CENTER FOR NUCLEAR WASTE REGULATORY ANALYSES

TRIP REPORT

SUBJECT: U.S. Department of Energy (DOE) Waste Package Materials Performance Peer Review Panel—Final Report Meeting (20.01402.571)

DATE/PLACE: March 18–19, 2002
Las Vegas, Nevada

AUTHOR: Gustavo A. Cragnolino

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PERSONS PRESENT: The meeting was attended by approximately 60 people. Tae Ahn from NRC was also present.

BACKGROUND AND PURPOSE OF TRIP:

The purpose of the trip was to attend the presentation in a public meeting of the final report prepared by the U.S. Department of Energy (DOE) Waste Package Materials Performance Peer Review Panel. The presentation by panel members was combined with presentations by technical staff from Bechtel SAIC Company (BSC) and Lawrence Livermore National Laboratory (LLNL). The agenda of the meeting is enclosed.

SUMMARY OF PERTINENT POINTS:

After brief introductory remarks, Joe Payer (Case Western Reserve University), the peer review chairman, presented an overview of the final report (available in www.ymp.gov) in which the main findings of the panel were covered. In his presentation he acknowledged the other members of the panel: John A. Beavers [CC Technologies Laboratory (CCTL)], Thomas M. Devine (University of California at Berkeley, UC), Gerald S. Frankel [The Ohio Sate University (OSU)], Russell H. Jones [Pacific Northwest National Laboratory (PNNL)], Robert G. Kelly [University of Virginia (UV)] and Ronald M. Latanision [Massachusetts Institute of Technology (MIT)]. He emphasized the broad range of expertise in corrosion science and engineering of the panel members, and noted that the panel was assisted by 12–15 subject matter experts from the U.S. and abroad on specific issues (i.e., geochemistry, hydrology, physical metallurgy, welding, etc). Among other contributions, Payer acknowledged the participation of NRC and CNWRA staff in the technical exchange that took place in Cleveland on July 24, 2001. The review considered the following areas: composition of aqueous environments, materials metallurgical stability, long-term uniform corrosion, localized corrosion, environmentally assisted cracking, including stress corrosion cracking and hydrogen effects, and contributing effects of design and fabrication factors on corrosion. The panel concluded that the current waste package design is likely to meet performance criteria for the repository, if some technical issues are favorably resolved. Although it was noted that the technical basis supporting the suitability of alloy 22 for the outer barrier of the waste package is substantial and growing, it was emphasized by the panel that there will always be uncertainty in the evaluation of the long term performance of the waste package in the repository as a result of the extremely

long life required. As an additional recommendation, the panel suggested the selection of a back-up alloy for the eventual case that following further testing alloy 22 does not perform as expected and the inclusion of a group of alloys more susceptible to various degradation modes than alloy 22 in specific tests for comparison purposes. The panel concluded also that Titanium Grade 7 might not be a suitable material of construction for the drip shield, mostly based on the reported observation of stress corrosion cracking in laboratory tests. Various technical concerns were presented emphasizing the need of a better determination of the realistic range of aqueous environments and a better integration between design and fabrication efforts and the testing and modeling of corrosion processes. Payer stated that uncertainties in the estimation of the long-term performance can be substantially reduced through further experiments and analysis, and that a large amount of necessary work, although already planned, remains to be completed to support performance assessment. The panel was concerned that adequate resources might not be allocated to complete the work needed to support design and fabrication of durable waste packages and encouraged the establishment of an External Advisory Board and the appointment of a senior, qualified individual with strong materials science and engineering background and management credentials to lead the waste package program.

This presentation was followed by brief remarks from Richard Spence (DOE) acknowledging the work of the panel and stating that the recommendations will be carefully evaluated and considered by the DOE.

In the public comments/questions period, Tae Ahn (NRC) asked Payer the following two questions: (i) Does the panel accept the TSPA analysis and results for alloy 22 and Ti Grade 7? and (ii) Did the panel consider the use of analog studies? Payer responded that (i) the evaluation of the model abstractions contained in TSPA was beyond the scope of the panel that was chartered to only evaluate corrosion process models and the supporting experimental methodology and data, and (ii) no evaluation of analogs was done by the panel with the exception of a report by a subject matter expert on corrosion in flue-gas desulfurization systems as industrial analog.

Alberto Sagues [Nuclear Waste Technical Review Board (NWTRB)] after commenting about the importance of analogs in supporting performance assessment of engineered materials, asked if the panel believed that the project is ready to extrapolate with reasonable assurance the behavior of the candidate engineered materials for many thousand of years. Payer responded that the panel was reasonably confident on the validity of the extrapolation but subject to two constraints: (i) no dramatic change of the expected environment and (ii) good control of microstructural effects that may become factors for premature damage.

Payer had a detailed presentation on repository conditions, degradation modes and waste package fabrication. In his overview of the evolution of the temperature, and physical and chemical conditions of the environment, he discussed the two operational modes under consideration: (i) the higher temperature operating mode (the current DOE base case) and (ii) the lower temperature operating mode (as evaluated by the DOE following recommendations of the NWTRB), as well as the impact of different preclosure times and ventilation conditions. No recommendation with respect to a preferred design was presented and several coupled processes of potential importance (e.g. drift collapse affecting temperature and relative humidity profiles) were not considered in the analysis. Nevertheless, Payer emphasized the strong need to redirect and balance the corrosion research effort to provide better technical support for the resolution of pending issues related to design and manufacture/fabrication operations. Specific

recommendations were listed, mostly related to welding operations and consideration of heat to heat variability on performance.

Kelly (UV) began his presentation on the composition of aqueous environments by stating that the prediction of corrosion behavior depends on the prediction of the environment. This apparently obvious statement reflects the prevailing concern about the evolution of the environment at Yucca Mountain, which is far less predictable than that of other potential repositories to be located in the saturated zone. The panel envisioned the aqueous environments formed on metal surfaces at Yucca Mountain as divided in three primary classes: (i) moist dust, (ii) scale and deposits, and (iii) crevice areas at metal-to-metal contact surfaces. The panel considered that full immersion of metal surfaces is a highly unlikely condition in the proposed repository. Kelly indicated that the panel found the approach used by the DOE project consistent with the current state of scientific understanding (i.e., the chemical divide concept to analyze the waters) but noted that a more comprehensive and in depth experimental and analytical modeling work is needed for defining the chemical composition of the three primary classes of environments listed above as related to their effect on various corrosion processes. The recommendation of the panel is to conduct a comprehensive experimental and modeling program for these three environmental conditions with the help of a task group of technical experts in the relevant disciplines (i.e., geochemistry, hydrology, corrosion, etc.). It appears that although the panel included the consideration of the source and effect of trace elements such as lead, mercury, arsenic, and bromide, it did not consider this aspect of the corrosion studies a high priority and recommended instead to evaluate how they may come into contact with the waste package surfaces. On the other hand, regarding microbial effects, the DOE approach of using a simple rate enhancement factor for passive corrosion was discouraged. Kelly, instead, recommended to focus the work in attaining an understanding of the effects of microbe metabolites on localized corrosion in welds and crevices.

Jones (PNNL) presented the panel findings regarding metallurgical stability and radiation effects. In the presentation he concluded that radiation damage is not a matter of concern even for the stability of passive films. The most significant potential degradation modes of the engineered barriers caused by metallurgical microstructural and microchemical changes were related to fabrication processes including both precipitation of intermetallic phases (the CNWRA work on thermal treatment at about 800 °C was quoted) and impurity segregation (sulfur and phosphorus were mentioned). The combined effect of segregation and long- range ordering was noted as matter of some concern that should be evaluated. Regarding the effect of repository operating conditions, the potential for anodic segregation of sulfur at the metal surface as a result of slow passive dissolution was suggested by Jones stating that a whole monolayer of adsorbed sulfur could occur in 180 yr assuming a corrosion rate of 0.01 μm/yr and a sulfur content of 100 atomic ppm in the bulk of the alloy. This surface segregation, observed in pure nickel, may lead to passivity breakdown of Ni -Cr- Mo alloys such as Alloy 22 as discussed by P. Marcus at the NWTRB Workshop on Long-term Passive Behavior.

The long- term uniform corrosion of passive metal was discussed by Devine (UC). After reviewing the DOE work in this area and other contributions reported in the literature, he indicated that the panel identified three factors that affect the stability of passive films, as follows: (i) changes in the intrinsic nature of the film, (ii) changes in the environment, and (iii) changes in the alloy itself. Devine discussed several aspects related to these three factors in relation to the DOE project current findings and plans, suggesting several areas of further work. They are: (i) to complete the characterization of the film structure and composition, (ii) to

investigate the effects of sulfur segregation and excursions of potential to transpassive conditions, (iii) to conduct an evaluation of temperature effects on corrosion rate at temperatures greater than 100 °C. Nevertheless, the panel concluded that failure of the waste package by uniform corrosion during the performance period is unlikely.

Jean Younker (BSC) provided a brief overview stating the BSC will convey to the DOE the results of its analysis of the peer review panel report by the end of April 2002 and introduced the various presentations by the BSC contractors dealing with the different topics of the report. She addressed the concerns of the panel regarding level of effort and allocation of resources emphasizing that the approach adopted is a risk-informed decision process to prioritize the overall work. Younker also noted that the transition from site recommendation to potential license application involves a focused effort on appropriateness of materials and conceptual designs/fabrication by using codes and standards whenever is necessary.

Greg Gdowski (LLNL) gave the presentation on the composition of the aqueous environment in contact with the WP and drip shield surfaces including the influence of dust, mineral deposits and their effect on heat transfer, and the deliquescence of salt mixtures. He defined a strategy to characterize the environment in aqueous solution "types' according to their corrosive nature and listed the parameters important for corrosion Calculations of relativity humidity as a function of temperature for various salt mixtures using the EQ3/6 code were presented indicating that a thermodynamic database based on Pitzer formulation is being developed for relevant species. Finally, he briefly described thin film corrosion testing using thermogravimetric analysis (TGA) and evaporative drip testing both extended to temperatures up to 170 °C.

Tammy Summers (LLNL) discussed the metallurgical issues related to potential degradation modes and the increased involvement of the staff in aspects related to waste package design and fabrication processes. She presented theoretical calculations and experimental results indicating that tetrahedrally close-packed precipitates and long-range ordering will not occur under projected repository conditions. In addition, Summers revisited calculations of phosphorus segregation in carbon steel prepared for the viability assessment indicating that similar calculations are planned for Alloy 22. Finally, she noted that the effects of processing on potentially detrimental metallurgical changes are being evaluated.

Joe Farmer (LLNL) presented the information on long-term uniform corrosion of passive metals by reviewing the approach and data from the Long-Term Corrosion Test Facility (LTCTF) at LLNL, data from some recent cyclic potentiodynamic polarization and potentiostatic tests, and studies of the passive film formed on alloy 22 and Ti grade 7 using the atomic force microscope and surface analysis techniques such as XPS. This discussion did not add much to previous presentations on the subject at the kick-off meeting of the panel or when the interim report was issued.

Frankel (OSU) presented a critical assessment of the approach used by the DOE for evaluating the possibility of localized corrosion and questioned both the validity of the corrosion potential values used by the DOE in the model abstraction for the TSPA, as well as the adoption as critical potentials of threshold potentials associated with current increases in cyclic potentiodynamic polarization obtained with non-creviced specimens. He noted that the criterium based on the corrosion potential and critical potential is valid if and only if the corrosion potential can be modeled and verified with long term experimental measurements and the critical potential is defined as the repassivation potential for crevice corrosion. He presented an

additional set of recommendations including the use of creviced samples, more aggressive solutions, oxidizing species, and higher temperatures to define regimes of susceptibility to localized corrosion. Frankel recommended to take into consideration various metal surface conditions and the classes of aqueous environments to be formed on waste package surfaces, as discussed by Kelly, including the effect of moist dust as well as that of scale and deposit formation. He recommended the development of more realistic models of localized corrosion propagation and arrest.

Beavers (CCTL) presented the panel findings regarding stress corrosion cracking. He questioned the validity of the slip dissolution/film rupture model for the repository conditions (predominance of residual stresses over cyclic or sustained applied loads) indicating that this model does not contain a threshold stress or stress intensity factor. Beavers also noted that the DOE approach of using a threshold stress instead of stress intensity may not be conservative because cracks usually initiate (with the exception of exposure to very potent environments) at preexisting defects. He recommended the consideration of alternative stress corrosion cracking (SCC) models, and the evaluation of Alloy 22 within and beyond the expected environmental bounds, in particular the role of lead. Concerns with fabrication and metallurgical changes as result of post-weld treatments were also raised and several recommendations were provided for further testing. Beavers expressed the view of the panel regarding Ti Grade 7 saying that the project should select an alternative material if it is confirmed that Ti Grade 7 is susceptible to SCC or hydride cracking under realistic repository conditions.

Latanision (MIT) make a brief presentation on hydrogen effects suggesting that detrimental effects can be promoted by solute segregation at grain boundaries or ordering reactions promoting planar slip. He noted that susceptibility of Alloy 22 to hydrogen embrittlement has not been studied and suggested that the project undertake some experimental work to explore the possibility of this phenomenon in cold worked, and cold worked and thermally aged Alloy 22.

Joe Farmer (LLNL) presented the planned program for the evaluation of localized corrosion resistance of the WP and DS materials. Most of the work previously conducted by LLNL was presented to highlight the extremely good resistance to localized corrosion exhibited by Alloy 22 under the assumed realistic environmental conditions used in the LTCTF. A limited number of electrochemical tests conducted using uncreviced specimens was also presented as an additional evidence. The inhibiting role of some anions was emphasizing but not in depth analysis of their role and their inhibition efficiency was described. The discussion on the validation of the crevice model was limited to some ad-hoc experiments. However, these experiments were not used to support the validity of the threshold potentials minus corrosion potential derived expressions used in the TSPA.

Gerald Gordon (Framatome) made the presentation on WP and DS stress corrosion cracking. The approach to mitigate the initiation and propagation of stress corrosion cracks was the main subject of his presentation. Data on SCC of Alloy 22 and Ti Grade 7 was presented , covering a range of material conditions in the case of Alloy 22 (i.e., cold worked, thermal treated, and welded tensile specimens were used). Whereas crack growth of Alloy 22 was observed under cyclic loading conditions on compact tension specimens in aerated and saturated alkaline solutions based on J-13 water at 105 °C, no crack growth was detected under constant load with the exception of a 20 percent cold worked compact tension specimen. No SCC was observed in constant loaded smooth tensile specimens at stresses well above the yield strength in the same type of environments. On the contrary, compact tension specimens of Ti grade 7

exhibited crack growth under constant load conditions and smooth tensile specimens showed SCC at 75–95 percent of the yield strength. Gordon presented recent results of slow strain rate tests of mill-annealed Alloy 22 in various simulated groundwater environments using an applied potential of about 400 mV ($Ag/AgCl/KCl_{sat}$) which is about 360 mV_{SCE}. Relatively severe transgranular SCC was observed only in simulated concentrated water (pH 10.3) at 73 °C. Although the applied potential is relatively high in these tests with respect to the expected corrosion potential, this observation reveals that the assessment of the environmental effects on cracking is by no means complete. Environments that are apparently innocuous may promote SCC of Alloy 22 within specific potential ranges not yet explored in the DOE studies. On the other hand, testing in specific environments does not imply that the whole possible range of realistic environments is well covered in the current DOE program.

In the closing remarks, Payer summarized the task of the peer review panel and provided the complete listing of the special topic reports prepared by members of the panel and the subject matter experts. These reports cover specific topics related to corrosion, chemistry and metallurgical factors, as well as design and fabrication issues. The reports should be already available because their publication was planned for the end of March 2002.

IMPRESSIONS/CONCLUSIONS:

Overall, attendance at this meeting was beneficial. The meeting provided an excellent opportunity to listen to the opinions of the panel members and to judge their confidence on the DOE approach. It is expected that the final report and the recommendations will help DOE to focus and prioritize the extensive work needed to closed pending CLST and ENFE technical issues according to the existing agreements. The recommendation on the need of a better integration between the staff involved in the corrosion testing and modeling program and the group in charge of the design and fabrication is extremely important in redirecting certain aspects of the DOE program. A parallel integration between the performance assessment staff and the one involved in testing and modeling, already initiated, should be reinforced because there is still a lack of connection between several process models and the corresponding abstractions.

PROBLEMS ENCOUNTERED:

None

PENDING ACTIONS:

None

RECOMMENDATIONS:

Obtain the special topic reports to evaluate the full range of studies conducted and the specific recommendations.

REFERENCES:

The handouts of the various presentations are available upon request from the author.

SIGNATURES:

Gustavo Cragnolino
Staff Scientist

4/23/02
Date

CONCURRENCE:

for V. JAIN

Vijay Jain, Manager
Corrosion Science & Process Engineering

4/24/02
Date

Budhi Sagar
Technical Director

4/24/2002
Date

GC:re

Waste Package Materials Performance Peer Review Panel
Presentation of Final Report
March 18-19, 2002
JW Marriott, Las Vegas, NV

Monday-March 18, 2002

8:15 a.m.	**Welcome and Introductions**
8:30 a.m.	**Introduction; Main Findings** Joe Payer, Case Western Reserve University & Peer Review Chairman
9:10 a.m.	**DOE Comments** Richard Spence - Dy mgr Office of Proj. Execution, YMP
9:20 a.m.	**Public Comments/Questions from the Audience**
9:40 a.m.	**Break**
10:00 a.m.	**Repository Conditions, Degradation Modes and Waste Package Fabrication** Joe Payer, Case Western Reserve University
10:40 a.m.	**Composition of Aqueous Environments** Robert Kelly, University of Virginia
11:20 a.m.	**Materials: Metallurgical Stability and Radiation Effects** Russell Jones, Battelle-Northwest
12:00 p.m.	**Lunch**
1:15 a.m.	**Long-Term Uniform Corrosion of Passive Metals** Thomas Devine, Jr., University of California, Berkeley
1:55 p.m.	**Public Comments/Questions from the Audience**
2:15 p.m.	**Break**
2:35 p.m.	**DOE comments** - Richard Spence
2:45 p.m.	**Overview** Jean Younker, Bechtel SAIC Company (BSC)
2:55 p.m.	**Composition of Aqueous Environments** Greg Gdowski, LLNL
3:30 p.m.	**Materials: Metallurgical Stability** Tammy Summers, LLNL
4:05 p.m.	**Long-Term Uniform Corrosion of Passive Metals** Joe Farmer, LLNL
4:40 p.m.	**Public Comments/Questions from the Audience**
5:00 p.m.	**Adjourn**

Waste Package Materials Performance Peer Review:
Presentation of Final Report
March 18-19, 2002
JW Marriott, Las Vegas, NV

Tuesday-March 19, 2002

8:00 a.m.	**Welcome and Introductions**
8:15 a.m.	**Localized Corrosion** ***(note cover 3.11 ennoblement here)*** Gerald Frankel, The Ohio State
8:55 a.m.	**Stress Corrosion Cracking** John Beavers, CC Technologies Laboratories
9:40 a.m.	**Hydrogen Effects** Ronald Latanision – Massachusetts Institute of Technology
10:10 a.m.	**Break**
10:30 a.m.	**Localized Corrosion** Joe Farmer, LLNL
11:05 a.m.	**Stress Corrosion Cracking** Gerald Gordon, BSC
11:40 a.m.	**Public Comments/Questions from the Audience**
12:10 p.m.	**Closing Remarks** Joe Payer, Peer Review Chairman
12:30 p.m.	**Adjourn**