Dissolution Ratio of Hydrogen Generated by Small-Leak Sodium-Water Reaction

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A study, both theoretical and experimental, was undertaken on the hydrogen dissolution ratio in liquid sodium, with the view of estimating the quantity of hydrogen that dissolves into sodium before reaching the free surface, following the occurrence of sodium-water reaction in a steam generator. Superheated steam was injected into the sodium in a reaction vessel of configuration simulating the steam generator. The hydrogen dissolution ratio was determined from analysis of the responses obtained on a hydrogen detector installed downstream of the reaction vessel.

The hydrogen dissolution ratio was found to vary from 5 to 15% over a leak rate ranging of 3~10 g/sec, as determined both from the transient response data given by the hydrogen detector and from the corresponding data obtained from tests on changes in hydrogen concentration upon complete mixture with the entire sodium inventory.

A theoretical model was assumed for estimating the hydrogen dissolution ratio, which yield results in good agreement with the experimental data, in respect of the rising tendency of the dissolution ratio with decreasing leak rate and with rising sodium temperature.

KEYWORDS: sodium, hydrogen, chemical reactions, dissolution ratio, leak detection, steam generators, hydrogen detectors, transient response, bubbles, mass transfer, solubility

I. INTRODUCTION

Rapid detection of water leakage is of critical importance for the sodium-heated steam generator of an LMFBR, in view of the severe wastage that would be caused on the adjacent heat transfer tubing by such leakage. It is widely recognized that for water-leakage detection, the most reliable instrument is the hydrogen detector, which measures the hydrogen produced by sodium-water reaction.

The two forms of hydrogen detectors in current use are those known as “in-sodium” and “in-cover gas” types. With both types, it is essential to know the quantity of hydrogen dissolving into the sodium, to permit the detector responses to be meaningfully related to all amounts of water leakage. For instance, if all the generated hydrogen dissolved into the sodium, the in-cover gas detector would be totally insensitive to water leakage.

Greene assumed the following four models to predict the order of magnitude of the ratio of hydrogen dissolution in sodium-water reaction\[^{15}\].

1. Total release model: All of the hydrogen is released into the cover gas space, upon its generation by the reaction

\[ 2\text{Na} + \text{H}_2\text{O} \rightarrow \text{Na}_2\text{O} + \text{H}_2. \]

2. Hydroxide model: The hydrogen gas is released to the cover gas space, upon its generation by the reaction

\[ 2\text{Na} + \text{H}_2\text{O} \rightarrow \text{Na}_2\text{O} + \text{H}_2. \]

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Na + H₂O → NaOH + 1/2H₂.

(3) Large-scale reaction test model: A fraction amounting to 60% of the hydrogen arising from the leak enters the cover gas space, according to large-scale tests conducted by U.K. AEA (2).

(4) Equilibrium pressure model (1): The quantity of hydrogen that enters the cover gas space varies from 0 to 1% of the maximum quantity available from the water, depending on the partial pressure of the hydrogen present in the cover gas.

Thus the ratio of hydrogen dissolution would vary from 0 to 100%, depending on the model considered.

Subbotin (3) has observed that the quantity of hydrogen entering the cover gas space was affected by the sodium flow rate, sodium temperature, pressure and bubble diameter. No quantitative comparison was, however, given between the model and experimental data.

Whittingham (4) has derived an empirical value for the dissolution rate constant of hydrogen gas into sodium, from which he calculated the duration of residence of a hydrogen bubble in sodium after sodium-water reaction.

Neither of the two authors cited above, however, presented any experimental data or theoretical model for estimating the quantity of hydrogen dissolving into the sodium or entering the cover gas space.

The present author conducted a series of experiments on sodium-water reaction, and measured the hydrogen dissolution ratio by means of a hydrogen detector. The theoretical model derived therefrom will be presented in this paper, together with experimental results.

II. EXPERIMENTAL

1. Test Apparatus and Procedure

The small-leak sodium-water reaction test loop SWAT-2 was used to conduct the experiment. This loop was made for the purpose of developing leak detectors and for performing wastage tests. It consists of a reaction vessel (40 cm I.D. x 230 cm height), main circulating system, cold trap line, in-sodium and cover gas hydrogen detectors, and also systems for reaction product release and steam injection. A flow diagram is shown in Fig. 1. Sodium is circulated through 2 in. diameter piping and enters from above the inner shell of the reaction vessel. The bottom of the inner shell connects with a flow shroud forming a passage downward for the sodium, and into which superheated steam is injected through a nozzle into the sodium at a point 780 mm below the free liquid surface in the reaction vessel. The nozzle opening diameter can be varied from 0.3 to 0.7 mm by changing the nozzle. The quantity of injected steam is calculated from the measured pressures in the steam reservoir before and after the injection.

![Fig. 1 SWAT-2 flow diagram and reaction vessel cross section](image)

The cross-sectional area of the sodium flow in the vicinity of the injection nozzle is 0.014 m². While not shown in Fig. 1, the injection nozzle is surrounded by a circle of 14 dummy tubes 26.5 mm O.D., lying parallel to the nozzle stem and normal to the sodium flow. These tubes and the flow shroud guiding the sodium stream are arranged to simulate the configuration of an LMFBR steam generator.

An in-sodium hydrogen detector was specially developed for this experiment (5). The detector is shown in Fig. 2. The hydrogen-
diffusing membrane is made of a nickel tube 0.5 mm thick and 200 mm long. The hydrogen concentration in sodium was determined from the reading of a high-vacuum gauge (B-A gauge) or else from the Noble pump ion current, using the relation given by Sieverts' law holding between the hydrogen concentration and the equilibrium hydrogen pressure. The hydrogen concentrations calculated from the detector reading were compared with the corresponding values determined from the cold trap exit temperature, assuming that the hydrogen was saturated in the sodium. Reasonable agreement was obtained.

Two different series of tests were performed for determining the hydrogen dissolution ratio. In the first series, the hydrogen concentration was measured after complete mixture of the injected steam with all the sodium present in the loop. This method is relatively simple, but the measured value may be affected by hydrogen transfer through the free surface into the cover gas in the reaction vessel while the loop sodium is being mixed.

The second series, conducted subsequently, consisted in analyzing the hydrogen detector signal immediately after the steam injection. While this method effectively obviates the above-mentioned drawback of the first method, it involves analysis of transient responses, for which account must be taken of differences in the reaction vessel flow pattern and of the diffusion behavior of hydrogen through the nickel membrane.

In both series of tests, superheated steam was injected into the sodium. The steam temperature was assumed to be equal to that of the sodium. The principal test conditions and the range adopted for the parameters are listed in Table 1.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sodium temperature</td>
<td>330~480°C</td>
</tr>
<tr>
<td>Sodium flow velocity</td>
<td>0.24 m/sec, at nozzle</td>
</tr>
<tr>
<td>Cover gas pressure</td>
<td>0~0.1 kg/cm²·g</td>
</tr>
<tr>
<td>Initial hydrogen concentration</td>
<td>&lt;0.1 ppm</td>
</tr>
<tr>
<td>Length of channel between injection point and free surface of liquid sodium</td>
<td>0.78 m</td>
</tr>
<tr>
<td>Sodium flow rate</td>
<td>200 l/min</td>
</tr>
<tr>
<td>Total sodium inventory</td>
<td>280 l</td>
</tr>
</tbody>
</table>

2. Results from Series I Tests

Figure 3 shows a typical example of hydrogen detector response after steam injection. The duration of injection was 20 sec, during which period the injection valve remained open. The hydrogen concentration is seen to have risen within 50 sec after the injection. Then the concentration dropped slightly when the portion of sodium that received the steam injection left the detector circuit, but increased again upon reentry into the detector of hydrogen-rich sodium after circulating through the loop.

The ratio of hydrogen dissolution is here defined to be the fraction of hydrogen that had dissolved in the sodium in reference to the total quantity available from the water.
The test results for Series I are summarized in Table 2 and Fig. 4. The horizontal axis represents the total quantity of hydrogen injected, and the vertical axis that of the dissolved hydrogen calculated from the hydrogen detector indication (corresponding to the value in Fig. 3 for the region beyond 300 sec) and the total sodium inventory. The hydrogen dissolution ratio $\phi_e$ can be determined from the slope formed by the plots:

$$\phi_e = \frac{9C_eQ_T}{W},$$

where $C_e$ is the equivalent hydrogen concentration, $Q_T$ the total sodium inventory, and $W$ the weight of injected steam. Almost all the plots lie between the two lines drawn in Fig. 4, representing dissolution ratios of 0.05 and 0.15. The values of $\phi_e$ are seen to be relatively small at the lower sodium temperatures.

### 3. Results from Series II Tests

The results obtained from the Series II tests are presented in Table 3. Here, $\beta$ means a transient dissolution ratio calculated from the initial response of the hydrogen detector after steam injection.

### Table 2 Series I test results

<table>
<thead>
<tr>
<th>No.</th>
<th>Steam leak rate (g/sec)</th>
<th>Injection duration (sec)</th>
<th>Injected steam weight (g)</th>
<th>Temperature (°C)</th>
<th>Concentration $C_e$ (ppm)</th>
<th>Dissolution ratio ($\phi_e$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>5.04</td>
<td>11.7</td>
<td>59.0</td>
<td>480</td>
<td>2.30</td>
<td>0.0823</td>
</tr>
<tr>
<td>2</td>
<td>3.21</td>
<td>9.0</td>
<td>28.9</td>
<td>480</td>
<td>1.96</td>
<td>0.143</td>
</tr>
<tr>
<td>3</td>
<td>3.73</td>
<td>9.6</td>
<td>35.8</td>
<td>480</td>
<td>1.11</td>
<td>0.0653</td>
</tr>
<tr>
<td>4</td>
<td>4.30</td>
<td>8.7</td>
<td>37.4</td>
<td>380</td>
<td>0.64</td>
<td>0.0361</td>
</tr>
<tr>
<td>5</td>
<td>2.53</td>
<td>19.5</td>
<td>49.3</td>
<td>290</td>
<td>1.32</td>
<td>0.0566</td>
</tr>
<tr>
<td>6</td>
<td>4.12</td>
<td>11.4</td>
<td>46.95</td>
<td>480</td>
<td>2.13</td>
<td>0.0958</td>
</tr>
<tr>
<td>7</td>
<td>1.25</td>
<td>14.7</td>
<td>18.4</td>
<td>280</td>
<td>0.511</td>
<td>0.0587</td>
</tr>
<tr>
<td>8</td>
<td>1.64</td>
<td>53.0</td>
<td>64.4</td>
<td>480</td>
<td>4.77</td>
<td>0.157</td>
</tr>
<tr>
<td>9</td>
<td>8.33</td>
<td>6.9</td>
<td>57.8</td>
<td>480</td>
<td>1.57</td>
<td>0.0576</td>
</tr>
<tr>
<td>10</td>
<td>2.41</td>
<td>8.8</td>
<td>21.2</td>
<td>480</td>
<td>0.915</td>
<td>0.0915</td>
</tr>
<tr>
<td>11</td>
<td>4.33</td>
<td>12.0</td>
<td>52.0</td>
<td>480</td>
<td>3.49</td>
<td>0.142</td>
</tr>
</tbody>
</table>

### Table 3 Series II test results

<table>
<thead>
<tr>
<th>No.</th>
<th>Steam leak rate (g/sec)</th>
<th>Injection duration (sec)</th>
<th>Injected steam weight (g)</th>
<th>Temperature (°C)</th>
<th>Dissolution ratio ($\beta$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>10.97</td>
<td>4.0</td>
<td>43.86</td>
<td>330</td>
<td>0.023</td>
</tr>
<tr>
<td>2</td>
<td>8.38</td>
<td>6.9</td>
<td>57.78</td>
<td>480</td>
<td>0.06</td>
</tr>
<tr>
<td>3</td>
<td>6.40</td>
<td>9.0</td>
<td>57.82</td>
<td>480</td>
<td>0.05</td>
</tr>
<tr>
<td>4</td>
<td>6.17</td>
<td>4.0</td>
<td>24.67</td>
<td>357</td>
<td>0.05</td>
</tr>
<tr>
<td>5</td>
<td>4.49</td>
<td>4.0</td>
<td>17.96</td>
<td>480</td>
<td>0.10</td>
</tr>
<tr>
<td>6</td>
<td>4.12</td>
<td>11.4</td>
<td>46.95</td>
<td>480</td>
<td>0.05</td>
</tr>
<tr>
<td>7</td>
<td>1.96</td>
<td>61.1</td>
<td>119.78</td>
<td>380</td>
<td>0.056</td>
</tr>
<tr>
<td>8</td>
<td>1.64</td>
<td>53.0</td>
<td>64.40</td>
<td>480</td>
<td>0.30</td>
</tr>
<tr>
<td>9</td>
<td>0.365</td>
<td>173.7</td>
<td>63.33</td>
<td>480</td>
<td>1.0</td>
</tr>
<tr>
<td>10</td>
<td>0.167</td>
<td>500.1</td>
<td>83.64</td>
<td>480</td>
<td>0.55</td>
</tr>
<tr>
<td>11</td>
<td>0.038</td>
<td>600.0</td>
<td>23.3</td>
<td>480</td>
<td>0.58</td>
</tr>
</tbody>
</table>
In these tests, the equivalent hydrogen concentration $C_e$ could not be determined, on account of unstable indications of the hydrogen detector often observed after lapse of some time after a steam injection. The instability is probably due to the effect of reaction product deposited in the cover gas space in the reaction vessel during the repeated sodium-water reaction tests. The reaction product is thought to have affected the hydrogen transfer through the sodium surface in the reaction vessel to or from the cover gas. This effect appears to have been relatively small in the case of the Series I tests which were conducted while SWAT-2 was still new and uncontaminated.

The transient hydrogen dissolution ratio $\beta$ was determined from the formula

$$QC_i = QC_i + \beta \frac{1}{9} G,$$

where $C_i$ and $C_e$ represent the hydrogen concentrations (weight fractions) in the upper part of the reaction vessel (inlet concentration) and in the lower part of the reaction zone, while $G$ and $Q$ are the rates of steam leakage and of sodium flow, respectively.

Upon issuing from the bottom of the flow shroud, the sodium enters a stagnant zone at the lower end of the reaction vessel. A parametric survey was conducted\(^{(14)}\) on the mixing effect in this zone, taking into account the effect of diffusion and turbulence present in the sodium flow\(^{(6)}\). The survey proved that no mixing took place such as to influence the final value obtained for the dissolution ratio. In other words, the sodium could validly be considered to traverse this zone in slug flow with negligible mixing.

After leaving the reaction vessel, the sodium was assumed to reach the hydrogen detector nickel membrane without changing the hydrogen concentration which was computed from the hydrogen detector indication by using the equations given in APPENDIX.

Finally, from the values of hydrogen concentration thus evaluated, the transient hydrogen dissolution ratio $\beta$ was determined as being that which best fitted the experimental data, as shown in Fig. 5. In the present instance, the value of $\beta = 0.05$ was adopted. The values of $\beta$ determined in this manner are plotted in Fig. 6 as function of the leak rate. For 480°C sodium temperature, $\beta$ decreases from 0.1 to 0.05 with an increase in leak rate from 3 to 10 g/sec; $\beta$ also decreases with lowering sodium temperature. The foregoing values of dissolution ratio coincide quite closely with those obtained in the Series I tests. The curves shown in Fig. 6 are theoretical values which will be discussed later.

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Fig. 5 Measured and calculated hydrogen detector responses

Fig. 6 Effect on transient hydrogen dissolution ratio brought by differences in leak rate and sodium temperature
III. THEORETICAL CONSIDERATIONS

The following assumptions were adopted for theoretically examining the experimental results obtained above.

1. The steam injected into the sodium converts to hydrogen gas, obeying the reaction equation

\[ 2\text{Na} + \text{H}_2\text{O} \rightarrow \text{Na}_2\text{O} + \text{H}_2. \]

2. The hydrogen gas instantly transforms into bubbles, all having the same radius \( r_0 \). The number of bubbles generated per second is denoted by \( n \).

3. The bubbles rise upward in sodium with a constant rise velocity \( U_B \) (cm/sec).

4. The hydrogen bubbles dissolve into sodium at a rate determined by the diffusion in liquid\(^{(b)}\).

5. The hydrogen bubble temperature is equal to that of the surrounding sodium.

6. Sodium in contact with the hydrogen bubbles is saturated with hydrogen.

The hydrogen gas generated by sodium-water reaction can be expressed by

\[ 4\frac{\pi}{3}nr_0^3\gamma_s = \frac{1}{9}G, \tag{3} \]

where \( \gamma_s \) is the specific weight (g/cm\(^3\)) of the hydrogen gas. The mass transfer of hydrogen through a bubble surface can be expressed by

\[ \frac{dm}{dt} = k\gamma_t(C_s - C)4\pi r^3, \tag{4} \]

where \( m \) is the weight of hydrogen dissolved in sodium, \( C \) the hydrogen concentration in sodium at some distance from the bubble, \( C_s \) the hydrogen concentration when saturated in sodium, \( \gamma_t \) the specific weight of sodium (g/cm\(^3\)), and \( k \) the mass transfer coefficient (cm/sec) for the liquid side of the bubble. The hydrogen mass balance between the sodium passing two planes separated by an infinitely small distance \( dx \), as shown in Fig. 7, is expressed by

\[ Q(C + \frac{dC}{dx} dx) - QC + \frac{dm}{dt} n \frac{dx}{U_B} = 0. \tag{5} \]

The first term on the left-hand side represents the balance between the quantity of dissolved hydrogen entering the upstream plane and that leaving the downstream plane with the sodium flowing at a rate \( Q \), while the last term gives the aggregate hydrogen mass transfer from the bubbles to the sodium during the travel of sodium through the distance \( dx \). Substituting Eq. (4) into (5), we have

\[ Q\frac{dC}{dx} dx + k\gamma_t(C_s - C)\pi 4r^3\frac{dx}{U_B} = 0. \tag{6} \]

We now consider a state of constant leakage, and assume that bubble dissolution does not affect the surrounding hydrogen concentration. In such a case, the relation between hydrogen mass transfer and the change in bubble radius can be expressed by

\[ \frac{dm}{dt} = -\frac{d}{dt} \left( \frac{4}{3}\pi \gamma_s r^3 \right) = -\frac{d}{dr} \left( \frac{4}{3}\pi \gamma_s r^3 \right) \frac{dr}{dx} \frac{dx}{dt}. \tag{7} \]

Here, \( dx/dt \) can be considered equal to the velocity \( U_B \) of bubble rise:

\[ \frac{dx}{dt} = U_B. \tag{8} \]

Substituting Eqs. (4) and (8) into Eq. (7), we have, after some rearrangement,

\[ \frac{dr}{dx} = \frac{k\gamma_t(C_s - C)}{\gamma_s U_B}. \tag{9} \]

We here consider \( C \) to be nearly constant during the steam injection and to be small compared with \( C_s \), since the cold trap will be operating to lower the hydrogen concentration in the period preceding steam injection. The foregoing assumption results in the right-hand side of Eq. (9) becoming constant during the bubble rise. Hence, integrating
where $\alpha$ is defined by
\begin{equation}
\alpha = \frac{k_0(C_s - C)}{\gamma_s U_B}.
\end{equation}

The hydrogen dissolution ratio $\phi$ is, as given earlier, the fraction of the generated hydrogen that dissolves into the sodium before reaching the free surface. We obtain
\begin{equation}
\phi = -\int_{r_0}^{r_s} 4\pi r^2 \gamma_s dr = \left(1 - \frac{r_s}{r_0}\right)^3 - 1 - \left(1 - \frac{aL}{r_0}\right)^3,
\end{equation}
where $L$ is the leak point-to-free surface depth (cm), and $r$ the bubble radius at the free surface.

Next, we consider large initial dissolution of the hydrogen in the immediate vicinity of the leak point, which would correspond to the transient hydrogen detector response. Substituting Eq. (7) into the second term on the left-hand side of Eq. (5), and integrating from $x=0$ to $x^*$, we have
\begin{equation}
QC_i - QC_s + \frac{4}{3} \pi n (r_s^3 - r^{*3}) \gamma_s = 0,
\end{equation}
where $r^*$ is the bubble radius at $x=x^*$. Upon substitution of Eq. (3) into Eq. (13),
\begin{equation}
QC_0 = QC_i + \frac{1}{9} G \left(1 - \left(\frac{r^*}{r_0}\right)^3\right),
\end{equation}
and from Eq. (10),
\begin{equation}
r^* - r_0 = -\alpha x^*.
\end{equation}
Further, Eq. (15) into Eq. (14) and comparison with Eq. (2) results in
\begin{equation}
\beta = 1 - \left(\frac{r^*}{r_0}\right)^3 = 1 - \left(1 - \alpha x^*/r_0\right)^3.
\end{equation}
This relation means that the transient dissolution ratio $\beta$ is equal to the dissolution ratio for the case where the free surface depth is located at a depth of $x^*$ from the leak point.

The mass transfer coefficient is assumed to be expressible by an equation that is generally applied to a single bubble rising in liquid
\begin{equation}
k = \sqrt{\frac{2D_L U_B}{\pi r_0}},
\end{equation}
where, to simplify the discussion in this case, the bubble radius $r_0$ is considered not to change during the rise. Here, $D_L$ is the diffusion coefficient of hydrogen in sodium, for which, Wilke$^{(8)}$ has given the formula
\begin{equation}
D_L = 7.4 \times 10^{-8} \left(\frac{aM}{V_s}\right)^{1/2} \frac{T}{\mu},
\end{equation}
where $D_L$: Diffusion coefficient of solute at infinite dilution (cm$^2$/sec), $M$: Molecular weight of solvent, $T$: Absolute temperature (°K), $V_s$: Molar volume of solute at normal boiling point (cc/g-atom), $a$: Association parameter, $\mu$: Solution viscosity.

The association parameter was assumed to be 1.0 in this case$^{(9)}$, and a value of 3.7 cc/g-atom was adopted for $V_s$, assuming that the hydrogen atom moves freely in sodium.

In sodium-water reaction, it is quite difficult to directly measure the hydrogen bubble diameter. In the present instance, it was estimated from the results obtained in a previous experiment on bubble rise velocity in sodium-water reaction, which had been conducted by the present author for examining the surface oscillation caused by rising hydrogen bubbles$^{(10)}$. The bubble rise velocities thus obtained are plotted in Fig. 8 as function of the leak rate, from which
\begin{equation}
U_B = 82.2 G^{0.317}.
\end{equation}

Here, the unit of $U_B$ is (cm/sec) and $G$ (g/sec).

On the other hand, in the range of fairly large Reynolds number based on bubble diameter, the bubble rise velocity in an ordinary liquid like water, is given by the equation
\begin{equation}
U_B = 1.74 \sqrt{gd},
\end{equation}
where $d$ is the bubble diameter and $g$ the
acceleration of gravity.

Assuming Eq. (20) to be applicable to the present data for hydrogen bubbles in sodium, we obtain from Eqs. (19) and (20) the bubble diameter

\[ d = 2.28 G^{0.634}. \]  

(21)

The actual values of \( d \) become 5.3 and 22.8 mm for leak rates of 0.1 and 1.0 g/sec, respectively.

Substituting Eqs. (11), (17), (20) and (21) into Eq. (12), the hydrogen dissolution ratio \( \phi \) is given by

\[ \phi = 1 - \left\{ 1 - 2.36 \times 10^{-6} \frac{G^{3/2} V L^{1/2} (C_s - C) L^3}{I} \right\}. \]  

(22)

The value of \( \beta \) can be obtained by using \( x^* \), instead of \( L \), in Eq. (22). For sodium temperatures of 480 and 350°C this value becomes as shown in Fig. 6 as function of the leak rate. For \( x^* \), we have here adopted the distance of bubble travel through the flow shroud (\( x^* = 45 \) cm, see Fig. 1). The theoretical curves in Fig. 6 present a configuration that as a whole, follows the measured plots, although the curve for the higher sodium temperature runs at a level higher than the plots.

The relationship between \( \beta \) and \( \phi \) is derived by eliminating \( a/r_0 \) from Eqs. (12) and (16):

\[ \phi = 1 - \left\{ 1 - (1 - \beta)^{1/3} \right\} \frac{L}{x^*}. \]  

(23)

Figure 9 shows this relation, with \( L/x^* \) as parameter. The value of \( L/x^* = 1.73 \) adopted for the third curve in this figure corresponds to \( L = 78 \) cm representing the distance between the leak point and the free surface and the \( x^* = 45 \) cm cited above, which was used to obtain the transient dissolution ratio \( \beta \). Thus, this curve for \( L/x^* = 1.73 \) theoretically represents the relation between \( \phi \) from the Series I and \( \beta \) from the Series II tests. The actual values of 0.05 to 0.15 obtained for \( \phi \) in Series I, are seen from this curve to correspond to \( \beta = 0.03 \sim 0.09 \).

Use of Eq. (23), will permit estimation of the dissolution ratio for an arbitrary leak point-to-free surface depth from experimental data obtained for a given depth. It can be seen that a 10-fold increase of depth raises the dissolution ratio by 5 to 10 times.

IV. CONCLUSION

(1) The dissolution ratio of hydrogen generated by sodium-water reaction was measured by two independent methods. The ratio was found to vary from 0.05 to 0.15 over a leak rate range of 3~10 g/sec.

(2) The dissolution ratio estimated from the transient response of the hydrogen detector showed a tendency to increase with rising sodium temperature and with decreasing leak rate.

(3) A theoretical equation (Eq. (22)) was derived for the hydrogen dissolution ratio by assuming that, in the course of its rise toward the free surface, the hydrogen bubbles dissolve gradually into the sodium at a rate determined by the diffusion of hydrogen in the liquid sodium.

(4) A tendency generally similar to this theoretical equation was shown by the experimental data, in respect of the variations in hydrogen dissolution ratio brought by changes in the sodium temperature and in the leak rate.

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--- REFERENCES ---


[A P P E N D I X]

According to Mecham\textsuperscript{12}, the relation between hydrogen concentration and hydrogen partial pressure in sodium is given by

\begin{equation}
P' = bC'^{2/3}, \quad (A1)
\end{equation}

where \( P' \) is the hydrogen partial pressure (atm), \( C' \) the hydrogen concentration in sodium (ppm) and \( b \) a constant \( 6.6 \times 10^{-5} \) (atm). The hydrogen concentration \( C_H \) on the surface of a nickel membrane in contact with sodium, is given by

\begin{equation}
C_H = k_N P'^{1/2}, \quad (A2)
\end{equation}

\begin{equation}
k_N = K / D_N, \quad (A3)
\end{equation}

where \( k_N \) is the solubility coefficient, \( D_N \) the diffusion coefficient \( \text{cm}^2/\text{sec} \), and \( K \) the permeability \( \text{cm}^3\text{mm/cm}^2\text{atm}^{1/2}\text{hr} \) whose values are\textsuperscript{13}

\begin{equation}
K = 1.000 \exp(-1.34 \times 10^4 / RT), \quad (A4)
\end{equation}

\begin{equation}
D_N = 5.22 \times 10^{-3} \exp(-9.56 \times 10^4 / RT), \quad (A5)
\end{equation}

where \( R \) is the gas constant.

The diffusion equation through the nickel membrane in unsteady state can be expressed by

\begin{equation}
\frac{\partial C}{\partial t} = D_N \frac{\partial^2 C}{\partial x^2}, \quad (A6)
\end{equation}

\begin{equation}
q = D_N \left( \frac{\partial C}{\partial x} \right)_{x=l_0}, \quad (A7)
\end{equation}

where \( C \) is the hydrogen concentration in the membrane \( \text{g/cm}^2 \), and \( q \) the hydrogen flux \( \text{g/sec/cm}^2 \) diffusing through the vacuum side of a membrane of thickness \( l_0 \).

The pressure in the vacuum system of the hydrogen detector can be obtained with the formula,

\begin{equation}
V_D \left( \frac{P_e + P_D}{2dt} \right) = A Q_H - S P_D, \quad (A8)
\end{equation}

\begin{equation}
C_{eD}(P_e - P_D) = S P_D, \quad (A9)
\end{equation}

where \( V_D \) is the vacuum system volume, \( A \) the nickel membrane surface area, \( P_e \) the pressure on the vacuum side of the membrane, \( P_D \) the pressure at the detector position (a vacuum gauge or noble pump), \( S \) the pumping speed, \( C_{eD} \) the conductance between the membrane and the vacuum gauge, and \( Q_H \) the hydrogen diffusing flux converted from \( q \), considering the temperature and pressure differences between membrane and vacuum system, namely:

\begin{equation}
Q_H = 9.354 \times 10^6 q \quad (\text{cc} \cdot \text{Torr/sec} \cdot \text{cm}^2). \quad (A10)
\end{equation}

The relationship between the hydrogen concentration \( C_0 \) at reaction zone exit and the detector pressure \( P_D \) can be calculated numerically by means of the foregoing equations, and adopting for the hydrogen detector used:

\begin{equation}
V_D = 2,700 \quad (\text{cm}^3), \quad A = 100 \quad (\text{cm}^2), \quad S = 16,000 \quad (\text{cm}^3 / \text{sec}), \quad C_{eD} = 1,870 \quad (\text{cm}^2 / \text{sec}).
\end{equation}

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