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THE INFLUENCE OF CHEMISTRY ON SEVERE ACCIDENT PHENOMENA IN INTEGRAL TESTS[a]

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ABSTRACT

The influence of chemistry on in-vessel severe accident phenomena in integral effects in-pile tests is reviewed. In-vessel severe accident chemistry involves high temperature interactions between the materials used in the tests; namely, fuel rods, control materials, spacer grids, and steam. The influence of chemistry on the release and transport of fission products has been found to be strongly dependent on core melt progression phenomena such as fuel liquefaction, control rod failure, and molten debris relocation. The extent of fuel irradiation, the presence of control materials and spacer grids, and the amount of steam available are important factors in determining the chemistry of the tests. Much has been learned about the influence of chemistry under PWR accident conditions, but additional information is needed on chemical interactions between fission product vapors and aerosols and on the influence of chemistry under BWR accident conditions

INTRODUCTION

The purpose of this paper is to examine the influence of chemistry on in-vessel severe accident phenomena observed in large in-pile integral tests and the TMI-2 accident. The paper is organized on the basis of chemical effects and the accident progression phenomena they affect. Much information is available on the influence of chemical processes on severe accident phenomena from small-scale separate-effects laboratory experiments, and this information has been essential to the interpretation of the more complex integral experiments. In some cases, additional small scale work is needed to help understand the integral test results, and in other cases the integral tests have not included materials and/or conditions needed to observe some important chemical effects.

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FUEL CHEMISTRY

The chemistry of uranium dioxide fuel changes as a function of burnup due to the production of fission products and the increase in the oxygen-to-metal ratio (O/M). More important to the behavior of fission products under severe accident conditions are changes in fuel morphology as a function of burnup and the H_2/H_2O ratio in the gas phase in the reactor vessel.

Effect of Burnup on Chemical State

The chemical state of fission products in irradiated fuel can be classified into four groups [1,2]:

1. fission gases and other elemental vapors: Kr, Xe, I, Br
2. fission products forming metallic precipitates: Cs, Rb, Te, Mo, Tc, Ru, Pd, Ag, Cd, In, Sn, Sb,
3. fission products forming oxide precipitates: Cs, Rb, Ba, Nb, Mo
4. fission products dissolved as oxides in the fuel matrix: Sr, Zr, Nb, and the rare earths Y, La, Ce, Pr, Nd, Pm, Eu.

More complex chemistry, such as the formation of uranates and molybdates, is also possible but is not represented in this treatment which is constrained to elements and oxides. Some elements, such as Mo, are listed in two groups because their chemistry is sensitive to the oxygen potential in the fuel. During normal operation, the oxygen potential of the fuel is determined by the O/M ratio which is a function of burnup. However, the O/M ratio is only a weak function of burnup [3], increasing by only about 0.006 at a burnup of about 43,000 MWd/tU.

Effect of Burnup on Morphology and Fission Product Release

Although fuel burnup has a relatively small influence on the chemical state of fission products under accident conditions, it has a strong influence on the concentration of fission products, the distribution of the fission products within the fuel, and the fuel structure itself. The uranium dioxide fuel undergoes restructuring as a function of burnup resulting in the formation of fission gas bubbles in grain boundaries which tend to interlink to form tunnels along grain edges at burnups above about 5,000 MWd/tU. This restructuring is enhanced by irradiation at higher powers and recent evidence has shown tunnel formation in grain boundaries of low burnup (<3,000 MWd/tU) fuel heated to elevated temperatures [4]. During irradiation, fission gases and vapors migrate to grain boundaries and, if tunnels have been developed, the gas bubbles can be released fairly easily to the gap outside of the fuel pellets. Grain boundary tunnels also provide a pathway for volatile fission product release during fuel heatup under accident conditions.

The effect of fuel burnup on fission gas release is illustrated in Figure 1 where the noble gas release rate is shown as a function of time for irradiated and unirradiated fuels tested in the Severe Fuel Damage (SFD) Test Series [5] in the Power Burst Facility (PBF). Both experiments experienced similar thermal transients. On heatup (between 1600 and

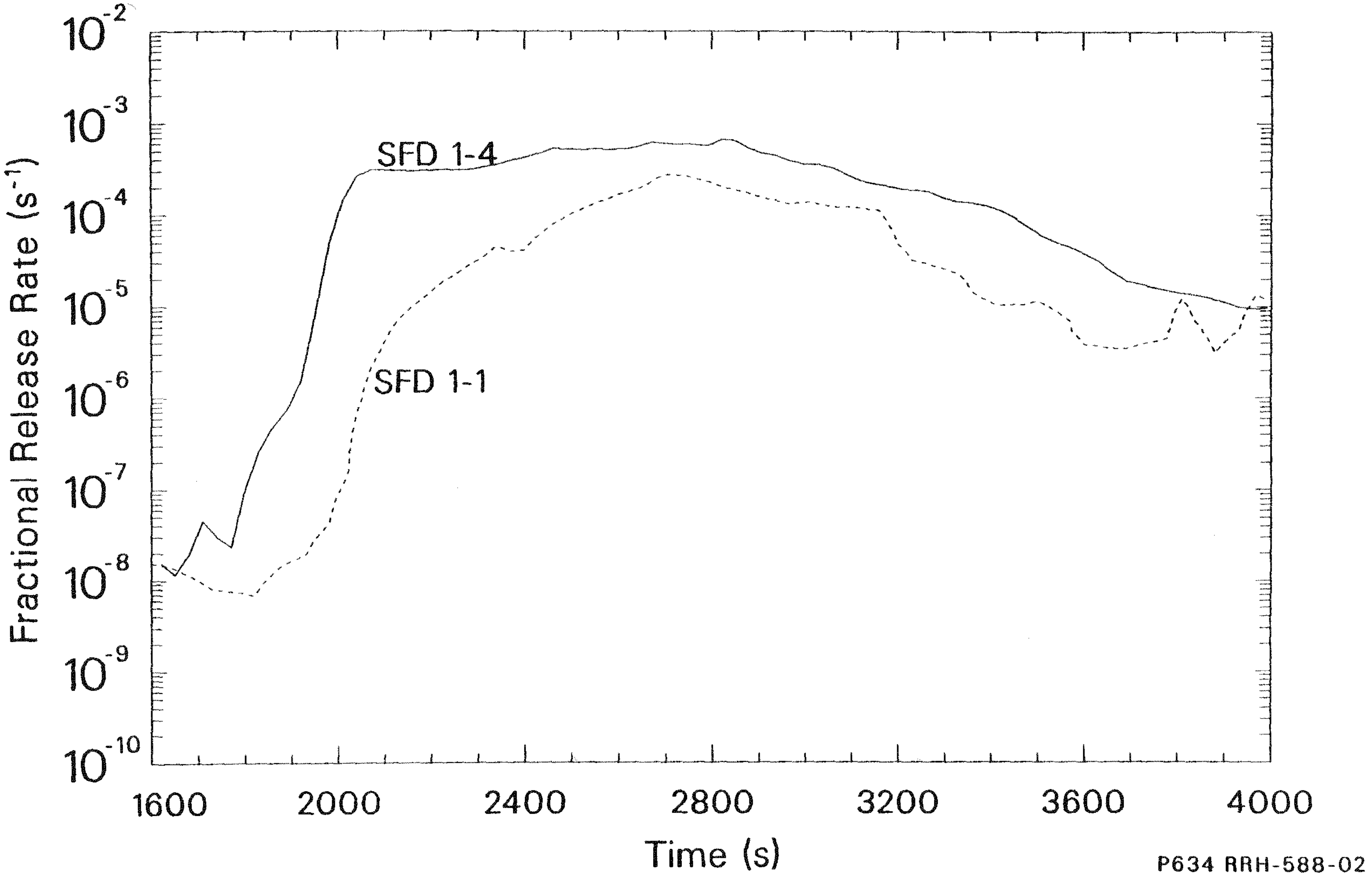

Figure 1. Noble gas release rates for trace-irradiated (SFD 1-1) and high burnup fuels (SFD 1-4).

2200 s) the release rate is one to two orders of magnitude lower for unirradiated fuel than for irradiated fuel. The differences in release rates tend to diminish at temperatures above about 2200 K (beyond ~ 2200 s) because fission product release becomes dominated by fuel liquefaction (dissolution of uranium dioxide by molten zircaloy cladding) which affects unirradiated and irradiated fuels similarly.

The effect of fission product distribution within the fuel structure is demonstrated by the releases measured in the SFD tests for fission gases of different half-lives. The results presented in Table 1 indicate that generally higher release fractions are measured for the isotopes with longer half-lives. These results can be explained by the fact that the longer-lived isotopes can migrate to grain boundaries from which release is relatively rapid, whereas shorter-lived isotopes reside primarily within the fuel grains from which release is more difficult.

TABLE 1. EFFECT OF HALF-LIFE ON FISSION PRODUCT RELEASE

Isotope	Half-life	Release Fraction
Xe-137	3.84 min	0.26 ± 25%
Xe-138	14.1 min	0.43 ± 25%
Kr-87	76 min	0.40 ± 25%
Kr-88	2.84 hr	0.35 ± 25%
Kr-85m	4.48 hr	0.36 ± 25%
Xe-135	9.1 hr	0.37 ± 25%
Xe-133	5.25 days	0.23 ± 18%
Kr-85m	10.72 yr	0.41 ± 13%
Cs-137	30.17 yr	0.51 ± 15%
Stable Xe	---	0.51 ± 100%
Stable Kr	---	0.52 ± 100%

Another feature of irradiated fuel structure that differs from unirradiated fuel is fuel pellet cracking. Fuel pellets crack as a result of thermal stresses caused by power changes during reactor startup and shutdown. These cracks tend to anneal out during operation at full power and re-form during shutdown, so that an approximately steady state crack pattern is developed independent of burnup. When debris beds are formed during the process of core damage progression by either cladding melting (SFD 1-4) or thermal shock of oxidized cladding (SFD ST and TMI-2), the particle size is two to four times smaller when irradiated fuels are present. The mass mean particle size in the upper debris bed in the TMI-2 core was measured to be about 1 mm, which is about one-third the size of particles formed from damaged fresh fuel. This difference in particle size is not expected to be large enough to cause significant differences in debris bed coolability and the dissolution rate by molten zircaloy.

Irradiated fuels have far greater concentrations of fission products than unirradiated fuels given trace irradiation prior to the start of integral in-pile testing. Calculations have shown a strong dependence of the chemical form of iodine on the concentrations of iodine and cesium in the gas phase relative to hydrogen and steam [6]. With trace-irradiated fuel, hydrogen iodide tends to be favored as the preferred iodine chemical form for gas phase transport, whereas with highly irradiated fuel, cesium iodide is favored.

It is important to use irradiated fuel operated under typical conditions for integral in-pile experiments in which the intention is to measure fission product release and transport. The concerns are fission product release pathways in the fuel, fission product concentrations in the gas phase outside the fuel, and coupling between core melt progression phenomena and fission product behavior. In experiments designed to measure only physical core melt progression phenomena, the use of irradiated fuel is less important.

Effect of Hydrogen-to-Steam Ratio

After fuel rod failure under accident conditions, the oxygen potential of the fuel is determined by the local H_2/H_2O ratio of the vapor in equilibrium with the fuel in the damaged reactor core. Chemical equilibrium is a reasonable assumption for all but the most dilute components of even solid phases at the high temperatures experienced by core materials during melt progression. Free energies of formation for oxides of various core materials are displayed in Figure 2. Also shown in the figure is the oxygen potential for hydrogen-to-steam ratios from ten to one-tenth. This range brackets many of the conditions anticipated in the reactor vessel during core melt progression in a severe accident. Toward the high oxygen potential end of this range, oxides can be formed by the fission products molybdenum and antimony. Tin from the zircaloy cladding, indium from silver-indium-cadmium control rods, and iron from structural stainless steel can also be oxidized. At oxygen potentials lower in the range, these materials would be expected to exist as elemental metals. The volatility of these oxides is very different than the volatility of the corresponding metals. The fission product cesium is stable as an oxide at low temperatures (<800 K) but should exist as elemental vapor at the fuel temperatures expected under accident conditions (>2000 K).

In the case of TMI-2, there is evidence from the temperature of control rod drive lead screw material in the upper plenum and the nature and thickness of the oxide on the surface of this material that the H_2/H_2O ratio was unity or less over most of the time that high temperature effluent was exiting the core [7]. Reference to Figure 2 indicates that gas with the H_2/H_2O ratio in the range 1.0 to 0.1 is capable of oxidizing UO_2 to $UO_{2.05}$. As indicated in a previous review [8], such oxidation is capable of increasing volatile fission product release rates by factors of four or more. Some evidence of localized fuel oxidation has been observed in the examination of materials from the TMI-2 core [9] and from the steam-rich test SFD-ST [10], but evidence of widespread fuel oxidation in TMI-2 or in the integral in-pile tests conducted to date (most of which have been steam-limited) has not been found.

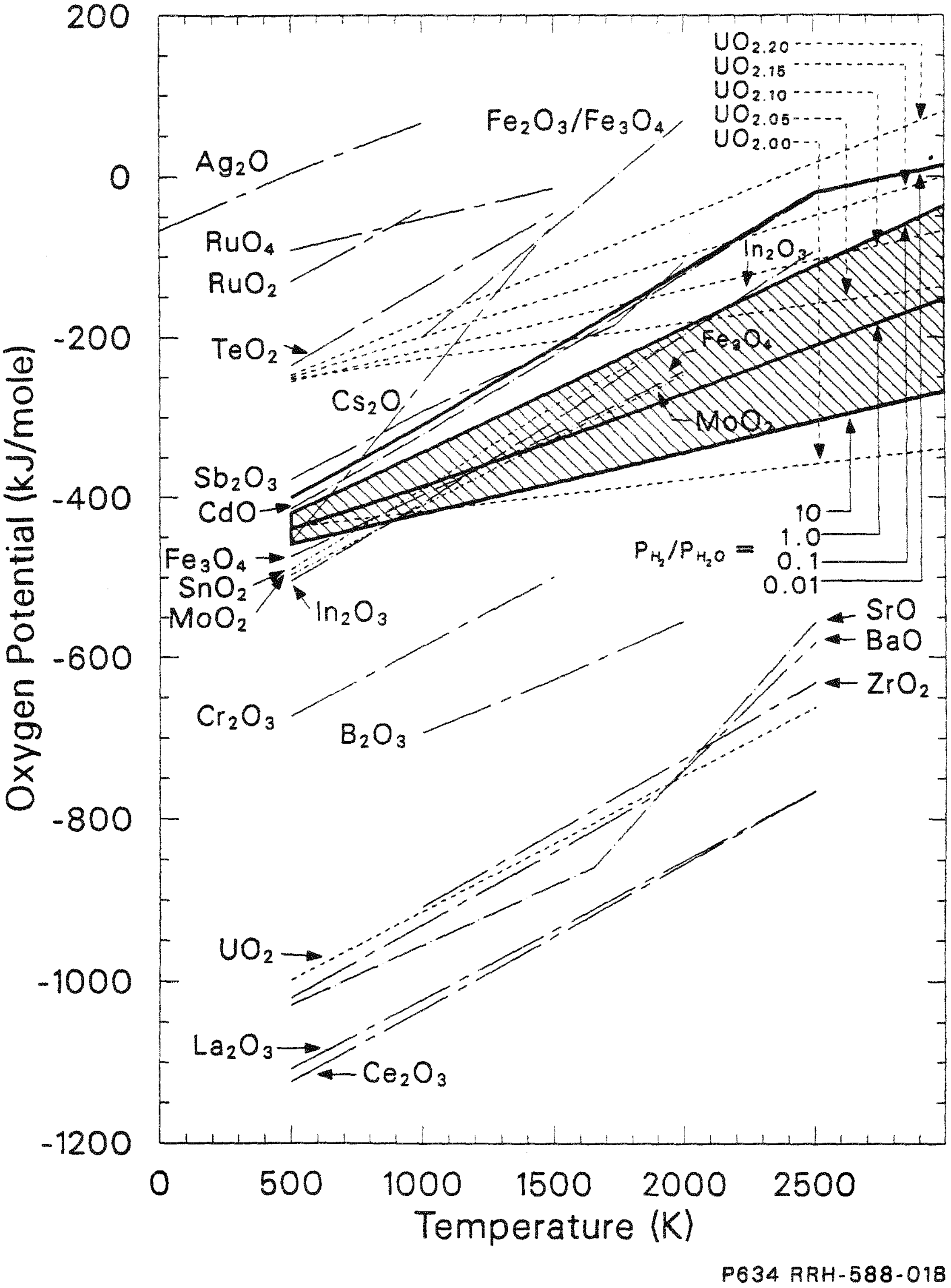

Figure 2. Free energies of formation of core material oxides.

A strong effect of the oxidation of zircaloy cladding on the release of tellurium has been observed in small scale laboratory tests at Oak Ridge National Laboratory [11] and confirmed in large scale in-pile tests in PBF [5]. The tellurium released from the fuel apparently reacts with zirconium and tin in the fuel rod cladding and is released when the cladding is oxidized enough to concentrate the tin in a thin unoxidized layer. The release data from TMI-2 [12] are consistent with this theory. In both SFD-ST and in the TMI-2 accident, the tellurium that exited the core transported far downstream suggesting a chemical form much less reactive than elemental tellurium, such as SnTe. SnTe has been found to be evolved from zircaloy doped with tellurium which was then oxidized [13], and furthermore is stable in the vapor state [14].

Very small releases of barium and strontium are expected when steam is present because these materials are expected to be in the form of oxides of low volatility in the fuel. Indeed, the releases of barium measured in the SFD tests in PBF are one percent or less [5] and the release of strontium from the fuel in the TMI-2 accident is similarly small [15]. Recent in-pile measurements of fission product release from highly irradiated fuel in a reducing atmosphere (hydrogen/inert gas mixture) in the ST-1 and ST-2 experiments in the Annular Core Research Reactor (ACRR) at Sandia indicate barium and strontium releases of several percent [16,17]. These results are consistent with those of the out-of-pile SASCHA experiments [18] and can be explained as the reduction of the oxides to elemental barium and strontium which have much greater volatilities.

According to Figure 2, ruthenium should exist in its elemental form under severe accident conditions and very small releases are expected based on results of laboratory tests with irradiated fuel [18]. It is interesting to note that large releases of ruthenium from the fuel, but negligible releases from the core, have been reported for ruthenium in the TMI-2 accident [7]. Examination of debris removed from the TMI-2 core is showing that ruthenium is retained in molten metallic materials [9] in agreement with earlier observations from out-of-pile meltdown studies [19].

Analysis of fission product release during the late phase of melt progression in the TMI-2 accident indicates that fission product chemical form and concentration dominate the release of medium and low volatile fission products and that bubble dynamics dominates the release of fission gases and volatile fission products [20]. The chemical forms of the medium and low volatility fission products were determined by the oxygen potential of the large consolidated melt region. The presence of iron oxides in the melt established a lower limit on the oxygen potential of about -150 kJ/mole at 2800 K. At this oxygen potential, rare earth elements such as europium and cerium would exist as oxides (Eu_2O_3 and Ce_2O_3 or CeO_2), strontium would exist as SrO, and ruthenium and antimony would be present as metals immiscible in the ceramic melt. Low releases were calculated for these materials primarily because of the low concentration and volatility of these species in the melt and the low surface-to-volume ratio of the consolidated region. Virtually all of the fission gases and volatile fission products should be released from the melt due to bubble coalescence and buoyancy. The presence of cesium in the molten debris transported to the lower plenum suggests the formation of some relatively stable chemical compound. Possible chemical forms include a zirconate, borate, molybdate, uranate, or chromate of cesium.

The foregoing discussion of the effects of steam oxidation is intended to demonstrate that the H_2/H_2O ratio in the gas surrounding the degrading fuel and the extent of zircaloy and UO_2 oxidation are important factors in determining the release of fission products from fuel, their volatility and their chemical forms in the gas and condensed phases. Results of equilibrium thermodynamic calculations of chemical species for vapor transport of fission products under postulated severe accident conditions are found to be dependent on, in addition to fission product concentrations, system temperature and pressure, H_2/H_2O ratio, and the variety of species considered in the calculations [21]. This realization leads to the consideration of the chemical effects of a variety of other materials in LWR cores on the transport of fission products.

CONTROL MATERIALS

In addition to the chemistry of the fuel and steam, the chemistry of other materials in the core, particularly control and spacer grid materials, is important in the course of core melt progression and the transport of fission products. If integral in-pile tests do not contain these materials, some important phenomena affecting core melt progression and fission product transport will not be represented.

Boron Chemistry

Separate effects tests [22,23] and calculations [21,24] have shown that boron chemistry can influence the vapor species of iodine and cesium in the reactor vessel. Boric acid in the reactor coolant and in the emergency coolant is used as a reactivity control material in PWRs, and boron carbide encased in stainless steel control blades is used in BWRs. Boric acid vapor reacts with cesium iodide in either the vapor or condensed states to form cesium borate and hydrogen iodide [22]. Boric acid vapor also reacts with cesium hydroxide in either the vapor or condensed phases to form cesium borate [22]. Separate effects tests have demonstrated that hydrogen iodide can be formed by reaction of boron carbide in steam with cesium iodide [23].

Hydrogen iodide and cesium iodide have very different transport properties: hydrogen iodide is much more volatile than cesium iodide but it is also much more chemically reactive with structural material surfaces, including aerosols. Without the influence of boric acid, cesium iodide is calculated to be the principal form of iodine for transport [21]. In this case, condensation onto structural surfaces and aerosols and subsequent aerosol deposition are the primary mechanisms expected for retention of iodine in the reactor coolant system. On the other hand, if hydrogen iodide is produced by reaction with boric acid, iodine retention will be dominated by chemical reactions with structural surfaces and aerosols and subsequent aerosol deposition. The role of aerosols in the transport of fission products is very important and depends on concentration and size distribution (which determines the surface area) of the aerosol particles, as well as temperature and chemical composition. The effect of cesium borate production on the transport of cesium is dramatic, since cesium borate is relatively non-volatile and cesium hydroxide will thus be removed from the vapor state.

The ability of boron compounds to interact with fission product cesium and iodine depends on the reactor type and the accident sequence. In-pile integral-effects Test DF-4 has shown that boron carbide in a BWR control blade may be prevented from interaction with steam and cesium iodide by preferential interactions with the stainless steel cladding of the blade [25]. This in-pile result confirms earlier results from small-scale out-of-pile laboratory experiments that showed boron carbide undergoes interaction with steel to form boron and carbon eutectics with iron and nickel [26]. These low melting eutectics relocated down to the bottom of the 0.5-m bundle in the DF-4 test. However, it is possible that in a full length core some boron carbide might be held up at blockages where hot, fuel-bearing material may subsequently relocate and cause interactions with steam and fission product vapors. Such blockages (involving silver-indium-cadmium control materials) were important in the SFD 1-4 test.

In a PWR, the availability of boric acid vapor to interact with cesium iodide and cesium hydroxide depends on the accident scenario. Large break loss-of-coolant accidents without emergency core cooling tend to result in nearly complete loss of water and thus boric acid before core heatup and fission product release, whereas small break loss-of-coolant accidents and station blackout entail slow boiloff of the coolant such that considerable steaming and boric acid vaporization could be present during core melt progression and fission product release from the fuel. Accumulator injection in a small-break loss-of-coolant accident would provide an additional source of boric acid during the damage progression phase of the accident.

The TMI-2 accident had some of the characteristics of a small break loss-of-coolant type of accident. Boric acid was present in the reactor coolant that boiled down and in the steam that was generated in the core. However, this accident was terminated by water injection and circulation so that evidence of water soluble chemical forms of iodine and cesium was not preserved. It is known from the ratio of iodide to iodate ions in the water in the reactor building sump that iodine was present as an iodide rather than in molecular form, but it is not possible to distinguish between cesium iodide and hydrogen iodide [7]. Had iodine been transported from the reactor core in the form of hydrogen iodide it would be expected to have reacted with any metal aerosols (such as silver, cadmium and tin) and structural surfaces (such as stainless steel) to form relatively non-volatile metal iodides. However, these compounds (with the exception of silver iodide) are water soluble and would not be preserved after the accident. Cesium is calculated to be transported in the form of cesium hydroxide in the TMI-2 accident, neglecting the influence of boric acid [7]. It would be expected that cesium hydroxide would react with structural surfaces in the upper plenum to form: (1) a cesium silicate within an inner oxide layer [27] that would be insoluble in reflood water, or (2) both water soluble and insoluble reaction products [28]. Results from the examination of two control rod drive leadscrews [29] indicate that < 1% of the core inventory of ^{137}Cs remained on structural surfaces in the upper plenum, although 55% was found in water repositories within the reactor building [30]. A possible explanation for this behavior is that cesium hydroxide reacted with boric acid to form cesium borate which is relatively unreactive but water soluble.

The OECD LOFT FP-2 experiment [31] simulated the early stages of an interfacing loss-of-coolant accident, or V-sequence, and utilized coolant containing boric acid. The release and transport of fission products were measured in this experiment in which a 101-rod fuel bundle containing eleven silver-indium-cadmium control rods was heated by decay heat and zircaloy-steam reaction to temperatures above 2100 K for 4.5 minutes. In addition to on-line gamma spectrometer measurements, the measurement system featured deposition coupons in the plenum, filtered sample lines and simulated low pressure injection system piping (the fission product transportation pathway) that were sealed off prior to reflood. The damaged bundle is undergoing postirradiation examination to address the relationships between core melt progression phenomena and fission product release and transport. This experiment should provide valuable information about the effects of core material chemistry on fission product release and transport because of its typicality to a full scale reactor.

Silver-Indium-Cadmium

Another control material whose chemical behavior is important to the release and transport of fission products is the silver-indium-cadmium control rods used in many PWRs. The control alloy is encased in a stainless steel tube and this rod moves within a zircaloy guide tube. Small scale laboratory experiments [19,26] have shown that control rod failure initiates at temperatures in the neighborhood of 1500 K at low system pressure (such as would be expected under large break loss-of-coolant accidents) by the formation of low melting eutectics (iron-zirconium and nickel-zirconium). These eutectics result from chemical interaction between the stainless steel and zircaloy which are placed in physical contact by the expansion of the stainless steel cladding due largely to the backfill pressure of the helium fill gas and the vapor pressure of cadmium. At high system pressure (representative of small break loss-of-coolant accidents and station blackout) control rod ballooning does not occur, and failure is initiated by melting of the stainless steel at about 1700 K. Following melting of the stainless steel, the zircaloy guide tube is quickly attacked and liquefied due to the formation of iron-zirconium and nickel-zirconium eutectics. Molten control alloy from above the control rod failure point drains out of the control rod and guide tube and runs down to lower elevations where it freezes. Small scale laboratory experiments [19,26] indicate that cadmium aerosols are produced after control rod failure. Aerosols are thought to play a significant role in fission product transport from the core and deposition within the reactor coolant system and are discussed in the next section.

AEROSOLS

The formation of cadmium aerosols observed in laboratory experiments [19,26] is confirmed by the SFD 1-4 in-pile experiment [5] in which cadmium (from silver-indium-cadmium control rods) and tin (from the zircaloy) were found to be the dominant aerosol sources. In this experiment, aerosol generation was found to be continuous over the entire time that fission products were being released from the test bundle. Iodine was transported considerably further downstream in SFD 1-4 than in a previous experiment (SFD 1-3) in which silver-indium-cadmium control rods were not present, perhaps as a result of aerosol-fission product vapor interactions [5].

As discussed previously, the interaction between fission product vapors and aerosols could be either physical (vapor condensation onto aerosols) or chemical (formation of stable chemical compounds on the surface of aerosols). Vapor condensation onto aerosols is modeled in fission product transport codes such as SCDAP/RELAP5 [32] and MELPROG [33], but chemical interactions are not yet modeled despite their potential importance. In-pile experiments that included silver-indium-cadmium control materials, in addition to SFD 1-4, are DF-3 (ACRR), STEP-4 (TREAT), and FP-2 (LOFT). The STEP-4 test [34] did not achieve temperatures sufficient to release fission products from the test bundle. Test DF-3 was designed to measure core degradation phenomena but not the transport of fission products and aerosols. In test FP-2, fission product and aerosol transport was measured and currently is being analyzed. Cadmium was found to be the dominant control rod material transported beyond the reactor vessel in the TMI-2 accident [7].

The STEP tests [34] consisted of two low pressure tests and two high pressure tests. Natural circulation prevented the two high pressure tests from achieving temperatures high enough to release significant amounts of fission products, except for cesium in test STEP-3. Aerosol releases in tests STEP-3 and -4 were dominated by iron and silicon from the structural materials of the test bundle, although some cadmium from the control rod in STEP-4 was measured. Peak temperatures of 2700 K or greater were calculated for tests STEP-1 and -2 and the chemical forms of fission products transported to an aerosol sampling system above the bundle of four irradiated fuel rods were studied. It was concluded from these tests with highly irradiated fuel and limited steam supply that cesium was transported principally as cesium hydroxide and iodine was probably transported as cesium iodide. Deposits of iodine without cesium on nickel and silver surfaces, and the formation of cesium-silver-iodine needles indicate that the iodine chemistry operative under the conditions of the STEP tests was complex. The most probable mode of transport of tellurium was given as cesium telluride, although another possibility suggested was cesium tellurite. Evidence is presented for cesium telluride in the relatively reducing conditions of the effluent in these steam-limited tests; however, it should be noted that this compound is not stable in steam [14], which is likely to be present in the effluent from the core in most severe accident scenarios. Molybdenum was observed located with cesium in deposits that appeared crystalline, suggesting cesium molybdate as the transporting species. This observation is of interest in regards to the question of stable cesium compounds mentioned earlier in connection with molten debris in the TMI-2 reactor.

Tin was a major constituent of the aerosol in STEP-1 and -2 and was second only to cadmium in SFD 1-4. This material was released from the zircaloy cladding, probably as it was concentrated in front of advancing oxide layers in molten zircaloy. Tin, like cadmium, was released throughout the high temperature portion of the SFD 1-4 test. The continuous aerosol generation and sustained fission product release during the high temperature portion of the SFD 1-4 experiment [5] are a result of the incoherent nature of melt progression. In this experiment melt relocations of varying compositions and, therefore, melting temperature, occurred over a range of times (as evidenced by multiple oxide layers within the relocated melts). Some of these relocations involved the interaction of high temperature ($T > 2200$ K) U-Zr-O melts with silver-indium-cadmium control alloy low in the bundle. These interactions caused heating of the control alloy to high temperatures and volatilization of cadmium over the period of the high temperature transient when volatile fission products were released.

FLOW BLOCKAGE

In addition to the formation of aerosols, the relocation of molten control materials leads to the formation of blockages low in the core by freezing of these materials. Such blockages are apparent in the SFD 1-4 and FP-2 tests. Continued accumulation of this material results in the formation of a lower crust as evidenced in the damaged TMI-2 core. In addition to the tendency to form blockages to coolant flow, the relocated control material permits the accumulation of relocating debris on top of it, including hotter, ceramic melts which can re-heat portions of the metals causing continued evolution of vapors such as tin, silver, and cadmium.

Evidence has been found in the TMI-2 accident, in in-pile tests (SFD 1-4, FP-2), and in out-of-pile tests [35] that spacer grids low in the core can act as molten debris collectors and thus promote blockage formation. These out-of-pile tests also demonstrate that inconel spacer grids higher in the core can initiate liquefaction by the formation of iron-zirconium and nickel-zirconium eutectics at temperatures of about 1500 K.

SUMMARY OF IN-PILE TEST CONDITIONS

Some of the important factors affecting chemistry in in-pile tests conducted to date are listed in Table 2. The tests are listed in order of their physical scale, with the smallest tests at the top of the list and the largest at the bottom. Nineteen in-pile tests have been performed to date, in addition to the examination of the damaged core from the TMI-2 accident. Analyses of the tests are in various stages of completion.

The table demonstrates that the in-pile testing has been very diverse. The experiments differ in test bundle configuration and size, rod length, steaming rate, the extent of fuel irradiation and the presence of control materials. The two source term tests (ST-1 and ST-2) consisted of four 15-cm long irradiated rod segments in a reducing atmosphere in the ACRR reactor at Sandia. These tests are in the process of being analyzed for fission product and aerosol release and transport behavior and bundle damage phenomena [16,17]. The STEP tests were conducted using four 1-m long irradiated fuel rods by Argonne National Laboratory in the TREAT reactor at the Idaho National Engineering Laboratory (INEL). These tests have been completed and the analysis presented in an EPRI summary report [34]. The debris formation (DF) test series was carried out with 0.5 m-long unirradiated fuel rods in the ACRR reactor at Sandia. Tests DF-1 and DF-2 used 9 fuel rods, DF-3 had 8 fuel rods and one silver-indium-cadmium control rod, and DF-4 contained 14 fuel rods and a portion of a B_4C control blade. The analysis is complete for DF-1 and DF-2 [36]. A report is in the process of being published on DF-4 [25] and DF-3 is still being analyzed. The severe fuel damage (SFD) test series carried out in the PBF reactor at the INEL utilized a 1-m long bundle capable of holding 32 fuel rods. In Tests SFD-ST and SFD 1-1 unirradiated fuel rods were employed and in Tests SFD 1-3 and SFD 1-4 irradiated rods were used. Test SFD 1-3 contained four empty control rod guide tubes and Test SFD 1-4 contained four silver-indium-cadmium control rods. Documentation of the first two tests is complete and is continuing on the last two [5]. Four tests have been performed in the full-length high-temperature (FLHT) test series being carried out in the NRU reactor at Chalk River Nuclear Laboratory in Ontario, Canada. These tests have used 4-m long bundles. Twelve unirradiated rods were used in the first two tests. The last two tests each contained one irradiated rod, ten unirradiated rods, and a gamma thermometer. These tests were directed primarily at the measurement of hydrogen generation during core melt progression in the full length bundles. Evaluation of these tests is being carried out at Pacific Northwest Laboratory [37]. The FP-2 test was carried out in the LOFT reactor at the INEL and contained 101 1.7-m long fuel rods and eleven silver-indium-cadmium control rods. Evaluation of the damage state in the test bundle and the fission product and aerosol transport data is continuing within the OECD LOFT program.

TABLE 2. SUMMARY OF IN-PILE TEST CONDITIONS

Experiment/ Accident	Fuel Irradiation	Control Materials	Spacer Grids	Steam Input	Time-Resolved Measurements	
					Fission Products	Aerosols
ST-1	30 GWd/tU	None	None	Ar/H_2	Yes	Yes
ST-2	30 GWd/tU	None	None	Ar/H_2	Yes	Yes
STEP-1	30 GWd/tU	None	None	Limited	No	Yes
STEP-2	30 GWd/tU	None	None	Limited	No	Yes
STEP-3	30 GWd/tU	None	None	Limited	No	Yes
STEP-4	30 GWd/tU	Ag-In-Cd	None	Limited	No	Yes
DF-1	Trace	None	Inconel	Limited	No	No
DF-2	Trace	None	Inconel	Limited	No	No
DF-3	Trace	Ag-In-Cd	Inconel	Limited	No	No
DF-4	Trace	B_4C	Inconel	Limited	No	No
SFD-ST	Trace	None	Inconel	Excess	Yes	No
SFD 1-1	Trace	None	Inconel	Limited	Yes	No
SFD 1-3	30 GWd/tU	None	Inconel	Limited	Yes	No
SFD 1-4	30 GWd/tU	Ag-In-Cd	Inconel	Limited	Yes	Yes
FLHT-1	Trace	None	Inconel	Excess	No	No
FLHT-2	Trace	None	Inconel	Limited	No	No
FLHT-4	1 - 30 GWd/tU 10 - Trace	None	Inconel	Limited	Yes	No
FLHT-5	1 - 30 GWd/tU 10 - Trace	None	Inconel + Zry	Limited	Yes	No
LOFT FP-2	0.45 GWd/tU	Ag-In-Cd +H_3BO_3	Inconel	Excess	Yes	Yes
TMI-2	3 GWd/tU	Ag-In-Cd +H_3BO_3	Inconel	Excess	No	No

Inspection of Table 2 reveals that relatively few of the nineteen tests have been performed with the full complement of materials and conditions typical of power reactors in severe accidents. Most glaring is the paucity of tests with BWR materials. Also few in number are tests with time-resolved measurements of fission products and aerosols utilizing realistic materials and conditions.

CONCLUSIONS

This review of the influence of chemistry on severe accident phenomena indicates that the release and transport of fission products is strongly coupled to core melt progression phenomena. The irradiation history of the fuel determines the microstructure of the fuel and influences the release rates of volatile fission products and noble gases prior to fuel liquefaction. The irradiation exposure of the fuel influences fission product chemistry by setting concentration levels which, in addition to the H_2/H_2O ratio in the gas phase, is important to determining the chemical speciation of the fission products released from the fuel. The influences of other materials in the core, such as control and structural materials, that can form vapors and aerosols and can interact with fission product vapors can be considerable. Incoherent core melting and relocation processes provide considerable time at high temperature and ample opportunity for chemical interactions such as steam oxidation, eutectic formation, and fission product vapor - aerosol reactions.

The preponderance of information generated to date on core melt progression and the release and transport of fission products and aerosols under severe accident conditions has been under PWR conditions. Much understanding of the relationship between core melt progression and fission product behavior has been obtained under these conditions. Lacking, however, is understanding of chemical interactions of fission product vapors and aerosols under integral effects conditions, especially with BWR materials.

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