

## ARGONNE NATIONAL LABORATORY

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Washington, D.C. 20555

Dear Mel:

You are to be commended for bringing the peer group together to review a problem of current interest to all that express interest in nuclear power. Certainly not everyone agrees that the report is ready for publication, even after the June review, but there are, I am sure, institutional needs that must be served. Obviously the contractor needs to address the report with even greater fervor to reduce/clarify the varied points of disagreement.

There are a number of problems dealing with chemistry that were highlighted and I shall not go over them again. However, a few additional items do need clarification:

- (1) The contention that control rod material will vaporize congruently (its stated composition) is in error. I would suggest that Cd will be vaporized first followed by Ag and then In as the expected result (this position is supported by ORNL). Recent post-irradiation examination studies at ANL of TMI-II filter material has identified particulate material of pure Cd, Ag, and In along with some alloys but never in the control rod (stoichiometric) composition.
- (2) There must be boundary limits placed on the I<sub>2</sub> limits that can be observed in any accident sequence. With CsI as the species being evolved in an accident the opportunities for molecular iodine formation is very limited and characteristically outside the bounds of the environments found in the stated accident scenarios.
- (3) In spite of the discussions there is still significant problems in the manner in which fission product tellurium is handled. Complex species have to be introduced into the release/transport scheme to ensure correctness. Further, Dana Powers is in error when he states that nickel telluride is the most stable species. There are others more stable and of greater importance, i.e., ZrI<sub>2</sub>, CrI<sub>2</sub>.

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- (4) Of a more generic nature is the problem of semantics in that non chemists are using chemical terms erroneously. In particular I am certain that the SANDIA work on chemisorption is really a corrosion problem that may have other materials/fission product behavior ramifications. This needs to be clarified.

On p. 5.7 an equation is given for the fractional release rate coefficient

$$K = Ae^{BT} \quad (1)$$

that does not have the proper form for the curves given in 5.2. This is demonstrated by the need to give three sets of values for A and B for the temperature range of interest. The given equations result in straight line segments. Since most of the data are for lower temperatures (Lorenz <1600°C, Parker <2000°C, and SASCHA <1800°C) there is practically no data to determine how these segments can be extrapolated to higher temperature. Most of the curves that are given in NUREG -772 and are used in this report fit an equation of the form

$$K = A \exp(B/T) \quad (2)$$

Fission product release from the fuel is generally assumed to involve diffusion to grain edges and to involve vaporization. Both of these processes can be represented by Eq. (2). Thus for most of the curves Eq. (2) can be extrapolated using a single set of A and B parameters. The Xe-Kr curve is more accurately fitted by the sum of two exponential curves like Eq. (2).

Further, in this section of the report it appears to have been made that release/vaporization/aerosolization of fuel, zircaloy cladding, and structural material can be treated in a manner similar to fission product release. In fact the coefficients used for Zr and Sn in the cladding are the same as that for fission product Zr and Sb, respectively. Structural materials are assumed to "vaporize" at .01 of the zircaloy cladding rate. The process for moving fission products out of the fuel into the steam environment is completely different from that for oxidation, vaporization, or aerosolization of cladding, structural material, and fuel into the environment as vapor or particle form.

If the equations for fraction release

$$K(T) = Ae^{B/T}$$

of Table 5.1 are used to fit  $K(T) = Ae^{B/T}$  then the value for B in the latter case is the heat of activation divided by the gas constant. Most of the fission product release data yield a heat of activation of 40-50 kcal. The numbers given in Table 5.1 for fuel "release" also give a heat of activation of 50 kcal. However, urania vaporization  $UO_2(s) \rightarrow UO_2(g)$  requires some 140 kcal and reaction of urania with water requires some 118 kcal. Similar deficiencies exist for zirconium. If therefore seems unlikely that the equations in Table 5.1 will accurately predict the "release" of fuel or cladding in an accident.

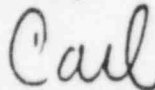
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A final thought reflecting on the meeting format that would allow more detailed discussion among the diverse group present for the peer review. Rather than debating specific issues (i.e., thermal hydraulics, chemistry, . . .) among all those assembled why not consider dividing up into smaller, more homogeneous groups to handle the specific issues. Debate is for a limited time and a "discussion" leader reports back the groups findings. This format could be an efficient way of resolving/handling specific issues very expeditiously. A disadvantage to this approach is that "cross-fertilization/education" of each others concern. However, if one is to produce a high quality report in a short period of time the focused activity may have some merit.

I appreciate the opportunity for participation in the peer review and would be happy to help in refining the chemical issues should the need arise. Our own studies on fission product release and thermodynamics and aerosol characterization may provide relevant background information to the development of this report.

Again plaudits for the good job under such a narrow time frame.

Sincerely,



Carl E. Johnson  
Chemical Technology Division

CEJ:kk