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The U.S. Nuclear Regulatory Commission
Office of the Secretary\
Washington D. C. 20555-0001

Dear Sir,

I have read in the February, 2000, issue of Nuclear News, page 11, that the Nuclear Regulatory Commission has issued a new rule that will allow the use of "an alternative source term for the accident analysis on which plant design and operations are based, replacing a source term (TID-14844, 1962) that has been in effect for more than 37 years." In addition, the article mentions "Revised Source Terms published by the NRC in 1995" and, more recently, the NRC has published a draft regulatory guide and a new section of the NRC Standard Review Plan.

I wish to receive copies of these documents, specifically, the new rule, the 1995 source term document, the draft regulatory guide, and the new section of the NRC Standard Review Plan.

My interest stems from the fact that on August 14, 1980, Dr. A.P. Malinauskaas, Dr. D.O. Campbell, both of Oak Ridge National Laboratory, and I , from the Los Alamos National Laboratory advised the Commission that the assumptions used in their required accident analysis were seriously in error and overestimated the released of iodine by a very large factor. I believe that the "updated research" mentioned in the article derived from the stimulus of our letter in 1980. I enclose a copy of our letter for your and others information.

I thank you for your help in this matter.

Sincerely,
William R. Stratton
William R. Stratton

TEMPLATE = SECY-067

SECY-02

August 14, 1980

Chairman John Ahearne
U.S. Nuclear Regulatory Commission
1717 H Street
Washington, D.C. 20555

Dear Chairman Ahearne:

We wish to bring to your attention a matter that may be a very important development in reactor safety analysis. We believe that sufficient evidence has accumulated to show that the behavior of iodine during nuclear reactor accidents is not correctly described by existing NRC models and Regulatory Guides. Iodine volatility is grossly overestimated by these models for accidents in which substantial amounts of water are present, and escape of iodine to the environment will be extremely small (as it was at Three Mile Island) as long as reasonable containment integrity is also maintained. As a consequence, the risk to the general public presented by iodine is lower than estimated, perhaps by orders of magnitude.

Our concern with this issue originated with our involvement in the several Technical Staff Analyses for the President's Commission on the Accident at Three Mile Island. The mechanism for the behavior of iodine that we propose here was derived from those analyses, from further examination of experimental and theoretical studies involving the chemistry of iodine and cesium fission products in light water reactor fuel and systems, and from the observed behavior of iodine subsequent to fuel failures during accidents and incidents at other reactor sites. We believe that the explanation presented here will change the present concepts of the hazards involved during and subsequent to reactor accidents and, therefore, will require a critical reexamination of how these hazards and risks are calculated, and the criteria to which engineered safeguards are designed and installed.

Although the Three Mile Island (TMI) reactor core inventories of xenon-133 and iodine-131 were comparable, between 2.4 and 13 million curies of xenon escaped to the environment during the accident, while only 13 to 18 curies of iodine similarly escaped! This great disparity was identified as a matter of crucial importance early in the investigation by the President's Commission, and an effort was made to find the explanation. It was clear that we could not claim to understand the accident until this discrepancy (a factor of 10^5 to 10^6) was explained satisfactorily. Further, it was recognized that the physical and chemical conditions during the accident at TMI may not have been unique. (We note that, generally, radioiodine is the controlling fission product species with respect to site safety analysis as well as the design and operation of certain engineered safeguards.)

The explanation for the very low escape of iodine that developed during the investigation by the President's Commission was that, as the temperature of the core increased, iodine diffused out of the fuel rods through the failed cladding and vaporized. The iodine escaping, if not already in the iodide form, then encountered a chemically reducing environment which converted it to iodide. The iodide subsequently went into solution as iodide ion when it contacted water. It was recognized that additional experimental work was needed to provide a quantitative description of the iodine behavior. Nevertheless, this explanation accounted for the much smaller escape of iodine that was observed at TMI compared to the amount predicted to escape if elemental iodide had been present, as is assumed in the Regulatory Guides.

We believe that this description can be strengthened and made more definitive. Although the present data are not absolutely conclusive, we believe that iodine emerged from the fuel as cesium iodide, already reduced to iodide. The reactor system environment then sustained this chemical state. Furthermore, it would have converted other iodine species, should they have been present, to iodide. Cesium iodide would be expected to condense or "plate-out" when it reached metal surfaces at temperatures at or below 400 to 500°C, and it would finally enter into solution as iodide ion as soon as water or condensing steam was encountered. The reactions of iodine species in water, and the fact that iodide ion is the dominant species, ensure that iodine volatility will be very small (compared to that implied by the Regulatory Guides, for example). A reaction causing oxidation of iodide would be necessary to increase the volatility of iodine. Additional experimental work is required to provide a quantitative description of iodine behavior, but this qualitative picture is consistent with the small escape of iodine observed in a number of incidents when water was present, such as at TMI.

This mechanism is supported by the following observations, as well as by measurements made at TMI:

1. Iodine and cesium are released congruently from PWR leakers during power transients (the iodine spiking phenomenon).

2. Thermodynamic calculations performed at several sites indicate that CsI is the stable form of iodine in LWR fuel. Further, the fission yield of cesium is larger than that of iodine, and cesium is always present in great (about tenfold) excess over iodine.

3. Irradiated fuel has been caused to fail in experiments performed under simulated accident conditions, and the iodine released is recovered predominantly as CsI rather than as molecular I_2.

4. The chemistry of iodine is such that, if water is accessible, iodine will interact with the water so that its concentration in the gas phase will be much smaller than its concentration in the water.

5. In other incidents that have led to the destruction of fuel in water systems (NRX, Spert-1, Snaptran-3, SL-1, MTR, ORR, and PRTR), we understand that a much smaller amount of iodine escaped from the systems than would be projected by the existing models. Data are hard to come by for many of these accidents and experiments, and our investigation is continuing. In marked contrast, a large fraction (20,000 curies) of the iodine escaped to the environment during the Windscale accident, which occurred under oxidizing conditions and in the absence of water.

The significance of this mechanism for iodine escape and transport can hardly be overemphasized. We assert that the unexpectedly low release of radioiodine in the TMI-2 accident is now understood and can be generalized to other postulated accidents and to other designs of water reactors. We believe that an accident involving hot fuel and a water or steam-water environment will have the same controlling chemical conditions as did the TMI-2 core and primary system. The iodine will emerge as CsI (and possibly some other iodides) and enter into the solution as soon as wet steam or water is encountered. It will persist in solution as non-volatile iodide ion as long as oxidizing conditions do not prevail.

Although we feel that the evidence is sufficiently strong to justify this letter, it is important to qualify our position. Iodine chemistry is very complex, and definitive experimental and analytical studies of iodine behavior during and following loss-of-coolant accidents are lacking. Nonetheless, it is clear that the behavior projected from the existing Regulatory Guides is wrong. The current NRC assumption, that elemental iodine is the chemical form of the radioiodine released, is regarded as a conservatism, but in this case the assumption of a wrong chemical form must be regarded as an error which has compounding effects.

If, after due consideration, the NRC is satisfied that our description of iodine behavior is valid, we recommend that an urgent study and assessment be made of all available information, and appropriate actions be undertaken. With due respect we point out four consequences should our position be correct:

1. The frequently quoted fission product escape assumptions (from TID-14844 in 1962 to the more recent Regulatory Guides 1.3 and 1.4, and the Reactor Safety Study, WASH-1400) should be reexamined. The present assumptions grossly overstate iodine release from a reactor site in many types of loss-of-coolant accident, and safety criteria based on these assumptions should be reevaluated.

2. The dispersal of radioiodine in the biosphere may no longer dominate and control consideration of accidents and the design of safety systems.

3. Many, if not most, accident sequences must be reexamined in detail. The iodine risk to the general public may, in fact, be lower than previously estimated, possibly by orders of magnitude. The impact of a reduction of iodine risk on the requirements for evacuation is particularly important at this time.

4. The engineered safeguards designed for iodine control should be reexamined to assure effectiveness and optimization for the actual iodine behavior rather than the behavior currently assumed.

Finally, we realize that a major revision of NRC assumptions relative to accident analyses, dose calculations, and design of safeguards should not take place without an adequate base of technology from both experiment and theory, and especially until the Commission itself is convinced that it is appropriate to accept a revised physical and chemical description of iodine transport from fuel to the environment. On the other hand, the impact of wrong assumptions is so serious that an intensive effort should be made to establish the facts.

We are ready to offer more detailed information or further assistance should the NRC request it. We will be pleased to brief the NRC staff or any review committees you may appoint.

Sincerely,

W. R. Stratton

W. R. Stratton
Los Alamos Scientific Laboratory

A. P. Malinauskas
Oak Ridge National Laboratory

D. O. Campbell

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cc: G. W. Cunningham, DOE-WASH
D. M. Kerr, LASL
H. Postma, ORNL