added to the system. So, the rate of change in concentration of corrosion products can be written:

$$\frac{dz}{dt} = \frac{px}{r+x} - \frac{pm}{r+m} \qquad (5)$$

From the explanation above, we get a metal corrosion rate model using inhibitors which is a system of first-order ordinary differential equations, namely:

$$\frac{dx}{dt} = by - ax \qquad (6)$$

$$\frac{dy}{dt} = ax - by - \frac{px}{r+x} - \frac{pm}{r+m} \qquad (7)$$

$$\frac{dz}{dt} = \frac{px}{r+x} - \frac{pm}{r+m} \qquad (8)$$

$$\frac{dm}{dt} = -\frac{pm}{r+m} \qquad (9)$$

with initial conditions:
x(0) = 1
y(0) = 0
z(0) = 0
m(0) = 1

with,
x : metal concentration in solution (variable)
y : concentration of metal ions (variable)
z : concentration of corroded product (variable)

m : inhibitor concentration (variable)
a : metal reduction rate (parameter)
b : rate of reduction of ions to return to metal (parameter)
p : maximum reaction rate (parameter) is 1
r : Michaelis Menten constant (parameter)
t : time

The simulation used to find a solution to the corrosion rate model is to apply the Predictor-Corrector method. The solution is a graph of the relationship between the concentration of corroded metal, the concentration of the corroded product, and the concentration of the inhibitor for time. The following are the steps in completing the model using the Predictor-Corrector method:

1) Determine the starting point. Determination of the starting point using the Euler method with the following formula:

$$v_{i+1} = v_i + f(x_i, y_i, z_i m_i]h, \qquad (10)$$

i = 0, 1, 2, … , n
v = variable (i.e. x, y, z and m)
f = function of the differential equation
x = initial metal concentration
y = metal ion concentration
z = concentration of corroded product
m = concentration of inhibitor
h = step for the time (in this study it is worth 0.03)

2) Predict the value of the function using the Adams-Basforth predictor method of order 3 with the following formula.

$$v_{i+1} = v_i + \frac{h}{12}[23f_{i+1}16f_{i-1} + 5f_{i-2}] \qquad (11)$$

3) Correcting the accuracy of the function values using the corrector method with the following formula.

$$v_{i+1} = v_i + \frac{h}{12}[5f_{i+1} + 8f_i - f_{i-1}] \qquad (12)$$

The parameter simulation was carried out in three stages :

1) The simulation determines the appropriate parameter values in the model by changing several parameter values with variations in the 0.1 M HCL solution so that the appropriate parameters are obtained according to the

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theory of the corrosion rate. The parameters that are simulated in the model are as follows: a = rate of metal loss with parameter value 0.4 – 0.95, b = the rate of reduction of metal ions back into the metal with parameter values of 0.4 – 0.95, p = maximum corrosion rate with parameter value 1 and c = the average rate of corrosion experiment multiple 0.0254 (convert mpy to mmpy), parameters b, and c are used to find the Michaelis Menten constant in the model.

$$r = \frac{b + c}{a}$$

r= Michelis Menten Constant
a= parameter of metal loss
b= parameter of corrosion product
c= average of corrosion rate (mmpy)

2) The simulation determines the steady time using suitable parameter values. After determining the suitable parameters and b in the model, these parameters will be simulated to obtain a stable time. Stable time is when all the metal has formed rust powder and precipitated at the bottom of the experimental solution. Stable time was obtained after the simulation run. Stable time is when the carbon steel reaches 0% or the corrosion product reaches 100% for the first time. We define stable time with code: if x=0, then t=stable time. We used the stable time to explain the lifespan for carbon steel as secondary pipe cooling in corrosive conditions (0.1 M HCl).

3) The simulation determines the corrosion slowdown due to the influence of the inhibitor. The slowing rate of corrosion events due to the addition of inhibitors can be analysed through the reduction rate of inhibitors. This is because when the inhibitor has coated the entire metal surface, the ions from the HCl solution will first corrode the inhibitor layer before corroding the steel surface.

RESULTS AND DISCUSSION

From several experiments, the data is obtained as in Table 1. In Table 1 shows the magnitude of the corrosion rate at each concentration of 0.1 M HCl which was carried out 5 times. At each salinity, the average corrosion rate will be sought as the parameter value of c. Parameter c serves to determine the value of r, mentioned (b+c)/a.

Table 1. Experimental data for corrosion rate of carbon steel on 0.1 M HCl.

Specimen	Corrosion rate Carbon Steel (mpy)
Blanko 1	19.38
Blanko 2	19.08
Blanko 3	17.88
Blanko 4	19.28
Blanko 5	18.28

The Simulation Determines The Appropriate Parameter Values in the Model

The parameter values are taken manually by entering the possible values one by one to form a graph that is in accordance with the chemical concept of corrosion rate. Parameters a, b, c, p may not be negative because the reaction for the formation of the corroded product takes place in one direction, which is only the reaction of increasing the concentration. Likewise, the parameter r which is the Michaelis Menten constant will have a non-negative value because it is obtained from (b+c)/a. The parameter p used for model simulation is 1, which is in accordance with the assumptions.

Parameter changes a were carried out for several values of inhibitor concentration (m), m = 0; m=0.3; m = 0.5; m = 0.7; and m = 0.9 which indicates that for different values of a, there is a significant change in the initial metal concentration and corroded product. The simulation of searching for parameter without using an inhibitor (0% ZnPO₄) can be shown in Figure 3.
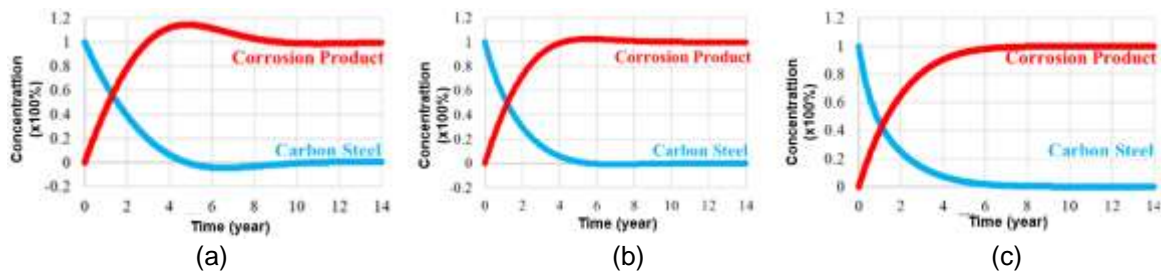

Figure 3. Simulation results for determining parameter without inhibitor with a value of a = 0.4 (a); a = 0.6 (b); and a = 0.95 (c)

The simulation of parameter search using ZnPO$_4$ inhibitor concentration of 0.9% can be shown in Figure 4. Figure 3 and 4 shows that the most suitable parameter a for the model is a=0.95 for without inhibitor model and a=0.95 for with 0.9% inhibitor model because in graph (Figure 3.c and 4.c) there is no longer an up and down rate.
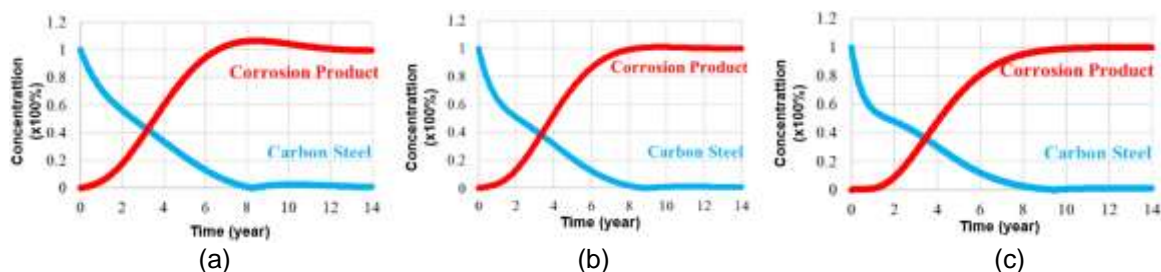

Figure 4.   simulation results for determining the parameter using a 0.9% inhibitor with a value of a = 0.4 (a); a = 0.6 (b); and a = 0.95 (c).

Simulation Determines Stable Time Using Appropriate Parameter Values

The value of the parameter that has been obtained through simulation, will look for the largest value to be used in the simulation to determine the stable time. This is because has the same parameter and fulfills the model requirements. In the simulation of determining, the stable time use suitable parameter values. It will be identified the time when a metal in the form of steel has turned into a corroded product (rust). The stable time indicates that all the metal has been oxidized resulting in iron rust that settles at the bottom of the brownish solution container[13]-[15].

The suitable parameter that has been obtained from the previous simulation is = 0.57, so it can be used for simulations to determine the stable time. The simulation to determine the stable time in the model without inhibitor in 0.1 M HCl medium can be shown in Table 2. While the simulation determines the stable time on the model using a 0.9% inhibitor can be shown in Table 3.

Table 2.   Simulation results of stable time for carbon steel corrosion without inhibitor with parameter a = 0.95

Simulation Curve	Parameters	value
	a	0.95
	b	0.95
	p	1
	c (mmpy)	0.0047
	m (%)	0
	t-stable (Year)	9.27

Table 3.   Simulation results of the stable time for carbon steel corrosion using 0.9% inhibitor with parameter a = 0.95

Simulation Curve	Parameters	value
	a	0.95
	b	0.95
	p	1
	c (mmpy)	0.0047
	m (%)	0.9
	t-stable (Year)	12.78

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Table 2 and 3 show the stable time for corrosion carbon steel, the initial metal concentration will decrease. The concentration of the reaction products (corroded products) will be higher. Here it can be seen that the corrosion compound continues to increase and will be stable when the metal compound is no longer corroded. The process of stabilizing the corrosion rate without inhibitors occurred at t = 9.27 years and stability using 0.9% inhibitors occurred at t = 12.78 years. This stability process is the same as the chemical equilibrium process where there will be no change in the macroscopic properties of the substance, but the decomposition reaction of the initial metal and the formation of corroded products will continue[14], [16], [17]. Compared to the time of stability of the corrosion rate without using an inhibitor, it turns out that the addition of 0.9% inhibitor has a positive effect on the system, slowing the corrosion rate by 3.51 years. The stable time of a corrosion process in 0.1 M HCl medium can be seen in Table 4.

Table 4. Effect of inhibitor on corrosion stability of in 0.1 M HCl medium

Inhibitor Concentration (%)	Stable Time (Years)
0 (without inhibitor)	9.27
0.3	10.71
0.5	11.48
0.7	12.06
0.9	12.78

Simulation Determines Corrosion Deceleration Rate Due to Inhibitor Effect

The slowing rate of the corrosion process can be determined through the inhibitor rate, because the HCl solution will corrode the inhibitor layer first before corroding the metal. Therefore, it can be said that the corrosion rate slows down because it is blocked by inhibitors[18]–[20]. The graph of the slowdown in corrosion of because the effect of adding an inhibitor to 0.1 M HCl medium can be seen in Figure 5.

Figure 5 shows that the greater the inhibitor concentration introduced into the system, the longer it takes for a metal to rust completely. The initial stable time is 12.6 months while the maximum stable time is 18.354 months, so the inhibitor can slow down the corrosion rate maximally for 5,754 months.

Then the efficiency of an inhibitor (EI, %) can be shown as follows:

$$EI(\%) = \frac{n_1 - n_0}{n_0} \times 100\%$$
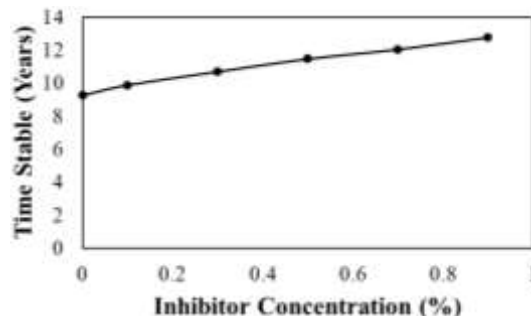
EI(%) = (12.78 – 9.27) / 9.27 x 100%
EI(%) = 37.86%

Figure 5. The curve of deceleration of stable time of corrosion on 0.1 M HCl medium due to inhibitor.

This result is in line with research conducted by Maman (2018) who also calculated inhibition efficiency based on the increase in impedance value on the surface of carbon steel. This explains that the higher the inhibitor concentration, the greater the inhibition efficiency with an increase in inhibition value of 63.31% at a ZnPO4 inhibitor concentration of 100 ppm[6].

CONCLUSIONS

The effect of $ZnPO_4$ inhibitor on the corrosion rate of carbon steel is seen as the relationship between inhibitor concentration and stable time, if the inhibitor concentration increases, the stable time will also increase. The greater the inhibitor concentration introduced into the system, the slower the corrosion rate (reducing the concentration of the corroded product). Therefore, the recommended inhibitor concentration value is 0.9% for the 0.1 M HCl medium model. This concentration is the most appropriate concentration to slow the corrosion rate at a minimum. The greater the concentration of inhibitor that is introduced into the system, the longer it will take to exhaust the entire inhibitor concentration before corroding the metal. The efficiency of the inhibitor to the corrosion rate is that it can slow down a maximum of 37.86% at 0.1 M HCl.

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