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April 30, 1987

Mr. K. C. Chang
623-SS
U.S. Nuclear Regulatory Commission
Washington, DC 20555

Dear Mr. Chang:

RESPONSES TO SANDIA COMMENTS ON DEMONSTRATION REPORT

Attached are our responses to the Sandia comments you sent us recently regarding the methodology demonstration report.

The comments reflect inputs from NRC consultants Carl Boyars, Gary Fuller, Robert Moler, Loren Zaremba, and myself.

If you have any questions related to the responses, please call us.

Very truly yours,

Kenneth W. Stephens

Kenneth W. Stephens

cc: C. Boyars
G. Fuller
R. Moler
L. Zaremba

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Response to March 1987 Sandia Comments on Methodology Demonstration Report 4/28/87

Background

From 1983 through 1986, a team at The Aerospace Corporation was involved in development of waste package performance assessment methodology for NRC. As a part of that work, the team conducted a preliminary methodology demonstration, which was summarized in an Aerospace technical report, WPR-86(6810-01)-01, "Demonstration of Methodology for Waste Package Performance Assessment", September 1986. In October 1986, the team members became NRC consultants and have now been asked to put the September report in the form of a NUREG or NUREG/CR document.

In late 1986, NRC gave the Aerospace methodology demonstration report to Sandia for review. At the end of March 1987, comments were sent to the NRC staff, which passed them on to us.

Disposition of the Comments

We have carefully reviewed all the comments and, where appropriate, will use them in our current revision of the report. Unfortunately, many of the comments reflect the reviewers' apparent misunderstanding of the methodology and the technical principles on which it is based. These items are addressed in our response.

The headings used below are keyed to the arrangement of the Sandia comments.

Bonano General Comments

In the general comments, the reviewer addresses three items: 1) his feelings regarding the purpose of the report, 2) criticism of the writing, and 3) an assertion that some of our statements are not technically correct.

The demonstration work covered in the report was conducted with the consultation and concurrence of the NRC staff. The purpose of the report is to summarize the results of a preliminary demonstration of the methodology, and we did just that. We make no pretense that this is the final demonstration that ever should be done.

While criticizing the writing quality in the report, the reviewer appears to expect a higher standard from us than he is willing to impose on himself. On the very page in which he accuses us of "numerous typographical errors", he commits two spelling errors and one grammatical error. We, and the NRC reviewers, tried to catch all the minor errors, but we realize

that in complex technical material, mistakes sometimes creep in. Nevertheless, we will attempt to make the current revision of the report "letter perfect".

The reviewer asserts that some of our statements are not technically correct. That comment is inappropriate, and we strongly disagree. Our rebuttal is included in specific responses below.

Bonano Specific Comments

Section 2.1

The reviewer takes issue with our handling of sampling for cases in which two parameters are correlated.

Our brief discussion of random parameter sampling appears to us to be quite straight-forward. If two random parameters are known to be highly correlated, either a priori or through specific studies such as sensitivity analyses, but are independent of other parameters, they should not be sampled independently. It seems pointless to belabor the obvious; parameters that have multiple correlations have to be sampled.

Section 3.2

We said:

> ...if a reaction mechanism involving formation of a protective coating is rate-controlling, average failure time might be greater at higher temperatures.

Our statement is correct. The reviewer changed our "might" to "will" in his comments and then criticized the altered wording.

Regarding another part of Section 3.2, the reviewer found our discussion of the temperature dependence of equilibrium constants to be "rather disturbing". We said:

> For many chemical reactions, the chemical equilibrium constants are likely to have a modest temperature dependence and are not well known in any case, so that many of the models available have no temperature dependence.

Our words are correct. To the degree that the appropriate chemical equilibria are understood and information about the temperature coefficients of their equilibrium constants are known, the effect generally is small. Our point was that without models that explicitly include chemical equilibria and their temperature dependencies, the modular approach is the preferred method.

However, the reviewer does have a point with regard to the need to explore the implications of temperature dependencies of reactions important to repository performance. In order to ensure that no-one mistakes the intent of the paragraph, we will include an appropriate qualifying statement such as:

> The effect of equilibrium-constant changes on repository performance is an important issue that should be explored.

Section 4.3

The reviewer disagreed with the way we accounted for the effect of temperature on the diffusion coefficients.

Our approach is approximate, but what makes it "inappropriate"? All prior approaches with which we are familiar used a room-temperature diffusion coefficient, which leads to under-estimation of the release rate. There are large variations in the expected temperature at the surface of individual containers, and using this value to infer the diffusion coefficient is a reasonable approach. The whole idea is to do a (conservative) probabilistic analysis, but the reviewer's suggestion of using the highest possible temperature that could be reached would amount to a single, absolute worst-case analysis.

Section 4.5

The reviewer says that our discussion of the limitations of the transport models seems to be "without physical basis".

He displays a serious misreading of the text and perhaps a poor understanding of the diffusion equations. Our statement that the 1- and 2-dimensional approximations understate actual release rates is absolutely valid and is based on fundamental physical phenomena.

We are not saying that data currently exist to verify this theoretical prediction. If and when such data become available, the statement can be experimentally verified as it has been verified abundantly in a host of other situations. (See for example the books of Crank on diffusion and Carslaw and Jaeger on heat flow.)

Chapter 5

The reviewer questions whether the temperature range used in our calculations is representative of the range expected in a repository.

We are not sure what the reviewer has in mind. It seems

transparently clear to us that the data used to calculate temperatures are the best available. It must be obvious that uncertainties were accounted for, otherwise why carry out a Monte Carlo calculation?

We clearly defined the limits of validity inherent in the work and made it clear that if models with greater temperature dependencies are found to be necessary, then the conclusions might not be valid, and further studies would be needed. We resent any implication that we did not recognize these issues in the report.

Chapter 6

In discussing analytical solutions for impulse sources, we said:

> If the source undergoes radioactive decay, the problem may become quite complex. Simple analytical approximations may have to give way to parametric numerical results in the form of response surfaces in that case.

The reviewer changed "may" in the first sentence to "are", took issue with the statement, and asserted that solutions exist. Solutions for the 1-dimensional case are well known, and there are solutions for certain other cases based on Danckwerts' method. However, the Danckwert solutions apply to some very specific boundary conditions that are different from the ones we need to address.

Perhaps there exist analytical solution to the 3-dimensional transport equations that include radioactive decay, but if so they seem to have escaped the notice of P. Chambré and other prominent experts. Such solutions would be welcome indeed, because they would greatly simplify the introduction of advection into the assessment.

The reviewer also considered our suggestion for possibly including groundwater flow through the repository as "not valid". We said:

> The repository is essentially a slab. If the groundwater is normal to the slab, as is suggested by the available evidence, and if edge effects are negligible, then it may be useful to treat the engineered barriers system as a 1-dimensional diffusion/advection problem. There are useful analytical approximations to this case.

Although such approximations may not be perfect, they could be useful for initial work. We feel they would be at least as valid as the Sandia mixing-cell model that homogenizes the entire repository. We happen to consider the mixing-cell

approximation to be appropriate, given the current state of knowledge.

Section 7.3.1

The reviewer accused us of confusing the terms "verification" and "validation". In fact, we did not use those terms at all. We said:

> For situations in which diverse, independently developed models are available, they can be compared with each other. The differences in outcome thus indicate the impact of model differences.

This statement is correct. In some cases, comparison of the outputs of diverse models is essentially the only way available to assess the degree to which the models represent the real world.

Section 7.3.6

The reviewer did not like our brief discussion of the necessity for probabilistic performance assessment. We agree that probabilistic approaches are highly desirable, but in our earlier work, some of the NRC staff expressed the feeling that they might not be mandatory.

It will be easy to expand the section to reflect the importance of probabilistic analysis and the current NRC staff thoughts on the subject.

Section A.1

The reviewer disagreed with the purpose of the methodology demonstration.

It is inappropriate for any reviewer to criticize the purpose of work structured by our team with the full concurrence of the NRC sponsors. The methodology involves new types of analysis, and it is entirely fitting that we demonstrate that the methodology can be applied successfully to waste package performance assessment. The report summarizes the progress in that direction. Complete and final demonstration will of course be completed only after there are improvements in data collection and advancements in the state-of-the-art for process modeling.

As clearly stated in the report, it is necessary to compare the modular and cascade approaches so that the computational efficiencies of the modular method can be utilized with confidence. Accordingly, the comparison is not only desirable, but also essential.

[Figure] A.1

The reviewer disagreed with our method for performing numerical integrations and mentioned Gaussian quadrature.

We chose the particular method that best met the requirements, all things considered. Our work showed that if non-uniformly-spaced basis points are used (as in Gaussian quadrature), integrations involving convolution equations become unwieldy.

Sections A.5 and C.2

The reviewer expressed reservations about the way we handled the corrosion analysis.

We believe we have used the available corrosion information in an appropriate manner, and we duly recognize the limitations.

Section D.1

Appendix D of the report describes and compares radionuclide transport models. The reviewer stated:

> The discussion that the available radionuclide transport models are nonconservative because they do not account for radioactive decay is simply incorrect.

We strongly disagree.

The reviewer attempts to demonstrate that this position is incorrect by straightforward reasoning based on the premise that "one can obtain the expression for the [radionuclide] concentration [as a function of time and position] without accounting for radioactive decay and multiply it by $\exp(-\beta t)$ to get the concentration profile with radioactive decay." Here, β = ln 2 / half-life.

This approach is certainly appealing because of its simplicity and the ease with which it can be implemented. Unfortunately, it is incorrect. It is invalidated by the reasonable boundary conditions of constant, solubility-limited concentration at the waste form surface.

The reviewer performs a simple calculation to demonstrate his point, in which he multiplies the average gradient between two values of the space coordinate, x, by $\exp(-\beta t)$, for some reasonable value of t, and thus reduces the gradient, since

$$0 \leq e^{-\beta t} < 1 \qquad \beta t > 0.$$

The average gradient would be, using the reviewer's figures,

$$\frac{c(x_1,t) - c(x_2,t)}{x_1 - x_2} = \frac{2 - 1}{x_1 - x_2} .$$

On multiplying, one obtains, using the reviewer's approach,

$$\frac{2 - 1}{|x_1 - x_2|} e^{-\beta t} < \frac{2 - 1}{|x_1 - x_2|}$$

(It is clear from the nature of the diffusion process in the present application that $x_2 > x_1$; the reviewer is really concerned with the absolute magnitude of the gradient, and the notation has been changed accordingly.)

If, however, the point x_1 is at the waste form surface, then this expression must be replaced by

$$\frac{c_s(t) - c(x_2,t)\, e^{-\beta t}}{|x_1 - x_2|} > \frac{c_s(t) - c(x_2,t)}{|x_1 - x_2|}$$

where $c_s(t) \geq 2$ and $c(x_2,t) = 1$, as before. Clearly, the gradient is increased, in this case, when decay is included.

In addition, for two points not at the waste form surface, the average gradient is given by

$$\frac{c(x_1,t_1)\, e^{-\beta t_1} - c(x_2,t_2)\, e^{-\beta t_2}}{|x_1 - x_2|}$$

where t_1, t_2 are measures of the elapsed times since departure of the small volumes of radionuclide at x_1, x_2 from the surface, where the concentration is constant. In particular, t_1 is not equal to t_2 for finite values of $x_1 - x_2$.

A mathematically rigorous approach for solubility-limited release and transport, with decay, must include effects related to these time differences. The expression provided in a Chambré article[1] does so. This expression, in the present notation, is

$$c(x,t,\beta) = \beta \int_0^t e^{-\beta t'} \, c(x,t',0) \, dt' + e^{-\beta t} \, c(x,t,0)$$

This equation differs from that of the reviewer by the addition of the integral term, accounting for the time differences.

In summary,

- Solubility-limited release precludes use of the simplistic approach put forth by the reviewer.
- The concentration gradient is increased when decay is included. (See the Chambré paper for sample calculations.)
- The models in common usage when the methodology demonstration report was published are non-conservative in that decay is not considered.

Section D.2.3

In Section D.2.3, we said that under certain circumstances, early times were of little interest because the release was not significant until much later. The reviewer expressed the hope that the lack of interest was not based only on analysis with this one model.

Our lack of interest was based on the collective consideration of all available information--not just on the outputs of the model in question. The text can be revised to reflect this.

Section D.2

The reviewer disagreed with our use of different porosities and the comparisons of the analytical and numerical models that were made.

[1] Chambre', P.L., et al, "Mass Transfer and Transport in a Geological Environment", LBL-19430, April 1985 (page 2, Equation 7).

The figures on pages D-22 through D-25 illustrate the comparisons done for different porosities.

As discussed on page D-21, the maximum releases are in relatively good agreement (except for the case shown on page D-25), and thus the linear model may be useful for "rough estimates" for early times (where the agreement is good).

Although numerical models may be subject to inaccuracies in principle, this is no reason to refrain from making comparisons. The good agreement of the models is only one type of information used in making judgments regarding the overall value of the models.

On another subject related to Section D.2, the reviewer suggested that our comparisons of the cylindrical and spherical models are not valid.

We disagree. Use of equivalent surface areas for diffusion modeling is quite reasonable, especially for solubility-limited releases in which the concentration at the surface of the waste form is held constant. Our work is consistent with the work by P. Chambré and T. Pigford.

Section D.3

The reviewer commented on the reasons why it is more difficult to predict release rates at large times using the linear model.

We agree. As discussed on page D-14, we took care to include a sufficient number of terms in the summation for later times. The program automatically tests for convergence of the series to 0.1 percent.

Section D.4

The reviewer commented on our observations related to the prolate spheroidal model. In addition, he suggested that a model could be easily developed using a finite cylinder geometry and compared to the prolate spheroidal model to determine whether end effects can bias the results.

Why would anyone go to the enormous effort of developing the prolate spheroidal solution if a finite cylindrical model could be easily developed? There are no known solutions for a finite cylindrical model for our boundary conditions. This subject is covered quite well in the work by Chambré and Pigford.

Guzowski General Comments

The reviewer disagreed with the title of the report. He apparently felt that the term "demonstration" should only be used once the methodology is more complete. We will consider retitling the revised report to reflect the preliminary nature of the demonstration.

The reviewer said, "Some conclusions are not adequately justified nor solutions or alternatives proposed". We believe we have handled the conclusions and discussion of issues in a responsible manner. Nevertheless, we will review the wording of the revised report to ensure that our intent is clear.

The reviewer noted that applicability of the methodology to the unsaturated zone (tuff) and salt sites was not addressed. We will consider revising the report to reflect that much of the methodology is generic and can be applied to all sites, but that successful application will require further development of site-specific process models and data.

The reviewer mentioned that previously developed computer codes were incorporated into this methodology and that adequate documentation for these codes must be available. The current revision of the report will discuss the user guide and other documentation that have been produced since the report was originally written.

Guzowski Specific Comments

NOTE: Responses are given below, without a restatement of the comments.

P. 2-2, para. 3

The issue is a matter of degree. Although simplifications are necessary for the modular and cascade approaches, they are much less extensive than those required for a full, global approach.

P. 2-3, para. 1, sent. 1

Because the waste package is composed of physical barriers, it is natural to consider whether the barriers can be modularized for analytical purposes. One of the central purposes of the demonstration was to examine the difference between the answers provided by the modular approach and the answers from the cascade approach. Issues associated with modularization and its appropriateness were discussed at some length in the Methodology Report (NUREG/CR-4477), which is referenced in the demonstration report.

P. 2-3, para. 1, last sentence

See above response.

P. 2-3, para. 3

See above response.

P. 2-3, para. 4

It is not possible to conduct a full, global investigation for the reasons stated earlier. The issue is whether the simplifications fairly represent the system as it is now understood. This too is addressed in the Methodology Report (NUREG/CR-4477).

P. 2-3, para. 5

The discussion covered in the paragraph is referring to the modular approach. One advantage of the modular approach is that pdf's calculated for the modules need not be recalculated if they involve a module that has not changed.

P. 2-4, para. 1

The text is correct as written. We said that premature failure and low probability events can be incorporated. It was not done in the demonstration.

P. 2-5, para. 1

We believe it is appropriate for our work to consider calculational efficiency.

P. 2-5, para. 3

We make no pretense that the methodology is complete and final for all time.

Subsection 3.2

Comment is self-explanatory.

P. 3-1, 3-2

The question of time-invariance is an important reason for comparing the modular and cascade methods. The demonstrations showed that the modular approach compared favorably with the cascade approach.

P. 3-2, para. 3, sent. 3

We will consider rewording the sentence for clarity.

P. 3-3

The paragraph at the top of page 3-3 will be reworded. The data used in the demonstration are a fair representation of the type of data available in the foreseeable future.

P. 4-1, para. 3 & 4

Validation of corrosion codes is beyond the scope of our work. These particular codes were used to demonstrate how our methodology will use corrosion calculations in the overall performance assessment.

P. 4-1, para. 5

Diffusion coefficient and porosity may be correlated. However, the solubility, porosity, and retardation were randomized in our work. Correlation information developed in the future can be incorporated into the methodology.

P. 4-4, para. 1

In the future, it will be desirable to conduct a number of sensitivity studies such as the one suggested by the reviewer.

P. 4-6, para. 3

The methodology has not yet been set up to handle the unsaturated zone.

P. 4-7, para. 1

We agree. The relative importance of pitting and uniform corrosion is one of the key issues in performance assessment.

P. 4-7, para. 2

All the relevant work known to us has been done under the assumption of diffusion-limited transport. The work by Chambré and Pigford is based on diffusion as is ours. Analytically, there is no current model to incorporate advection for our purposes. Until there is better information on the actual conditions in the repository, it would be difficult to model advection with confidence.

P. 4-7, para. 4

We agree. Water chemistry is important.

P. 4-7, para. 5

We agree. The methodology is not final.

P. 5-3, para. 2

Comment is self-explanatory.

P. 5-3, para. 3

The methodology will continue to evolve. Nevertheless, the methodology represents reasonable use of information available now.

P. 5-4, para. 3

Organic complexing was addressed in Appendix E.

P. 6-3, para. 1

SWIFT was mentioned only as an example. If the code is not suitable for our purposes, it will not be used.

P. 6-4, para. 4

Comment is self-explanatory.

P. A-9, para. 1

The BWIP project assumed resaturation at 100 years and made the indicated assumptions about temperature. Our model can randomize resaturation times and compute temperature histories. As shown on page A-13, our simulation randomized resaturation times over a range of 1 to 300 years. Page 3-2 will be revised to reflect the actual values used in the simulations.

P. A-11, Section A.4

There may be physical mechanisms that would keep the water from contacting the waste. However, until those mechanisms are understood, it is appropriate to assume that the water is available to the waste.

P. A-11, para. 2

As discussed in Section D.1, calculations for the demonstration used two models--one that uses diffusion through infinitely long cylinders, and a linear 1-dimensional model. The prolate spheroidal model was not available to us early enough to incorporate it into the simulations. However, Appendix D does discuss the relative merits of all these models. The transport

model to be used in the future is yet to be determined.

P. B-1, para. 3

Specifics of the use of TEMP3D are included in the user manual for our programs, which is under development. Comparisons of results from TEMP3D and other codes are included in Appendix B.

P. B-3, para. 3

The Altenhofen work, which used the more elegant code HEATING5, enabled us to assess whether TEMP3D can be used with reasonable confidence. We have no reason to doubt the validity of the Altenhofen work.

The 150° temperature is an approximate median value.

P. B-3, para. 5

The comparison was not for a basalt site vs a salt site. The results from TEMP3D (for a salt site) were compared with the results from a salt site EA.

P. B-6 and B-7

The figures will be reoriented in the revised report.

P. B-8, Table B.3

The data used were for the salt site.

P. B-9, para. 1

Virtually all the data needed to run TEMP3D were available from the EA. The only exception was a small amount of information on the temperature dependence of thermal conductivity. That was obtained from the Westinghouse report referenced in the EA.

P. B-9, para. 2

The final basalt EA was checked to see if there were any significant differences compared to the draft EA. There were none that affected these calculations.

P. C-1

We have researched the little available work on the corrosion layer ("crust"). There is no definitive work that would enable quantification of any reduction in corrosion. The state of knowledge regarding basic corrosion modeling is still developmental. We would imagine that it will be some time before the effect of the "crust" is explored.

Expansion of the corroded material has been hypothesized, but we can find no work that has experimentally or theoretically examined the issue in sufficient depth to draw any conclusions.

P. C-1, para. 4

We will consider expanding the text.

P. C-2, para. 1

The lack of water movement may limit the amount and rate of corrosion, but that was not assumed in the models used.

The Greek letter mu was inadvertently missing from the reproduction copy of this page. When the mu is added, the corrosion-rate units are correct.

P. C-3, para. 3

We share the reviewer's concerns about the state of corrosion modeling in general and these two models in particular. It was necessary, however, to use some corrosion model for the demonstration. We chose to use two to illustrate possible effects of model differences. The development of better corrosion models is outside the scope of our work.

P. C-4

Pitting is a complex phenomenon, and it is difficult to explain the mechanisms without a lengthy discourse.

P. C-9

The objective was to demonstrate the methodology for a basalt site. The specifics of corrosion for tuff and salt were not addressed

P. C-10, para. 1

The Greek letter mu is missing from the units.

P. C-11, para.2

We have addressed this comment in earlier responses.

P. D-3, para. 2

Documentation for the codes will be included in the user manuals.

P. D-9, para. 4

We merely stated our observations and indicated that the analytical model should be carefully compared with numerical code results before proceeding.

P. D-14, para. 3

Documentation will be included in the user manuals.

P. D-21, para. 4

The porosity and other parameters for a "real site analysis" are yet to be determined and will depend on the models used at that time.

P. D-34, para. 1

The models to be used in the "final methodology" have not been chosen. The choice of which transport model to use will require further work. The demonstration report describes the current state of knowledge regarding the methodology, which will evolve as better information is obtained.

Appendix E

The reviewer said that we should make proposals on how to resolve the uncertainties involving processes and materials such as those included in Appendix E.

This was done in Sections E.3, E.4, and E.5.