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MEMORANDUM FOR: John T. Greeves, Acting Chief
Engineering Branch
Division of Waste Management

FROM: Michael Tokar
Engineering Branch
Division of Waste Management

SUBJECT: TRIP REPORT - BATTELLE COLUMBUS LABORATORIES ACRS INFORMATION MEETING ON NRC HLW MATERIALS PROGRAM - DECEMBER 6, 1983

Background

On December 6, 1983, I attended the subject meeting at Battelle Columbus Laboratories (BCL), Columbus, Ohio. The meeting was held at the request of Dr. Paul G. Shewman, ACRS member and Chairman of the Metallurgical Engineering Department at Ohio State University. The meeting was characterized by Dr. Shewman as an "information gathering." On that basis, it was not posted in the Federal Register and was not open to the public or other governmental agencies.

The subject of the meeting was the NRC-BCL program on Long-Term Performance of Materials Used for High-Level Waste Packaging (FIN B-6764) for which Dr. K. S. Kim is the NRC-RES Project Officer and Dr. D. Stahl is the BCL Project Manager. This program, scheduled to be a five-year study, is currently in its second year. The program has been subject to adverse criticism from ACRS and others.

Dr. Shewman was accompanied by two consultants: (1) Dr. Thomas Kassner, a metallurgist from Argonne National Laboratory and (2) Dr. Dennis Readey, Ceramics Department Head at Ohio State. The NRC participants included, in addition to myself, Dr. Kim, Dr. Michael McNeill (the NRC-RES Project Officer for the Container Materials Task), and Dr. Frank Arsenault (RES Director of the Division of Health, Siting and Waste Management).

A meeting agenda is attached. Following opening remarks by Dr. Frederick Milford (BCL Director of Research), Dr. Arsenault and Dr. Shewman, presentations were made by the individual task leaders for six of the seven principal programmatic tasks; no presentation was made on the task

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on Accelerated Testing/Statistics due to a lack of new information. Some brief comments about each presentation are provided below, followed by some general impressions of the meeting and program.

Comments on Specific Tasks

Waste Forms - The two major waste form task goals were stated to be (1) to define the status of the waste form at the time of canister failure (i.e., at 300 to 1000 years), and (2) to achieve a better understanding of the mechanisms for release of radioactive material to repository water. The general approach taken was to (a) identify, (b) evaluate experimentally, and (c) rank in order of importance the parameters critical to the two major areas of concern. Borosilicate glass has received about 85% of the effort so far. Four glass compositions (PNL 76-68, PNL 77-260, SRL-31, and SRL-165) are under investigation. Several leaching/corrosion variables of importance have been identified, among them the Eh, which is very difficult to measure, and which, in fact, the BCL team seems not to comprehend very well. Both Dr. Shewman and Dr. Readey raised questions concerning the temperatures used in the leaching/corrosion experiments relative to expected temperature conditions at the time of container breach (~250°C vs.<<100°C, respectively). Dr. Readey asked why leaching experiments had been conducted in reagent water and basalt/water conditions but not in brine. Other questions focused on the fact that the glasses under investigation did not contain any waste material and that the data being taken were, therefore, on nonprototypic materials in which radiation effects could not be quantified directly.

Overpack Corrosion - The two systems that have received attention so far are Ti Grade 12-salt and Cast Steel-basalt. In the Ti Grade 12-salt system, it was said that an objective was to reproduce BNL and other corrosion data. The effects of iron contamination and heat transfer and temperature gradients on corrosion performance were studied using autoclaves. As a result of crevice corrosion and stress corrosion cracking (SCC) problems with Ti alloys, work is now being focussed on the low carbon (1018) steel-basalt system. Electrochemical test results (polarization curves) were presented for various (doped and undoped, cast and wrought) 1018 steels. Dr. Kassner raised several technical questions regarding the details of the experimental procedure and the advisability of performing electrochemical rather than purely mechanical tests. There was considerable debate on this point. Dr. Kassner also questioned whether the test conditions were representative of repository conditions. It was implied that the results might not be applicable. Dr. McNeill

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pointed out that we did not know what the exact repository conditions would be.

Internal Corrosion - In this program element the focus has been on initial high temperature contact (between the glass and canister materials) effects, long-term intermediate temperature contact phenomena, and diffusion of corrosive components. The experimental procedure involves immersing coupons of steel in molten glass at various temperatures and measuring parameters such as corrosion pit depth, weight loss, general corrosion product thickness and composition, etc. Most tests have so far been conducted at fairly high temperature (900°C). At these temperatures and test conditions, there are readily observable and measureable effects, but their relevance to long-term performance was not clear.

Hydrogen Embrittlement of Container Materials - This task currently involves the investigation of hydrogen effects on cast and wrought low-carbon steels that are candidate overpack materials. A major portion of the effort so far has gone into the preparation and characterization of clean and doped (with S and P) castings. Some tensile and fracture-toughness tests have been conducted. The ductility of the doped steel is low, apparently due to the presence of manganese sulfide inclusions at the grain boundaries. Future work will concentrate on fracture-toughness tests to be conducted in hydrogen (and nitrogen) to observe the effects of structure (cast versus wrought) and chemical composition (clean versus doped).

Separate-Effects Analysis - The stated objective of this task was to develop a quantitative understanding of individual waste package degradation processes using "physically based models," an example of which is a Battelle water-chemistry/glass-dissolution model that uses temperature, concentrations of elements, water flow rate, etc. as input and glass-dissolution rate, glass-saturated concentrations, speciation, and pH as output. Two-way interaction with the experimentalists was also said to be a major objective. Current activities involve the development of models for waste form degradation, localized and general corrosion of the container, and water chemistry (including radiolysis). Dr. Digby D. MacDonald, Professor of Metallurgical Engineering of Ohio State University and Director of its Fontana Corrosion Center, has served as a consultant in the development of a corrosion model. He was not present at this meeting, however. Some modeling results were presented for glass dissolution, general corrosion, and pitting corrosion. As yet there appears to have been little verification of the models with data.

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System Performance - The stated objectives of this were to (1) develop an improved understanding of phenomena that affect waste package performance at the system level and (2) to provide technical and analytical support to the separate effects task and experimental effort. The work has centered on studies of water radiolysis mechanisms, using the WR-20 code developed at ANL several years ago. WR-20 is a very slow running code. Therefore, BCL is attempting to acquire a Chalk River code called MAKSIMA-CHEMIST. Energy deposition rates are being calculated using a code called ANISN. Dr. Kassner suggested using radiolysis data now being generated at Dresden to check the calculations.

General Impressions

As an information exchange, I believe the meeting served its purpose well. I personally came away from the meeting with a much better understanding and impression of the program than I had acquired initially from the annual report (NUREG/CR-3405). Dr. Stahl offered to send me a copy of the work plan for year two. That should be helpful in following future progress.

It is clear that there has been a significant change in the overall program objective. In the annual report (and I understand also in the original program Statement of Work), it is indicated that the ultimate objective of the program is to "develop a method for predicting the containment life and subsequent radionuclide release rate of the waste package for very long times under expected repository conditions." The current stated objective is to "develop a sufficient understanding of the processes which can lead to waste package containment failure and subsequent release of radionuclides in support of the NRC's compliance assessment of candidate license applications." The current objective is more likely to be realized, taking into account the available resources and program schedule.

It is also clear that there has been good progress made since the annual report. This is quite important to the ultimate success of the program inasmuch as there had seemed to be insufficient progress made, or inappropriate emphasis placed, on some program elements in the first year. For example, a lot of resources (staff time and money) had been spent during the first year on melting, casting, and characterizing low carbon steel ingots when steels having a range of sulfur and phosphorous contents probably could have been purchased commercially. This is a matter of emphasis, however, rather than a glaring weakness. Other areas that might be de-emphasized somewhat are the high temperature corrosion

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of stainless steel in molten glass, the high pressure testing in hydrogen, and the mechanical property measurements in nitrogen. Basically, however, the overall program seems to be moving along reasonably well at this time, considering the fact that it faces a moving target because of the changing or evolving DOE waste package design, experimental data base, and analytical development. To provide maximum benefit from this program, BCL must strive to stay abreast of DOE test results and waste package design plans.

As indicated in the meeting agenda, there was a tour of the laboratory facilities during the early afternoon, followed by a brief "concluding discussion" with Dr. Shewman, his consultants, and the BCL task leaders. Dr. Shewman stated that the meeting had been informative, but he offered no specific conclusions or criticisms. He indicated that he would meet later with his consultants, Dr. Readey and Dr. Kassner, to discuss their reactions to what had been presented. It is my understanding that some sort of report will be provided by Dr. Shewman to the ACRS, but it is not at all clear what form the report will take or what date it will be issued by. Inasmuch as Dr. Shewman will be starting a sabbatical leave around the first of the new year, I would expect the report to be readied before then. It is impossible at this point to speculate with any confidence on what the report will say. However, because many of questions raised during the meeting concerned the usefulness or applicability of accelerated or high temperature testing to actual repository conditions, it would not be surprising if some comments addressed that issue (if any specific technical issues are mentioned). I also came away from the meeting with the impression that Dr. Shewman wants NMSS to monitor this program more actively to ensure that the information generated will be useful in licensing.

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Michael Tokar
Engineering Branch
Division of Waste Management

Enclosure:
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TENTATIVE AGENDA

FOR THE ACRS TECHNICAL REVIEW MEETING

December 6, 1983

8:15 - 8:30 am	Introductions/Coffee	Conference Room B
8:30 - 8:35 am	Welcome to Battelle	Dr. Frederick J. Milford
8:35 - 8:40 am	Opening Remarks: NRC	Dr. Frank Arsenault Dr. Kyo S. Kim
8:40 - 8:45 am	Opening Remarks: ACRS	Dr. Paul G. Shewmon
8:45 - 9:00 am	BCL Program Overview	Dr. David Stahl
9:00 - 9:30 am	Waste Forms	Mr. Emilio Spinosa
9:30 - 10:10 am	External Corrosion	Dr. John A. Beavers
10:10 - 10:30 am	Internal Corrosion	Dr. H. H. Krause (Beavers)
10:30 - 10:45 am	Break	--
10:45 - 11:15 am	Hydrogen Embrittlement	Dr. Henry J. Cialone
11:15 - 11:45 am	Separate Effects	Dr. Alan J. Markworth
11:45 - 12:15 pm	System Performance/Integration	Mr. Stephen L. Nicolosi
12:15 - 1:15 pm	Lunch	--
1:15 - 2:15 pm	Tour of Facilities	--
2:15 - 3:00 pm	Concluding Discussions	All