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Dear Mel

Enclosed are comments on the draft of NUREG-0772 "Technical Bases for Estimating Fission Product Behaviour during LWR Accidents". They have been gathered from a number of UKAEA staff at various establishments who have expertise in appropriate areas, and are mainly on Chapters 4-6 of the draft. Some of the points were raised during the Washington meeting and you are no doubt already taking account of them. Nevertheless I believe they merit stating again as the people concerned had not, at the time of writing, had any significant feed-back from me concerning the meeting.

1.5 The fact that the chapters need linking to ensure consistency, especially between Chapters 4 and 5 (and their related appendices) and the later chapters, is made by several people. Another general comment concerns the need to spell out as clearly as possible the input data to and the major assumptions implicit in the various codes on which so much reliance is, and indeed must be, placed. The mitigating factors for iodine as caesium iodine which appear in conclusion 3 (page ii) are thought to be surprisingly low in relation to the new approach to fission product chemistry contained in the Report.

1.1 Having made these comments and criticisms, we all congratulate those responsible for the Report for the excellent job which they have done in a very short time. To single out one particular feature is perhaps invidious but the approach of collating the release from fuel data in terms of rate coefficients of individual fission products applicable to modelling the time/temperature history of accident development represents a very significant step forward from the approach used in WASH-1400.

Yours sincerely

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CHAPTER 4 (and related Appendix B)

As was pointed out at the meeting by, for example, Malinauskas, a stronger recommendation on the release of iodine as caesium iodide seems justified. The weight of the evidence now points strongly to caesium iodide as the major chemical form of iodine.

It would be of help in modelling the stages of fission product transport subsequent to release from the fuel if the expected physical forms e.g. vapour, aerosol, particulate, of the released species could be stated.

The data on which Fig. 4.3 are based are scattered especially at temperatures $> 1600^{\circ}\text{C}$. Further experimental work is needed in this area. Whilst it is made clear in the text, it would be worth re-stating in the legend to this figure that the burst release, which will be irradiation history-dependent is excluded.

In the temperature range $1300-1800^{\circ}\text{C}$ the calculated releases of the less volatile fission products depends critically on the results of Parker et al. (ORNL-3981). Since his experiments were on unclad fuel, the release from a 'real' fuel rod should be less.

It is not clear why the SACHSA experiments in which no noble fission gases were present should give high releases of Cs and I (Fig. 4.3) at $> 1600^{\circ}\text{C}$. This perhaps emphasises the need to give greatest weight to results obtained on irradiated fuel rather than on UO_2 containing simulated fission products.

The calculations summarised in Tables B.2 and B.3 represent an excellent approach to the problem of calculating whole core release. However, some measure of caution is needed in applying the results in view of the uncertainties in the data base and the limitations of the MARCH code.

In pressurised sequences especially, liquid phases could form at low temperatures, say $\sim 1400^{\circ}\text{C}$, as a consequence of UO_2 reacting with the Zircaloy. Consideration needs to be given to the rate of release from low temperature melts.

The anomalies reported by Osetek and King concerning the release of certain barium and lanthanum isotopes in RIA experiments needs to be explained.

We are not surprised at the low burst releases from the H B Robinson fuel in view of its low operating temperature. On page 4.13 it is stated that typical burst releases might be 3%. It would be interesting to compare this with calculations of releases from modern PWR fuel operating at current low ratings. Our own calculations would suggest 0.5% as more likely than 3%, at least for the majority of the rods.

CHAPTER 5 (and related Appendices C1-C9)

Whilst the vapour phase chemistry of Cs, Te Sr, Ru and I and the solution chemistry of iodine are treated comprehensively, the material is not presented in such a way as to be readily applicable to accident conditions. More attention might usefully be devoted to identifying the likely conditions of, for example, temperature, pressure and pH in specific accident sequences.

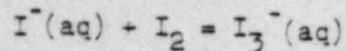
The approach which has been used is essentially that of classical equilibrium chemical thermodynamics. We share the reservations concerning application of this in the assessment of the species released into steam atmospheres at temperatures below 600-800°C where kinetic factors may be dominant. The kinetic aspects of behaviour and release of caesium and iodine in oxide fuel on the lower temperatures must be examined. There is no indication of what accident circumstances, if any, will involve vapour regime temperatures > 600°C. Similarly, in the treatment of aqueous iodine chemistry, more consideration could be given to the effects of the large differences in temperature, pressure and pH expected between primary circuit water and water released into the containment building.

Vapour-surface and solution-surface chemistry are largely neglected, probably because of an absence of data. Nevertheless, we feel that it is important that greater recognition of the role of surface processes in disturbing equilibria is needed. In particular, the chemistry of iodine compounds on steel surfaces needs detailed consideration and probably experimental investigation.

We endorse the comments made at the meeting that tellurium chemistry is a major area requiring further experimental work. We also agree with the arguments

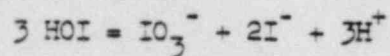
for treating ruthenium as a refractory material with low release fractions. The discussion of strontium chemistry is rather inconclusive and needs a recommendation as to the most probable release and transport form of the element.

It is not clear why this chapter omits consideration of the equilibrium



in the treatment of iodine hydrolyses. This reaction was considered to be important in earlier work; e.g. Eggleton Ref. 5.34. Including it has a significant effect when incomplete equilibrium, with small or zero iodate formation, is assumed for kinetic reasons.

So long as reliable kinetic data on reaction 5.4



under accident conditions are lacking it may be preferable to accept Eggleton's model, in which iodate formation is omitted, in the calculation of partition coefficients. Kinetic studies on this reaction are needed.

The hydrolysis of methyl iodide (section 5.3.6) is barely worth including in any model. The grounds for re-assessing and reducing the organic iodine fraction are strong but the quantitative results can be no more than illustrative since the necessary input data do not exist.

The computed iodine equilibrium data could, with advantage, be applied to specific accident sequences with the appropriate conditions of temperature, pH etc.

Chapters 4 and 5 indeed the whole document, make one aware of the great amount of thought and effort which has and is being deployed in very relevant areas, many of which we consider as being very important. It is apparent, however, that future endeavours, possibly as a collaborative programme, should aim at:

- 1) Providing a good base of chemical thermodynamic data to the appropriate species.
- 2) Developing other descriptions of property chemical equilibria under

relevant conditions of temperature, concentration and pressure using codes such as SOLGASMIK and SOLGASWATER.

3) To apply the information from (1) and (2) to the analysis of the required accident sequence step.

CHAPTER 6 (and the related Appendix D)

The input assumptions are critical in assessing results from the TRAP code and more discussion and justification of the input data would enhance the value of the data which are presented. Whilst the work reported concentrations on the release mitigation aspects of TRAP more detail of the influence of the input parameters on the characteristics (e.g. particle size distribution) of the release into the containment would be useful as would clarification of the relationship between this Chapter on the one hand and Chapter 4 (and Appendix B) on the other.

The TRAP runs usually stop at relatively short times after the start of an accident. No guidance is given as to whether this relates to assumptions in the model or the source term. The importance and validity of modelling primary circuit retention at longer times could perhaps be considered.

On page 6.10 (third paragraph) an example is presented of the growth of aerosol particles. Whether such high concentrations would arise during core heat up and melt down is highly questionable and the results of small scale experiments suggest that these values are too high. However, it is agreed that in a dry regime the particle size range of interest is 0.1 - 1 μm .

The view of the iodine in the various accident sequence has to be considered in more detail and with more clarity. Under various sequences the proportion of CsI, molecular iodine and organic iodides must be assessed. There is little mention of other fission product elements, for example, tellurium.

CHAPTER 7

It would be interesting to include in the Report a comparison of the computed accumulated mass leaked as a function of time for the various codes which have been used in order to show the effect of the various models and the consequence

and the effect of time to failure.

In section 7.5.1, steam condensation is dismissed as a minor effect. Such a small effect on particulate transport and deposition is surprising (and arguable) and further studies are required to assess the magnitude of steam condensation effects.

Both iodine and caesium iodide are considered. The relationship between these forms is not clearly explained. It is noteworthy that there are significant quantities of caesium iodide remaining as aerosols in the containment of several hours in certain sequences. This would not support Levenson's conclusion. The presence of CsI is not beneficial in all sequences.

CHAPTER 8

Quantitative details of the analysis of ESF systems together with some statement of current thinking on fission product removal by sprays and/or coolers would be of interest.

For the accidents which involve limited fuel damage with a low aerosol source in the containment and little particle agglomeration it is stated that removal of vapour phase iodine would be favoured rather than removal of particulate-associated caesium iodide. Our view is that caesium iodide is by no means a less restrictive form of fission product iodine than elemental iodine under all circumstances.

Editorial Points

CHAPTER 4

- p 4.23 Table. Release coefficient units should be fraction/min.
- p 4.25 Following Table 4.4. Sentence at beginning of next paragraph should refer to Figures B3 and B4 (not B2 and E3).
- p 4.25 Table 'Comparison of Fractional Release Estimates'. Why is Cs omitted from the RSS fission product group column?
- p 4.27 Three unrelated lines inserted before beginning of last paragraph.
- p 4.27 Fourth line from bottom. Figure 4.5 (not 4.4) should be referenced.
- p 4.29 Figure 4.5. Units of ordinate temperature scale not given.
- p 4.30 Section 4.5. Several errors in editorial assembly here resulting in repetition and discontinuities.

CHAPTER 5

- p 5.14, 5.15 Section 5.3.5 repeats much of section 5.3.4. Equation 5.1 is repeated, and the relationship to 5.3 and 5.4 should be clarified. Different values for the equilibrium constant applying to 5.1 are quoted at top and bottom of page.
- p 5.23 Reference to Eggleton should be 5.34 (not 5.22).
- p 5.23 and Figure 5.4 Does 'Total Iodine' mean total aqueous-phase iodine?