

May 13, 2015

MEMORANDUM TO: Timothy J. McGinty, Director  
Division of Safety Systems  
Office of Nuclear Reactor Regulation

FROM: Paul M. Clifford, Senior Technical Advisor */RA/*  
Division of Safety Systems  
Office of Nuclear Reactor Regulation

SUBJECT: ACCEPTABLE FUEL CLADDING HYDROGEN UPTAKE  
MODELS

The purpose of this memorandum is to provide acceptable fuel rod cladding hydrogen uptake models for the current commercial zirconium alloys to aid in the implementation of the hydrogen-dependent ECCS performance requirements. Figure 2 of DG-1263 provides an acceptable analytical limit on peak cladding temperature and integral time-at-temperature (expressed as equivalent cladding reacted calculated using the Cathcart-Pawell correlation (CP-ECR)) as a function of pre-transient cladding hydrogen content. To support implementing these new requirements, steady-state cladding waterside corrosion and hydrogen pickup models are needed to translate these analytical limits to fuel burnup.

These models are also acceptable for implementing other hydrogen-dependent fuel performance requirements, e.g. reactivity initiated accident (RIA) pellet-to-cladding mechanical interaction (PCMI) cladding failure thresholds.

Enclosure:  
Fuel Rod Cladding Hydrogen Uptake Models

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## Fuel Rod Cladding Hydrogen Uptake Models

### 1.0 SCOPE AND PURPOSE

The purpose of this memorandum is to provide acceptable fuel rod cladding hydrogen uptake models for the current commercial zirconium alloys to aid in the implementation of the hydrogen-dependent ECCS performance requirements. Figure 2 of DG-1263 provides an acceptable analytical limit on peak cladding temperature and integral time-at-temperature (expressed as equivalent cladding reacted calculated using the Cathcart-Pawell correlation (CP-ECR)) as a function of pre-transient cladding hydrogen content. To support implementing these new requirements, steady-state cladding waterside corrosion and hydrogen pickup models are needed to translate these analytical limits to fuel burnup.

These models are also acceptable for implementing other hydrogen-dependent fuel performance requirements, e.g. reactivity initiated accident (RIA) pellet-to-cladding mechanical interaction (PCMI) cladding failure thresholds.

### 2.0 DISCUSSION

Pacific Northwest National Laboratory (PNNL) recently compiled publically available cladding hydrogen measurements and revised the hydrogen uptake models in the FRAPCON fuel rod performance code. In addition, PNNL quantified the standard deviation in these model predictions relative to the database. Table 1 of Reference 1 (shown below) summarizes the recommended changes to the FRAPCON-3.4 hydrogen uptake models for Zircaloy-2, Zircaloy-4, M5™, and ZIRLO™ based upon the expanded hydrogen database. The revised corrosion and hydrogen uptake models are documented in NUREG/CR-7022, Volume 1, Revision 1 (Reference 2).

Table 1 Hydrogen models in FRAPCON-3.4 and new model

Alloy	FRAPCON-3.4		New Model	
	Model	Std. dev.	Model	Std. dev.
<b>BWR</b>				
Zry-2 pre 1998	Eq. 2	10 ppm <sup>1</sup> NA <sup>2</sup>	Eq. 2	10 ppm <sup>1</sup> 54 ppm <sup>2</sup>
Zry-2 post 1998	Eq. 3	11 ppm <sup>1</sup> 61 ppm <sup>2</sup>	Eq. 3	13 ppm <sup>1</sup> 60 ppm <sup>2</sup>
<b>PWR</b>				
Zry-4	15% <sup>3</sup>	40 ppm	15.3% <sup>3</sup>	94 ppm
ZIRLO™	12.5% <sup>3</sup>	162 ppm	17.3% <sup>3</sup>	110 ppm
M5™	10% <sup>3</sup>	20 ppm	10% <sup>3</sup>	23 ppm

<sup>1</sup> standard deviation below 50 GWd/MTU

<sup>2</sup> standard deviation above 50 GWd/MTU

<sup>3</sup> pickup fraction

**ENCLOSURE**

### BWR Zircaloy-2

For boiling water reactor (BWR) conditions, a constant hydrogen pickup fraction does not fit the observed cladding hydrogen data. As a result, FRAPCON-3.5 (Reference 2) uses a burnup-dependent hydrogen concentration model. In addition, the recommended Zircaloy-2 model is divided between modern alloys (with tighter control of composition and second phase precipitation particle size) and legacy alloys. The best-estimate hydrogen uptake models are listed below:

Legacy alloys:

$$\begin{array}{ll} H = 47.8 \exp[-1.3/(1+BU)] + 0.316BU & BU < 50 \text{ GWd/MTU} \\ H = 28.9 + \exp[0.117(BU-20)] & BU \geq 50 \text{ GWd/MTU} \end{array}$$

Modern alloys:

$$H = 22.8 + \exp[0.117(BU-20)]$$

Where:

$$\begin{array}{l} H = \text{total hydrogen, wppm} \\ BU = \text{local axial burnup, GWd/MTU} \end{array}$$

Given the allowable range in composition within the Zircaloy-2 ASTM specification and the degree of flexibility and variability in manufacturing procedures between the fuel vendors, the staff has elected to adopt the more conservative legacy hydrogen uptake model. For this model, Table 1 of Reference 1 provides a standard deviation on the model prediction of 10 wppm below 50 GWd/MTU and 54 wppm above 50 GWd/MTU. To account for variability and uncertainty, the staff decided to use a +2-sigma on the model prediction. Figure 1 illustrates the best-estimate and +2-sigma model predictions along with the entire database. Examination of this figure reveals a discontinuity at 50 GWd/MTU where the larger standard deviation is first applied. In addition, application of the same standard deviation to even higher burnup suggests that the relative scatter in hydrogen content is becoming smaller. This is not likely the case. As a result, the staff developed a 1.40 multiplier of the model prediction which is approximately equal to 2-sigma at the lower burnup. This new model is shown on Figure 2. The application of this multiplier removes the discontinuity and ensures that the model reflects a larger uncertainty at higher concentrations of hydrogen.

An acceptable BWR Zircaloy-2 hydrogen uptake model is provided below.

$$\begin{array}{ll} H = (47.8 \exp[-1.3/(1+BU)] + 0.316BU) * 1.40 & BU < 50 \text{ GWd/MTU} \\ H = (28.9 + \exp[0.117(BU-20)]) * 1.40 & BU \geq 50 \text{ GWd/MTU} \end{array}$$

Where:

$$\begin{array}{l} H = \text{total hydrogen, wppm} \\ BU = \text{local axial burnup, GWd/MTU} \end{array}$$

References 3 and 4 describe an independent Zircaloy-2 hydrogen uptake model along with hydrogen data from various sources. A comparison of best-estimate predictions with these two models is provided in Table 1 below. Examination of this table reveals reasonable agreement up to 60 GWd/MTU. At higher exposures the FRAPCON model accelerates relative to the Heck model. Given the lack of data in this region, the more conservative FRAPCON model is

preferable. In addition, examination of the data scatter shown in References 3 and 4 supports the 1.40 multiplier on the model prediction.

### PWR Zirconium Alloys

Corrosion rates and the amount of corrosion at discharge vary widely across the PWR fleet due to alloy composition, operating conditions, and residence time. Fuel vendors have approved fuel performance analytical tools along with corrosion models. In general, these corrosion models are capable of predicting a best-estimate corrosion thickness as a function of residence time (EFPD) and local operating conditions (fuel duty).

Hydrogen data collected on PWR zirconium alloy cladding does not exhibit the same breakaway hydrogen uptake at higher fluence levels as observed in the BWR Zircaloy-2 data. However, the pickup fraction does appear to be alloy-specific. As a result, a constant hydrogen pickup fraction will be proposed for each zirconium alloy.

These hydrogen pickup fractions should be used, along with a best-estimate prediction of the peak oxide thickness using an approved fuel rod thermal-mechanical model, to estimate the cladding hydrogen content.

Table 1 of Reference 1 defines the following best-estimate hydrogen pickup fractions and standard deviation relative to the hydrogen database for Zircaloy-4, ZIRLO™, and M5™ cladding.

Zircaloy-4	- 15.3% pickup , 94 wppm standard deviation
ZIRLO™	- 17.3% pickup , 110 wppm standard deviation
M5™	- 10.0% pickup , 23 wppm standard deviation

Figure 4, 6, and 8 of Reference 1 show predicted versus measured hydrogen concentration along with a  $\pm 2$ -sigma bound for Zircaloy-4, ZIRLO™, and M5™ cladding respectively. Similar to the above BWR model, the staff has decided to apply a +2-sigma on the model prediction to account for variability and uncertainty in the database. However, the application of a constant, additive standard deviation has negative attributes: (1) overly conservative when applied to low burnup, low corrosion fuel rods and (2) no recognition for larger scatter in highly corroded fuel rods.

With consideration of the extent and variability of the supporting database, the staff developed upper bound pickup fractions. As described in Reference 1, the expanded Zircaloy-4 hydrogen database has over 280 measurements. Figure 3 shows predicted versus measured hydrogen concentration using the above 15.3% pickup fraction. With over 280 data points, a 95/95 non-parametric statistical upper bound could be derived. However, given all of the variables (e.g., alloy content, operating conditions) and uncertainties, there is no guarantee that the data is actually poolable. Instead, the staff elected to iterate on pickup fraction until a reasonable upper bound prediction was obtained. Figure 4 shows predicted versus measured hydrogen content assuming a 20% pickup fraction. Examination of the figure reveals that a vast majority of the data is conservatively predicted.

For ZIRLO™ cladding, the hydrogen database is limited to 60 data points. As such, a 95/95 non-parametric statistical upper bound would need to bound 100% of the data and likely be

overly conservative. Figure 5 shows predicted versus measured hydrogen content using PNNL's recommended 17.3% pickup fraction. For the reasons stated above, the staff elected to iterate on pickup fraction until a reasonable upper bound prediction was obtained. Employing a bounding pickup fraction of 25% shifts the predictions, as shown in Figure 6. Examination of this figure reveals that a reasonable majority of the data is conservatively predicted.

For Optimized ZIRLO™ cladding, applicants may use the bounding 25% pickup fraction along with an approved alloy-specific corrosion model.

For M5™ cladding, the hydrogen database is limited to less than 20 data points. As such, a 95/95 non-parametric statistical upper bound of this database is not possible. Figure 7 shows predicted versus measured hydrogen content using PNNL's recommended 10.0% pickup fraction. For the reasons stated above, the staff elected to iterate on pickup fraction until a reasonable upper bound prediction was obtained. Employing a bounding pickup fraction of 15% shifts the predictions, as shown in Figure 8. Examination of this figure reveals that a reasonable majority of the data is conservatively predicted.

Based on the above discussion, the staff finds the following bounding hydrogen pickup fractions acceptable.

Zircaloy-4	- 20% hydrogen absorption
ZIRLO™	- 25% hydrogen absorption
Optimized ZIRLO™	- 25% hydrogen absorption
M5™	- 15% hydrogen absorption

These hydrogen pickup fractions should be used, along with a best-estimate prediction of the peak oxide thickness using an approved fuel rod thermal-mechanical model, to estimate the cladding hydrogen content.

#### Applicability

The hydrogen models are applicable to currently approved commercial alloys up to their respective limits on fuel rod burnup, corrosion, and residence time.

The hydrogen models are not applicable to fuel rods which experience oxide spallation.

TABLE 1: Comparison of Hydrogen Uptake Models

Local Exposure (GWd/MTU)	Best-Estimate Hydrogen Prediction	
	FRAPCON-3.4 Legacy	Heck (2008)
0	15	18
10	46	42
20	51	48
30	55	48
40	59	55
50	62	80
60	137	150
70	376	260

FIGURE 1: Zircaloy-2 Hydrogen Model, +2-Sigma Prediction Versus Data

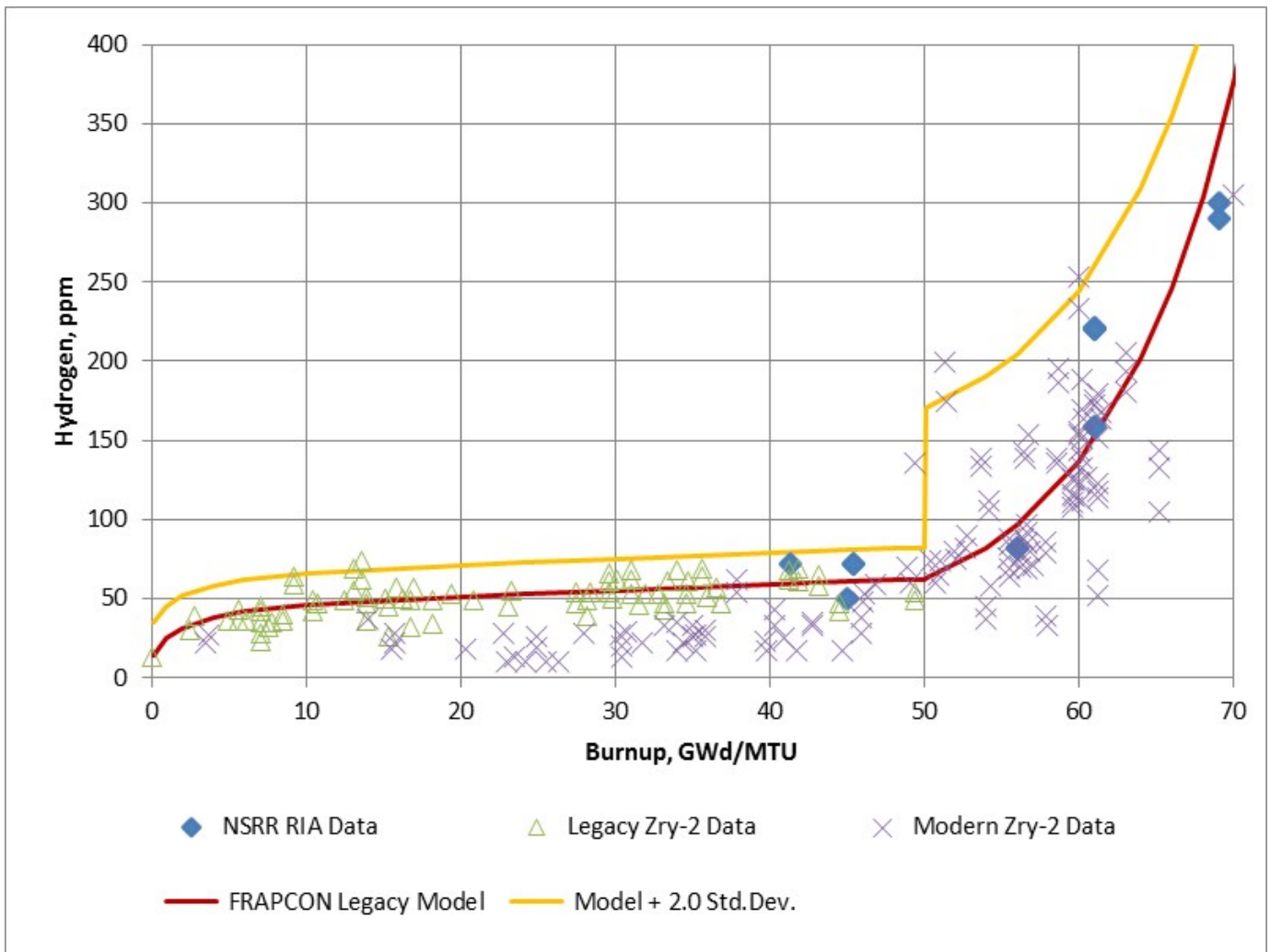




FIGURE 2: Zircaloy-2 Hydrogen Model, 1.40 Multiplier Prediction Versus Data

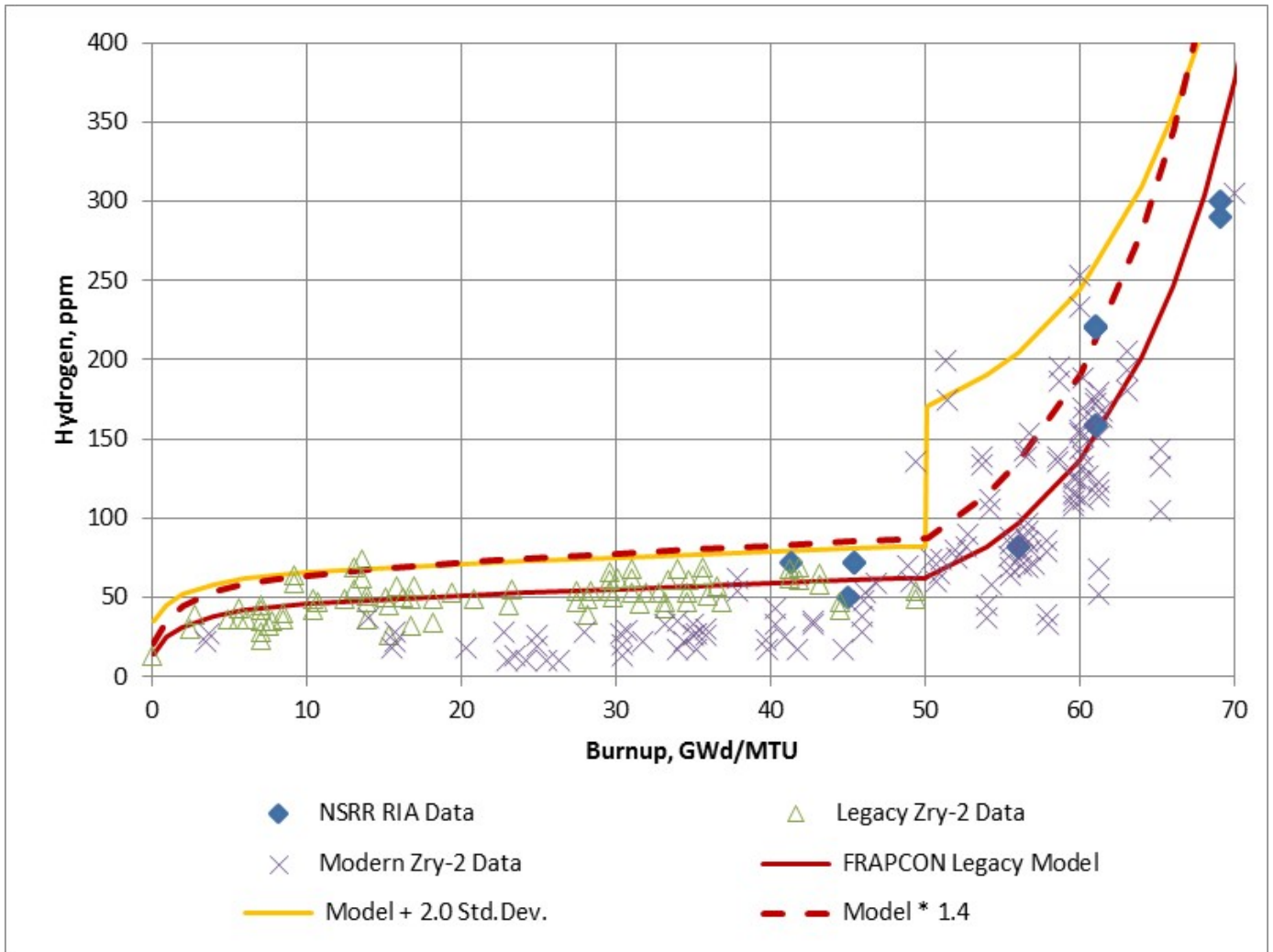


FIGURE 3: Zircaloy-4 Predicted Versus Measured Hydrogen Content, 15.3% Pickup  
(Figure 4 of Reference 1)

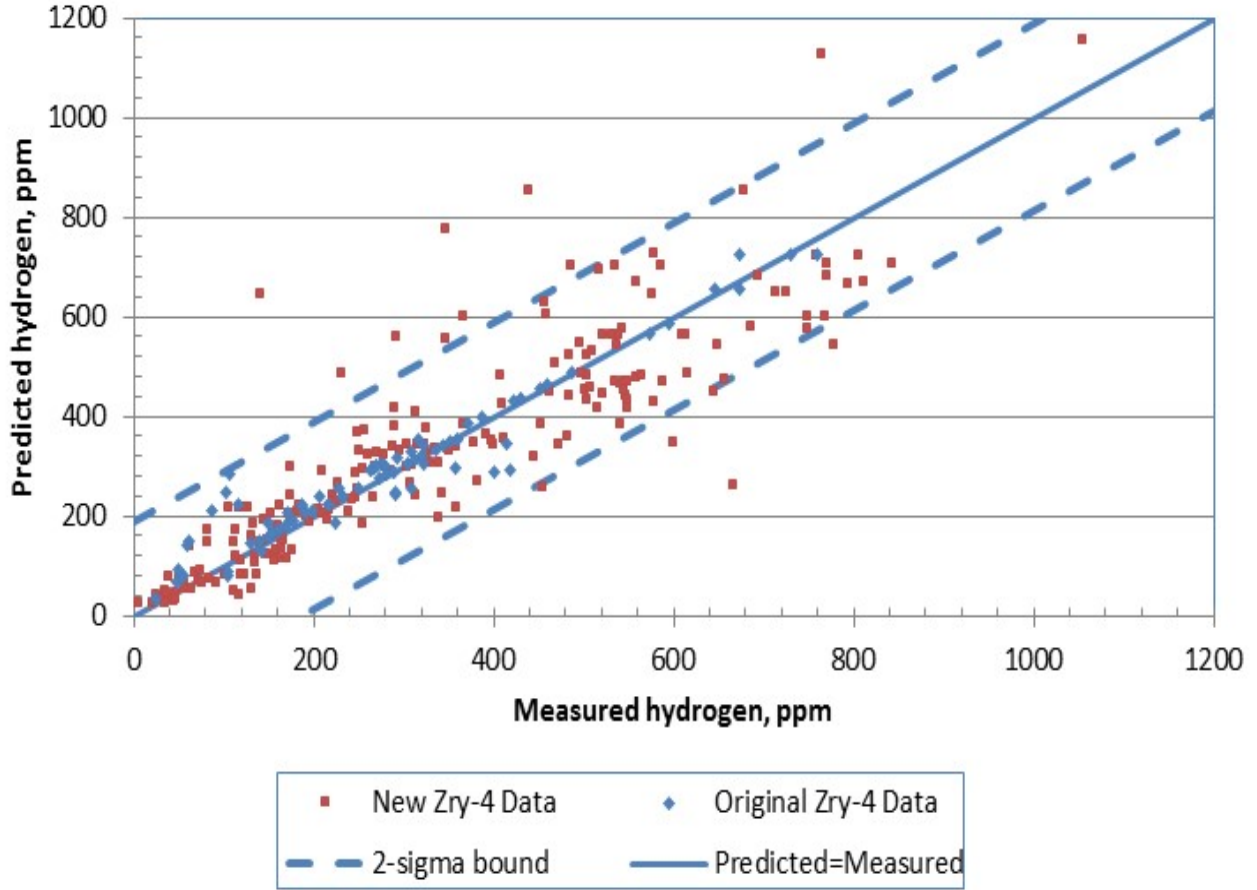


FIGURE 4: Zircaloy-4 Predicted Versus Measured Hydrogen Content, 20.0% Pickup

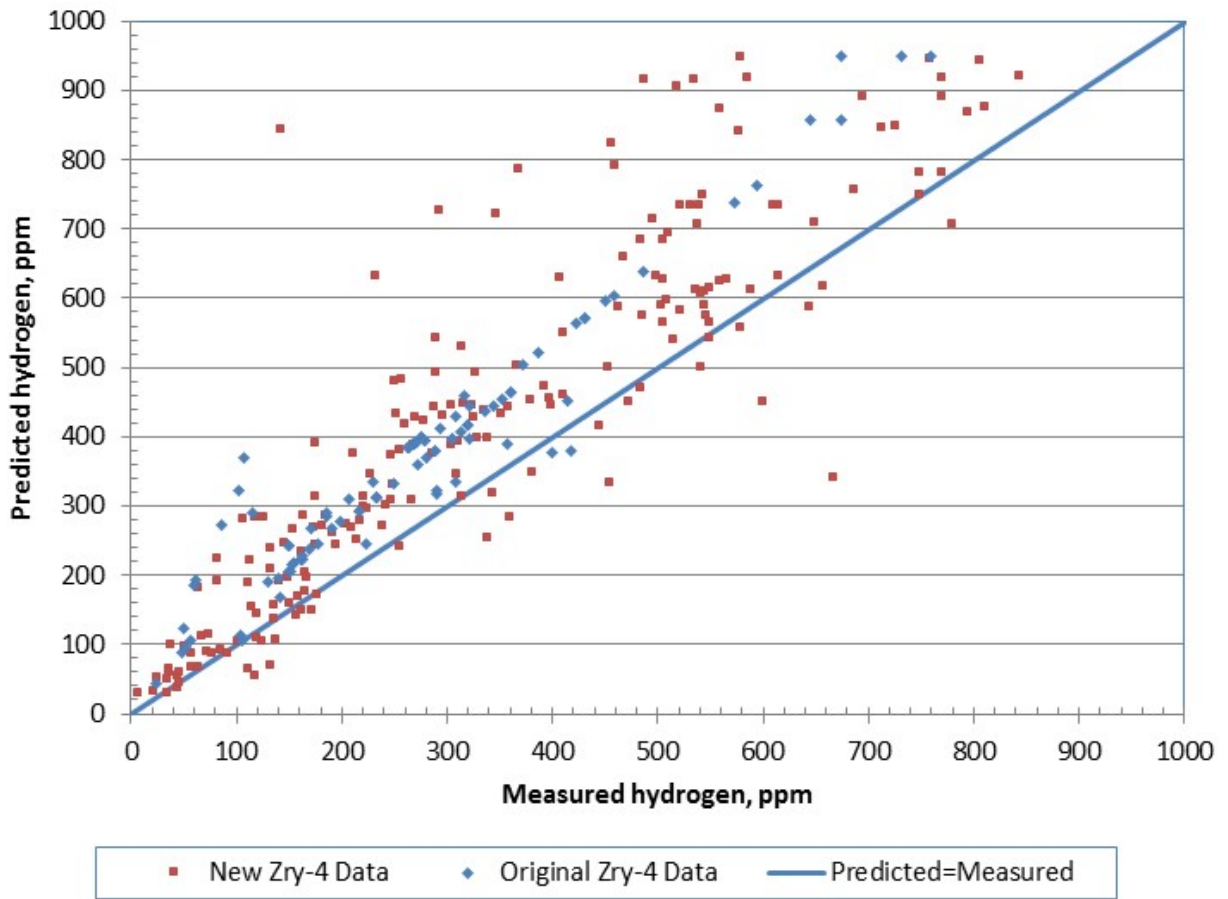


FIGURE 5: ZIRLO™ Predicted Versus Measured Hydrogen Content, 17.3% Pickup  
(Figure 6 of Reference 1)

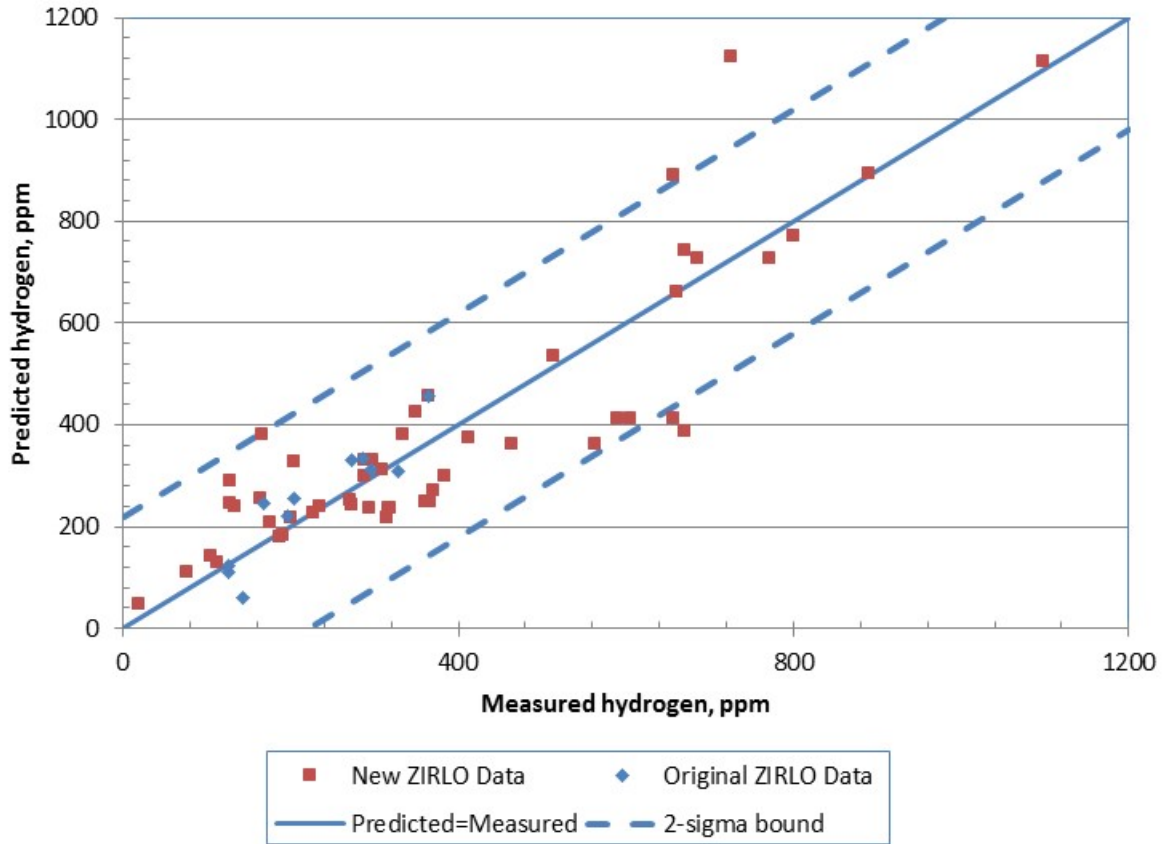


FIGURE 6: ZIRLO™ Predicted Versus Measured Hydrogen Content, 25.0% Pickup

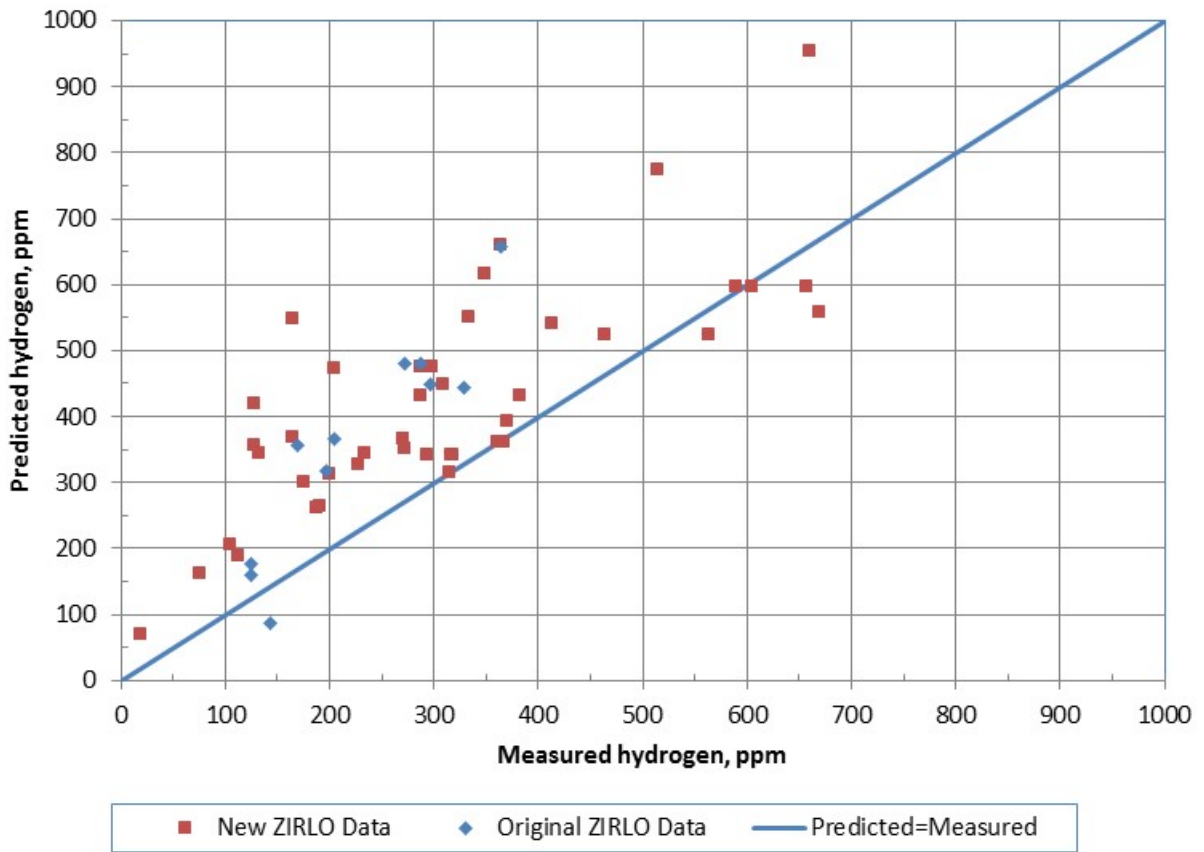


FIGURE 7: M5™ Predicted Versus Measured Hydrogen Content, 10% Pickup  
(Figure 8 of Reference 1)

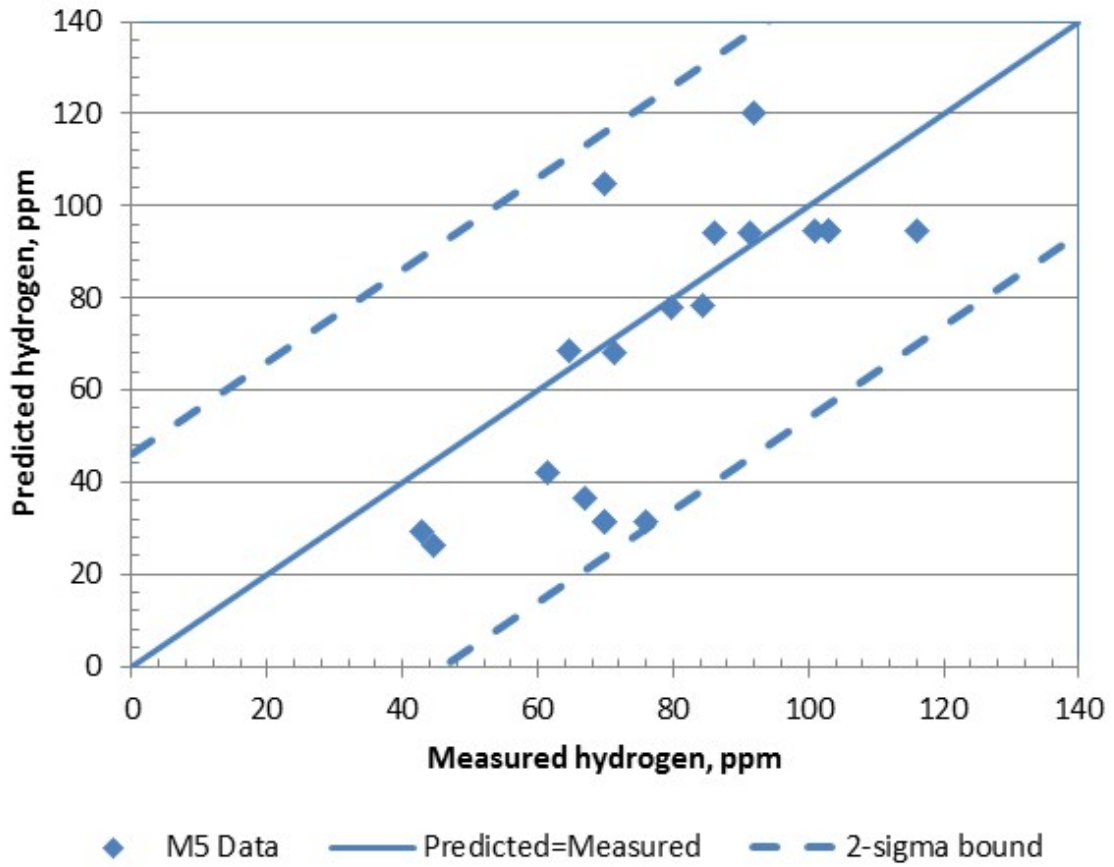
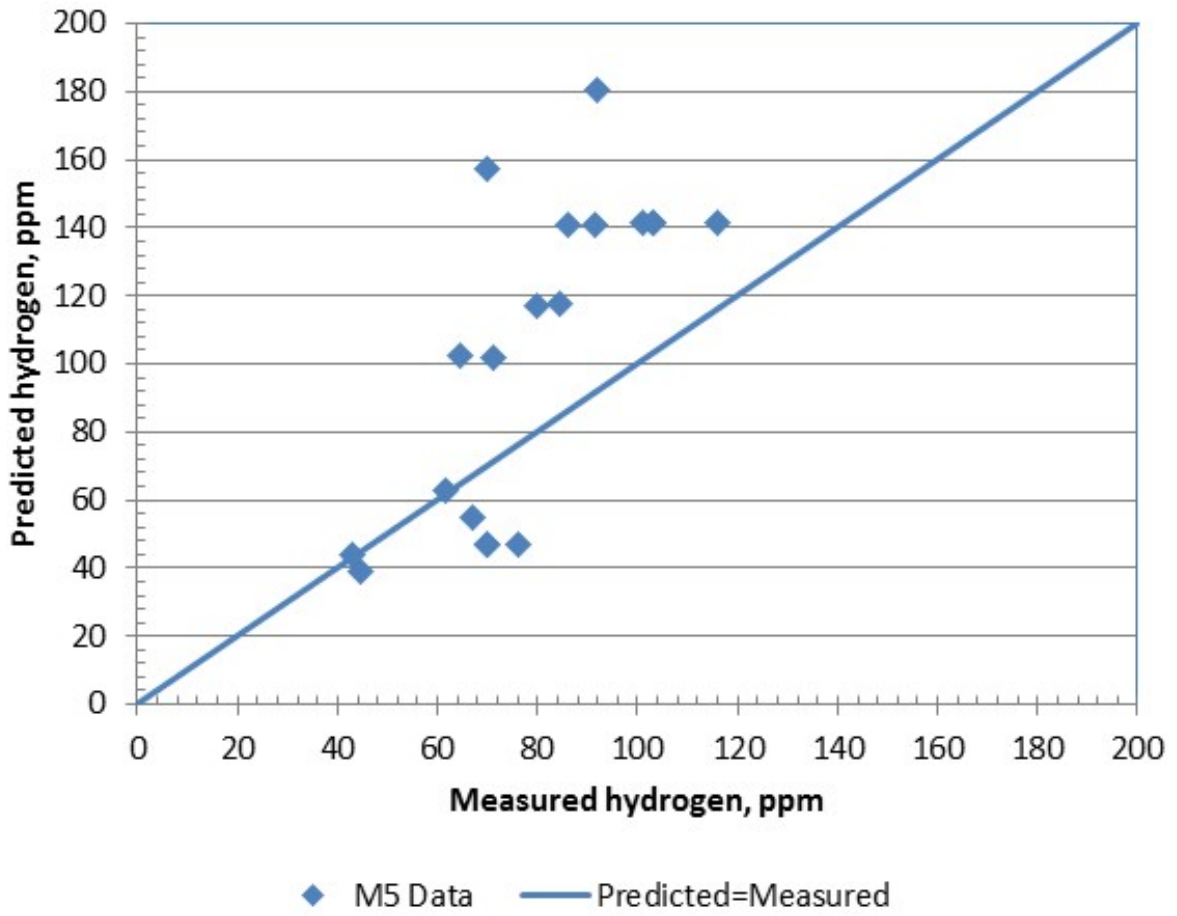


FIGURE 8: M5™ Predicted Versus Measured Hydrogen Content, 15% Pickup



### 3.0 **REFERENCES**

1. K. Geelhood and C. Beyer, "Hydrogen Pickup Models for Zircaloy-2, Zircaloy-4, M5<sup>TM</sup> and ZIRLO<sup>TM</sup>," 2011 Water Reactor Fuel Performance Meeting, Chengdu, China, September 11-14, 2011.
2. NUREG/CR-7022, "FRAPCON-3.5: A Computer Code for the Calculation of Steady-State, Thermal-Mechanical Behaviour of Oxide Fuel Rods for High Burnup," Volume 1, Revision 1, October 2014.
3. P. Rudling, "Zr alloy corrosion and hydrogen pickup," ANT International, December 2013.
4. C. Heck, "BWR Control Rod Drop Accident: Methodology, Application and Regulatory Compliance," NRC Workshop, September 24-25, 2008.