From: Richard Lee/RES
To: Jason Schaperow/RES
Date: 3/7/01 3:37PM
Subject: Fwd: Ruthenium release white paper

Jason:

I thought we have model for Ru release in VICTORIA. Please take a look at this paper. I don't think the authors know the latest info. on Ru release under air ingress (e.g., European test).

Richard

G12

From: Cecil Parks <cvp@ornl.gov>
To: <fxe@nrc.gov>
Date: 3/1/01 4:22PM
Subject: Ruthenium release white paper

Farouk:
You may remember that on January 25 we met briefly to discuss an earlier informal white paper prepared at ORNL to identify potential experiments on Ru release following an accident scenario in which damaged fuel is exposed to air. You had indicated the estimated expense associated with those experiments appeared to substantially exceed the resources NRC could justify expending given the nature of the identified safety issue. Attached is a white paper entitled "Summary White Paper on Assessing Fission Product Release Of Damaged LWR Fuel in Air". The paper proposes an effort to use existing experimental information together with our understanding and modeling experience in fission product release phenomena as a means to predict the Ru release rate and associated uncertainty. Although the uncertainties resulting from this work may be large in the absence of additional experiments, the uncertainties may be adequate to address the safety concerns presently being considered by the NRC. Follow-on experiments to reduce the uncertainty could be identified as part of the project discussed in the attached paper.

Please contact me if you have questions. I will try to contact you next week to briefly discuss and get your reaction as to whether you think the paper proposes an approach that can adequately address the NRC issues.

Cecil

CC: <ryl@nrc.gov>

Summary White Paper on Assessing Fission Product Release Of Damaged LWR Fuel in Air

J. L. Collins and C. F. Weber
Oak Ridge National Laboratory
March 1, 2001

Background

For more than three decades, the U.S. Nuclear Regulatory Commission (NRC) has conducted thorough experimental and modeling programs, designed to quantitatively estimate the behavior, risks, and consequences of nuclear reactor accidents. Such studies have followed the release of radioactive species from fuel, transport in primary system and containment, and subsequent release and transport in the environment. Several generations of experimental results have been achieved, and a number of computer models constructed to accurately and mechanistically simulate physical and chemical behavior of many elements in various reactor systems.

Most of the studies (and modeling efforts) have focused on standard hypothesized loss-of-coolant accidents in a functioning reactor. The fission products are released from overheated (and possibly melted) fuel in the primary system, with a steam or steam-hydrogen atmosphere. The released species are then transported through the primary system and into the containment, whose atmosphere is usually saturated in steam, may contain some hydrogen, and may even be inert (free from oxygen). Most of the fission products are retained in these regions through physical and chemical processes, although a small fraction may be released to the environment.

Recent evaluations by the NRC staff have indicated the possibility exists for accidents that do not reflect the standard conditions present in most earlier studies.[1] The primary emphasis has been on spent fuel stored on site in which a low-probability, catastrophic event occurs, resulting in rapid oxidation of cladding and cladding fires. This sequence would alter fission product release by removing an important barrier. Direct exposure to an air environment creates more strongly oxidizing conditions for both release from fuel and chemical behavior of released species. In addition, the time delay - release would not occur until months or even years after reactor discharge - produces a different fission product inventory; iodine is no longer a primary concern, but other fission products may present hazards.

The release and transport of fission product ruthenium has been identified as a major concern under the conditions noted above. Under oxidizing conditions, the RuO_4 can form in the fuel pellets, and this species is quite volatile at temperatures above 500°C. If cladding were reduced or eliminated, this oxidation and subsequent release could occur easily and rapidly. As was mentioned recently by the Advisory Committee on Reactor Safeguards,[2] the biological consequences of ^{106}Ru releases are equivalent to those for ^{131}I.

If runaway cladding oxidation occurs, the fuel temperature could rise as high as 2000°C, which could also release large amounts of other volatile fission products: Ba, Sr, Cs, I, Te, Ag, and Sb. These elements would be of concern even if conditions were not strongly oxidizing. However, in a strongly oxidizing atmosphere, release rates could be accelerated by formation of U_3O_8 which is less dense than UO_2. The resulting fuel expansion would create more pores and cavities in the fuel, thus creating additional pathways for volatile release of fission products.

Under standard accident conditions, most fission products are expected to travel as aerosol particles in the containment. It is likely that similar behavior would ensue in postulated fuel pool accidents. Apart from iodine, very little consideration has been given to the chemical interactions that could enhance or impede transport of various fission product species through containment systems and into the environment. For conventional accidents, this has been reasonable, since iodine has presented both the greatest contribution to dose and the most varied chemical behavior. Under the conditions of this postulated accident in a spent fuel pool, other fission products would replace iodine in importance, and therefore their transport and interaction pathways merit greater attention. The phenomena of interest (release and transport of fission products in air environments) may also be relevant to reactor accidents that cause exposure of damaged spent fuel to air.

Proposed Work

We propose to construct a comprehensive model of fission product release and transport under conditions represented by an accident in a spent fuel pool that causes exposure of damaged fuel to an air environment. The focus would be on the behavior of Ru and other fission products deemed important by the NRC (such as those mentioned above). Through literature searches, reviews of previous modeling efforts, and consideration of general chemical principles, the initial model development would reflect the present state of understanding. The model will consider chemical form in degraded fuel, mechanistic release from fuel, interactions in the containment, and release from containment under accident sequences specified by the NRC. Such a model will be rigorously assessed with regard to 1) ability to simulate actual experimental data, 2) uncertainty due to missing information or uncertainty in data, and 3) sensitivity to modeling assumptions and events in postulated accident sequences.

Ruthenium is present in light-water reactor (LWR) spent fuel as RuO_2 that is very stable in a reducing or inert atmosphere. Even in air at temperatures below 500°C, it is stable. However, in air or under oxidizing conditions at elevated temperatures (above 500°C), the dioxide oxidizes to the tetraoxide (RuO_4) which is very volatile. Both the oxidation and vaporization rates increase considerably with increased temperature. At 1200°C the rate of vaporization is about 4000 times faster than at 700°C. Gaseous RuO_4 decomposes back to the dioxide as it cools to temperatures below 500°C. Once RuO_4 vapor transports from the hot zones of a hypothetical accident, it would be expected to transport primarily

as fine particulates of RuO_2 associated with aerosol. Since ruthenium dioxide is insoluble in water, it would have a high health risk if inhaled.

As a part of the modeling process, we anticipate the need for additional information to enhance and verify model predictions with experimental data. Work at this stage would only involve small-scale, bench-top experiments to evaluate reaction kinetics and equilibrium behavior for Ru reactions. If the estimated model uncertainties are larger than that needed to effectively address the safety concerns, then ORNL will propose additional experiments deemed necessary to clarify the model and reduce the uncertainties to an acceptable range.

Principal Investigators

C. F. Weber has participated in fission product transport modeling efforts for nearly 20 years, for both NRC[10-13] and DOE.[14] His efforts have focused on transport and speciation in containment of major fission products, most notably iodine. J. L. Collins was a major participant in NRC tests of fission-product release from LWR fuel conducted at ORNL from 1981-1995.[3-9] He has considerable expertise in fission-product chemistry and in release from fuel. In addition, ORNL retirees R. A. Lorenz, M. F. Osborne, and E. C. Beahm have extensive experience and are available for consultation. Excellent facilities are available at ORNL for bench-top experiments and hot cell tests. Computational capability and software support are ideal for implementation of the modeling effort.

Schedule and Funding

Review of available data	October 2001–March 2002
Model construction and testing	April 2002–September 2002
Supporting experiments	January 2002–September 2002
Draft report	December 2002
Final report	four weeks after NRC review of draft

Support Required: 2 FTEs

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