Analysis of Self-wastage Phenomena of Micro Leak Caused by Sodium-water reaction in Sodium-cooled Fast Breeder Reactor through Simulant Experiment

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ABSTRACT

Self-wastage phenomena are an enlargement of a leak on the heat transfer tube caused by a corrosive sodium-water reaction (SWR) in a steam generator (SG) of sodium-cooled fast breeder reactor (SFR). If the steam generator operates for sometimes under this condition, the self-wastage phenomena start from the sodium side and advance through the tube thickness. The leak rate stays almost constant level until the wastage reaches the sodium side, however, when the thin diaphragm of the tube wall is removed, the leak rate sharply increase, and it may bring a secondary failure of the surrounding heat transfer tubes.

The design and safety concern is a possibility of the secondary failure of nearby SG tubes that could cause undesirable development of the accidents. One needs to evaluate the increased resultant leak rate due to the self-wastage phenomenon. Therefore, a quantification of the diameter of enlarged leak is needed to estimate the resultant leak rate. For this purpose, a simulant self-wastage experiment was proposed to investigate the self-enlargement of the leak so that evaluate the mechanism of the Self-wastage. In the experiment, high concentrated hydrochloric acid (HCl) is injected to the reaction tank that is filled sodium hydroxide (NaOH) solution through a nozzle made by paraffin wax. The self-enlargement of the leak was evaluated by considering the melted nozzle due to the reaction heat released from the Neutralization reaction. Also, a numerical investigation has been carried out to evaluate the enlarged nozzle and validate the results of experimental methodology. Based on the experimental and computational results, it is found that despite initial leak rate, there is an upper limit in the enlarged nozzle. These results show a similar tendency with the experimental result of SWAT-4 experiment carried out by Power Reactor and Nuclear Fuel Development Corporation (PNC), Japan. Furthermore, the increased resultant leak rate is evaluated using the enlarged leak size from the experiment and computational results.

KEYWORDS

Self-wastage phenomena, sodium-water reaction, CFD.

1. INTRODUCTION

Sodium-water reaction (SWR) is a design basis accident of a Sodium-cooled Fast breeder Reactor (SFR). A breach of the heat transfer tube in a steam generator results in contact of liquid sodium with water vapor. Typical phenomena are deterioration of the tube wall material and its removal due to the corrosive SWR and the erosive effect caused by high-pressure vapor jet. As results, the breach is enlarged at its initial site and it brings a sharp increase of
leak rate. The enlargement of leak caused by the SWR is called Self-wastage phenomena. According to experimental researches, it has been observed that even though the initial leak rate was very small (less than 0.05 g/sec), the leak rate increases more than two orders of magnitude in a period of about ten seconds [1,2]. Thus, there is a danger that the resultant leak rate caused by the self-wastage might be in the region of a small leak, or even an intermediate leak which would then give rise to problems of target wastage. The Phenomena of self-wastage have been studied experimentally by several investigators [3-6]. Sandusky explained the Self-wastage starts from the sodium side and advances through the tube thickness. The leak rate stays almost unchanged until the thin edge of the wall is removed, then the leak rate sharply increases [7]. Quantification of the diameter of the enlarged leak and the time to sudden enlargement under various steam generator operating conditions are necessary to evaluate the leak detection capability of small leaks before they become larger leaks and, consequently, damage the surrounding tube. However, there exist limitations in the experimental approach to evaluate the self-wastage phenomenon since the SWR is complex, and the experimental measurement technology is not well matured enough to unravel the reaction mechanism in detail.

So we suggested a new method to assess the self-wastage phenomena through a simulant experiment. This new method focuses on the quantification of the diameter of the enlarged leak by the self-wastage phenomena. Since the self-wastage phenomena are attributed to a chemical reaction takes place near the outside of the leak. We assumed that it was able to evaluate the damage on the surface of the leak by making the Neutralization reaction with hydrochloric acid (HCl) solution and sodium hydroxide (NaOH) solution.

In this paper, a preliminary numerical investigation was carried out to validate the feasibility of a new method and to decide the conditions for the experiment. In the analysis, governing equations of concentrations of acid and alkali were implemented into a commercial CFD tool, Fluent Ver.6.3.26. The reaction rate was calculated based on the Arrhenius law.

2. SKETCH OF THE SIMULANT EXPERIMENT

2.1. Simulant Material

The Self-wastage is the result from the interaction of the corrosive and erosive effect of the SWR. This combination of the erosive and corrosive effects of the SWR is so complicated that it is difficult to evaluate the self-wastage phenomena quantitatively. However, from a phenomenological viewpoint, it can be said that, the self-wastage phenomena is contributed to an exothermic reaction which takes places around the outside of the leak. Thus, the idea of adopting an exothermic reaction was proposed to simulate the self-wastage phenomena. The neutralization is an exothermic reaction when HCl and NaOH aqueous solution are mixed the following exothermic reaction takes place

\[
HCl_{aq} + NaOH_{aq} \rightarrow NaCl_{aq} + H_{2}O \Delta
\]

As a result of the reaction, the reaction heat (\(= 55.6 \text{ kJ/mol}\)) releases. Paraffin wax 115F, which has a relatively low melting point (115 F, 319.2 K), is adopted as simulant to represent the tube wall. When the neutralization reaction takes places around the outside of the leak, it is expected that the temperature in the reaction zone increases beyond the melting point of the wax so that the wax in that area is melted by the reaction heat. As the reaction continues, the nozzle enlargement advances through the nozzle wall. This procedure reproduces the crack
enlargement by the self-wastage phenomena.

2.2. Experimental Procedure

Brief depiction of self-wastage phenomena and the concept of the simulant experiment are described in Fig. 1.

(a) The self-wastage phenomena are attributed to a chemical reaction, which is followed by reaction heat, takes place at the initial leak site.

(b) The wastage phenomena start from the sodium side and advance through the tube thickness. As the leak gets enlarged by the self-wastage the reaction zone advances toward the water/vapor side. As a result of the enlargement, the corrosive reactants become easily reach the tube wall. Also, the swirl flow accelerates the transport of the corrosive reactant in the reaction zone. Thus, the sodium water reaction will be continued. The leak rate has stability in this stage.

(c) If the wastage reaches the water side, resultant leak rate will be sharply increased then the sodium water reaction zone migrates to the sodium side so that the self-wastage is mitigated.

(d) Hydrochloric acid solution (HCl) and sodium hydroxide solution (NaOH) are used as a simulant. Paraffin wax 115 is chosen as a simulant for the nozzle which represents the heat transfer tube. In the experiment, HCl solution is injected to the reaction tank which is filled with NaOH solution through the paraffin wax nozzle, so that the neutralization reaction would occur in the outside of the nozzle and the reaction heat will release.

(e) If the temperature of the surface of the wax exceeds the melting point of the paraffin wax, the surface of the wax in that area will be melted. Thus, the size of the nozzle would be enlarged. The resultant leak rate is affected by the diameter of the nozzle in sodium side. By investigating the diameter and the shape of the enlarged nozzle hole, the effect of the self-wastage on the heat transfer tube wall could be evaluated.

Fig. 1. Overview of Self-wastage and Simulant experiment
Figure 2 shows a schematic experimental apparatus. Experimental setup consists of a reaction tank, a paraffin wax nozzle and a solution supply system. In order to evaluate the feasibility of the simulant experiment and to decide the experimental conditions, numerical analyses were carried out using commercial CFD software ANSYS Fluent 6.3.36.

3. NUMERICAL FEASIBILITY ANALYSIS

3.1. Governing Equations

A two-dimensional unsteady flow of mass transfer with chemical reaction has been calculated in this study. The conservation equation for chemical species is given by

\[
\frac{\partial}{\partial t} (\rho Y_i) + \nabla \cdot (\rho \vec{v} Y_i) = -\nabla \cdot \vec{J}_i + R_i
\]  

(2)

Where \( R_i \) is the net rate of production of mass of species \( Y_i \) per unit volume by chemical reaction. \( \vec{J}_i \) is the diffusion flux of species \( i \), which arises due to concentration gradients. For the net reaction rate, the laminar finite-rate model is adopted. Chemical species are computed based on the Arrhenius expression [7]. The net source of chemical species \( i \) due to reaction is computed as the sum of the Arrhenius reaction sources over the \( N_r \) reaction which the species participate in

\[
R_i = M_{w,i} \sum_{r=1}^{N_r} R_{i,r}
\]  

(3)

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 \( r \). Reaction may occur in the continuous phase at wall surfaces. The molar rate of creation/ destruction of species \( i \) in reaction are given by

\[
R_{i,r} = k_F \left[ C_{j,r}^{n-j_i+r_i} \right]^{n_j+r_j}
\]  

(4)

where

\( k_F = \) rate constant for reaction

\( C_{j,r} = \) molar concentration of species \( j \) in reaction \( r \)
\( \eta^r_{j} \) = rate exponent for reactant species \( j \) in reaction \( r \)

\( \eta^r_{j'} \) = 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

\[
R_{i,r} = k_r [HCl][NaOH]
\]

(5)

The rate constant depends on only temperature and it can be computed from the Arrhenius equation shown as below

\[
k_r = A_r e^{-E_a/RT}
\]

(6)

where

- \( A_r \) = pre-exponential factor (mol\(^{-1}\) m\(^3\) s\(^{-1}\)), \( E_a \) = Activation energy for the reaction (Jmol\(^{-1}\))
- \( R \) = universal gas constant (Jmol\(^{-1}\)K\(^{-1}\))

### 3.2. Numerical Approach

Numerical approach to evaluate the self-enlargement of the leak consists of the two parts, (a) construction of analytical model (b) wastage rate evaluation using local thermal hydraulic properties around the initial crack on the heat transfer tube. The remeshing procedure was followed by evaluating the wastage rate calculated from the local thermal properties.

#### 3.2.1 Numerical Procedure

Since the phenomena require a long period until the heat transfer tube enlargement occurs after the initial leak initiates, there are limitations to carry out a computation during the whole time span of the phenomena. It requires a large computation load, and it has complexity to develop a computational model to reproduce continuous wastage phenomena. Thus, instead of transient numerical calculations which consider the whole series of event at one time, a transient calculation was carried out during a certain period time until it reaches the metastable state. The amount of the wastage rate is evaluated from the thermal properties from the numerical calculation and then a new analytical mesh model is reconstructed. Figure 3 shows the numerical procedure of the simulant experiment.

Step I) Determining an initial nozzle size: To determine the nozzle size SWAT-2 experiment data was used. In order to achieve dynamic similarity between the experiment in real condition and the numerical analysis, Reynolds number of the numerical analysis was decided to be equal to that of the SWAT-2 experiment. The nozzle size was decided to have the same aspect ratio of the real nozzle.

Step II) Investigation local thermal properties and evaluate the amount of the melting wax: Carry out an unsteady numerical simulation for a certain period until it reaches metastable state. Then obtain thermal properties such as temperature contour in the wax and species distribution around the exit of the nozzle. In real phenomena, crack progresses according to the crystal grain boundary of the material. It is assumed that the wax also melted according to the contour line of the melting point of the wax.
Step III) Remeshing based on the melting wax: The new mesh grid was produced by changing solid cells to fluid cells according to the temperature contour of the melting point which was obtained from the step III.

Step IV) Iteration from the step II to III: Iterate from the step II to step II until the thickness of the remained wax on HCl side become 10 % of the original thickness. In real condition, due to high pressure of water vapor, the opening of the remained tube wall is expected to happen when the wastage reaches about 90 % thickness of the initial thickness.

Step V) Evaluating the minimal size of the enlarged nozzle: The resultant leak rate would be almost constant until the remained diaphragm of the wall is removed. Therefore, it is expected that the leak rate is controlled by the minimal diameter of the enlarged nozzle. When the thickness of the region of melted wax reaches the 80 % of the initial thickness, obtain the minimal size of the enlarged nozzle on the sodium side.

![Numerical Procedure for Simulant Experiment](image)

Fig. 3 Numerical Procedure for Simulant Experiment

4. NUMERICAL ANALYSIS

4.1. Analytical model and conditions

Table 1 shows the experimental conditions of the SWAT-2 and the analytical conditions for simulant experiment [8]. The SWAT-2 experiment adopted a crevasse-shaped artificial crack which was manufactured mechanically by opening a hole with a drill. Figure 4 shows two-dimensional analytical region and its mesh arrangements. The region contains the reaction tank, the paraffin wax and the nozzle. The thickness of the wax in the simulant material was decided as 2 mm from the viewpoint of the convenience in manufacturing. The width of the nozzle was determined to have the same aspect ratio with the crack of the SWAT-2 experiment so that the geometrical similarity was achieved between the real prototype and the simulant material. As shown in Fig.4, the width of the initial nozzle is
divided equally into 11 cells (I). The meshes of the other region are divided into 111 (I) × 59 (J) for Reaction tank, 50 (I) × 42 (J) for the wax, and each element increases with the ratio of 1.1. The Total number of the meshes is approximately 10,000. In order to attain the dynamic similarity of the flow between the two models, the initial leak rate in the simulant experiment was decided to have the same Reynolds number with the real condition. For the inlet boundary, constant inlet velocity was adopted, and the inlet velocity was decided to have the same Reynolds number with the real condition had constant flow rate (2.35×10⁻⁵ g/s) and the outlet boundary, constant pressure was used. The side walls of the reaction tank are the free-slip condition and the heat exchange between the fluids, and the surface of the wax on the top of the wax was considered.

![Fig. 5 Analytical Region for Initial Calculation](image)

<table>
<thead>
<tr>
<th>Table 1. Experiment and Analytical Conditions of SWAT-2 and Simulant Experiment</th>
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<tbody>
<tr>
<td><strong>(a) SWAT-2 Experiment Conditions</strong></td>
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<tr>
<td><strong>Experimental Conditions</strong></td>
</tr>
<tr>
<td>· Experiment Number : 2020</td>
</tr>
<tr>
<td>· Tube Material : 2.25 Cr-1Mo</td>
</tr>
<tr>
<td>· Tube Thickness : 3.84 mm</td>
</tr>
<tr>
<td>· Sodium Temperature : 470 °C</td>
</tr>
<tr>
<td>· Sodium Pressure : 1.47×10¹ MPa</td>
</tr>
<tr>
<td>· Steam Temperature : 470 °C</td>
</tr>
<tr>
<td>· Steam Pressure : 12.8 MPa</td>
</tr>
<tr>
<td>· Initial crack width : 15 μm</td>
</tr>
<tr>
<td>· Initial leak rate : 3.4×10⁻⁴ g/s</td>
</tr>
<tr>
<td>· Average leak rate : 2.5×10⁻⁵ g/s</td>
</tr>
</tbody>
</table>

| **(b) Analytical Conditions for Simulant**                     |
| · Initial Condition                                           |
| · · Temperature : 298K                                       |
| · · Fluid : NaOH                                              |
| · · Concentration : 5.0 mol/l                                 |
| · Boundary condition                                          |
| · · Inlet                                                    |
| · · Fluid : HCl                                              |
| · · Velocity : 0.05 m/s                                       |
| · · Concentration : 5.0 mol/l                                 |
| · Analytical Condition                                        |
| · Number of cells : 9961                                      |
| · Time step : 5.0 μs                                           |
| · Analysis time : 10.0 sec                                    |

4.2. Results and Discussion

4.2.1 Analysis of the initial model
Figure 5 shows the numerical results of the initial model at several different seconds. The red line on the temperature contours indicates the melting point of the paraffin wax (319.26K). The temperature of the area inside the red line exceeds 319.26 K. Accordingly it is expected that the wax in this area would be melted by the reaction heat. It is shown that as time passes, the area where temperature is higher than the melting point become smaller and located beside the nozzle outside. In order to check that the calculation reaches meta-stable state, transient temperature distribution on the top of the wax was obtained in Fig. 6. The temperature has slight changes less than 0.5 sec; however, temperature changes on the top of the wax do not distinguish from 0.5 sec to 10.0sec. Molar concentration, reaction rate and velocity vector around the outside of the nozzle was given in Fig. 7. It can be recognized that the neutralization reaction take place at the almost same area, right beside of the nozzle in which the temperature increases more than the melting point. From the velocity vector of the flow, it can be said that the calculation reached the meta-stable state in a few seconds. Using the thermal properties at 10 sec, evaluation of the melting wax was conducted.

Fig. 5. Temperature around Nozzle at Different Times
Figure 8 shows the temperature distribution on the top of the wax at 10 secs. To estimate the width of the melted area the temperature distribution toward x-direction on the surface wax (Y=2.0 mm) was obtained. In this figure, the red dot-dashed line indicates the melting point of the paraffin wax, and black dotted area indicates the width of the nozzle (0.2mm). It is shown that the temperature of the wax surface right beside the nozzle increased over the melting point. The width of this area is approximately 0.035 mm. Since the wax would be melted along the temperature contour line of the melting point, it is assumed that the thickness and shape of the melted area are regarded the same as that of the contour line. So the shape of the surface of wax after being melted by the reaction heat would be represented as black-dotted line in Fig. 9-(a). A newly remeshed grid was obtained as shown in Fig. 9-(b) by exchanging the solid(wax) cells to fluid cells in the melted area. These procedures mentioned as the step II and III were iterated until the remaining thickness of the wax become 10 % of the initial thickness.
4.2.2 Further numerical analysis

Using the newly remeshed grid in Fig. 9 further calculations were performed under the same numerical condition of the initial calculation. In total 75 calculations until the thickness of the remained wall on sodium side become 4 mm. Figure 10 shows the outline of the remeshed grid that were taken among the 75 calculations. As shown in the early states, the enlargement of the nozzle is noticeable in the both x and y-direction as calculation goes further. It seems that the enlarged nozzle has a symmetric concave shape that means the wax is melted by reaction heat in meta-stable state like shown in the initial calculation. The results of the further calculations show that the nozzle enlarges in an unsymmetrical way. In other words, one side of the wax at the top of the wax has more enlargement than the other side. This result was brought due to the direction change of flow. At first as shown in Fig 7, in early stage of the self-enlargement, the injected flow has a stable flow pattern. However as the wax is enlarged, the injected flow of HCl was fluctuated by the reaction, these bring the unsymmetrical shape of the enlarged nozzle. As the calculation proceeds, it seems that the shape of the enlarged nozzle looks like almost symmetric. It is expected that until the enlargement of the nozzle reach to a certain size, the nozzle would be melted in having a symmetric shape. However, when the enlarged nozzle exceeds a certain size, the flow of injected HCl was disturbed, and it produces an unsymmetric further enlargement of the nozzle. Hence, as calculation goes on, the disturbance of the flow brings another almost symmetric surface of the enlarged nozzle. Accordingly from the 40th calculation, even though the estimated enlarged nozzle shape is unsymmetrical, it is assumed that the nozzle enlarged to have a symmetric shape. Through 75 calculations, it is found that the enlargement advances until 10 % of the initial tube thickness. So it is regarded that the self-wastage phenomena were suppressed. Therefore, the numerical calculation was terminated.
4.2.3 Evaluation of the width of the enlarged nozzle

As mentioned earlier, in the real condition high pressure is working on the steam side (inside of the tube), so that when the remaining tube wall reaches certain thickness, the remaining diaphragm of the wall would be removed by the steam jet. As a result, the width of the crack will be enlarged, and the remaining wall thickness will also be increased. In this study, it is assumed that the when the remaining wall thickness become 10% of the original thickness, nozzle enlargement on HCl side occurs, thus the new remaining thickness of the wall become 20% of the initial value (Fig. 11).

By changing the solid cells to fluid cells in the colored area in Fig. 11, the final shape of enlarged nozzle obtained. Figure 12 shows the result of the estimation. The width of the complete enlarged nozzle was 3.52 mm which is 17.6 times as bigger than the initial width of the nozzle (0.2 mm). The shape of the enlarged nozzle is like a conical with openings toward the NaOH side.

Another numerical calculation was carried out with the same condition to assure that the self-enlargement of the nozzle will be suppressed after the nozzle enlarged on HCl side using the complete mesh grid in Fig. 12. Fig. 13 shows the temperature contour of the calculation result. It is found that the high-temperature area does not exist on the wax anymore. Therefore, it is expected that the self-enlargement of the nozzle will be mitigated.
4.2.4 Comparison with the SWAT-2 Experimental Data

Kuroha reports that the shapes observed in the SWAT-2 test are paraboloid or conical with openings toward the sodium side [9]. Figure 14 shows the sectional morphology on complete self-wasted crack. Similarity in the shape of completely enlarged nozzle and crack is identified between the numerical analysis and the experimental data [8,9].
Fig 14. Sectional Morphology on Complete Self-wasted Crack

Experimental data about the ratio of the diameter of the enlarged crack and the initial crack has ranged from 3.30 to 34.52. In this study, the ratio of the enlarged nozzle width and the initial width was 17.6. It is hard to say that the process of simulant experiment and that of a real experiment are exactly same, but from the ratio of the enlarged nozzle width and the initial width, it can be expected that the simulant experiment shows similarity with the SWAT-2 experimental data.

5. CONCLUSIONS

The simulant experimental methodology of leak enlargement by the Self-wastage phenomena was proposed. In the experiment, HCl, NaOH aqueous solution and the paraffin wax were used as a simulant. The reaction heat released by the neutralization reaction with HCl and NaOH is used to reproduce the self-wastage phenomena. By injecting the HCl solution through a narrow wax nozzle, making the neutralization reaction near the outside of the nozzle. As a result of the release of the heat, the paraffin wax in the reaction zone would be melted, and the enlargement of the nozzle takes place.

To evaluate the feasibility of this methodology a computational analysis procedure was devised and a preliminary numerical calculation was carried out. The enlargement of wax nozzle was estimated by using the temperature contour in the wax around the outside of the nozzle. Through the series of calculations, it was proved that the propagation of the nozzle enlargement takes place due to the reaction heat of the neutralization and the enlargement advances toward HCl side. It was assumed that when remained wall thickness becomes 10 % of the initial thickness, the enlargement was terminated. The shape of the complete enlarged nozzle was a conical pit, and this shows a similarity with the cross section of the complete enlarged crack of SWAT-2 Experiment. The ratio of the width of the enlarged nozzle and that of the initial nozzle was 17.6, and it also has a similar tendency with the real ratio that is from 3.30 to 34.52. These results assure the validity of the simulant experiment to reproduce the self-wastage phenomena in sodium fast breeder reactor.

The experimental condition was determined through this study. HCl solution was injected with 0.5 m/sec velocity through the wax nozzle (width = 0.2 mm), and the flow has dynamic similarity with the flow in the real experiment (No. 2020) of SWAT-2 experiment. Also, it was proved that there is a possibility to evaluate the effect of parameters such as injection rate, nozzle geometry in simulant experiment.
REFERENCES


