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**Comment on PRM-50-73A**

**In 1942, the heat transfer expert, W. H. McAdams, observed, "The small amount of scale necessary to reduce a high (heat transfer) coefficient by a substantial amount is not generally realized." This appears to be the case today with the structure of Appendix K in which there is no recognition of the potential impact of scale deposits on emergency core cooling processes. There is also no recognition in Appendix K of the potential impact of scale or crud deposits on zirconium-water reactions during LOCAs.**

**The following sections, 1.1, 1.2, 1.3 and 1.4 detail specific aspects of this comment. Section 1.1 details the limitations of the Baker-Just equation that is prescribed in Appendix K, including the investigators' admission that, "This discussion is of a preliminary nature: work in this area is continuing." Section 1.2 details the limitations of the document NUREG –17 that is prescribed in Regulatory Guide 1.157, including the investigators' admission that, "Scoping tests of the effect of steam pressure are presently in progress and their results will be described in a subsequent report." Section 1.3 discloses the relevance of FLECHT Zircaloy bundle tests that are reported in WCAP-7665. Section 1.4 briefly discusses the tight controls by the federal agencies during the era when most of the test programs were conducted.**

**1.1 Inapplicability of ANL-6548 to zirconium-water reactions during LOCA**

**Appendix K to Part 50, I. 5 does not accurately describe the extent of zirconium-water reactions that may occur during a loss-of-coolant accident. The Baker-Just equation (Baker, L., Just, L.C., "Studies of Metal Water Reactions at High Temperatures, III. Experimental and Theoretical Studies of the Zirconium-Water Reaction," ANL - 6548, page 7, May 1962) does not include any allowance for the complex thermal-hydraulic conditions during LOCA including the potential for very high bulk fluid temperatures within the cooling channels of the zirconium clad fuel elements.**

**Quoting from the abstract of ANL-6548: "Further studies of the zirconium-water reaction by the condenser-discharge method are reported. The reaction was studied with initial metal temperatures from 1100 to 4000 C with 30 – 60-mil wires in water from room temperature to 315 C (1500-psi vapor pressure). Runs in heated water showed markedly greater reaction. This was explained in terms of a 2-step reaction scheme in which the reaction rate is initially controlled by the rate of gaseous diffusion of water vapor toward the hot metal particles and of hydrogen, generated by reaction, away from the particles. At a later time, the reaction becomes controlled by the parabolic rate law, resulting in rapid cooling of the particles."**

**Thus, the Baker and Just equation is based on data from tests in which the bulk water temperature was no greater than 315 C (599 F). Furthermore, the volume of the water within the test apparatus was substantially greater than the volume of the zirconium**



Template = SECY-067 SECY-02

**specimens. Thus, the capacity to quench (cool) the heated zirconium particles of the Baker-Just experiments was vastly greater than the capacity to quench (cool) the zirconium-clad fuel rods within a nuclear reactor fuel assembly under LOCA conditions. In addition, the zirconium specimens were exposed to water only, while LOCA conditions include steam and non-equilibrium water-steam mixtures that reach substantially higher bulk fluid temperatures.**

**The reaction cells that were used in the Argonne investigations are described on pages 15 through 17 of report ANL-6257. The dimensions of the apparatus are not disclosed, however, the illustrations appear to be drawn to scale in which case the volume of the water is several hundred times the volume of the zirconium wires. For example, the text describes the zirconium wires as either 30 or 60 mils in diameter by 1 inch long. In that case, the diameter of the cylindrical reaction vessel illustrated on page 17 of ANL-6257 is about 1 inch. It follows that the volume ratio of water to zirconium is over 250 for the I inch length alone. Including the water volumes above and below the specimen triples this ratio. And for 30 mil specimens, the ratio exceeds 1000 for the I inch length alone.**

**On page 58 of ANL-6548, Baker and Just disclose Part VII. APPLICATION TO REACTOR HAZZARDS ANALYSIS*. The asterisk leads to the following footnote: <u>*This discussion is of a preliminary nature: work in this area is continuing.</u> Clearly, the Baker and Just admission that , "This discussion is of a preliminary nature..." obviates the application that is prescribed as follows in Appendix K. I. A. 5. "** ***Metal -- Water Reaction Rate*****. The rate of energy release, hydrogen generation, and cladding oxidation from the metal/water reaction shall be calculated using the Baker-Just equation (Baker, L., Just, L.C., "Studies of Metal Water Reactions at High Temperatures, III. Experimental and Theoretical Studies of the Zirconium-Water Reaction," ANL - 6548, page 7, May 1962)."**

**1.2 Inapplicability of NUREG-17 to zirconium-water reactions during LOCA**

**Regulatory guide 1.157, BEST-ESTIMATE CALCULATIONS OF ECCS PERFORMANCE, Paragraph 3.2.5.1, allows the use of the data of NUREG-17 for calculating the rates of energy release, hydrogen generation, and cladding oxidation for cladding temperatures greater than 1900 degrees F. However, the very limited test conditions and results that are disclosed in NUREG-17 obviate its use in LOCA calculations. The Zircaloy-4 specimens were not exposed to LOCA fluid conditions. Only steam was applied, steam was applied at only very low velocities for the main test series. There was no documented heat transfer from the zircaloy surface to the slow-flowing steam, and the conditions of the very small scale laboratory test were thus not typical of the complex thermal-hydraulic conditions that prevail during LOCA. The authors of NUREG-17 only partially admit the inapplicability of their laboratory work to LOCA analyses on page 39 where they state, "Scoping tests of the effect of steam pressure are presently in progress and their results will be described in a subsequent report." This quotation applies to the test series at very low steam**

**velocities.**

**NUREG-17 describes two test series: One test series was conducted in apparatus called MaxiZWOK and covered the temperature range from 1652 to 1832 degrees F. Regulatory guide 1.157 does not explicitly recommend NUREG-17 for this temperature range. Instead, Paragraph 3.2.5.1 states:**

***Correlations to be used to calculate metal-water reaction rates at less than or equal to 1900 F should:***

*a. **Be checked against a set of relevant data and***
*b. **Recognize the effects of steam pressure, pre-oxidation of the cladding, deformation during oxidation, and natural oxidation from both steam and UO2 fuel.***

**The second test series was conducted in apparatus called MiniZWOK and this covered the temperature range from 1832 to 2732 degrees F. NUREG-17 states that these data are considered acceptable for calculating the rates of energy release, hydrogen generation, and cladding oxidation for cladding temperatures greater than 1900 degrees F.**

**The test specimen in the MaxiZWOK apparatus was an18 inch length of PWR tubing that was heated by superheated steam flowing at about 60 feet per second. The specimen was heated by the superheated steam, that is, in order to collect data at 1832 degrees F the steam was injected at 1832 F and approximately 60 PSIG. Although the MaxiZWOK data are not acceptable in Regulatory Guide 1.157, it is revealing to examine the data of Figure 35 on page 78 of NUREG-17. Quoting from the description on page 79, *"For the reaction at 1832 F, the exothermic heat of reaction is sufficient to drive the specimen temperature above that of its environment, creating the overshoot that was typical of MaxiZWOK experiments in this temperature range. In the particular case illustrated in Fig. 35, an overshoot of 32 F was observed before the specimen temperature began to return to its steady-state value, and several minutes were required for the effects of specimen self-heating to be dissipated."***

**Next, consider the MiniZWOK apparatus that was employed for investigations in the temperature range from 1832 to 2732 F. In this case the PWR tubing specimen was only 1.18 inches long, the steam temperature was only about 212 F, and the steam flow rate was only about 1 foot per second (NUREG-17 erroneously reports the flow rate as approximately 3 feet per second). The specimen was not heated by superheated steam as was the case with MaxiZWOK. Instead the specimen was heated by radiant heating from infrared radiant heaters that were located outside of the quartz flow tube.**

**Why did the investigators shift from MaxiZWOK to MiniZWOK for the determinations beyond 1832 F? This is not disclosed. However, the sustained temperature overshoot that was observed in the MaxiZWOK run with 1832 F superheated steam provides the answer. At higher temperatures of injected**

**superheated steam, the temperature overshoots would have been substantially greater than the sustained temperature overshoot of the run at 1832 F. Furthermore, there would likely have been a temperature runaway with destruction of the zircaloy tubing at the higher temperatures of the MiniZWOK runs. The MaxiZWOK runs had a very substantially greater mass transfer coefficients between the steam and the zircaloy than was the case in the MiniZWOK runs. This greater mass transfer coefficients delivered steam at relatively high pressure to the zircaloy surface. In addition, the high velocity swept away hydrogen from the surface of the zircaloy. In contrast, with the low pressure and low velocity of the MiniZWOK runs, the mass transfer coefficients at the heat transfer surface were very substantially less than with MaxiZWOK. It is likely that the investigators ran some tests with the MaxiZWOK at superheated steam temperatures substantially greater than 1832 F and it is unfortunate that none of that expedience was reported.**

**In their Conclusion 5, page 118 of NUREG-17, the investigators misleadingly state,** ***"Neither steam flow rate (above levels leading to steam starvation); steam temperature, ....significantly influence the isothermal rate of oxidation of Zircaloy-4."*** **The statement is misleading because it is not possible to achieve an isothermal rate of oxidation of Zircaloy-4 if the Zircaloy-4 is exposed to LOCA fluid conditions at elevated temperatures. The run at 1832 F proved that and that is why the investigators shifted to the MiniZWOK apparatus. The investigators hedge their opening sentence of Conclusion 5 with the following qualification:** ***"Obviously, however, both steam temperature and flow rate are important parameters in heat transfer calculations, and any failure to remove the heat of the Zircaloy-steam reaction from the fuel cladding can result in an increase in the temperature of the cladding."*** **Next, the investigators close Conclusion 5 with another misleading sentence,** ***"We have shown that the extent of this temperature increase can be calculated with the SIMTRAN computer code given an input of appropriate heat transfer coefficients."*** **The sentence is misleading because it overlooks the very substantially greater mass transfer coefficients that accompany the so-called appropriate heat transfer coefficients. It is those very substantially greater mass transfer coefficients that led to the temperature overshoot of the MaxiZWOK test at 1832 F, and that would have led to very substantially greater temperature overshoots and likely destruction of the Zircaloy tubing if MaxiZWOK has been operated over the temperature range of the MiniZWOK runs.**

**The introductory page to NUREG-17 includes the following warning:**

***This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the Energy Research and Development Administration/United States Nuclear Regulatory Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.***

Inasmuch as the investigators do not warrant their work, and inasmuch as they specifically assume no responsibility for the accuracy of their work, the NUREG-17 is clearly inapplicable to the regulation of nuclear power reactors in the U. S. A.

1.3 Relevance of data reported in WCAP - 7665

The following report is cited in Appendix K. I. D. 3 and Appendix K. I. D. 5: PWR FLECHT (Full Length Emergency Cooling Heat Transfer) Final Report, April 1971. Although WACP-7665 is not cited in K. I. A. 5., Metal-Water Reaction Rate, the discussion of the Zircaloy bundle test, Run 9573, page 3-97, is applicable. The certified Run 9573 includes the complex thermal-hydraulic conditions and zircaloy-water reactions that characterize reflood. These conditions are not found in the narrow test procedures of ANL-6548 or NUREG-17. At the low flooding rate of water at 140 F, only 1 inch per second, the fluid temperature at the 7 foot elevation exceeded 2500 F at 16 seconds into the run. The fluid temperature at the 7 foot elevation likely exceeded 3000 F when heaters began failing 2 seconds later. Prior to the test, cladding temperatures were predicted to reach 2400 F at about 30 seconds.

There is no doubt that the data collected during the first 18 seconds of Run 9573 is valid data. Nevertheless, in WCAP-7665, page 3-98, the following is reported: "...anomalous (negative) heat transfer coefficients were observed at the midplane for 5 of the 14 thermocouples during this period." The negative heat transfer coefficients were not anomalous. The negative heat transfer coefficients were calculated as a result of a heat transfer condition during which more heat was being transferred into the heater than was being removed from the heater. And the reason for that condition was that the heat generated from zircaloy-water reactions at the surface of the heater added significantly to the linear heat generation rate at the location of the midplane thermocouples. This increasing linear heat generation rate is also revealed in the time-temperature plot on Page C-42 of WCAP 7665. The increasing rate is apparent from the increasing rate of temperature increase during the first 18 seconds. Moreover, the increasing rate of linear heat generation occurred during the time span when the electrical power input to the FLECHT assembly steeply and smoothly decreased to 80 percent of the initial power (See Figure 2-12, page 2-17 of WCAP-7665).

Extensive failure of the heat transfer assembly in the severe damage zone (the severe damage zone was approximately 16 inches long centered at the 7 foot elevation) was attributed to electrical arcing from failed heaters since power was not turned off until 36 seconds after the first heater failure. However, there is no proof that electrical arcing occurred. In contrast, the inadvertent overheating of a FLECHT assembly with stainless steel cladding on April 18, 1969, had no gross failure of the cladding during reflood although two-thirds of the heaters failed and there was melting of the braze alloy of some grids. Unfortunately, a detailed thermal-hydraulic analysis of Run 9573, including evaluation of the heating from zircaloy-water reactions, was likely never performed; certainly none is reported in WCAP-7665.

**A pertinent description of the complexities of thermal-hydraulic conditions during reflood, including negative heat transfer coefficients, is included in Part 3.2.3 of WACP-7665. This description applies to data collected with FLECHT bundles with stainless steel cladding. Note that in this description, the negative heat transfer coefficients are not described a "anomalous."**

*Heat Transfer Behavior at Different Elevations*

*Figure 3-24 shows the behavior of typical low and high flooding rate heat transfer coefficients at different elevations. At low flooding rates (2 in./sec or less), the heat transfer coefficient at any given time decreased with increasing elevation. This trend was different for high flooding rate cases (6 in./sec or greater) at early times. For high flooding rates at early times, the heat transfer coefficient at high elevations (6 ft or above) increased with elevation. At later times the trend reversed, and was similar to the low flooding rate behavior.*

*There were two important competing fluid property effects on heat transfer coefficient with elevation: 1) increasing void fraction with elevation, tended to decrease the heat transfer coefficient; and 2) acceleration or increased velocity of the two-phase mixture with elevation, tended to increase the heat transfer coefficient. At low flooding rates, the first effect dominated. At high flooding rates and early times, the second effect was dominant. The dominance of the second effect at high flooding rates and early time was due to the availability of larger amounts of liquid for evaporation, hence acceleration, of the two-phase mixture above the bundle midplane.*

*The negative heat transfer coefficient for the 10-foot elevation at early times indicates heat transfer into (rather than out of) the rod. This was caused by the presence of superheated steam having temperatures above the clad temperature at the 10-foot elevation. Data verifying the presence of highly superheated steam is presented in Section 3.6. Negative heat transfer coefficients were generally found at the 10-foot elevation for low flooding rate runs (2 in./sec or less) at early times (from around 5 up to a maximum of about 120 sec after flood).*

**Another FLECHT Zircaloy bundle test, Run 8874 is pertinent to this petition. In this case, the initial midplane temperature was 2325 F and the flooding rate was 6 inches per second for 8 seconds followed by a step reduction to 1 inch per second for the balance of the run. The time-temperature history of Run 8874 indicated generally consistent and reliable behavior of the instrumented heater rods although there were five heater failures at 9.6, 18.5, 19.3 29.3 and 55.5 seconds after flood.**

**The very high initial flooding rate of 6 inches per second for 48 seconds was extremely effective in the initial cooling of the assembly and the maximum steam probe temperature at the 7 foot elevation remained under 2000 F. This is in marked contrast to Run 9573. The initial heat transfer coefficients were substantially greater than those of a corresponding FLECHT run with stainless steel cladding (Run 6155). This was likely because hydrogen evolved from Ziracaloy-water reactions improved**

enhanced the heat transfer at the zircaloy interface with the water-steam mix. Of course, hydrogen was also present at the zircaloy-steam interface of Run 9573, but in that case, at the low flooding rate, there was not sufficient mass flow of water-steam to yield effective heat transfer and that was evidenced by the negative heat transfer coefficients in Run 9573. In Run 8874 there was a substantial amount of Zircaloy oxidation and debris formation as a result of zircaloy-water reactions, however, the amount of failure was substantially less than in Run 9573.

The following misleading statements appear in WCAP-7665, Part 5.6 on page 5-3:

*1.6 ZIRACLOY-STAINLESS STEEL COMPARISON*

*Heat transfer coefficients for Zircaloy clad appeared to be somewhat greater than for stainless steel. The difference is believed due to observed differences in quench behavior, differences in clad emissivity and possibly the effects of hydrogen produced by the Zircaloy-water reaction. Because the amount and consistency of the data did not permit a quantitative assessment of the magnitude of the difference , it is recommended that stainless steel clad heat transfer coefficients be used as a conservative representation of Zircaloy behavior.*

The above paragraph is misleading because it implies that stainless steel heat transfer coefficients may be used as a conservative representation of Zircaloy behavior. The stainless steel heat transfer behavior is certainly not a conservative representation of Zircaloy behavior. The data for the first 18 seconds of Run 9573 are real and certified. There is no basis for rejecting the negative heat transfer coefficients in run 9573. The higher values of the heat transfer coefficients of Run 8874 are also valid. The differences in the behavior between these runs are explained by the differences in the thermal hydraulic conditions that led to a different combination of heat transfer and mass transfer factors; the differences are not explained on the basis of inconsistency of the data.

The following section, part 5.11 of CONCLUSIONS, WCAP-7665, is also misleading:

*5.11 MATERIALS EVALUATION*

*The Baker-Just parabolic rate equation appears to provide a satisfactory basis for determining the overall extent of metal-water reaction. However, the formation of an oxide film and an oxygen containing alpha-zirconium layer beneath the oxide film should be accounted for in determining the metal-water reaction energy release and oxide film thickness.*

*In the investigations reported here, the extent of metal-water reaction was basically homogeneous with no major variations in oxide film thickness at given cross-sectional locations in a fuel rod. The amount of hydrogen absorption was a low proportion (less than 10 percent) of the available hydrogen resulting from the metal-water reaction.*

***Even though the specimens examined reached temperatures as high as 2545 F, there was no evidence of clad shattering or failure as a result of being exposed to typical loss-of-coolant accident environments."***

**The foregoing Part 5.11 is misleading in view of the total experience with FLECHT Run 9573. The experience with Run 9573, with the fluid temperature at the 7 foot elevation exceeding 2500 F at 16 seconds (and likely exceeding 3000 F when heater failures began at 18 seconds) does not justify the assertion that, "*The Baker-Just parabolic rate equation appears to provide a satisfactory basis for determining the overall extent of metal-water reaction.*" Furthermore, there was extensive clad oxidation and clad shattering during Run 9573.**

**1.4 Control of the testing programs by the AEC, ERDA and the NRC.**

**The test programs of 1.1, 1.2 and 1.3 were funded by government agencies. Most of these test programs were firmly controlled by those who were indoctrinated in the methods of the tightly regimented Naval Reactors Program. This mindset dominated AEC, ERDA and NRC programs during the era of the test work that is referenced in Appendix K and Regulatory Guide 1.157. In particular, the biased reporting of WCAP-7665 may be traced to these controls. The lack of application of the MaxiZWOK apparatus beyond 1832 F in NUREG-17 may likely be traced to rigid restrictions by management at the NRC. The Argonne work of ANL-6548 was likely less impacted by these controls.**

**This Commenter has made several requests to the Knolls Atomic Power Laboratory (KAPL) for Report KAPL-1534, <u>Some Qualitative Observations of the Zirconium-Water Reaction at Atmospheric Pressure</u>, (April 1956). These requests have been ignored by KAPL. Only a fragment of the KAPL work is discussed by Baker and Just in ANL-6357 as follows: *Layman and Mars demonstrated a self-sustaining reaction between Zircaloy and water, by condenser-discharge heating to simulate exponential periods of 2 to 29 msec. Chemical reaction was initiated very close to the melting point. Some self-heating was noted in every run, and reaction in some cases was sustained to completion.***

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