Concentration profile and water level effect on the evaporation in the spent nuclear fuel storage pool using a diffusion approach

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Abstract. In the spent nuclear fuel storage pool (SFSP) of GA Siwabessy Multipurpose Reactor (GAS-MPR), there are three status of pool water level with unknown effect of water level on evaporation rate. This study aims to determine the effect of pool water level on the evaporation rate in the SFSP, so that it can be a consideration in minimizing the evaporation rate. A prediction of water evaporation rate can be explained as a diffusion model through a stagnant gas film. A numerical method, 4th order Runge-Kutta was used to describe the concentration profile of water and air over a SFSP column to the top of the pool. The effect of the water level can be explained then. The various distance of the water to the pool top at constant water temperature and constant air temperature shows a result that the evaporation rate is decreasing while the distance of the water to the pool top is increasing. The investigation results conclude that the closer of the liquid-water surface to the gas-air flow, increases the evaporation rate. This approach can be applied in a SFSP that has an abundant amount of water, the water temperature remained constant by operation of the water-cooling system, air temperature, pressure, and relative humidity remained constant as well by the operation of ventilation and air conditioning (VAC) system. Those constant conditions were stated as normal operation in SFSP of GAS-MPR. In the case of abnormal conditions such as the VAC not operating or the water-cooling system not operating, there will be dynamics of pool water and air temperatures which require further modeling research.

Keywords: concentration profile, evaporation, spent nuclear fuel, water level effect.

1. Introduction
Spent Nuclear Fuel Storage Pool (SFSP) is a storage pool designed to temporarily store spent nuclear fuel for a certain period to reduce radioactive decay heat before being stored in a dry storage facility [1]. GA Siwabessy Multipurpose Reactor (GAS-MPR) spent nuclear fuel storage pool is rectangle in shape with 14 m length, 5 m width, and -9.1 m depth [2]. Spent nuclear fuel is stored in demineralized water as deep as 3.6 meters from the top of the fuel so that the water can provide sufficient radiation shielding.
to workers and the environment. The Safety Analysis Report (SAR) of the SFSP explains three-step of the safety warning system, that is Normal Condition (NC), Safety System Setting (SSS), and Safety Limit (SL). The water temperature is classified as a normal condition when it is no more than 35°C, as safety system setting when it is around 36-40°C, and as safety limit when it reaches 67°C. The water level is classified as a normal condition when it has a minimum of 4.8 m from the pool bottom, as safety system setting when it is less than 4.8 m but still more than 4.6 m, and as safety limit when it starts going through 4.2 m. In every three steps of this safety warning system, there is not a description of how the water pool behaves especially for evaporation occurrences due to water diffusion into the air.

The study of the evaporation rate has been conducted by many researchers in some applications such as swimming pools, water storage, tanks, and nuclear spent fuel storage pools. Carrier [3,4], Lovell et al. [5], Shah [6,7], Boelter et al. [8], Lecoq et al. [9], Asdrubali [10], Blazquez et al. [11] investigated factors that influence the evaporation rate of swimming pool with and without ventilation and air conditioning (VAC).

For water evaporation in SFSP, some investigation is well presented by Shah [12], Hugo [8,13], Brewster [14] in various cases such as a normal condition and Station Blackout (SBO). However, the concentration profile and the effect of water level have never been explained. In this paper, the concentration profile and the effect of water level on the water evaporation rate were investigated as a diffusion approach using numerical methods and being compared with the analytical methods.

2. Methodology
This study was conducted with real SFSP design data from GAS-MPR under normal operating conditions and steady-state processes. The air temperature and humidity were maintained constantly due to VAC operation, air pressure is maintained at 759.44 mmHg. To maintain the water level and replace the loss of pool water due to evaporation, demineralized water was supplied as make-up water to the SFSP. In the investigation of the water level effect, the water temperature was maintained constantly by circulating pool water through a cooling system.

The concentration profile of water and air mixture as a mass transfer process or evaporation were studied using numeric methods and compared with the analytical method. A simple description of a spent fuel storage pool and its associated systems is shown in Figure 1.

![Figure 1. Spent nuclear fuel storage pool at normal operation condition.](image-url)
Figure 1 shows the diffusion system in which pool water (liquid A) is evaporating into room air (gas B). This study adopted the theory of evaporation as a mass transfer diffusion of liquid surface to stagnant gas film by Bird et al. [15]. The combined flux consists of molecular flux and convective flux. The number of moles of pool water (liquid A) that go through a unit area in unit time is defined as \( N_A \). The unit area which is the cross-sectional area of the pond filled with pool water is fixed in space. The distance of the pond water surface to the upper end of the pond construction is determined by the Safety Analysis Report (SAR) that the water level at normal operation is 4.8 m. Right at the pool water–room air interface, the concentration of pool water in vapor phase is expressed as mole fraction \( X_{A1} \), \( X_{A1} \) is the vapor pressure of pool water divided by the total pressure in the system, \( P_{A}^{vap}/P \).

A gas mixture pool water–room air flows slowly past the top of the pool to maintain the mole fraction of pool water at \( \dot{X}_{A2} \) for \( z = z_{2} \). The pool water system was kept at constant temperature by operation of water-cooling system to remove the spent nuclear fuel heat generation in the bottom of the pool. The vapor velocity at the pool wall and in the center of the pool were assumed to be uniform. By assuming the evaporation process attained in a steady-state system, there was a net motion of pool water (A) away from the interface, and the species of room air (B) was stationary. With \( N_{B,z} = 0 \), solving for \( N_{A,z} \), the rate of mass transfer at the pool surface along z-coordinate is defined as Eq.2 [15].

\[
N_{A,z} = -c D_{AB} \frac{dx_{A}}{dz} \left( \frac{1}{1 - x_{A}} \right) \frac{dz}{dz}
\]

Mass balance at steady state over the increment \( \Delta z \), states that water vapor entering is equal to the leaving ones. Accordingly, in cross-sectional area \( S \) of the pool column, the mass balance was defined as Eq.3.

\[
SN_{A,z} \bigg|_{z} - SN_{A,z} \bigg|_{z + \Delta z} = 0
\]

Division by \( S \Delta z \) and taking the limit as \( \Delta z \rightarrow 0 \) [15] gives:

\[
\frac{-dN_{A,z}}{dz} = 0
\]

Substitution of Eq.2 into Eq.4 gives:

\[
\frac{d}{dz} \left( \frac{c D_{AB}}{1 - x_{A}} \frac{dx_{A}}{dz} \right) = 0
\]

The gas phase of pool water and room air were assumed to be ideal gases. The solubility of air in pool water is negligible. For the ideal gas mixture, \( p = cRT \), so that \( c \) must be constant at constant pressure and temperature. Gases \( D_{AB} \) is very nearly independent of the composition. Therefore, \( cD_{AB} \) can be moved to the left of the derivative operation and be as follows,

\[
\frac{d}{dz} \left( \frac{1}{1 - x_{A}} \frac{dx_{A}}{dz} \right) = 0
\]

The concentration profile as moles fraction of pool water \( (x_{A}) \) expressed in Eq.6 and it is a second-order differential equation. The boundary conditions were:
Boundary condition 1: At \( z = z_1 \), \( x_A = x_{A1} \)
\[ z_1 = 0 \]
\( x_{A1} \) is the vapor pressure of A divided by the total pressure = \( \frac{P_{A\text{ vap}}}{P} \).

Boundary condition 2: At \( z = z_2 \), \( x_A = x_{A2} \)
\[ z_2 = 280 \text{ cm} \]
\( x_{A2} = 0 \)

As the mole fraction in a mixture, the relation of \( x_A \) and \( x_B \) is \( x_A + x_B = 1 \). The pressure, \( P \), in the pool atmosphere, is defined as the pressure in a normal operating system at 759.44 mmHg. For the vapor pressure of water, \( P_{A\text{ vap}} \), which is a function of temperature, it uses data in Table 1 [16].

| Temperature [°C] | Pressure [mmHg] |
|------------------|-----------------|
| 26               | 25.2            |
| 35               | 42.2            |
| 40               | 55.3            |
| 67               | 205             |

2.1. Concentration profile using 4\textsuperscript{th} Runge-Kutta numeric method

The second-order differential equation (Eq.6) was solved using the fourth-order Runge-Kutta (RK4) method with the two boundary conditions. Firstly, the second-order differential equation Eq.6 was simplified into a first-order differential equation as Eq.7 and Eq.8 as follows:

\[
\frac{dx_A}{dz} = x_A' \tag{7}
\]

\[
\frac{d}{dz} \left( \frac{1}{1 - x_A} \right) = 0 \tag{8}
\]

The first-order differential equation (Eq.8) was solved then as follows:

\[
\frac{1}{1 - x_A} \frac{dx_A'}{dz} + x_A' \frac{d}{dz} \left( \frac{1}{1 - x_A} \right) = 0
\]

\[
\frac{dx_A'}{dz} = -(1 - x_A) x_A' \frac{d}{dz} \left( \frac{1}{1 - x_A} \right) = 0
\]

\[
\frac{dx_A'}{dz} = -(1 - x_A) x_A' \left( \frac{d(1 - x_A)}{dz} \right) = 0
\]

\[
\frac{dx_A'}{dz} = \left( \frac{1 - x_A}{(1 - x_A)^2} \right) x_A' \frac{dx_A}{dz} = 0
\]

\[
\frac{dx_A'}{dz} = \frac{1}{(1 - x_A)} (x_A')^2 = 0 \tag{9}
\]

The Eq.7 and Eq.9 then were solved using fourth-order Runge-Kutta (RK4) in Spreadsheet, to get the mole fraction of water (\( x_A \)) and mole fraction of air (\( x_B \)) in each increment of \( \Delta z \). The calculation takes increment, \( \Delta z = 2.8 \text{ cm} \) as the distance from the water surface to the top of the pool (280 cm).
divided by 100 points. The calculation of error between the model of RK4 and the analytical method was calculated then.

2.2. Concentration profile using analytical method

The analytical solution for the problem as Eq.6 and the two boundary conditions was described by Bird et al. [15] for the concentration profile of water (A) as follows:

\[
\frac{(1 - x_A)}{(1 - x_{A1})} = \left(\frac{1 - x_{A2}}{1 - x_{A1}}\right)^{\frac{(z-z_1)}{(z_2-z_1)}}
\]  

(10)

Or the concentration profile of air (B) as follows:

\[
\frac{x_B}{x_{B1}} = \left(\frac{x_{B2}}{x_{B1}}\right)^{\frac{(z-z_1)}{(z_2-z_1)}}
\]

(11)

2.3. Water evaporation rate in a stagnant air/gas film

The rate of mass transfer at the water-air interface, that is the evaporation rate, was obtained from Eq.2 as follows:

\[
N_{A,z} = -c \frac{D_{AB}}{(1 - x_A)} \frac{d x_A}{d z} = c \frac{D_{AB}}{x_A} \frac{d x_B}{d z} = c \frac{D_{AB}}{x_{B1}} \ln \left(\frac{x_{B2}}{x_{B1}}\right)
\]

(12)

Where \( c = \frac{P}{RT} \), \( P \) is the total pressure, \( R \) is the gas constant, \( T \) is the air temperature and \( D_{AB} \) is diffusion constant of water vapor into air that is a function of temperature. The values of diffusion constant of water vapor into the air at temperature 26, 35, 40, and 67°C can be obtained from linearization from reference data in Table 2 [17].

| Temperature, \( T_w \) [°C] | Diffusion constant, \( D_{AB} \) [cm²/sec] |
|-----------------------------|-------------------------------------------|
| 20                          | 0.246                                     |
| 30                          | 0.260                                     |
| 40                          | 0.275                                     |
| 60                          | 0.305                                     |
| 70                          | 0.321                                     |

The evaporation rate was calculated then in the mass of water transferred per time using Eq.13.

\[
W = N_{A,z} M_A S
\]

(13)

Where \( W \) is the mass of water evaporate (g/hr then will be converted to kg/hr), \( M_A \) is the mass molecule of water (18.02 g/mole), and \( S \) is the pool water surface exposed to evaporation (cm²).

3. Results and Discussions

In this study, the concentration profile and rate of mass transfer were calculated in various water temperatures and also in various water level in the next. First, 26°C is the water temperature in the reality at that time (August 2020), 35°C is the water temperature at the maximum limits of normal operating conditions criteria, 40°C is the water temperature specified as the safety system setting, and 67°C is the water temperature specified as the safety limit.
3.1. Concentration profile
In this section, the profile of water vapor concentration and room air was modeled first to determine the effect of pool water temperature. The In the liquid-vapor interface maintained at a fixed position, the diffusion model of pool water (A) through a stagnant room air (B) in a steady-state system is shown in Figures 2 to 5.

Figures 2 to 5 show how the concentration profile varies with different water temperature variations. The red lines are a mole fraction of water and the green lines are a mole fraction of air. Both of them were distributed in a pool column as z-coordinate from $z_1$ (pool water surface) to $z_2$ (top of the pool).

![Figure 2](image)

**Figure 2.** Concentration profile in z-coordinate using 4th order Runge-Kutta at $T_w = 26^\circ C$.

![Figure 3](image)

**Figure 3.** Concentration profile in z-coordinate using 4th order Runge-Kutta at $T_w = 35^\circ C$.

![Figure 4](image)

**Figure 4.** Concentration profile in z-coordinate using 4th order Runge-Kutta at $T_w = 40^\circ C$.

![Figure 5](image)

**Figure 5.** Concentration profile in z-coordinate using 4th order Runge-Kutta at $T_w = 67^\circ C$.

With the same $\Delta z$ ($z_2-z_1$), various water temperatures were resulting in differences in initial mole fraction ($x_{A1}$ and $x_{B1}$). This is because different water temperatures caused different water vapor pressure...
and $x_{A1}$ is the water vapor pressure divided by the total pressure. Increasing water temperature increased the water vapor pressure then increased the mole fraction of water. The final mole fraction ($x_{A2} = 0$ and $x_{B2}=1$) in various water temperatures is the same since it is the boundary conditions. This case demonstrates that the water level, water temperature, air temperature, air pressure, and air humidity were maintained at its value (constant).

The water evaporated that passed the top of the pool (at $z_2$, $z = 280$ cm) was removed by airflow (gas B) of the pool top continuously to maintain the same boundary conditions. From the calculation of the error between the RK4 numerical method and the analytical method, the error for each water temperature of 26, 35, 40, and 67 is 0.017%, 0.059%, 0.130%, and 1.610% respectively.

3.2. *Water evaporation rate in various water level*

When a mixture has concentration differences at two points, there will be a migration of molecules from the high to the low concentration region. The water molecules migrate from $z_1$ to $z_2$ reaching the top of the pool. Assuming the air humidity was maintained at the same value by removing the humid air, there will be a steady-state water mass transfer from the pool water surface that is called water evaporation. The rate of evaporation is the rate of mass transfer at the liquid water-air gas interface. The effect of water temperature on the water evaporation rate is shown in Figure 6.

![Figure 6](image.png)

**Figure 6.** Evaporation rate as a rate of mass transfer in various water temperature.

Figure 6 shows that the increase in water temperature will increase the evaporation rate. Water temperature at 26°C, 35°C, and 40°C show a low increasing gradient. However, there is a significant increase in the evaporation rate from water temperature at 40°C to 67°C. This is because the water vapor pressure was also increasing significantly from 55.3 to 205 mmHg. The amount of water evaporated in pool water should be replaced by make-up water (demineralized water) to maintain the water level. The production capacity of the demineralized water system was about 0.8 m$^3$/hr or about 800 kg/hr. As the limit of safe operation, the water temperature at 67°C caused the evaporation reaching 0.661 kg/hr.

According to the Safety Analysis Report (SAR) of the spent nuclear fuel storage pool, there are three-step conditions for level warning system: (1) normal operating conditions; (2) safety system setting; and (3) safety limit, that are shown in Table 3.

| Step conditions   | Water level [cm] | Distance from water level to pool top, $\Delta z$ [cm] |
|-------------------|------------------|---------------------------------------------------|
| Normal operation  | 480              | 280                                               |
| Safety system setting | 460            | 300                                               |
| Safety limit      | 420              | 340                                               |
The concentration profile and rate of mass transfer (evaporation rate) were calculated in each step conditions above at constant water temperature (for 67°C) to predict the effect of the water level towards evaporation that occurs. In the same way, the concentration profile was calculated using the 4th order Runge-Kutta in various Δz. The concentration profiles are shown in Figure 7 and the evaporation rate predictions are shown in Figure 8.

The concentration profile for various Δz in Figure 7 shows that the initial mole fractions are at the same point, but the mole fraction ended at a different point because of the different distance from the water surface to the pool top. The mole fraction in each step of increments changed gradually for each \( x_B \) and \( x_A \) in every Δz. The average error between the model and the analytical calculation for each Δz 280, 300, and 340 cm is 1.61%, 1.60%, and 1.58%. The prediction of evaporation rates for various Δz at \( t_{\text{ref}} = 67^\circ\text{C} \) in Figure 8 shows that the evaporation rate is decreasing while Δz is increasing. This explains that the closer the water surface A to the gas flow B, the greater the evaporation rate that occurs.

![Figure 7](image-url)  
**Figure 7.** Concentration profile for water temperature 67°C in various Δz.

![Figure 8](image-url)  
**Figure 8.** Water evaporation rate for water temperature 67°C in various Δz.

However, this approach only applies when all parameter conditions such as water temperature, air temperature, pressure, and humidity are remained constant and the amount of water is abundant. Those constant conditions and abundant water were stated as normal operation in actual SFSP of GAS-MPR. The amount of water is kept abundant within normal conditions to provide sufficient radiation shielding.
The water temperature is remained constant by operation of the water-cooling system. The air temperature, pressure, and relative humidity are remained constant as well by the operation of VAC system. In the case of abnormal conditions such as the VAC not operating or the water-cooling system not operating, there will be dynamics of pool water and air temperatures which require further modeling research temperature.

4. Conclusions
A numerical method is used to show the concentration profile of water and air over a spent nuclear fuel storage pool. This paper demonstrates the water level, water temperature, air temperature, air pressure, and air humidity maintained at its value (constant) then the water evaporated, passed the top of the pool, and removed by the airflow of the pool top continuously to maintain the same boundary conditions. A prediction of water evaporation rate for a variable distance of the water to the pool top at constant water temperature and constant air temperature shows that the evaporation rate is decreasing while the distance of the water to the pool top is increasing. The closer a liquid-water surface to the gas-air flow, the greater the evaporation rate that occurs. It can be applied in a spent nuclear storage pool which has an abundant amount of water, the water temperature is remaining constant by operation of the water-cooling system, the air temperature, pressure, and relative humidity are remaining constant by operation of the VAC system. This circumstance is approaching the spent nuclear storage pool at normal operation.

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