A sodium–water reaction takes place when high-pressured water vapor leaks into sodium through a tiny defect on the surface of the heat transfer tube in a steam generator of the sodium-cooled fast reactor. The sodium–water reaction brings deterioration of the mechanical strength of the heat transfer tube at the initial leakage site. As a result, it damages the crack itself, which may eventually enlarge into a larger opening. This self-enlargement is called the "self-wastage phenomenon." In this study, a simulant experiment was proposed to reproduce the self-enlargement of a crack and to evaluate the mechanism of the self-wastage. The damage on the surface of the crack was simulated by making the neutralization reaction with hydrochloric acid solution and sodium hydroxide solution. A numerical investigation was carried out to validate the feasibility of the approach and to determine experimental conditions. From the computation results, it is observed that when 5M HCl is injected into 5M of NaOH with 0.05 m/s inlet velocity, the temperature at the surface near the crack increased over 319.26 K. The computational results show that the self-wastage phenomenon is capable of being reproduced by the simulant experiment. Copyright © 2016, Published by Elsevier Korea LLC on behalf of Korean Nuclear Society. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/4.0/).
So far, self-wastage phenomenon has been studied by experimental approaches such as mock-up tests [1–5]. For example, Kuroha et al.’s [6] experimental result showed that even though the initial leak rate is very small (<0.05 g/s), the leak rate remains at a constant level for some time (from several minute to several days). The leak rate then sharply increases up to several grams per second, which could induce the secondary failure of the adjacent heat transfer tubes by target wastage. In addition, a small leakage is the most probable for steam generator operation and a large leakage, as a rule, is the consequence of subsequent failures of the heat exchange surface in the area of a small leak, and very seldom appears as an initial leakage.

Therefore, a quantification of self-wastage phenomena is important from the viewpoint of not only safety assessment but also the elucidation of the self-wastage mechanism in the steam generator.

However, it is hard to evaluate the self-wastage phenomena quantitatively due to experimental difficulties in treating liquid sodium, which possesses high chemical activity and opacity when in a liquid state.

As an alternative approach to assess the self-wastage phenomenon, an experiment using simulated material was devised. This new method focuses on reproducing the crack enlargement and its progression, which are caused by a chemical interaction between the SWR and the tube wall material.

Since the self-wastage phenomenon is attributed to a chemical reaction that takes place near the outside of the crack, we assumed that the damage on the surface of the crack can be evaluated by making the neutralization reaction with hydrochloric acid (HCl) solution and sodium hydroxide (NaOH) solution.

In this study, a numerical investigation was carried out to validate the feasibility of a new method and to decide on the conditions for the experiment. In the analysis, governing equations of concentrations of acid and alkali were implemented into a commercial Computational Fluid Dynamics (CFD) tool, Fluent version 6.3.26 by ANSYS Inc. The reaction rate is calculated based on the Arrhenius law.

### 2. Experimental methodology for simulating self-wastage phenomena using simulants

#### 2.1. Schema of SWR

When pressurized water leaks from a heat transfer tube, it will vaporize immediately due to adiabatic expansion. Hence, it is expected that water reacts with sodium under a gas state in the SWR. With respect to sodium, it reacts with water vapor under a liquid state in an early stage of the SWR as shown in Eq. (1). According to the water-leakage test that models the steam generator of Japanese prototype fast breeder reactor “Monju”, the maximum temperature measured in the reaction region was of 1,100–1,200 °C [7]. Thus, sodium vigorously evaporates, and the gas-phase sodium reacts with water vapor. At the second stage, the chemical interaction of products from the first stage (NaOH and hydrogen gas) takes place with an excessive sodium condition, as shown in Eqs. (2)–(4) [8].

\[
2\text{Na}(l) + 2\text{H}_2\text{O(g)} \rightarrow 2\text{NaOH(g)} + \text{H}_2(g) \tag{1}
\]

\[
\text{Na}(g) + \text{H}_2\text{O(g)} \rightarrow \text{NaOH(g)} + \text{H}(g) \tag{2}
\]

\[
2\text{Na}(l) + \text{H}_2\text{O}(g) \rightarrow \text{Na}_2\text{O(s)} + \text{H}_2(g) \tag{3}
\]

\[
\text{Na}(l) + \text{NaOH(s)} \rightarrow \text{Na}_2\text{O(s)} + 1/2\text{H}_2(g) \tag{4}
\]

It is expected that the wastage rate is related to the reaction rate. In the gas-phase reaction, the reaction rate is evaluated by the Arrhenius equation as follows:

Chemical reaction (\(\text{Na} + \text{NaCl} \rightarrow \text{C} + \text{D}\))

\[
\gamma = k(T)[A]^n[B]^m \tag{5}
\]

\[
k(T) = A_0(T)^{k_0} \exp\left(\frac{E_a}{T}\right) \tag{6}
\]

Here, \(\gamma\) is a reaction rate, and the \(k(T)\) is the rate constant. Eqs. (2)–(4) are considered as the gas-phase reaction. The rate constant can be derived experimentally and numerically.

The mechanism of self-wastage phenomena can be simplified by focusing on the following features. Firstly, the self-wastage phenomena contributed to an exothermic chemical reaction, and the of self-wastage rate is related to the reaction rate that is expressed by the Arrhenius reaction equation. To reproduce the enlargement of a crack from the interaction between the SWR and the tube-wall material, the following simulant experiment was designed.

#### 2.2. Simulant material

When HCl solution and NaOH solution are mixed, the following neutralization reaction occurs, and the reaction heat is released [9].

\[
\text{HCl(aq) + NaOH(aq)} \rightarrow \text{NaCl(aq)} + \text{H}_2\text{O} + \Delta H \tag{7}
\]

In an aqueous solution, each reactant is dissociated fully so that the above reaction can be written in the following form:

\[
\text{H}_2\text{O}^+ + \text{OH}^- \xrightleftharpoons[k_f]{k_b} 2\text{H}_2\text{O} \tag{8}
\]

where \(k_f\) and \(k_b\) are the rate constants for the chemical reaction. The rate constant \(k\) is also expressed by the Arrhenius equation, as follows:

\[
k_f = A_te^{-\frac{E_a}{RT}} \tag{9}
\]

where, \(A_t\) = pre-exponential factor (mol\(^{-1}\) m\(^3\) s\(^{-1}\)); \(E_a\) = activation energy for the reaction (J mol\(^{-1}\)); and \(R\) = universal gas constant (J mol\(^{-1}\) K\(^{-1}\)).

In Eq. (8), the rate constant \(k_b\) is a few orders of magnitude smaller than \(k_f\), therefore, the effect of backward reaction could be neglected.

One similarity exists in the reaction mechanism of the SWE and the neutralization reaction: the reaction rate is expressed by the Arrhenius equation. The reaction rate is closely related to the self-wastage phenomena.
2.3. Experimental procedure

Fig. 1 explains the process of the self-wastage phenomenon. (1) The self-wastage phenomena are attributed to a chemical reaction followed by a reaction heat and a corrosion that takes place at the initial crack site. (2) The crack enlargement starts from the sodium side and advances through the tube wall. As the crack enlargement advances by the self-wastage, the reaction zone would migrate toward the water/vapor side. The corrosive reactant easily reaches the tube wall. Thus, the self-wastage phenomena would be accelerated. At this stage, the leak rate would be stable at a certain level. (3) If the wastage reaches the water side, the resultant leak rate will sharply increase. The SWR zone then goes downstream so that the corrosive reactants seldom reach the tube wall. Thus, the self-wastage phenomenon is mitigated. (4) In the experiment, HCl and NaOH were used as the simulant. Paraffin wax (C25H52) which has a low melting point (115 °F, 319.26 K) was chosen as simulated material which represents the heat transfer tube. In the experiment, HCl solution was injected into the reaction tank that is filled with NaOH solution so that the neutralization reaction takes place in the nozzle exit and the reaction heat is released. (5) The temperature of the wax surface increases. When the temperature exceeds the melting point of the paraffin wax, the surface melts. Thus, the nozzle is enlarged. The nozzle enlargement proceeds until nozzle penetration. (6) When the enlargement reaches the inlet, the injection rate will increase. Thus, the reaction zone moves downstream. Therefore, the nozzle enlargement is mitigated.

When the nozzle is penetrated, the phenomenon is regarded as terminated. Then the geometry of the enlarged leak is evaluated. Schematic experimental apparatus is shown in Fig. 2. The experimental setup consists of a reaction tank, a paraffin wax nozzle, and a solution supply system.

3. Numerical procedure of the methodology

A two-dimensional preliminary numerical investigation was carried out to evaluate the feasibility of the experimental procedure. Commercial Computational Fluid Dynamics software (Fluent version 6.3.26 by ANSYS Inc.), was used for the numerical analysis.

3.1. Governing equations

In this study, a transient two-dimensional flow was considered. The conservation equation for chemical species is given in the following form:

$$\frac{\partial}{\partial t}(\rho Y_i) + \nabla \cdot (\rho \mathbf{v} Y_i) = -\nabla \cdot \mathbf{J}_i + \mathbf{R}_i$$  \hspace{1cm} (10)

where $\mathbf{R}_i$ is the net rate of production of a mass of species; $Y_i$ is the per unit volume by chemical reaction; and $\mathbf{J}_i$ is the diffusion flux of species $i$, which arises due to concentration gradients, under the diffusion flux. This can be written as:

$$\mathbf{J}_i = -D_{i,m} \nabla Y_i$$  \hspace{1cm} (11)

Here, $D_{i,m}$ is the molecular diffusion coefficient for species $i$ in the mixture. In aqueous solution, the diffusion coefficient $D_i$ (the electrolyte diffusion coefficient), can be given as:

$$D_i = \frac{|z_1|^2 + |z_2|^2}{|z_1||z_2|} \left( \frac{1}{D_1} + \frac{1}{D_2} \right)^{-1}$$  \hspace{1cm} (12)

where $D_1$ and $D_2$ are the molecular diffusion coefficients of the individual ions, and $z_1$ and $z_2$ are the valences of the ions. $D_i$,
and $D_2$ can be obtained from the Stokes–Einstein equation as shown below [10]:

$$D_{(1.2)} = \frac{kT}{6\pi \mu R_i}$$  \hspace{1cm} (13)

where $k$ is the Boltzmann constant, $T$ is the absolute temperature, $\mu$ is the dynamic viscosity of the m (in this study, it is water), and $R_i$ is the radius of the solute.

For the net reaction rate, the laminar finite-rate model is adopted. The model computes the chemical source terms using the Arrhenius expression. The net source of chemical species $i$ in the reaction is given by:

$$R_i = M_{w,i} \sum_{r=1}^{N_r} R_{i,r}$$  \hspace{1cm} (14)

where $M_{w,i}$ is the molecular weight of species $i$ and $R_{i,r}$ is the Arrhenius molar rate of creation/destruction of species $i$ in reaction $r$. The reaction may occur in the continuous phase at wall surfaces.

The molar rate of creation/destruction of species $i$ in the reaction is given by:

$$R_{i,r} = k_F[C_{j,r}]^{\eta_j}$$  \hspace{1cm} (15)

where $k_F = $ rate constant for reaction; $C_{j,r} =$ molar concentration of species $j$ in reaction $r$; $\eta_j =$ rate exponent for reactant species $j$ in reaction $r$; and $\eta_{i,r} =$ rate exponent for product species $j$ in reaction $r$.

For the neutralization reaction with HCl and NaOH, the rate exponents are 1, respectively. Thus, the molar rate of creation/destruction can be written as:

$$R_{i,r} = k_i [\text{HCl}][\text{NaOH}]$$  \hspace{1cm} (16)

The rate constant depends on only temperature, and it can be computed from the Arrhenius equation shown as the previous chapter. The pressure-based solver is adopted, and the Simple algorithm is chosen to enforce mass conservation and to obtain the pressure field.

3.2. Modeling and analytical conditions

3.2.1. SWAT-2 experiment

Kuroha et al. performed a mock-up experiment using the Sodium-Water Reaction Test Rig (SWAT-2) test rig and studied the behavior under the same conditions as the Japanese prototype fast breeder reactor Monju’s steam generator.

The slit-type nozzle, which was manufactured by pressing a square plate with a drilled hole at its center, was used in the experiment. Each slit had a high aspect ratio. The crack length was approximately 0.05 mm and the crack width was in the range of 10–100 μm. The behavior of the vapor blow-off through the slit nozzle differs according to nozzle direction. As shown in Fig. 3, the vapor was blown off directly from the front view. However, the vapor was ejected radially from the side view. The estimated direction of self-wastage is depicted as a dotted line in the figure. The nozzle will enlarge dominantly in the direction of the nozzle thickness and the width. The same tendency was observed from the SWAT Experiment that initially the nozzle had a slit shape; however, the enlarged nozzle became to have a pit-hole shape. The dominant enlargement took place in the nozzle width direction. It can be said that the self-wastage phenomenon mainly takes place toward both the nozzle thickness and width directions. In other words, the nozzle enlargement in the nozzle-length direction can be negligible. Therefore the two-dimensional model is adopted as the analytical model.

Table 1 shows the #2021 experimental condition of the SWAT-2 Experiment. The aspect ratio of the nozzle width (24 μm) and the depth (3.48 mm) is 1:166 and the equivalent diameter of the nozzle is 0.046 mm.  

<table>
<thead>
<tr>
<th>Experiment no.</th>
<th>Tube material</th>
<th>Tube thickness (mm)</th>
<th>Sodium temperature (°C)</th>
<th>Sodium pressure (MPa)</th>
<th>Steam temperature (°C)</th>
<th>Steam pressure (MPa)</th>
<th>Initial nozzle width (μm)</th>
<th>Equivalent diameter of leak (mm)</th>
<th>Initial leak rate (g/s)</th>
<th>Average leak rate (g/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2021</td>
<td>SUS321</td>
<td>3.58</td>
<td>505</td>
<td>1.47 × 10^-2</td>
<td>505</td>
<td>12.8</td>
<td>15</td>
<td>0.046</td>
<td>7.1 × 10^-3</td>
<td>2.4 × 10^-3</td>
</tr>
</tbody>
</table>

Table 1 – SWAT-2 experimental conditions.

3.2.2. Numerical procedure

The numerical approach, estimating the self-enlargement of the nozzle, consists of the two parts: (1) construction of the

![Fig. 3 – Configuration of injected vapor and slit wastage direction.](image-url)
analytical model and (2) the wastage rate evaluation. The analytical model was derived to achieve the geometric and dynamic similarity between the analytical model and the prototype experiment. In order to estimate the wastage rate (amount), a temperature contour in the nozzle was used. Since the calculation is transient, the temperature contour is obtained after several seconds when the temperature around the nozzle exit becomes stable.

Self-wastage requires several days to penetrate the tube wall after its initiation. Therefore, the heavy computational load is necessary to cover the whole picture of the phenomena at once. During the self-wastage advance through the tube wall, the leak rate is almost constant. It is therefore assumed that the self-wastage rate is also constant during the same period. The self-wastage phenomena can be assumed to have a steady condition. Thus, a stepwise numerical procedure is proposed to represent the steady period of the self-wastage phenomena. Fig. 4 explains the numerical procedure.

Step I: determine an initial nozzle size. To determine the nozzle width, the SWAT-2 experimental data is used. In order to achieve dynamic similarity between the SWAT Experiment and the numerical analysis, the Reynolds number of the numerical analysis was determined to be equal to that of the SWAT-2 Experiment. The aspect ratio of the nozzle has the same value as the SWAT Experiment.

Step II: perform a transient calculation up to a few seconds until the reaction gets to be stable and obtain thermal properties such as temperature contour, heat flux, and species' distribution around the exit of the nozzle.

Step III: to evaluate the amount of the melted wax, the heat flux at the surface and the surface temperature are necessary. Melting rate (mm/s) can be obtained by dividing the heat flux by the latent heat and the density of the wax.

Step IV: the new mesh grid was produced by eliminating the area where the temperature goes over 319.26 K along the isothermal line of the wax melting point.

Step V: iterate the computation from Step II to Step IV until the enlargement reached the HCl side.

Step VI: Evaluate the size of the enlarged nozzle.

3.3. Numerical conditions

Fig. 5 shows the two-dimensional analytical area and its mesh arrangements. The region contains the reaction tank, the paraffin wax, and the nozzle. The height of the computational region (reaction tank) was decided on to take a sufficient margin to evaluate the phenomena. Thus, the height of the computational domain of the reaction tank is 150 mm and the width of that is 100 mm. Also, the width (0.2 mm) and the height (20 mm) of the nozzle were determined to have a similar aspect ratio as the nozzle used in the SWAT-2 Experiment. The analytical conditions are listed in Table 2.

To evaluate the mesh sensitivity of the analytical grid, three different mesh grids were used, as shown in Fig. 6. All these meshes have the same mesh size for the nozzle (10 meshes). However, each mesh has six, 10, and 16 meshes for a section that has the same length as the nozzle width in the vicinity of the nozzle exit.

Fig. 7 shows the temperature distribution at the surface of the nozzle top of the three meshes. In this figure, the red dot-dashed line represents the melting point of the paraffin wax. The temperature is averaged over 5 s. For all meshes, the temperature at the surface has almost the same distribution.
All meshes have the same maximum temperature (331.2 K) at the same place. Therefore, the minimum mesh size for the feasibility analysis was decided as the same value with the six mesh grid in Fig. 6A.

The width of the initial nozzle is divided equally into 10 (l). The cells of the other region are divided into 111 (l) × 59 (l) for the reaction tank, 50 (l) × 59 (l) for the wax, and each element increases with the ratio of 1.1. The total number of the cells is ~12,000.

For the boundary condition, the constant inlet velocity is applied to the inlet and the constant pressure condition is adopted for the outlet boundary. The inlet velocity is determined to have the same Reynolds number like that in the SWAT-2 Experiment so that the similarity of the flow behavior in the computation is ensured to the experiment. The heat conduction between the fluid and the paraffin wax is considered to evaluate the temperature increase by the reaction heat. Further computational conditions are summarized in Table 1.

---

### Table 2 – Analytical conditions.

<table>
<thead>
<tr>
<th>Initial condition</th>
<th>298 K</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fluid</td>
<td>NaOH</td>
</tr>
<tr>
<td>Concentration</td>
<td>5.0 mol/L</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Boundary condition</th>
<th>Inlet</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fluid</td>
<td>HCl</td>
</tr>
<tr>
<td>Velocity</td>
<td>0.05 m/s</td>
</tr>
<tr>
<td>Concentration</td>
<td>5.0 mol/L</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Analytical condition</th>
<th>Mesh arrangement</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nozzle</td>
<td>11 × 42</td>
</tr>
<tr>
<td>Reaction tank</td>
<td>111 × 59</td>
</tr>
<tr>
<td>Wax</td>
<td>50 × 59</td>
</tr>
<tr>
<td>Time step</td>
<td>5.0 μs</td>
</tr>
</tbody>
</table>

---

![Fig. 6 – Different mesh grids for mesh sensitivity evaluation. (A) Six mesh. (B) Ten mesh. (C) Sixteen mesh.](image_url)
4. Results and discussion

4.1. Temperature distribution near surface of wax nozzle

As shown in Fig. 7, from 0.15 mm to 0.45 mm, the surface temperature increased over the melting point of paraffin wax. The temperature is high enough to melt wax in this area. To decide the amount of melting wax by reaction heat, the following equation was used:

\[ M = \frac{H_f}{\rho L} \]  

(17)

where \( M \) is the melting rate (m/s); \( H_f \), the heat flux at the surface (W/m²); \( L \), the latent heat of fusion; and \( \rho \), the density of paraffin wax. Since the analysis model deals with the stepwise procedure, the maximum melt amount is set to be 5% (1 mm) of the initial nozzle wall thickness for each calculation. The maximum melt amount is located where the melting rate is maximum. The melt amount on another meshes is decided by multiplying the ratio with the maximum melt rate.

Fig. 8 shows the surface heat flux at the nozzle top. Heat flux has a similar distribution to temperature distribution as shown in Fig. 7. To decide the melting amount on the heating surface, we divided the width of the melting area into two parts, which are designated A and B, and investigated the average heat flux in the areas. The average heat flux of A is about 5,000 W/m², and the \( H_f \) of B is 39,000 W/m².

Therefore the melting amount on A and B is 1 mm and 0.8 m, respectively. By eliminating the depth of the melting wax on each mesh, the new mesh arrangement was obtained, as shown in Fig. 9.

The new calculation is performed using the new mesh grid shown in Fig. 9. The above procedures (obtaining temperature distribution at the surface of the wax in both width and thickness directions of the nozzle, investigating the surface heat flux, then evaluating the melting area and obtaining the new mesh grid by removing the melting area from the mesh) are iterated until the melting propagation reaches the inlet side.

4.2. Development of self-enlargement of nozzle

A series of computations were carried out to evaluate the development of the self-enlargement of nozzle caused by the reaction heat. In each calculation, a new remeshed computational grid was generated by eliminating the melted zone according to the temperature contour and temperature profile around the leak exit.

Fig. 10 shows the microscopic morphology on completely self-wasted leaks [11]. Both leaks have a similar funnel shape, as shown in Fig. 11. More similarities are found as a result of the calculation (Fig. 10) and experimental results (Fig. 11). Both the cross sections have a larger opening at the exit of the leak and its diameters are about the same as the thickness of the tube wall. From the computational results, it is shown that the simulated experiment and the self-wastage phenomenon have a similar behavior. However, the reason the outer diameter of an enlarged leak becomes comparable to the thickness of the nozzle is not clear. More parametric analysis are necessary to evaluate to make clear that which parameter affect the nozzle enlargement.

Conventional mock-up tests for the self-wastage phenomena have the difficulty that they require a high operating cost. Thus, only a limited number of experiments were conducted, which was not enough to evaluate the effect of all parameters on the phenomena.

However, the simulant experiment has the advantage that it can easily modify the experimental conditions, such as leak
geometry and leak rate. Therefore, parametric analysis can be carried out.

The concept of the simulant experiment, which reproduces the self-wastage phenomena, was proposed in this paper. In the experiment, HCl, NaOH aqueous solution, and paraffin wax were chosen as simulant. The reaction heat released by the neutralization reaction with HCl and NaOH is used to simulate the corrosion effect of the self-wastage phenomena. By injecting the HCl solution through a narrow wax nozzle, the neutralization reaction occurs near the leak exit. As a result of the release of the heat, the paraffin wax in the reaction zone would melt. Thus, the enlargement of the nozzle takes place. This procedure represents the self-enlargement of the leak.

To evaluate the feasibility of this methodology, a numerical analysis procedure was devised, and a two-dimensional numerical calculation were performed. When 5M of HCl and NaOH solution are used, the temperature at the surface of nozzle increases above the melting point of paraffin wax. The melting rate is obtained from the surface heat flux and is used to evaluate the melting amount on each mesh for every calculation. By removing the wall according to the melting amount, a mesh grid for the enlarged nozzle is obtained. The cross section of a completely enlarged nozzle can be obtained by iterating these procedures. The shape of the cross section is like a funnel, and it shows good agreement with the SWAT experimental results. Therefore, it can be concluded that the feasibility of the simulant experiment is assured from a phenomenological viewpoint.

**Conflicts of interest**

All authors have no conflicts of interest to declare.

**REFERENCES**