

ZIRCALOY-OXIDATION AND HYDROGEN-GENERATION RATES
IN DEGRADED-CORE ACCIDENT SITUATIONS*

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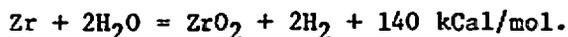
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ABSTRACT

Oxidation of Zircaloy cladding is the primary source of hydrogen generated during a degraded-core accident. In this paper, reported Zircaloy oxidation rates, either measured at 1500-1850°C or extrapolated from the low-temperature data obtained at <1500°C, are critically reviewed with respect to their applicability to a degraded-core accident situation in which the high-temperature fuel cladding is likely to be exposed to and oxidized in mixtures of hydrogen and depleted steam, rather than in an unlimited flux of pure steam. New results of Zircaloy oxidation measurements in various mixtures of hydrogen and steam are reported for >1500°C. The results show significantly smaller oxidation and, hence, hydrogen-generation rates in the mixture, compared with those obtained in pure steam. It is also shown that a significant fraction of hydrogen, generated as a result of Zircaloy oxidation, is dissolved in the cladding material itself, which prevents that portion of hydrogen from reaching the containment building space. Implications of these findings are discussed in relation to a more realistic method of quantifying the hydrogen source term for a degraded-core accident analysis.

INTRODUCTION

The rate of high-temperature Zircaloy oxidation during a severely undercooled degraded-core accident in a light-water reactor has a major influence on core heat-up, related damage, and hydrogen generation. Oxidation of Zircaloy fuel cladding by steam molecules (contained in the reactor coolant) is the predominant source of hydrogen generation, in contrast to the reactions of water with other core materials, i.e., stainless steel, Inconel, UO₂, and control rod materials. Two moles of hydrogen are generated for each mole of zirconium oxidized, as shown in the equation



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Kinetics of the reaction at $\sim 1500^{\circ}\text{C}$ have been extensively investigated under conditions of sufficient supply of pure steam.¹⁻¹² The sufficient steam supply in those investigations means that the incident rate of steam molecules per unit area of zirconium-oxide surface is such that the overall kinetics is controlled only by the oxygen diffusion in Zircaloy material. Under such a condition, the oxidation kinetics are parabolic and the rate constant is a function of Zircaloy temperature only. The uncertainty limit of the reported rate constants for $\sim 1500^{\circ}\text{C}$ is fairly significant, and is usually attributed to uncertainties in Zircaloy specimen temperature⁷ or temperature gradient across the specimen thickness.¹²

Under a degraded-core accident situation, kinetics for $>1500^{\circ}\text{C}$ are relatively more important, because most oxidation and hydrogen generation takes place in the high-temperature region. However, the literature regarding rate constants for $>1500^{\circ}\text{C}$ is relatively limited.^{1-4,10} A number of aspects must be carefully examined with respect to the reported rate constants for $>1500^{\circ}\text{C}$. For the high-temperature region, uncertainty associated with inaccurate temperature measurement is likely to be inherently large. Rate constants measured at $>1580^{\circ}\text{C}$ may be influenced by the tetragonal-to-cubic transformation of the oxide, as originally suggested by Klepfer⁴ and later shown by Urbanic and Heidrick.¹⁰ Another important consideration is that, under a degraded-core accident situation, Zircaloy fuel cladding at $>1500^{\circ}\text{C}$ is likely to be exposed to and oxidized in mixtures of hydrogen and depleted steam, rather than in an unlimited flux of pure steam. Examples of lesser oxidation in a hydrogen-steam mixture, compared with the case of pure steam, have been reported by Westerman¹³ for 750°C , by Furuta and Kawaskai¹⁴ for $1000-1100^{\circ}\text{C}$, and by the authors¹⁵ for 1375 and 1500°C . Attention has been called to two possible mechanisms for the oxidation-rate limitation when the hydrogen fraction in the coolant mixture is high: (1) chemical reduction of adsorbed oxygen on the zirconium oxide surface by gaseous hydrogen, and (2) retardation of the rate of oxygen transport in oxide that may contain a significant amount of dissolved hydrogen.¹⁵ In view of these complexities associated with the kinetics at $>1500^{\circ}\text{C}$, it is necessary to examine critically the oxidation rates reported for the high-temperature range, with emphasis on their applicability to a degraded-core accident.

In analyzing hydrogen behavior during a degraded-core accident, Zircaloy fuel cladding is usually considered only as a source of hydrogen production through oxidation with steam. However, it can also act as a hydrogen sink, because hydrogen is highly soluble in β -phase Zircaloy.¹⁶ Hydrogen absorption by Zircaloy cladding under accident situations can take place via two routes, i.e., from the outer surface of the cladding, by penetrating the growing oxide layer, and from the inner surface of the ballooned and ruptured cladding. The oxide barrier in the latter process is negligible, except in the limited area near the rupture opening. Information on the hydrogen absorption by deformed and ruptured cladding during an accident situation is limited.^{16,17} In this paper, available data on the subject are analyzed, to provide a better method of quantifying the hydrogen release term. New results on hydrogen absorption by Zircaloy-4 tubes oxidized in hydrogen-steam mixtures are also shown and discussed relative to similar data obtained in pure steam environments.

OXIDATION RATES MEASURED IN STEAM

There have been reported indications that the oxidation rates determined for $\gtrsim 1600^{\circ}\text{C}$ are significantly higher than the values extrapolated from the data obtained at lower temperatures.⁴ Bittel and Sjordahl⁴ suggested that the higher rates they observed for $\gtrsim 1600^{\circ}\text{C}$ could be explained on the basis of tetragonal-to-cubic transformation of the zirconium oxide phase. However, the early day oxidation rates¹⁻⁴ apparently reflect significant uncertainties associated with temperature measurements and limited data base. Later works⁵⁻¹² (prior to the Three Mile Island reactor accident) were directed primarily to rate constant measurements for $\gtrsim 1500^{\circ}\text{C}$. The more extensive data base obtained from those later works clearly indicated that the high-temperature rates obtained in the early days are not consistent with values extrapolated from the data for lower temperatures. Thus, in contrast to the single Arrhenius-type rate equation reported by Baker and Just,³ Urbanic and Heidrick¹⁰ represented rate constants by two equations, one for $1580\text{--}1850^{\circ}\text{C}$ and the other for $1000\text{--}1580^{\circ}\text{C}$.

Urbanic and Heidrick showed that the higher rates at $\gtrsim 1580^{\circ}\text{C}$ are associated with the cubic crystal form of the oxide that is stable at the high-temperature range. The transport rate of oxygen ion is therefore, expected to be higher in the cubic oxide than in the tetragonal form. In this regard, the discontinuity at $\sim 1580^{\circ}\text{C}$ can be best shown by oxide layer growth rate constants, rather than by rate of total weight gain. Figure 1 shows oxide layer growth rate constants (plotted as a function of inverse temperature) that were determined recently for $1200\text{--}1705^{\circ}\text{C}$ in unlimited flux of pure steam. The details of experimental procedures and oxide layer thickness versus isothermal-oxidation-time plots have been reported elsewhere.¹⁸ For comparison, the Arrhenius equations for rate constants reported by Urbanic and Heidrick¹⁰ and Cathcart and Pawel⁷ are plotted in the same figure. Although the present rate constants are somewhat smaller than the ones reported by Urbanic and Heidrick, the results of Fig. 1 confirm their finding that there is a discontinuous jump of oxidation rates at $\sim 1580^{\circ}\text{C}$. It appears that the rate constant determined by Urbanic and Heidrick was obtained from single oxidation test for each temperature, in which specimen weight gain was continuously recorded as a function of time.¹⁰ Thus, an oxide layer growth rate constant for a given temperature may have been determined from the slope of the line connecting the origin and the one data point obtained for the temperature, i.e., the oxide layer thickness measured for the single oxidation test.

OXIDATION RATES MEASURED IN HYDROGEN-STEAM MIXTURES

In a prolonged core uncover, exemplified by events during the Three Mile Island reactor accident, a large amount of hydrogen can be generated and can blanket the high-temperature ($\gtrsim 1500^{\circ}\text{C}$) fuel cladding in the upper level of the core.¹⁵ Under such a situation, the oxidizing environment is likely to be a mixture of hydrogen gas and depleted steam, rather than an unlimited flux of pure steam. High-temperature oxidation kinetics of Zircaloy-4 in hydrogen-rich steam-hydrogen mixtures have been measured from simulated oxidation tests, and reported recently.¹⁸ Zircaloy tube specimens described in the report were oxidized at $1200\text{--}1705^{\circ}\text{C}$ by steam molecules that impinged on the oxide surface through natural convection in a static hydrogen overpressure of

~33 kPa. Steam impingement rate was varied by changing the steam supply rate into the reaction chamber. Because the hydrogen overpressure was kept constant, a decreased steam supply rate corresponds to an increased hydrogen mole fraction at the oxide/mixture boundary for a given test temperature. Under these experimental conditions, the oxide layer growth rate constant was a function of the steam supply rate as well as of the Zircaloy temperature. However, it was not possible to quantify in the experiments the fundamental thermal-hydraulic parameters that influence the overall oxidation rate in the hydrogen-steam mixture, i.e., the local partial pressures of hydrogen and steam at the oxide/mixture boundary.

Both linear and parabolic oxidation regimes were observed from the oxidation tests in a hydrogen-steam mixture.¹⁸ The linear regimes were observed only for the initial period, and attributed to overall kinetics controlled by the net rate of dissociation of steam molecules on the oxide surface, minus the rate of chemical reduction of the adsorbed oxygen by the hydrogen gas molecules. In the parabolic regime, oxide layer growth rate constants were smaller than those obtained in pure steam, and decreased with decreasing steam supply rate and, hence, increasing hydrogen mole fraction at the oxide/mixture boundary. Figure 2 shows the ratio of the oxide layer growth rate constants for the hydrogen-steam mixture to those in pure steam, plotted as a function of steam supply rate and Zircaloy temperature.

Similar results were obtained under somewhat different thermal-hydraulic conditions, and are shown in Fig. 3. The results in Fig. 3 were obtained from oxidation tests in which the Zircaloy-4 tube specimens were surrounded by a quartz flow shroud that simulated a fuel rod channel. Under those conditions, the hydrogen-steam mixture flow was confined within a well-defined boundary. Therefore, it was possible to determine the average hydrogen mole fraction of the input mixture. Hydrogen flow was kept constant at 10 standard liter⁵/min. Steam flow rate was varied between 0.5 and 3.2 g/min. to establish input hydrogen mole fractions of between 0.94 and 0.71 at the mixture inlet plane of the shroud. The Reynolds number for the mixture flow was in the range similar to those calculated for the Three Mile Island reactor accident.

The zirconium oxide formed during oxidation in a hydrogen-steam mixture may contain a significant amount of dissolved hydrogen as either interstitial or substitutional impurity. The smaller parabolic rate constants observed for the hydrogen-steam mixtures (see Figs. 2 and 3) can be interpreted in terms of lower transport rates of oxygen ions in such oxide.¹⁸ To evaluate the model in more detail, fine powders were obtained from the "surface oxide skin" by mechanical abrasion and were analyzed for oxygen and hydrogen content by the inert-gas fusion technique.

The powder specimens obtained in this manner contain only the surface oxide layer that was exposed to the hydrogen-steam mixtures. Figure 4 shows the results obtained from several specimens oxidized at 1705°C for ~120 s in various hydrogen-steam mixtures that were described in association with Fig. 2. Weight percentages of hydrogen and oxygen in the "surface oxide skin" are plotted as a function of steam supply rate. For comparison, similar results obtained from comparable specimens oxidized in pure steam are also shown in the figure. The oxide formed in pure steam contained less than

0.03 wt % hydrogen and corresponds to a composition close to ZrO_2 . However, as the steam supply rate (hence, the steam mole fraction) decreased in the test, the hydrogen content dissolved in the oxide increased and the oxygen content decreased in proportion. The composition of the oxide skin observed for the smallest steam supply rate in Fig. 4 corresponds to $ZrO_{0.996}H_{0.965}$, which is close to $ZrOH$. For this composition, it is very possible that the crystal structure is different from the structure of the ZrO_2 that is normally produced in pure steam. The result of Fig. 4 supports the validity of the model¹⁸ for the smaller rate constants of Fig. 2, in which oxygen transport rate in the oxide was assumed to be modified by the significant amount of dissolved hydrogen.

In view of the results shown in Figs. 2-4, it appears unrealistic to assume the applicability of the oxidation rates obtained under conditions of unlimited flux of pure steam (e.g., Fig. 1) to a degraded-core accident situation, in which a fuel cladding is likely to be exposed to mixtures of hydrogen and depleted steam at $>1500^\circ C$.

ZIRCALOY CLADDING AS HYDROGEN SINK

Some fraction of hydrogen gas produced as a result of Zircaloy-steam reaction has been known to be dissolved in the cladding material itself. Lemmon² reported that 1.5% and 12.5% of produced hydrogen were dissolved in Zircaloy-2 specimens at oxidation temperatures of 1000 and 1600°C, respectively. Dissolved hydrogen contents of up to 0.22 and 0.4 wt % have been reported, respectively, by Uetsuka et al.¹⁷ for ballooned and ruptured Zircaloy-4 tube specimens oxidized in steam, and by Furuta and Kawasaki¹⁴ for short undeformed tube specimens oxidized in hydrogen-steam mixtures at 950-1100°C. Although corresponding oxygen contents have not been reported for the tube specimens and, hence, no fraction of dissolved hydrogen can be determined from those test results,^{14,17} it appears that the fractions are considerably large for the large hydrogen uptakes.

Oxygen as well as hydrogen contents in oxidized Zircaloy-4 specimens have been analyzed and reported from a recent investigation¹⁶ in which Zircaloy-4 tubes were internally pressurized, ballooned, ruptured, and oxidized isothermally in a steam environment, in simulation of an accident situation. The report shows that the hydrogen uptake was strongly influenced by the morphology and thickness of the inner surface oxide layer, which indicates that the primary route for the absorption was through the inner surface of the ruptured cladding tube. From the oxidation and hydrogen uptake data in the report, the fraction of the dissolved hydrogen (i.e., the ratio of hydrogen atoms dissolved in Zircaloy cladding to hydrogen atoms produced as a result of Zircaloy-steam reaction) was calculated and plotted (Fig. 5) as a function of oxidation temperature. In the same plot, similar values obtained from the similar tube specimens oxidized in hydrogen-steam mixtures¹⁸ and described in association with Fig. 2 are also shown. For the latter data set, oxygen and hydrogen weight contents were determined by inert-gas fusion techniques, as for the case of Fig. 4.

From Fig. 5, it seems clear that the hydrogen absorption fraction is not negligible. The apparent maximum of the fraction at $\sim 1140^\circ C$ is believed to be associated with the formation of porous breakaway-type oxide layer, primarily

at the cladding inner surface, which has been reported¹⁶ to be limited to temperatures $\lesssim 1140^{\circ}\text{C}$. Under transient oxidation conditions during a degraded-core accident, formation of the porous oxide is not likely. For temperatures $\gtrsim 1200^{\circ}\text{C}$, in which no porous oxide was observed, the absorption fraction is still considerable, i.e., as much as $\sim 20\%$.

Because the steam molecules are the only source of oxygen during oxidation in a hydrogen-steam mixture, each dissolved oxygen atom in the Zircaloy (in either oxide, α -, or β -phase) still corresponds to one molecule of steam dissociated or two atoms of produced hydrogen. Therefore, a number of data obtained from specimens oxidized in hydrogen-steam mixtures and shown in Fig. 5 still signify the fraction of hydrogen atoms absorbed per unit number of hydrogen atoms produced by oxidation of the specimen in the mixtures. The specimen showing as much as 18.5% of absorption at 1705°C (see Fig. 5) contained 0.1993 and 8.63 wt % hydrogen and oxygen, in that order. It was oxidized in a hydrogen-steam mixture for 180 s and had an oxide layer of 0.214 mm at the outer surface.

The information contained in Fig. 5 clearly demonstrates that Zircaloy cladding itself can act as a considerable sink for hydrogen. A better understanding in this area, which requires a more comprehensive data base, would be helpful for a more realistic quantification of hydrogen release rate during a degraded-core accident situation.

SUMMARY AND CONCLUSIONS

- (1) Zircaloy-oxidation and corresponding hydrogen-generation rates reported in the literature have been critically reviewed with respect to their applicability to the analysis of hydrogen behavior during a degraded-core accident situation.
- (2) New oxidation rate constants obtained for $>1500^{\circ}\text{C}$ in mixtures of hydrogen and steam are shown. The rate constants are a function of thermal-hydraulic parameters of the mixture flow as well as of the Zircaloy temperature.
- (3) Zircaloy cladding can act as a considerable sink of hydrogen during high-temperature oxidation in either steam or hydrogen-steam environments. An absorption fraction of as much as $\sim 20\%$ of the produced hydrogen was observed for 1200 - 1700°C .

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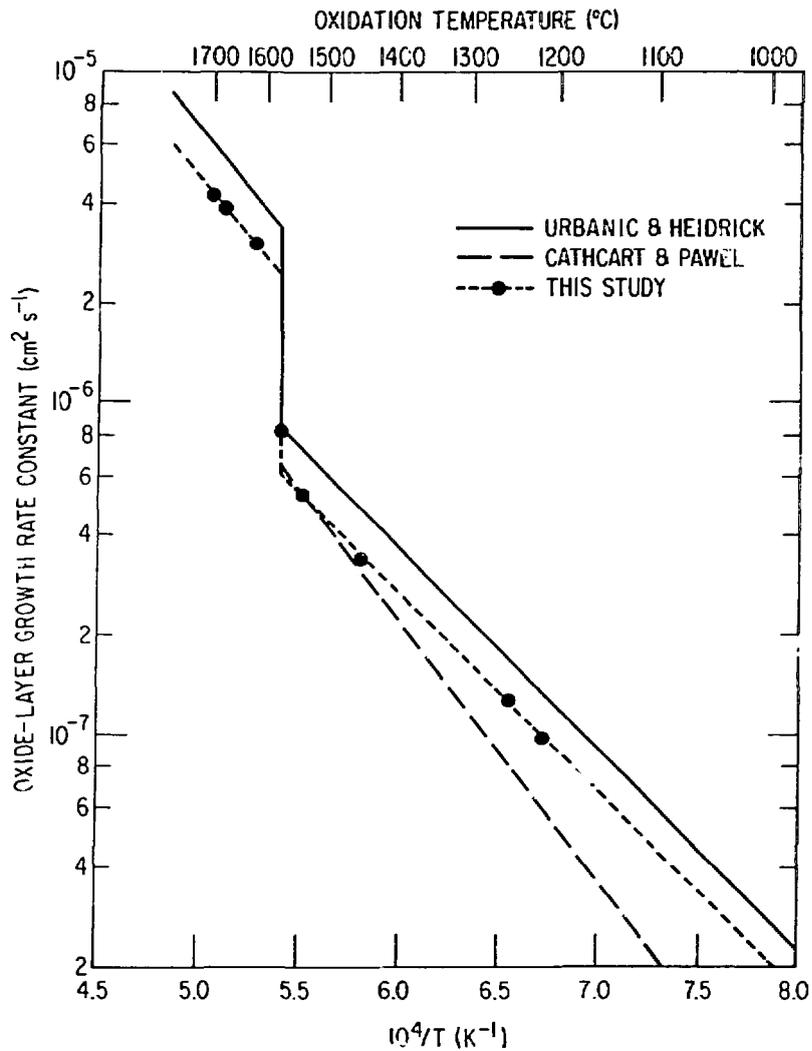


Fig. 1. Parabolic Oxide Layer Growth Rate Constants Obtained for Pure Steam Environments vs Inverse Temperature. Similar results reported by Cathcart and Pawel⁷ and Urbanic and Heidrick¹⁰ are shown for comparison.

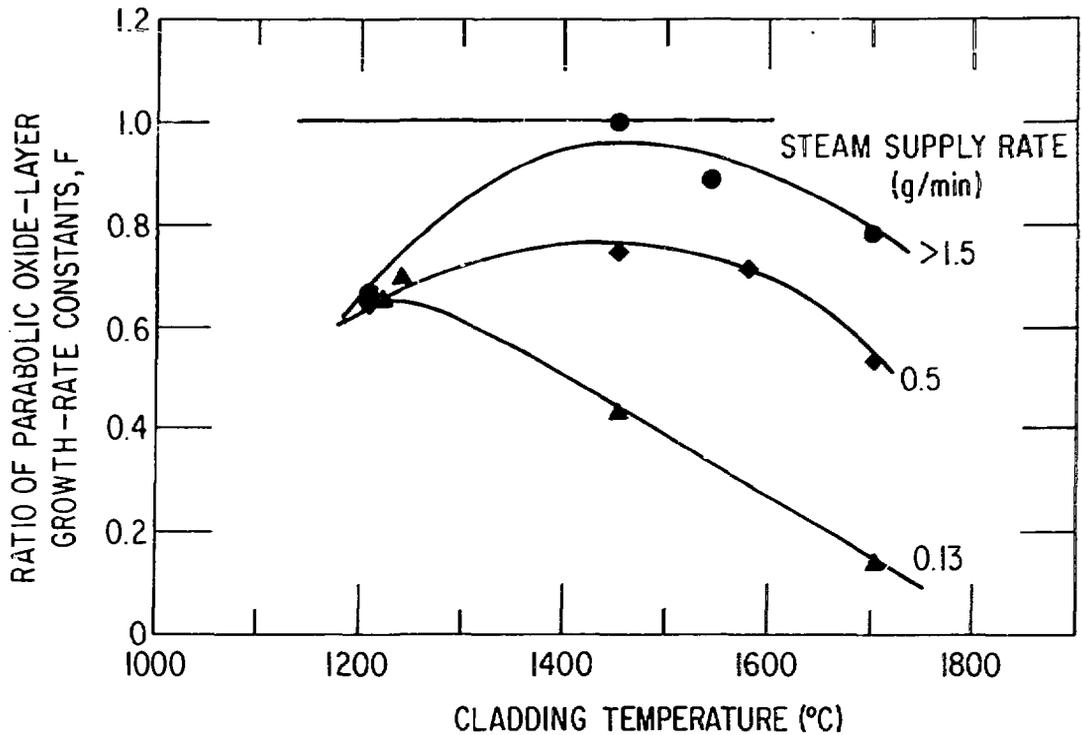


Fig. 2. Ratio of Parabolic Oxide Layer Growth Rate Constant Obtained in Hydrogen-Steam Mixtures to that Obtained in an Unlimited Flux of Pure Steam as a Function of Zircaloy-4 Temperatures and Steam Supply Rate. The static hydrogen overpressure was kept constant at ~ 36 kPa and the specimen was oxidized by steam molecules impinging the oxide surface through natural convection. The parabolic growth rates obtained without the hydrogen overpressure, but with steam supplies of similar rates, are shown in Fig. 1.

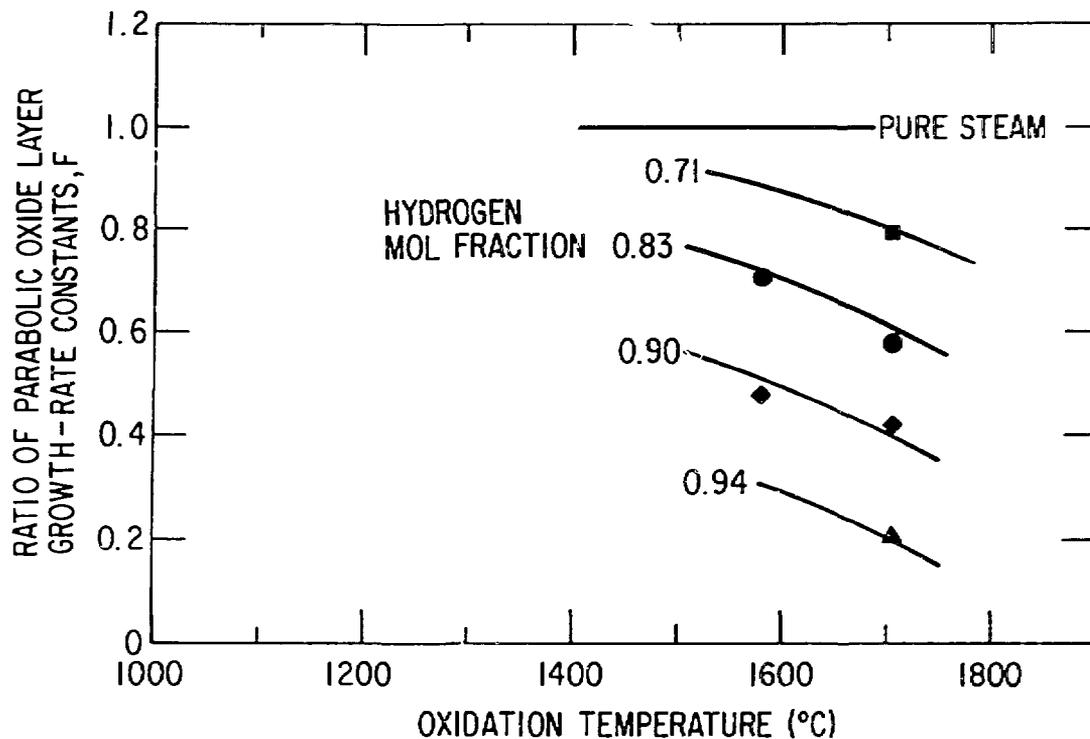


Fig. 3. Ratio of Parabolic Oxide Layer Growth Rate Constant Obtained in Flowing Hydrogen-Steam Mixtures to that Obtained in an Unlimited Flux of Pure Steam as a Function of Zircaloy-4 Temperature and Average Hydrogen Mol Fraction in the Inlet Mixtures. Hydrogen input to the shrouded reaction channel was 0.446 mol/min and steam flow was varied 0.5-3.2 g/min.

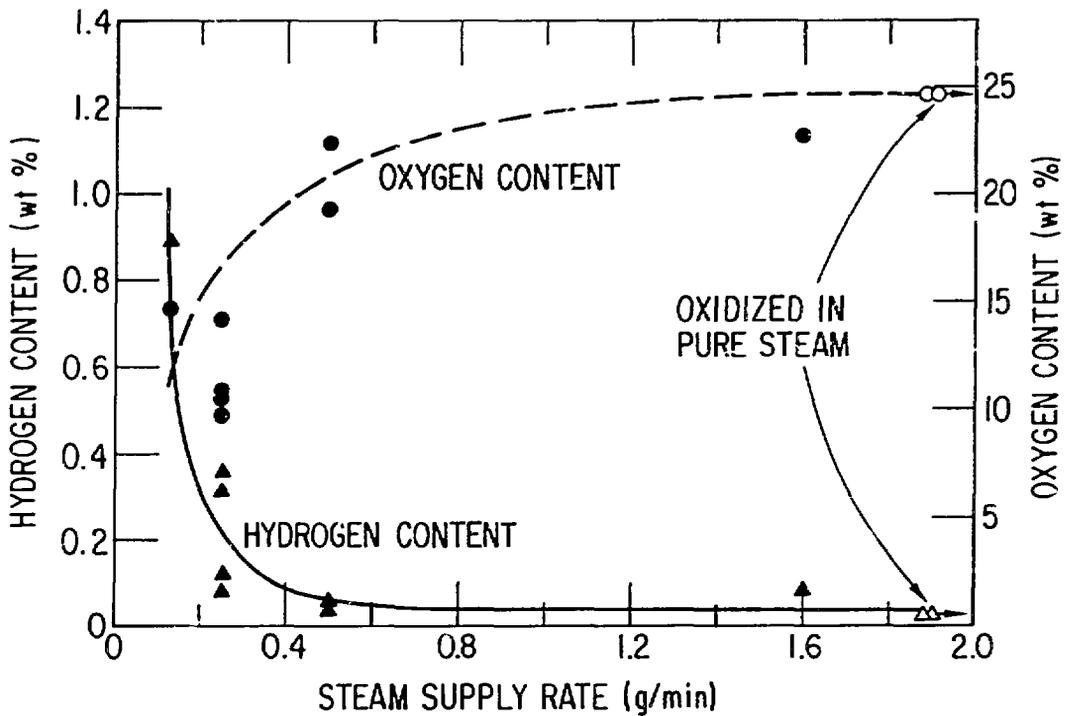


Fig. 4. Hydrogen and Oxygen Contents of the Fine Powders Obtained from Surface Skin of the Oxide Formed During Oxidation at 1705°C for ~120 s in Various Hydrogen-Steam Mixtures Described in Association with Fig. 2. For comparison, similar contents for specimens oxidized in pure steam are shown.

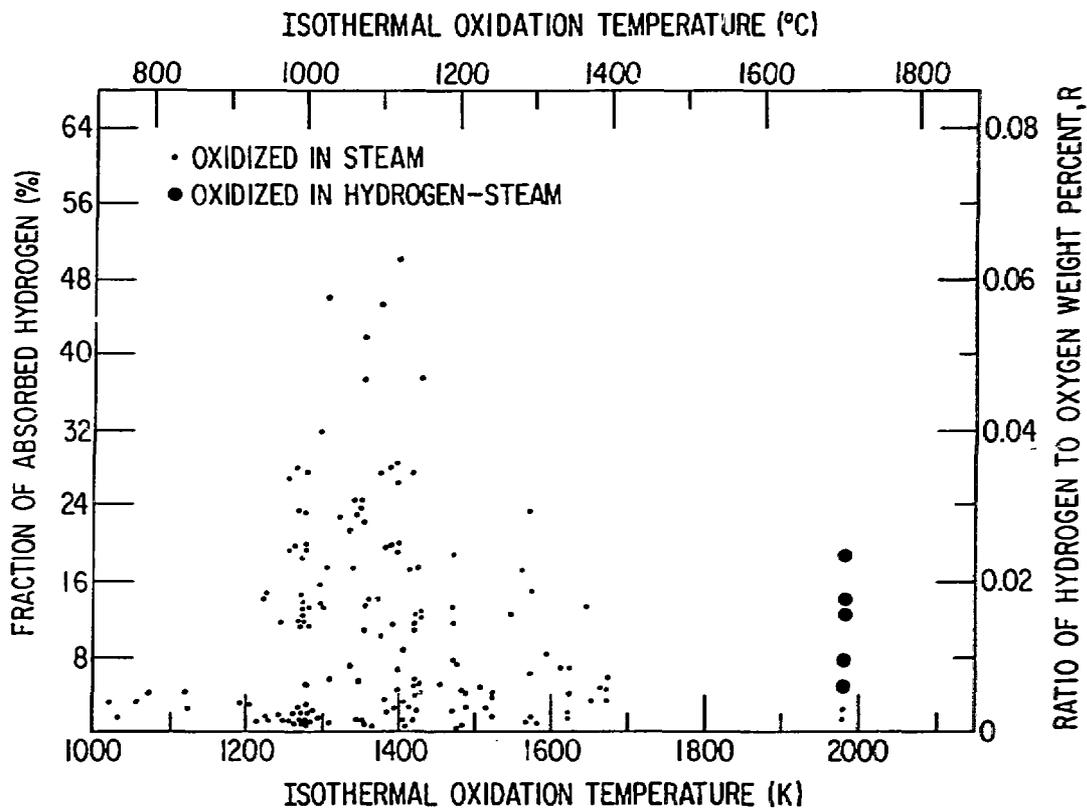


Fig. 5. Fraction of Hydrogen Atoms Dissolved in a Cross Section of Zircaloy-4 Cladding Tube Out of Total Hydrogen Atoms Produced as a Result of Zircaloy-Steam Reaction at Isothermal Oxidation Temperatures Between 1000 and 1700°C. The fractions from oxidation tests in pure steam were obtained from Ref. 16 and similar values for oxidation in hydrogen-steam mixtures were obtained from specimens described in association with Figs. 2 and 4.