Draft Interim Review of PRM-50-93/95 Issue Related to Experimental Methods Used to Derive the Baker-Just Metal-Water Oxidation Reaction Correlation

Disclaimer:

Public availability of this interim report is intended to inform stakeholders of the current status of the NRC's review of the issues raised in PRM-50-93/95. This interim report is subject to further revisions during resolution of PRM-50-93/95. The NRC is not soliciting public comments on these interim conclusions, and will not provide a formal response to any comments received. The NRC's findings on PRM-50-93/95 issues will not be final until the NRC publishes a notice of final action on this PRM in the Federal Register.

1.0 Issue Raised in the Petitions and Associated Comments

A petition for rulemaking was docketed as PRM-50-93 on November 17, 2009 (Leyse, 2009). The petition is requesting revisions to §10 CFR 50.46 "Acceptance Criteria for Emergency Core Cooling systems for Light Water Nuclear Power Reactors" and to 10 CFR Part 50, Appendix K "ECCS Evaluation Models" as well as associated regulatory guidance.

This interim report is the NRC staff's interim evaluation of certain assertions in the Petition for Rulemaking PRM-50-93/95 (Leyse, 2009) regarding experimental measurements of oxidation kinetics. One of the submissions (pages 12-14 of Leyse, 2010) raised the issue of oxidation measurements with inductive heating and radiative heat losses with respect to the Baker-Just correlation (Baker, 1962). The submission contends that the experiments did not replicate reactor LOCA behavior of cladding because of the radiative heat losses from the sample inherent in using inductive heating, and so proposes that the Baker-Just correlation is non-conservative. NRC shows in this interim report that the questioned data are similar to data obtained by other methods, and so the heat losses are not relevant in correlating the data.

2.0 Considerations Regarding Inductive Heating

A recent paper by French experts (Vandenberghe, 2012) on cladding behavior shows that heating mode (resistance furnace heating and induction heating) does not affect the oxidation behavior at a temperature near 1000 ºC. Oxide layer thickness measurements were quite similar for each heating mode, thus showing oxidation kinetics is similar.

3.0 Considerations Regarding Radiative Heat Losses

The submission (Leyse, 2010) asserts the following: "So the radiative heat losses of the zirconium specimens in Bostrom and Lemmon's induction heating experiments would have affected the oxidation kinetics that Bostrom and Lemmon measured. Bostrom and Lemmon's experiments certainly did not replicate the oxidation kinetics that would occur in a nuclear power plant's core, in the event of a LOCA."

With respect to radiative heat losses, there are the following considerations:

1) The Bostrom data (Bostrom, 1954) were obtained for temperatures at and above 1300 ºC (1573K), so they are not pertinent for consideration here.
2) The Lemmon data (Lemmon, 1957) at 1100 and 1200 ºC , although reasonable, have been superseded by numerous measurements (see below).

3) The crucial aspect of oxidation measurements is knowing the temperature of the oxidizing surface and having adequate steam. Heat losses are only important to the extent that they affect temperature control, or are so large that the sample has significant temperature gradients.

The table below shows the Lemmon values of rate constant (using induction heating) that were used to set the low temperature end of the Baker-Just correlation. The table also shows values determined by other experimenters using resistance heating (Cathcart, et al., 1977) and resistance and induction heating (Leistikow and Schanz, 1987). The Cathcart report describes in detail how uncertainty was considered. Note that the Lemmon values are conservative (higher), as are the Baker-Just values, when compared to the more recently determined values. Thus, there is no indication in the data that induction heating with assumed radiative heat losses leads to non-conservative reaction rate values. With regard to the assertion that the test conditions were not representative of LOCA behavior, it is noted that the metal-water reaction rate constant correlation is a function of temperature only. It must be used in conjunction with a cladding oxidation correlation and other system-level thermal hydraulic models in order to determine the instantaneous cladding temperature.

Values of total oxidation (weight gain) rate constant for temperatures between 1000 and 1200 ºC
Rate Constant in $(gm/cm^2)^2$/second. The computed values are based on correlation coefficients from a recent state-of-the-art report (OECD, 2009), except for Lemmon, based on Figure 1 in (NRC, 2004).

Experimenter (lab)	Rate Constant $(gm/cm^2)^2$/sec (times e+08)			Q/R (K)	Heating mode
	1200 ºC	1100 ºC	1000 ºC		
Cathcart (ORNL)	21.5	7.94	2.51	20100	Resistance
Leistikow (KfK)	17.2	6.09	1.83	20972	Resist. & Induc.
Lemmon (BMI)	25	11.5	4	17111	Induction
Baker (ANL)	36.3	11.6	3.15	22900	N/A

4.0 Summary and Conclusions

This interim report has evaluated the petition's contention that the Baker-Just oxidation kinetics correlation is non-conservative because it is partly derived from experimental data asserted to be not representative of reactor LOCA behavior. The metal-water reaction rate constant correlation is a function of temperature only, and must be used in conjunction with system-level thermal hydraulic models in order to determine the instantaneous cladding temperature. The staff's evaluation of data and induction heating method described in the Lemmon report (Lemmon, 1957), does not support the petition's assertion that use of such data by Baker and Just resulted in a non-conservative correlation.

5.0 References

Baker Jr., L., and Just, L. C., Studies of Metal-Water Reactions at High Temperatures, III. Experimental and Theoretical Studies of the Zirconium-Water Reaction, ANL-6548, May 1962, ADAMS Accession No. ML050550198.

Bostrom, W. A., The High Temperature Oxidation of Zircaloy in Water, WAPD-104, March 1954, ADAMS ML100900446.

Cathcart, J. V. et al, Zirconium Metal-Water Oxidation Kinetics IV. Reaction Rate Studies, ORNL/NUREG-17, August 1977, ADAMS Accession No. ML052230079.

Leistikow, S., and Schanz, G., "Oxidation kinetics and related phenomena of zircaloy-4 fuel cladding exposed to high temperature steam and hydrogen-steam mixtures under PWR accident conditions," Nucl. Eng. Des. 103, 65-84, 1987.

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Vandenberghe, et al., Sensitivity to Chemical Composition Variations and Heating/Oxidation Mode of the Breakaway Oxidation in M5 Cladding Steam Oxidized at 1000 ºC (LOCA Conditions), TOPFUEL 2012, Manchester, UK, September 2012.