U.S. DEPARTMENT OF ENERGY

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March 30, 1981

MEMORANDUM FOR: M. Silberberg, Chief
Experimental Advanced Safety Technology Branch
U.S. Nuclear Regulatory Commission

FROM: Carl E. Johnson
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SUBJECT: PEER REVIEW OF DRAFT NRC REPORT ON TECHNICAL BASES FOR ESTIMATING FISSION PRODUCT BEHAVIOR DURING LWR ACCIDENTS

You and your staff should be commended for drafting NUREG 0772 under such a constrained time table. I have focused my attention on Chapters 4 and 5 and, therefore, most comments are restricted to same. However, first some generic comments applicable to the whole document.

Foremost, is my concern that all who made thermochemical or modeling calculations used the same fission product properties data base. There were several instances in reviewing Chapters 4 and 5 that my own calculations did not give the stated result. Data evaluation and interpretation must be made against the best available properties data base, else the understanding may be wrongfully directed.

Further, frequently throughout the report the discussion includes a statement "..... in more reducing atmospheres." (For example, p. 15, 5th paragraph, "The results of the chemical thermodynamic analyses performed for this report indicate that CsI becomes the dominant species at higher fission product concentrations, lower temperatures, and in more reducing atmospheres.") This is a very loose description of a situation that must be defined rigorously if it is to have meaning. A reducing atmosphere must be defined in relation to the oxygen potential within an LWR fuel element. The environment within an LWR fuel element is already reducing with regard to that of the water-steam environment which is also reducing with regard to a steam-air environment, etc. Only by rigorously defining the condition of the calculation (or experiment) can a true meaning be attached to its interpretation.

In Section 1 (p. 7) a comment is made concerning the appropriateness of TMI-II data that appears reminiscent of the mind-set position so widely discussed in the Kemeny report. While the data from TMI-II are not quantitative, certainly their qualitative nature can be invaluable in testing certain positions as to the absence or presence of iodine in waste gases and waste streams emanating from the reactor building.

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Paragraph 5 on page 4.3 appears to be extremely speculative. There are ways of calculating population probabilities that can identify more rigorously whether Cs and I are in atomic or combined form. If the author is just making an assumption it should be clearly stated.

Page 4.3, Section 4.1.3. The possibility (probability) that iodine can exist in the fuel as ZrI_4 is so low that it should not even be discussed. Zirconium prefers to be stabilized as an oxide much more readily under all circumstances within the fuel.

Page 4.5, middle. Our calculations show that even for a fuel of O/M = 2.03 (extremely high and probably never attainable in a urania fuel), the partial pressure of iodine would be about 10^{-10} atmospheres. Further, to get to UO_3 would require direct oxidation of fuel. Further, we know of no-known evidence for liquid Cs_2UO_4; this compound apparently decomposes as a solid never reaching liquid state.

Table 4.2, page 4.6 needs significant revision and maybe outright deletion because of numerous errors therein. For example, as the standard state for cesium and iodine is the gas at 1000 K, the entries should be zero in each case.

Page 4.7, section on results needs to be revised based upon the more recent work of Fee and Johnson (J. Nucl. Mater., In Press, preprint attached). Discussions concerning 50-50 steam/air mixtures should always be made relative to the total environment, i.e., fuel, fission products, and cladding. Such calculations are best assessed using an oxygen potential versus temperature plot covering the ranges and redox couples expected in the total system.

Table 4.3, page 4.12. The data of Table 4.3 need some corrections, i.e., for HBU-1, -2 etc., the amount of iodine released is in μg not mg. Further, the discussion concerning the HBU experiments gives a very misleading result when compared with that of NUREG/CR-0722, ORNL/NUREG/TM-287/R1. In that document a better prespective is given as to the importance of the formation of cesium silicate in releasing iodine. (The silicate liner within the furnace apparently caused complications.) Also, it should be made significantly clearer that if and when molecular iodine was observed, the magnitude found was extremely small. The handling (in this report) of this particular data set bespeaks of a lack of understanding or lack of interest on the part of the author in applying sound thermodynamic and chemical principles in evaluation of the experimental data.

Jumping ahead to Section 4.4 on the Zircaloy-UO_2 Interaction discussion, I would urge strong review of the work of H. Chung* on matters relating to this topic. What appears to be overlooked is the rapidity of zircaloy oxidation with respect to the transients in a LOCA situation. Depending on the rate of increase in temperature, it is likely that zircaloy can be changed

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*J. R. Honekamp, H. M. Chung, "A Zircaloy Oxidation Model for the TMI Heatup Code," ANL/LWR/SAF 80-2, September 1980.

significantly - if not completely to ZrO_2 - such that temperatures well in excess of 2000°C will be needed before melting of either fuel or zirconium oxide will occur.

In Chapter 5 there are several calculations of the Cs, I, O, H system made and data presented. Recognizing the complexity of the aqueous environment as shown by the data of Table 5.1, how are the data of Fig. 5.1 (and others in Appendix C) related to the real situation. I would feel much more comfortable if the various equilibria were presented in an Ellingham-type plot with an overlay of H_2O/H_2 ratios which appears to be the controlling situation in an accident scenario. I cannot urge too strongly that the calculations be made for the total system for the explicit characterization of each subset (Cs, I, H, O) is extremely difficult.

Page C.3. It appears unrealistic to assume that as CsOH disappears I_2 will remain in molecular form. I_2 is an oxidizing agent and reaction to iodate or other iodides is likely to occur.

Your effort in drafting NUREG 0772 has helped clarify areas of experimental work that need immediate attention. I have listed several that appear to be of highest priority.

1) Characterize relevant factors describing the behavior of fission products in LWR fuels so as to improve our predictive capability for transport and release. This should include the development of a thermochemical data base of appropriate temperature regime applicable to accident scenarios.
2) Determination of the high temperature vapor phase chemistry of cesium, iodine, and tellurium so as to identify the predominant atmospheric species during a reactor accident.
3) Determination of the diffusion (transport) characteristics of cesium and iodine in UO_2.

Do not hesitate to contact me if I can be of any further help in completing the report or consulting on fission product chemistry for irradiated oxide fuels.

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Enclosure: (1)