CASTLEMAN

THE PENNSYLVANIA STATE UNIVERSITY

152 DAVEY LABORATORY
UNIVERSITY PARK, PENNSYLVANIA 16802

College of Science
Department of Chemistry

January 31, 1983

Mr. M. Silberberg
Assistant Director
Research & Technical Support
U.S. Nuclear Regulatory Commission
7915 Eastern Avenue
Silver Springs, MD 20910

Dear Mel:

As a follow-up to our recent Peer Review meeting on NUREG-0956 (Draft) Radionuclide Release Under LWR Specific Accident Conditions, I am writing a few comments as additions to the many ones which I made during the course of the meeting. It was apparent that as a result of the program under your direction, much progress has been made during the last several years in being able to quantify the consequences of a reactor accident under specific pre-stated conditions. Nevertheless, it is evident that there is still much more to do in this area and I am sure that the groups at Oak Ridge, Sandia, and Battelle will contribute substantially in this regard.

My major overall concern is that through the use of highly developed computer programs we, as a technical community, may lose sight of the fact that the output is only as good as the input and there are numerous assumptions made in order to predict the consequences of various accident scenarios. It is easy to forget that there is still much experimental data needed in order to quantify the behavior of fission products and aerosols and that the research needs in this area are far from being complete. Those of us who are experimentalists realize that even in the simplest experiment it is often difficult to control all of the various parameters and in very complex situations, such as those leading to the release of fission products and the generation of aerosols, full control of all the variables is often an unobtainable goal. Hence, parameterization is often necessary in order to interpret the measurements. Incorporating the results into a mathematical model in order to predict the outcome of events having even greater complexities leads to numbers which may have very large limits of uncertainty. For these reasons, I strongly urge you to insist that the final report contain extensive sensitivity calculations with full presentation of the range of uncertainties in the final predicted consequences. This will not only lend more credibility to the report itself, but will also guide those groups which are engaged in experimental work in making measurements which are most crucial in furthering to define source terms. In my opinion, inadequate attention to this matter is the major shortcoming in the work presented in NUREG-0956.

There are clearly a number of experimental problems which warrant further investigation. Among those for which there seems to be an urgent need of additional experimental investigation include (1) a study of water condensation on aerosols, (2) scale up experiments for fission product release, (3) the potential for aerosol generation from the silver component of the control rods under a wider range of chemical environments and temperatures than investigated heretofore, and (4) a more extensive investigation of fission product deposition and reevaporation to more clearly define this in terms of physical condensation and chemisorption mechanisms. In this regard it is clearly important to have a realistic determination of the influence of decay heat on the temperature of the deposition surfaces. A case is made in the report that the release of certain fission products, lanthanium species in particular, may be influenced by the degree to which they form ideal solutions with the fuel matrix. Later in the report it is stated that the experimental data would indicate that this assumption is "not too bad." Nevertheless, from the material presented it is difficult to ascertain whether the agreement is fortuitous because of the nature of the release studies or whether this is really verification of the validity of the assumption of ideality.

Another point concerns the temperature of the fuel. As a result of the exponential dependence of aerosol generation on fuel temperature, it is imperative that additional attention be given to the definition of the maximum attainable temperature during an accident event in order to calculate more realistic numbers. One final general comment is that, despite statements to the contrary, the release and aerosol generation calculations are still not as mechanistic as would be desirable.

Some specific comments by page number are given on the attached sheets. I look forward to contributing to a further definition of the source term and reviewing subsequent drafts of this document.

Best regards.

Sincerely,

A. Welford Castleman, Jr.
Professor of Chemistry

AWC:bai

Enclosure

A. W. Castleman, Jr.

Pages 2-2, 3-5, and C-8 are inconsistent concerning their statement of the mechanistic approach utilized in the treatment presented. In particular, the rate coefficients on page 3-5 are not mechanistic in the sense implied in the report.

Page 3-3, 3rd paragraph. Some discussion of sensitivity calculations is warranted here.

Page 3-5, item 1. The implication of a "single melting temperature" needs to be discussed.

Page 3-5, item 2. The statement here is inconsistent with others elsewhere in the report.

Page 3-6, item 10. It is not clear what influence the chosen deposition velocities have on the final results. Clearly a sensitivity calculation is required in order to assess the effect of picking values in the mid-range of those deduced experimentally.

Page 4-10, 2nd paragraph. Discussion of the hydrogen combustion should be given at this point.

Page 5-2. A more extensive discussion of the codes mentioned on this page is warranted for each case. It is impossible for the reader to ascertain how adequately each individual one describes the event being treated.

Page 5-5, 2nd paragraph. It is the "timing" of the release which determines which species are available to interact with which other ones.

Page 5-6, 2nd paragraph, last sentence. The basis for the gap release values should be stated.

Page 5-8, 1st paragraph. The experimental conformation of the silver release warrants attention.

Page 5-8, 2nd paragraph. A determination of the maximum fuel temperature should be given more consideration.

Page 5-7, 1st paragraph, last sentence. Clearly more work needs to be done to put these release values on a firmer foundation. In this regard, despite the fact that the lower temperature release rates for cesium, strontium and zirconium are negligible compared to those at higher temperature, it is still misleading to infer that the parameters can be treated in the manner given in Table 5.1.

Page 5-13, 2nd paragraph. The point concerning the thermochemical limits of vaporization from the melt needs to be discussed more completely.

Page 5-14, 5th line. I believe the third word should be or.

Page 5-15, 3rd paragraph. The "competitive condensation on, or... tellurium" should be discussed more completely.

Page 5-16, next to the last sentence. The details need to be given.

Page 5-16, 2nd paragraph. The last sentence needs to be clarified.

Page 5-17. The first paragraph, last sentence needs to be discussed.

Page 5-17, 2nd paragraph. It is not clear why this number is conserved.

Page 5-17, 4th paragraph. There is a lot contained in this paragraph which is not available to the reader of the document.

Page 5-18, 3rd paragraph. What error does this introduce?

Page 5-19, last sentence. What implication does this have to the final results?

Page 5-22. The details of the steam condensation warrant more attention both theoretically and experimentally.

Page 5-25. How is the boundary layer thickness treated and how is its dependence on temperature ascertained? This same point should be discussed when, and if, the mechanism of particle deposition due to thermophoresis is treated quantitatively.

Page 6-31, 1st paragraph. How dependent were the tests on the geometry? Can these same findings be extrapolated to a reactor accident situation?

Page 6-31, last sentence. What is the basis for the 2×10^{-7} fraction per hour release figure for the iodine?

Page 6-45 (Table 6.8). It is surprising that the silver figure is so large here compared to no "release" in the case of the control rod simulation tests.

Page 6-53, next to the last line. (1608 seconds) is 1 correct for the value of Cs/I ratio.

Page 7-6, 3rd paragraph. How is Vap handled quantitatively? The same comment applies to the first paragraph on page 7-9 (new 7-11).

Page 7-13, last paragraph (new 7-18). The potential effect of aerosol resuspension needs to be treated in a sensitivity calculation.

Page 7-18, Table 7.7 (new 7-23/Table 7.9). The results require discussion.

Page 7-25 (new 7-35), last paragraph. One wonders what influence the assumption of the species being "evenly distributed for all particle sizes" has in the final calculated results.

Page 7-26 (new 7-36), 1st paragraph. The fraction per hour of 2×10^{-7} warrants justification. The discussion on pages 7-26 and 7-27 (new 7-36 and 37) is very confusing for the reader and should be completely rewritten and clarified.

Page 7-28 (new 7-49). The treatment of water condensation onto aerosol particles warrants more discussion and experimental verification.

Page 7-31, Table 7.13 (new 7.17). It is not clear why these values for iodine release exceed those given on page 7-26 (new 7-36).

Page 7-44 (new 7-70). A clear presentation of the difference in the calculated values using the NAUA code and the CORRAL code should be given.

Page C-7, 2nd paragraph. The statements contained here suggest that the vaporization of 200 species are well understood. I doubt whether this is the case.

Page C-8, 2nd paragraph. The technical basis for the statements made herein warrant considerable discussion and detailed presentation.

Page C-9. Items 2, 3, and 4 warrant discussion in detail. The technical basis for the equation shown at the bottom of the page should be presented.

Page C-15, last paragraph, first sentence. The stated inference concerning unit activity coefficients does not necessarily follow due to inadequate knowledge of the other parameters influencing the observed release rates.

Page C-16, last sentence; Page C-17, first sentence. I doubt whether this sentence is true. The entire issue concerning the scaling of experiments has been glossed over in this report. To me this is one of the major experimental factors warranting attention. Figure 4 in Appendix C, for instance, must be influenced by a gas boundary layer over the surface from which the aerosol formation is resulting. This and fission product release fractions must be greatly influenced by scaling factors since gas boundary layers, diffusion within various phases, etc., must bear on the release and vaporization factors.