Summary of Mark Leyse's June 24 Afternoon Presentation for the Public Meeting on the 50.46c Proposed Rule and Draft Regulatory Guides

The presentation mainly focused on two topics: 1) the thermal resistance of crud deposits and oxide layers on fuel cladding and how that would increase the peak cladding temperature in the event of a loss-of-coolant accident ("LOCA") and 2) breakaway oxidation and the affects of the hydrogen content in the fuel cladding.

Argonne National Laboratory's Experimental Boiling Water Reactor

Problems with the thermal impact of crud on zirconium alloy fuel cladding (fuel plates, not fuel rods) were documented as far back as 1959.

At Argonne National Laboratory, Robert Leyse initiated studies of the thermal impact of scale (crud) on the zirconium alloy cladding of the Experimental Boiling Water Reactor (EBWR). For example, in June 1959 he wrote a paper titled, "Effect of Scale Deposits on Fuel Element Temperatures with EBWR at 100 Megawatts."

The EBWR had zirconium alloy fuel plates and there were aluminum alloy filler pieces in the core; aluminum oxide deposits ended up accumulating on the zirconium alloy fuel plates. The maximum scale thickness (crud) was measured at about 200 microns. Heat transfer calculations based on the crud's low thermal conductivity indicated that maximum fuel and fuel-cladding temperatures substantially increased during operation. So much so that increasing the power level of the EBWR up to 100 Megawatts was expected to cause crud-induced fuel growth and fuel element distortion and damage.[1]

PRM-50-84

In 2003, Robert Leyse submitted a couple of rulemaking petitions to the NRC on how crud deposits would increase the severity of a LOCA at a commercial light water reactor. Those petitions were denied.

Then in March 2007, I submitted a rulemaking petition to the NRC, which was docketed as PRM-50-84.[2] PRM-50-84 requested new regulations: 1) to require licensees to operate light

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[1] C. R. Breden, I. Charak, R. H. Leyse, Argonne National Laboratory, "Water Chemistry and Fuel Element Scale in EBWR," ANL-6136, November 1960, Abstract, p. 9.

[2] Mark Leyse, PRM-50-84, March 15, 2007 (ADAMS Accession No. ML070871368).

water reactors under conditions that effectively limit the thickness of crud (corrosion products) and/or oxide layers on fuel cladding, in order to help ensure compliance with ECCS acceptance criteria; and 2) to stipulate a maximum allowable percentage of hydrogen content in fuel cladding.

PRM-50-84 also requested that the NRC amend Appendix K to Part 50—ECCS Evaluation Models I(A)(1), *The Initial Stored Energy in the Fuel*, to require that the steady-state temperature distribution and stored energy in the fuel at the onset of a postulated LOCA be calculated by factoring in the role that the thermal resistance of crud and/or oxide layers on cladding plays in increasing the stored energy in the fuel. PRM-50-84 also requested that these same requirements apply to any NRC-approved best-estimate ECCS evaluation models used in lieu of Appendix K to Part 50 calculations.

In 2008, the NRC decided to consider the issues raised in PRM-50-84 in its rulemaking process.[3] And in 2012, the NRC Commissioners voted unanimously to approve a proposed rulemaking—revisions to Section 50.46(b), which will become Section 50.46(c)—that was partly based on the safety issues raised in PRM-50-84.[4]

Considering the Thermal Resistance of Crud in LOCA Analysis

The thermal resistance of crud deposits has not been considered in LOCA Analysis. An attachment to a 2003 letter from Gary W. Johnsen, RELAP5-3D Program Manager, Idaho National Engineering and Environmental Laboratory, to Robert Leyse states:

> [W]e are not aware of any user who has modeled crud on fuel elements with SCDAP/RELAP5-3D. … We suspect that none of the other [accident analysis] codes have been applied to consider [fuel crud buildup] (because it has not been demonstrated conclusively that this effect should be considered). … SCDAP/RELAP5-3D *can* be used to consider this effect, it is simply that users have not chosen to consider this phenomen[on] [emphasis not added].[5]

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[3] Federal Register, Vol. 73, No. 228, "Mark Edward Leyse; Consideration of Petition in Rulemaking Process," November 25, 2008, pp. 71564-71569.

[4] NRC, Commission Voting Record, Decision Item: SECY-12-0034, Proposed Rulemaking—10 CFR 50.46(c): Emergency Core Cooling System Performance During Loss-of-Coolant Accidents (RIN 3150-AH42), January 7, 2013, (ADAMS Accession No. ML13008A368).

[5] From an attachment of a letter from Gary W. Johnsen, RELAP5-3D Program Manager, INEEL to Robert H. Leyse, June 17, 2003, (ADAMS Accession No. ML032050508).

Not considering the thermal resistance of crud deposits is a problem. A paper I coauthored, "Considering the Thermal Resistance of Crud in LOCA Analysis,"[6] which was presented at the American Nuclear Society's 2009 Winter Meeting, provides evidence that crud needs to be considered in LOCA analysis. The paper discusses RELAP5-3D computer simulations that were performed by Rui Hu in 2009 when he was a graduate student at MIT.

A RELAP5-3D model of a reference Westinghouse four-loop PWR plant that MIT developed for a previous study was used. RELAP5-3D simulated a large break LOCA—a double-ended guillotine break—at the modeled plant: surface temperatures of clean fuel cladding were compared to those of fuel cladding with a 100 μm thick crud layer. For both cases, surface temperatures of the hottest fuel rod of the hottest assembly were examined.

(The crud layer was assigned a thermal conductivity of 0.8648 Watts per meter Kelvin (W/m·K); the heat capacity of the crud layer was assigned the same value as that of the fuel cladding: these values would be close under the high-temperature conditions of the postulated LOCA.)

The RELAP5-3D analysis demonstrated that the peak cladding temperature ("PCT") of the crud case was 77 Kelvin (139 Fahrenheit) higher than that of the clean-cladding case, for the postulated LOCA. Hence, the thermal resistance of crud deposits on fuel cladding needs to be considered in LOCA analysis for licensing and other related activities.

(I have also sent "Considering the Thermal Resistance of Crud in LOCA Analysis," and a second paper, "Assessment of Fuel Rod Performance by Consideration of Crud Deposition," by Joosuk Lee and others of the Korea Institute of Nuclear Safety ("KINS"). In the KINS paper, values of 0.8648 and 0.4324 W/m·K were used for the thermal conductivity of crud.)

The Proposed Rule

As published in the Federal Register, regarding the thermal effects of crud and oxide layers, the proposed rule for Section 50.46(c), Paragraph (g)(2)(ii), states:

> The thermal effects of crud and oxide layers that accumulate on the fuel cladding during plant operation must be evaluated. For the purposes of this paragraph, crud means any foreign substance deposited on the surface of fuel cladding prior to initiation of a LOCA.

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[6] Rui Hu, Mujid S. Kazimi, Mark Leyse, "Considering the Thermal Resistance of Crud in LOCA Analysis," American Nuclear Society, 2009 Winter Meeting, Washington, D.C., November 15-19, 2009.

Paragraph (g)(2)(ii) needs to be augmented with additional instructions; it should have an additional sentence, inserted after the first sentence, stating:

> The thermal effects of crud and oxide layers must be evaluated based on the observed crud and oxide layers that are present on the fuel cladding at the start of the forthcoming operating cycle, and in addition, the projected changes in the crud and oxide layers during the course of the forthcoming operating cycle must also be included in order to provide an accurate evaluation.

This information is in comments on the proposed rule submitted by Robert Leyse on April 23, 2014, in ADAMS at ML14115A463.

Clearly, there are a number of factors that could play a role in how much crud would be present in any forthcoming operating cycle. Ultrasonic fuel cleaning could be used to remove a portion of the existing crud; and of course more crud would accumulate on the fuel cladding. But it's important to note that there are models of crud and oxide deposition that have been developed that are intended to predict of the thicknesses of crud deposits and oxide layers on the fuel cladding. That information is mentioned in a 2003 paper "Taming the Crud Problem: The Evolution."[7]

(**NOTE:** In the question-and-answer session after the presentation, someone made what I believe is an important point. He pointed out that during a fuel cycle there could be "fluffy," non-tenacious crud on the fuel rods, which would not be observed at the end of the fuel cycle. Non-tenacious crud can be released from fuel rods during refueling outages; this is sometimes termed a "crud burst." Regarding some observed crud bursts, Electric Power Research Institute ("EPRI") states that "[s]everal PWRs have experienced anomalous crud releases during refueling outages, characterized by unexpectedly high particulate crud releases followed by deposition or by abnormally high activity releases after peroxide addition (or release after floodup)."[8]

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[7] Yovan D. Lukie, Jeffrey S. Schmidt, "Taming the Crud Problem: The Evolution," Advances in Nuclear Fuel Management 2003 Hilton Head Island, South Carolina, USA, October 2013.

[8] EPRI, "Product Abstract: High Activity Crud Burst Impacts and Responses," available at: http://www.epri.com/abstracts/Pages/ProductAbstract.aspx?ProductId=000000000001016766

It is pertinent that EPRI has developed a BWR crud-deposition model called the Crud DepOsition Risk Assessment ModeL ("CORAL"),[9] "[t]o facilitate improved management of any crud-related fuel performance risk."[10]

Regarding CORAL, EPRI states:

> The BWR Fuel Crud Model provides *a prediction of the crud deposition and tenacious crud layer thickness* both radially and axially within a selected fuel assembly throughout the entire operating history of the assembly. The analysis inputs include the fuel assembly geometry and actual or projected operating history, as well as crud inputs determined from a reactor system mass balance. A boiling deposition model, in conjunction with mechanistic crud release models, defines the deposited inventory along the length of all fuel rods within the fuel assembly. *The deposited crud material is separated between an outer loose, fluffy layer and an inner tenacious layer*. The thickness of the tenacious layer is determined [emphasis added].
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> The BWR Fuel Crud Model has been extensively validated through benchmarking of the model using data taken on fuel rods operated in actual commercial BWRs. These benchmarking measurements include (1) poolside fuel deposit sampling of *both total and tenacious crud layer*, (2) laboratory examination of crud flakes and tenacious deposits on irradiated fuel rods to determine composition, thickness, density, and structure, and (3) poolside eddy current lift-off and cladding diametral profilometry data. This successful benchmarking activity provides confidence in the model's ability to capture and quantitatively describe crud deposition behavior, as well as quantifying the inherent variability in the crud deposition and release processes and resultant tenacious crud layer thickness[11] [emphasis added].

Such models need to be used to predict the thicknesses of crud deposits—for both outer loose, fluffy layers and inner tenacious layers—that would be present on the fuel cladding during each fuel cycle. And such models need to be used for both PWRs and BWRs. Clearly, the thermal effects of crud—for fluffy layers and tenacious layers—and oxide layers need to be evaluated in LOCA analysis; and the thicknesses of such layers need to be modeled conservatively.)

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[9] EPRI, "Product Abstract: Technical Basis and Benchmarking of the Crud Deposition Risk Assessment Model (CORAL)," available at: http://www.epri.com/abstracts/Pages/ProductAbstract.aspx?productId=000000000001025189

[10] EPRI, "Product Abstract: Fuel Reliability Program: BWR Fuel Crud Modeling," available at: http://www.epri.com/abstracts/Pages/ProductAbstract.aspx?ProductId=000000000001021034

[11] *Id*.

Breakaway oxidation

The proposed rule for Section 50.46(c) states:

> *Breakaway oxidation,* for zirconium-alloy cladding material, means the fuel cladding oxidation phenomenon in which weight gain rate deviates from normal kinetics. This change occurs with a rapid increase of hydrogen pickup during prolonged exposure to a high-temperature steam environment, which promotes loss of cladding ductility.

And Draft Regulatory Guide 1261 states that breakaway oxidation is an instability phenomenon that can spread rapidly in the axial and circumferential directions of fuel rods and that there is a criterion of 200-weight parts per million (wppm) for hydrogen pickup. It says that fuel-cladding ductility is maintained as long as the average hydrogen content is below 435 wppm.

Draft Regulatory Guide 1261 states:

> [T]he 200-wppm hydrogen pickup criterion is conservative by a factor of at least two. However, it is not overly conservative for high oxidation temperatures because the time needed to increase from 200 wppm to >400 wppm hydrogen pickup could be as low as 100 seconds.

When high burnup and other fuel rods are discharged from the reactor core, the fuel cladding can have local zirconium dioxide (ZrO_2) "oxide" layers that are up to 100 microns (µm) thick (or greater); there can also be local crud layers on top of the oxide layers. And according to NUREG/CR-6851, medium to high burnup fuel cladding typically has a "hydrogen concentration in the range of 100-1000 wppm [weight parts per million];" it adds that "[z]irconium-based alloys, in general, have a strong affinity for oxygen, nitrogen, and hydrogen…"[12]

NRC's conclusions on how hydrogen content affects fuel cladding ductility are based on the results of isothermal experiments conducted with small specimens. These were experiments in which a tiny section of a fuel rod was held at a constant temperature. I think most of this program was done at Argonne; and there were some tests done with pre-oxidized fuel cladding.

The PHEBUS B9R-2 test is an integral experiment conducted with pre-oxidized fuel cladding, which I think the NRC should study. PHEBUS B9R-2 was conducted in a light water reactor—as part of the PHEBUS severe fuel damage program—with an assembly of 21 UO_2 fuel

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[12] K. Natesan, W.K. Soppet, Argonne National Laboratory, "Hydrogen Effects on Air Oxidation of Zirlo Alloy," NUREG/CR-6851, October 2004, (ADAMS Accession No: ML042870061), p. iii, 3.

rods.[13] A 1996 European Commission report states that the B9R-2 test had an unexpected fuel-cladding temperature escalation in the mid-bundle region; the highest temperature escalation rates were from 20°C/sec (36°F/sec) to 30°C/sec (54/°F/sec).[14]

Discussing PHEBUS B9R-2, the 1996 European Commission report states:

> The B9R-2 test…illustrates the oxidation in different cladding conditions representative of a pre-oxidized and fractured state. … During B9R-2, an unexpected strong escalation of the oxidation of the remaining Zr occurred when the bundle flow injection was switched from helium to steam while the maximum clad temperature was equal to 1300 K [1027°C (1880°F)]. The current oxidation model was not able to predict the strong heat-up rate observed even taking into account the measured large clad deformation and the double-sided oxidation (final state of the cladding from macro-photographs).
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> … No mechanistic model is currently available to account for enhanced oxidation of pre-oxidized and cracked cladding.[15]

On the ninth slide from my presentation there is a graph with a plot of fuel-cladding temperature values at the 0.6 meter "hot level" of the PHEBUS B9R-2 test bundle.

As stated, the cladding-temperature escalation commenced at approximately 1027°C (1880°F). That is thermal runaway. The fact that PHEBUS B9R-2 was conducted with a pre-oxidized test bundle makes its results pertinent to the cladding of medium and high burnup fuel rods.

The hydrogen content of the cladding of PHEBUS B9R-2 test bundle most likely played a role in the test results. I think the results indicate what could happen in a LOCA; the test was conducted under conditions far more representative of LOCA conditions than the Argonne tests with tiny specimens.

I recommend that the rulemaking branch study the results of the PHEBUS B9R-2 test. In the statement on breakaway oxidation from Draft Regulatory Guide 1261 quoted above there is the observation that breakaway oxidation deviates from normal kinetics.

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[13] G. Hache, R. Gonzalez, B. Adroguer, Institute for Protection and Nuclear Safety, "Status of ICARE Code Development and Assessment," in NRC "Proceedings of the Twentieth Water Reactor Safety Information Meeting," NUREG/CP-0126, Vol. 2, 1992, (ADAMS Accession No: ML042230126), p. 311.

[14] T.J. Haste *et al.*, "In-Vessel Core Degradation in LWR Severe Accidents," European Commission, Report EUR 16695 EN, 1996, p. 33.

[15] *Id.*, p. 126.

I guess that normal oxidation kinetics are supposed to be those observed in the tests with tiny zirconium specimens held at a constant temperature. In such tests the rate of steam flow is controlled. And different adjustments can influence oxidation rates. This is discussed in a paper by Gerhard Schanz titled "Recommendations and Supporting Information on the Choice of Zirconium Oxidation Models in Severe Accident Codes." The Schanz paper states that an investigator "reached an important improvement of the specimen temperature homogeneity by only optimizing the geometry of the specimen and registered considerably increased reaction rates."[16]

I think that any honest, objective study of a number of integral experiments conducted with multi-rod bundles of fuel rod simulators or real fuel rods with UO_2 fuel would reveal many deviations from so-called normal oxidation kinetics. The reaction rates have been rapid in a number of the large-scale, integral experiments. In those cases thermal runaway is more of an issue than cladding embrittlement. Preventing thermal runaway could be a more important safety issue than preventing excessive cladding embrittlement.

The 2200°F Peak Cladding Temperature Limit

I also think the results of the PHEBUS B9R-2 test should be looked at, along with other integral experiments to help determine if the proposed rule for Section 50.46(c), Paragraph (g)(1)(i) is non-conservative. That is, the test results of integral experiments may indicate that the 2200°F PCT limit needs to be lowered. In a large break LOCA there could be steam-binding conditions that would not allow much coolant to be injected, so if the fuel-cladding temperature were increasing at say a rate of 10°F per second; that would mainly be from the stored energy (heat) in the fuel, at the beginning of the LOCA. So if the fuel cladding temperature were to reach around 1832°F in a steam environment; and there were little or no coolant injection, there would probably be results similar to those of the PHEBUS B9R-2 test.

Please See Papers I Attached to My E-Mail: 1) "Considering the Thermal Resistance of Crud in LOCA Analysis" and 2) "Assessment of Fuel Rod Performance by Consideration of Crud Deposition."

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[16] Gerhard Schanz, "Recommendations and Supporting Information on the Choice of Zirconium Oxidation Models in Severe Accident Codes," FZKA 6827, 2003, p. 5.