



# Benton City Technology

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NUREG-0956

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Ms. Jocelyn Mitchell  
U.S. Nuclear Regulatory Commission  
Washington D.C. 20555

Dear Jocelyn:

Attached is a brief write up entitled "Discussion of Organic Iodide Formation Under Severe Accident Conditions". This information documents the views stated in our recent telephone conversations.

As expressed earlier, I conclude that a best estimate conversion factor for organic iodides is in the neighborhood of 0.5%. This number derives from the Reactor Safety Study, and is based on a realistic evaluation of all available data.

As noted in the following discussion it might be worthwhile to look again in detail at organic formation to determine if information published during the past 13 years would change our perspective.

Please get in touch if you wish to discuss any of the information reported herein.

Very truly yours,

Arlin K. Postma

CC: G.R. Hulman

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DISCUSSION OF ORGANIC IODIDE FORMATION  
UNDER SEVERE ACCIDENT CONDITIONS

OBJECTIVE

The objective of this discussion is to describe the technical approach used to arrive at an estimate of the fractional conversion of fission product iodine to organic iodides under postulated severe accident conditions in water-cooled reactors.

BACKGROUND INFORMATION

Pertinent background information that helps characterize the formation of organic iodides under severe accident conditions is described as follows.

Phenomenological Basis

The postulate that a fraction of iodine released from over-heated fuels will appear in containment in the form of organic iodides is based on experimental results. Numerous tests were conducted in the 1960's in which iodine was released into simulated containment atmospheres. It was invariably found that a small fraction of the iodine would appear as an organic specie, airborne in the containment atmosphere. Various iodine species were used in the experiments; while a majority of tests used elemental iodine, a significant number of the tests used irradiated  $\text{UO}_2$  fuel as the iodine source. The conversion to organic iodides was similar for iodine evolved from fuel and for iodine released in the elemental form. Therefore, the formation of organic iodides under reactor accident conditions can not be discounted by the claims that the iodine species released from overheated fuel will be different than the form used in some experiments.

The data base on organic iodide formation available in 1972 is reviewed in detail in WASH-1233<sup>(1)</sup>.

### Dominant Regions for Formation Processes

Organic iodides are expected to be formed mainly in containment spaces outside the reactor coolant circuit. In containment spaces, iodine will encounter organic materials under thermal and radiation conditions that permit organic iodides to be formed and to persist for significant time periods.

Thermal and other conditions inside the primary coolant circuit tend to minimize the formation of organic iodides and maximize its destruction. Therefore it is expected that only the iodine released from the primary system would be susceptible to conversion to organic forms.

It was concluded from the 1972 review<sup>(1)</sup> that organic iodides could be formed both by non-radiolytic reactions and by radiolysis. Unfortunately, theoretical, mechanistic studies had not evolved to a level that allowed organic iodide formation processes to be quantified by means of mechanistic theoretical models. Therefore, predictions for accident conditions must be made on the basis of the available data set itself.

### Earlier Projections of Fractional Conversion to Organic Iodides

#### Early Projections by AEC Directorate of Licensing

A formal source term was developed in the 1960's by the AEC Directorate of Licensing for use in reactor licensing procedures. An important part of the formal source term was iodine. It was assumed that 25% of the core inventory of iodine was airborne in the containment atmosphere, and that of the airborne iodine, 10% and 8% were present as organic iodides in the PWR and BWR respectively<sup>(2,3)</sup>. These fractional conversions to organic iodine were postulated to be conservative, i.e., upper limit values.

WASH-1233 Review<sup>(1)</sup>

Available data was reviewed in 1972 to provide a better technical basis for the assumptions used in the early Regulatory Guides<sup>(2,3)</sup>.

Based on a review of all available data, it was concluded that for the hypothetical accidents postulated as a basis for containment system design, that no more than 3.2% of iodine released to the containment atmosphere could be converted to organic iodides. Thus, the 3.2% conversion was identified as a conservative upper bound for organic iodides.

The WASH-1233 review<sup>(1)</sup> identified 69 experiments in which sufficient data were taken to allow quantification of the airborne iodine mass concentration and the fraction of the airborne iodine that became converted to organic species. The experiments covered a broad range of containment vessel sizes, atmospheric compositions and iodine concentrations. While a majority of the tests used elemental iodine as the fission product simulant, a significant number of tests used irradiated  $UO_2$  fuel as the source of iodine. The apparent agreement of results from tests that used  $I_2$  and those which used  $UO_2$  fuel indicated that  $I_2$  was a valid simulant for fission product iodine released from overheated nuclear fuel.

Results of the WASH-1233 review<sup>(1)</sup> were incorporated into AEC licensing procedures. The second edition of applicable Regulatory Guides<sup>(4,5)</sup> listed the assumed organic iodide fraction as 4%, a rounded approximation to the 3.2% figure projected in WASH-1233.

Reactor Safety Study ,

Organic iodides were one of the source term species considered in the Reactor Safety Study<sup>(6)</sup>. The 4% conversion identified in the Regulatory Guides<sup>(4,5)</sup> was not used in the Reactor Safety Study because a realistic, as opposed to conservative, estimate was needed. A realistic estimate was deduced from the data base reviewed in WASH-1233.

It was projected that 0.7% or 0.4% of iodine injected into the containment atmosphere could be converted to organic iodides. The higher number applied if sprays did not operate and the lower number applied if containment sprays did operate.

The average conversion, 0.5%, is roughly one order of magnitude lower than the conservative number of 4% used in licensing procedures.

#### NUREG-0722

A more recent estimate of organic iodide formation was presented in NUREG-0722<sup>(7)</sup>. The stated objective was to arrive at a less conservative estimate than was given in WASH-1233. The same data base presented in WASH-1233 was used. The resulting projections vary from essentially zero to 0.03%. The higher number, 0.03%, was said to apply only to a hypothetical case in which the iodine is released as elemental iodine.

It can be argued that these estimates of organic iodide formation are unrealistically low. First, the postulate that organic iodide conversion would be negligible if one considered a realistic iodine form released from  $UO_2$  fuel is not substantiated by the data base. As noted earlier, a significant number of tests employed real iodine released from irradiated  $UO_2$ , and results from these tests generally agreed with those that employed  $I_2$  simulants. When the tests that employed highly irradiated fuel were separated out and compared with those that did not use highly irradiated fuel, the trend was that iodine emitted from  $UO_2$  fuel underwent equal or greater conversion than iodine injected as  $I_2$  or emitted from trace irradiated fuel. Some 10 of the 69 tests described in WASH-1233 employed highly irradiated fuel, and there is no obvious reason for these tests to be totally discounted. Therefore, it is difficult to accept the lower limit postulate given in NUREG-0722, i.e., that negligible conversion to organic iodides would be expected when realistic iodine forms are considered.

Second, one should consider whether the 0.03% conversion is a realistic estimate if it is assumed that iodine is released in the elemental form. Several comments aimed at judging this estimate are listed as follows.

In the NUREG-0772 estimate of organic iodide fraction, the fractional conversion resulting from non-radiolytic processes was arrived at on the basis of a predictor line drawn roughly a factor of five lower than the least squares fit of all of the data. There is little technical justification for the use of a predictor line significantly lower than the average.

The method used to arrive at the 0.03% figure leads to roughly an order of magnitude under-prediction of the organic iodide fraction observed in the PRTR incident<sup>(8)</sup>. Because the PRTR incident involved realistic simulations of all aspects of a real accident (it was a real accident) projection methods that greatly underpredict the PRTR result are not very satisfying.

The 0.03% projection is not consistent with the TMI-2 experience. According to the measurements reported by Pelletier<sup>(9)</sup>, approximately 0.003% of the core inventory of iodine became airborne in the containment atmosphere of TMI-2 as organic iodides. If one uses the best estimate assumption that 0.03% of iodine is converted to organic forms, then one can back-calculate the amount of  $I_2$  initially airborne. The  $I_2$  airborne is predicted to be 0.003/0.03 or 0.1 of the core inventory. This estimate of 10% of the core inventory of iodine airborne as  $I_2$  is orders of magnitude higher than would be expected for the TMI-2 scenario.

One must conclude that either: (1) there was a large quantity of  $I_2$  airborne in the TMI containment atmosphere or (2) that the fractional conversion of airborne iodine to organic iodides was much higher than the 0.03% figure. The second of these two possibilities appears to be the more likely.



### FILTRA Project

A project has recently been undertaken in Sweden to provide mitigation for hypothetical core melt accidents at nuclear power plants. The effectiveness of the gravel bed filter system identified in the FILTRA project was evaluated under the assumption that 1% of iodine entering the containment atmosphere would be converted to organic forms. Since organic forms were not expected to be trapped efficiently in the gravel bed, the organic iodides constituted the main iodine source term to the environs. The 1% conversion factor appears to be a rounded value of the 0.7% specified in the Reactor Safety Study<sup>(6)</sup>.

### RECOMMENDATIONS

Based on the foregoing review of organic iodide formation, it is recommended that the projections made in the Reactor Safety Study (RSS) be used for the present time. If only a single fraction is to be used, then a rounded average of the two fractions specified in the RSS, 0.5%, appears to be reasonable. The projections used in the RSS are judged to be the best option because:

- the entire available data base was used,
- the approach was designed to yield a realistic estimate,
- the projections are consistent with results from both the PRTR and TMI-2 incidents.

It should be recognized that the use of a single number for organic iodide conversion is a gross simplification in itself. It would be more realistic to use a conversion factor that applies to each scenario, accounting at least for potentially different airborne iodine concentrations.

Also, it has been some 13 years since the organic iodide issue has been

studied in detail. It would probably be worthwhile to have another look at organic iodides to factor in information that has been generated during the last 13 years.



LITERATURE CITED

1. A.K. Postma and R.W. Zavadoski, "Review of Organic Iodide Formation Under Accident Conditions in Water-Cooled Reactors", WASH-1233, USAEC, Washington, D.C. (1972).
2. Assumptions Used for Evaluating the Potential Radiological Consequences of a Loss of Coolant Accident for Pressurized Water Reactors. Safety Guide 4, November 2, 1970. Division of Reactor Standards, USAEC.
3. Assumptions Used for Evaluating the Potential Radiological Consequences of a Loss of Coolant Accident for Boiling Water Reactors. Safety Guide 3, November 2, 1970. Division of Reactor Standards, USAEC.
4. Assumptions Used for Evaluating the Potential Radiological Consequences of a Loss of Coolant Accident for Boiling Water Reactors. Regulatory Guide 1.3, Revision 2, June 1974, Division of Regulatory Standards, USAEC.
5. Assumptions Used for Evaluating the Potential Radiological Consequences of a Loss of Coolant Accident for Pressurized Water Reactors. Regulatory Guide 1.4, Revision 2, June 1974, Division of Reactor Standards, USAEC.
6. Reactor Safety Study, WASH-1400, Appendix VII (1975).
7. Technical Bases for Estimating Fission Product Behavior During LWR Accidents, NUREG-0722, U.S. Nuclear Regulatory Commission, Washington, D.C. (1981).
8. R.W. Perkins, C.W. Thomas, and W.B. Silker. "Fission Product Aerosol Behavior in the PRTR Fuel Rod Failure of September 29, 1965" BNWL-SA-668 REV, Battelle Pacific Northwest Laboratory, Richland, Washington, 1965.
9. C.A. Pelletier. "TMI-2 Evaluations and Modeling", NSAC/14, Nuclear Safety Analysis Center, Palo Alto, California, November, 1980.