Original Article

Influence of hydrogen concentration on burst parameters of Zircaloy-4 cladding tube under simulated loss-of-coolant accident

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1. Introduction

Zircaloy-4 is used as a nuclear fuel cladding in water reactors. Zirconium transforms from the hexagonal close-packed α-phase to the body centered cubic β-phase at 865 °C and atmospheric pressure. However, the alloying elements that convert zirconium to Zircaloy-4 alters the transformation temperature as well as introduce a temperature region of about 150 °C in which the α and β phases coexist [1]. Zircaloy-4 fuel cladding undergoes water-side corrosion during service and as a result hydrogen is produced [2,3].

\[
\begin{align*}
Zr + 2H_2O & \rightarrow ZrO_2 + 4H \\
4H + 2Zr & \rightarrow 2ZrH_2
\end{align*}
\]

A fraction of hydrogen diffuses into Zircaloy-4 since hydrogen is more stable as solution in the α-phase of zirconium alloy matrix than in gaseous form. Hydrogen in solution in the α-zirconium lattice occupies the tetrahedral interstices in the hexagonal α-zirconium unit cell. The hydrogen in solid solution form in the α-zirconium matrix is highly mobile at nuclear reactor operating conditions, it migrates down both the concentration and the temperature gradients while migrates up a stress gradient [4–6]. The diffused hydrogen precipitates and forms brittle zirconium hydride phase after exceeding a limiting concentration for the given conditions of temperature and hydrogen pressure. It is well established that precipitation of hydride alters the critical properties of fuel cladding like dimensional stability, mechanical strength, corrosion, and creep behaviour of the cladding [7].

During normal operation of the reactor, cladding tube is subjected to temperatures in the range of 290–350 °C and a compressive hoop stress of 40–80 MPa because of pressurised coolant overpressure. The cladding tube is also internally pressurised with helium of the order of 2 MPa to improve thermal conductivity at the fuel pellet-cladding interface, and to reduce the probability of mechanical interaction between pellet and cladding during service [8–10]. In a loss-of-coolant accident (LOCA), there is a decrease in the system pressure outside of the cladding and the heat transfer from the fuel. The decrease in the outside coolant pressure gives rise to hoop stress while decrease in the heat transfer rate causes a rapid increase in the temperature of the cladding. As a result, the creep deformation or ballooning of the fuel cladding occurs which may cause its bursting. Moreover, ballooning of the fuel cladding may result in a blockage of the coolant sub-channel that in turn may impair the fuel coolability [11].

The basic parameters controlling deformation of the cladding during the first phase of loss-of-coolant accident (LOCA) are tensile hoop stress, temperature ramp, and creep strength. Single-tube
burst test performed outside of reactor constitutes the bulk of the available open literature and it has largely served to elucidate the effects of these basic parameters on the burst behaviour of the cladding during LOCA as well as in the development of the burst criterion. Swelling or ballooning of the cladding which eventually leads to bursting during LOCA is primarily a function of creep strength which is influenced by the hydride precipitation. In addition, the presence of hydrogen—a $\beta$-phase stabiliser—also shifts the phase transformation temperature of the Zircaloy cladding. Brachet et al. [12] investigated the influence of hydrogen content on the phase transformation temperature using calorimeter and burst behaviour using single-tube burst test approach. The tests were performed on pre-hydrided Zircaloy-4 cladings wherein hydrogen concentrations ranged from 100 ppm to 1000 ppm. It was reported that $\alpha/\beta$ phase transformation temperature decreases with the increase in hydrogen content as well as hydrogen decreases both creep strength and ductility of the material, the effect being greater for higher hydrogen content. They propounded that these results are not only due to the effect of hydrogen on the $\alpha/\beta$ phase transformation shift but also by an intrinsic effect of hydrogen on the creep behaviour, especially in the $\alpha$-phase and lower $\alpha + \beta$-phase temperature ranges. Thus, they emphasised the need to assess burst criterion incorporating the role of hydride during LOCA for Zircaloy-4. Another important finding of their study was that deformation behaviour is mainly linked to the hydrogen uptake associated with the cladding oxidation, and that the irradiation defects annihilate out and play no significant role during the first phase of LOCA. Kim et al. [13] studied the individual effects of the surface oxide (20 $\mu$m and 50 $\mu$m thick) and the absorbed hydrogen (300 ppm and 1000 ppm) on the behaviour of Zircaloy-4 cladding during LOCA. Their findings about the effects of hydrogen were similar to the findings of the Brachet et al. [12]. This investigation added that oxide layer also affected the high temperature ballooning behaviour by arresting the intra-granular deformation of the $\alpha$-grain up to 700 °C. Uetsuka et al. [14] conducted integral tests of rod-burst, oxidation and thermal-shock with the objective of understanding the failure behaviour of Zircaloy-4 cladding during LOCA, especially during quenching stage. Even though the primary focus of their study was to evaluate the failure boundary of oxidation condition of the cladding during quenching stage of LOCA, it was established that hydrogen absorbed by the Zircaloy-4 cladding played a dominant role in the failure and major of the cladding failed at the location of the maximum hydrogen concentration. In their investigation, hydrogen was picked up by the cladding during the oxidation process and, thus role of hydrogen could not be identified with distinctiveness. Nagase and Fuketa [15,16] in their two investigations similar to the study of Uetsuka et al. [14] used pre-hydrided Zircaloy-4 cladding tube to understand the exclusive role of hydrogen on its failure bearing capacity during LOCA. It was concluded that both rupture temperature and circumferential burst strain decreased with an increase in the initial hydrogen concentration. Moreover, fracture threshold for the amount of oxidation was also reduced by an increase in the initial hydrogen concentration.

It is evident from the succinct literature review that there is no clear understanding about the effects of hydrogen on the deformation behaviour of Zircaloy cladding during the first phase of LOCA. There has been no investigation reported in the open literature except that of Brachet et al. [12] which emphasised the need of a new burst criterion incorporating the effects of hydrogen. In the present research work, an extensive single-clad burst test on pre-hydrided Zircaloy-4 fuel cladding, whose as-received composition is provided in Table 1, has been conducted to understand the role of hydrogen concentration on the burst behaviour during the first phase of LOCA. In addition, burst correlation for as-received Zircaloy-4 is also developed for the reference and developing distinctive understanding about the role of hydrogen on burst parameters.

2. Experimental setup and procedure

This section describes the indigenously designed and developed clad burst facility. It has the following main components as shown in Fig. 1: cladding heating setup, pressurisation setup, arrangement for maintaining inert atmosphere inside the burst chamber, temperature and pressure measuring instruments, and high-speed data acquisition system.

The burst chamber (1), a cuboidal enclosure having dimensions 1.0 m × 0.5 m × 1.0 m, is mounted on heavy mild steel frame. Its rear, top, and bottom sides are made up of 5 mm thick mild steel plate while other three sides are made up of 10 mm thick transparent acrylic plates. The heating arrangement for the clad specimen (2) at a required rate consists of 64 kVA rectifier (3) with a control unit (4) and a display unit (5). In order to avoid the development of axial stresses in clad specimen, copper clamps of the specimen are connected to copper bus bars (6) by means of flexible electric cables (7) having high current carrying capacity. An argon gas cylinder (8) is connected to the cladding specimen through a control valve for internal pressurisation. Once the clad tube is internally pressurised at the desired pressure, the valve is closed. In order to conduct burst experiment in inert atmosphere, an additional argon gas cylinder (9) is used to purge argon inside the burst enclosure through a perforated copper tube right under the cladding specimen. This ensures the presence of an inert atmosphere near the clad specimen. A vent is provided at the top of the enclosure to evacuate the air and excess argon. The temperature field of clad specimen is captured by thermal imaging camera (10). The online temperature and pressure data have been acquired with high speed data acquisition system (11). The burst video is recorded using high speed camera (12). The data is stored in the computer using LabVIEW 2017 program.

The experimental procedure involves three major activities, namely, specimen preparation, leak testing and the burst test. The sample preparation requires: (a) charging the as-received cladding specimen with desired hydrogen concentration (b) spot welding of two thermocouples at its outer surface and (c) fixing ferrule joint between the open end of clad specimen and pressurised gas connecting line. The cladding specimen is hydrogenated using the gaseous hydrogen charging facility. This gaseous charging facility, based on a modified Sievert’s apparatus, consists of a cylindrical glass chamber placed inside the furnace. The clad specimen is weighed and then placed inside the chamber. The glass chamber is evacuated to create a vacuum of the order $10^{-5}$ Pa. Subsequently, the specimen chamber is heated to the temperature of 363 °C. Depending upon the weight of the sample and target hydrogen concentration, hydrogen is released into the specimen chamber up to a pre-computed pressure. The amount of hydrogen picked up by the sample is calculated from difference between the initial and the final pressure readings recorded at ambient temperature. Two K-type thermocouples are spot welded on the outer surface of specimen before being installed in the enclosure for internal pressurisation and heating. The specimen is connected to pressurising line using brass ferrule and nut-connector assembly. Pressure inside the cladding specimen is set to a desired value by operating the regulator mounted on argon cylinder. Once the required internal pressure is achieved, the gate valve present just before the cladding-pressurising junction is shut-off which disconnects the specimen from pressurising gas line. It ensures that even a small variation in the internal pressure during heating of the cladding is recorded. The pressure transducer reading is monitored for at least of 300 s to...
identify any leakage in the test section. Another hole on the rear wall right below cladding specimen is provided for purging argon inside the chamber using another argon gas cylinder. The gas is purged into the chamber through a perforated copper tube. The perforated tube is placed at a distance below the clad specimen. The idea is to create an argon rich (inert) atmosphere around the clad specimen. Moreover, perforated tube is positioned and flow rate of argon gas is adjusted in such a way that convection near heated clad specimen remains as small as possible. The argon purging is initiated a few minutes before the test. A vent is provided on chamber’s roof to facilitate the escape of air and excess argon. Thermal imaging camera and high-speed camera are started. Rectifier is switched ON and the required magnitude of direct current is applied (to achieve a particular heating rate) to the cladding specimen by operating control unit’s knob. The temperature starts rising at a particular rate corresponding to the set current value. Cladding specimen undergoes ballooning leading to its burst as the clad attains a certain temperature. The burst video is recorded using high speed camera at 5000 frames per second. Time, pressure, and temperature at the instant of burst are termed as burst time, burst pressure and burst temperature, respectively. Burst data is acquired and stored in computer using high speed data acquisition system. Burst specimen is disassembled and high resolution image is captured, especially of the burst location for further image processing. Then after the post-burst measurements of thickness and circumference at burst location are performed. Finally, a small piece is cut from the burst site and the hydrogen concentration at the burst location is determined using the LECO RH IE inert gas fusion analyser. This procedure is repeated for each burst test.

The range of operating parameters has been provided in Table 2. The heating rate, internal overpressure, and hydrogen concentration in the Zircaloy-4 cladding specimen are varied. Two tests are conducted for the identical set of input parameters, in other words, repeatability is kept as 2.

### Results and discussion

Each cladding specimen is subjected to a given heating rate and internal pressure. Both temperature and pressure increase during

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Clad tube outer diameter</td>
<td>13.08</td>
<td>mm</td>
</tr>
<tr>
<td>Clad tube wall thickness, $S_0$</td>
<td>0.41</td>
<td>mm</td>
</tr>
<tr>
<td>Clad tube total length</td>
<td>150</td>
<td>mm</td>
</tr>
<tr>
<td>Clad tube effective heating length</td>
<td>100</td>
<td>mm</td>
</tr>
<tr>
<td>Internal overpressure, $p_0$</td>
<td>5, 20, 40, 80</td>
<td>bar</td>
</tr>
<tr>
<td>Heating rate, $\eta$</td>
<td>4–150</td>
<td>K/s</td>
</tr>
<tr>
<td>Hydrogen concentration, $H_c$</td>
<td>0–2000</td>
<td>wppm</td>
</tr>
</tbody>
</table>
heating of the cladding specimen. The internal pressure rises due to heating and then there is a slight decrease in it due to increase in volume caused by the ballooning of the cladding. Eventually cladding bursts and it is characterised by the sudden drop of internal pressure to zero. The instant at which this sudden drop in pressure occurs is taken as burst time. The temperature and the internal pressure corresponding to this time is called burst temperature ($T_B$) and burst pressure ($p_B$), respectively. The burst hoop stress, and burst hoop strain are evaluated as described in Ref. [17].

3.1. Effects of internal pressure ($p_0$) and heating rate ($\eta$)

While heating as well as during ballooning, the internal pressure of the cladding does not remain constant. Variations in pressure are non-linear given the simultaneous increase in temperature and non-symmetrical increase in volume due to ballooning. Fig. 2 shows the effects of heating rate ($\eta$) and internal pressure ($p_0$) on rate of pressure rise ($\gamma$) for as-received cladding with assumption that variations was linear. It is seen that rate of pressure increase is higher for high internal pressure and high heating rate.

For low internal pressure or hoop stress, the burst happens at higher temperature corresponding to $\beta$-phase. With increase in internal pressure, the burst temperature shifts towards lower temperatures corresponding to $\alpha$-phase of the Zircaloy-4 cladding as evident from post-burst pictures A and C in Fig. 3. The burst temperature decreased to 936 K from 1177 K on increasing the initial internal pressure from 2 MPa to 8 MPa. Fig. 4 shows the relationship of burst stress with burst temperature for both as-received and hydrogenated claddings at a heating rate of 5–10 K/s. The bursts stress is significantly lower for cladding hydrogenated with 450–600 wppm compared to as-received cladding in $\alpha$-phase. This reduction in failure bearing stress is exhibited because of hydrogen in the cladding and this reduction in burst stress is larger for higher hydrogen contents. However, as burst temperature shifts to mixed $\alpha + \beta$-phase regime the effects of hydride reduce and in $\beta$-phase the burst stress is similar to as-received cladding or higher than as-received cladding. Similar observation of higher burst strength for hydrogenated cladding at higher burst temperatures was also reported earlier by Brachet et al. [8].

Burst strain for as-received cladding attains a maxima in $\alpha$-phase and dip sharply in mixed $\alpha + \beta$-phase regime given change in material property due to phase transformation. For low internal pressure when burst temperatures start shifting towards higher temperatures, another peak in burst strain is observed, as shown in Fig. 5. The burst strain for Zircaloy-4 cladding hydrogenated up to 600 wppm, there is remarkable decrease in burst strain. This loss in ductility is observed in $\alpha$-phase and lower $\alpha + \beta$-phase which eliminated the first maxima peak observed for as-received cladding. This reduction in ductility is more pronounced for higher hydrogen content. The burst strain decreased by 27% for 600 wppm while it was decreased by 63% for 2000 wppm compared to as-received cladding. For 2000 wppm, even the second maxima peak is eliminated and loss of ductility is observed up to the end of $\alpha$-phase to $\beta$-phase transformation. In $\beta$-phase, the hydrogenated cladding shows more ductility compared to as-received cladding. At higher temperatures corresponding to $\beta$-phase, the more concentration of hydrogen is in solid solution form rather than precipitated hydride. This hydrogen enhanced ductility may be attributed to the effects of hydrogen atmospheres and not of precipitated hydride which reduces ductility like in $\alpha$-phase. Hydrogen atmospheres reduces the elastic interactions between dislocations and other internal stress fields by a mechanism known as elastic shielding and soften the material [16].

3.2. Rupture area

The burst opening area is evaluated using a java based open source image processing program named ImageJ. The high resolution image of cladding post-burst is taken against a standard environment [10,17,19–21] and it is given as:

$$
\sigma_B = a \exp (-b T_B)
$$

where $a$ and $b$ are the experimentally determined material-dependent parameters. $\sigma_B$ and $T_B$ are burst stress and burst temperature respectively. Nevertheless, these experimentally determined parameters may be influenced by other operating variables such as the heating rate or test atmosphere like inert, air or steam [21].
Dependence of burst stress on burst temperature for as-received cladding obtained during the present investigation as well as from study conducted on Zircaloy-4 having similar chemical composition [17,19] is shown in Fig. 8. The value of material dependent constants \(a'\) and \(b'\) are evaluated using the regression analyses on the burst data. The values of these constants are phase dependent and thus have different values in \(\alpha\)-phase, \(\alpha + \beta\)-phase, and \(\beta\)-phase. For hydrogenated cladding tubes, the burst stress is found to be dependent not only on burst temperature but also hydrogen concentration. Thus, a new correlation incorporating the effect of hydrogen concentration, in accordance with correlation given by Erbacher et al. [20] for oxidized Zircaloy-4 cladding, is proposed as:

![Fig. 3. Visual examination of the Zircaloy-4 cladding tubes after burst test at different conditions.](image)

![Fig. 4. Effects of hydrogen on burst stress of Zircaloy-4 cladding.](image)

![Fig. 5. Effects of hydrogen on burst strain of Zircaloy-4 cladding.](image)
\[
\sigma_B = a \cdot \exp(-b T_B) \cdot \exp(-c H_c)
\]

where \(c\) is hydrogen influenced material parameters and \(H_c\) is hydrogen concentration in the cladding. The burst stress obtained during this study is plotted against burst temperature in Fig. 9 and the material dependent parameters is again evaluated using regression analyses. The burst correlation curves corresponding to limiting range of hydrogen concentration, that is 0 ppm and 2000 ppm, are also drawn in Fig. 9. Majority of data lies within these curves attesting the accuracy of the proposed correlation. There is too less data in mixed \(\alpha + \beta\)-phase for hydrogenated cladding to form any conclusive understanding. However, the burst characteristics in \(\alpha\)-phase and \(\beta\)-phase may be interpolated to form some understanding about rupture behaviour of hydrogenated Zircaloy-4 cladding in mixed \(\alpha + \beta\)-phase. Another noteworthy remark regarding the developed burst correlation for hydrogenated cladding is that phase transformation of cladding influenced by hydrogen concentration is not considered and this may also influence the burst parameters to a certain extent.

The burst correlations obtained for Zircaloy-4 cladding in the present research is provided in Table 3. Burst correlation obtained by Erbacher et al. [20] based on oxygen concentration in Zircaloy-4 cladding has also been included in Table 3 for the sake of comparison and completeness since correlation for hydrogenated cladding was proposed in accordance with this.

### 4. Conclusions

Single-clad burst tests are performed on both as-received and hydrogenated Zircaloy-4 fuel cladding under postulated first phase of loss-of-coolant accident conditions. The as-received cladding is hydrogenated using gaseous hydrogen charging method and the concentration of hydrogen in the Zircaloy-4 cladding specimens is in the range of 10 wppm to 2000 wppm. The heating rate is varied from 4 K/s to 150 K/s while initial internal overpressure is in the range of 5 bar–80 bar. Following conclusions are drawn from the present burst investigation on both as-received and pre-hydrided Zircaloy-4 cladding specimens:

- For as-received cladding specimens, burst strain is higher for the burst temperature falling in \(\alpha\)-phase region, attains minima in \(\alpha + \beta\)-phase, and again increases in \(\beta\)-phase. The phase transformation of Zircaloy-4, from an anisotropic hexagonal close-packed \(\alpha\)-phase to an isotropic body centered cubic \(\beta\)-phase, may be attributed for this behaviour.
- Hydrogenated Zircaloy-4 cladding exhibits reduced ductility in the \(\alpha\)-phase region, the effect being greater for high hydrogen concentration.
The increase of hydrogen content eliminates the minima of burst strain observed in the mixed $\alpha + \beta$-phase for as-received claddings.

- Influence of hydrogen concentration is significantly reduced in $\beta$-phase, and the burst strain value is comparable to that of as-received claddings.

- Burst opening area is strongly dependent on the initial stress irrespective of the hydrogen concentration in the Zircaloy-4 cladding. For higher internal pressure, the burst area is larger for hydrogenated cladding.

- Burst stress is sensitive to hydrogen concentration and a new burst correlation incorporating hydrogen concentration is proposed.

### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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### Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.net.2020.02.009.

### References


