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CHEMICAL CONSIDERATIONS IN SEVERE ACCIDENT ANALYSIS\*

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ABSTRACT

The Reactor Safety Study presented the first systematic attempt to include fission product physicochemical effects in the determination of expected consequences of hypothetical nuclear reactor power plant accidents. At the time, however, the data base was sparse, and the treatment of fission product behavior was not entirely consistent or accurate.

Considerable research has since been performed to identify and understand chemical phenomena that can occur in the course of a nuclear reactor accident, and how these phenomena affect fission product behavior. In this report, the current status of our understanding of the chemistry of fission products in severe core damage accidents is summarized and contrasted with that of the Reactor Safety Study.

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## I. INTRODUCTION

The Reactor Safety Study<sup>1</sup> (WASH-1400) represents the first major systematic attempt to estimate the escape of radionuclides into the biosphere in the event of a nuclear reactor accident involving severe core damage. It is also the first effort to consider chemical influences on the behavior of fission products under such accident conditions in a detailed manner.

Since the issuance of the WASH-1400 report, two major accidents have occurred and considerable research has been performed to develop a clearer understanding of fission product release from overheated fuel and subsequent transport within the primary system and reactor containments. Hence, although some of the models that had been developed in the course of the Reactor Safety Study are now regarded as crude or deficient, it is nonetheless instructive to review the WASH-1400 approach, if only as a means of gauging the current state of the technology.

## II. THE WASH-1400 APPROACH

### A. FISSION PRODUCT RELEASE MECHANISMS

The Reactor Safety Study identified four conditions or times in the course of a severe core damage accident for which major driving forces exist for fission product escape from irradiated UO<sub>2</sub> fuel. These conditions were then identified in terms of the corresponding release components:

1. Gap release: release of fission products that are associated with the interconnected void volume in a fuel rod at the time of initial failure of the fuel cladding. The fuel-clad gap region constitutes the major component of this volume.
2. Meltdown release: escape of fission products as the fuel becomes molten.
3. Vaporization release: release that occurs from the molten mass when penetration of the reactor vessel has occurred and the material comes into contact with concrete structures. Release results from sparging of the molten core material by the gases formed upon decomposition of the concrete.
4. Oxidation release: escape from finely divided fuel particulates that are formed as the result of a steam explosion. This release is due to extensive oxidation of the droplets upon their dispersal into an air atmosphere.

#### 1. Gap Release

The gap inventory is developed during normal reactor operation; it is largely determined by the heat rating of the rod and, to a lesser degree, the extent of irradiation.<sup>2</sup> In the Reactor Safety Study, three independent sets of calculations were performed; the results for selected long-lived nuclides, expressed as fractions of total core inventory, are presented in Table I. Considering that somewhat different calculational approaches were

employed by the three groups, the results are in remarkably good agreement for the noble gases and the halogens, and are in reasonably good agreement for the alkali metals, but diverge for the alkaline earths. This divergence arises from a thermodynamics constraint within the ANC approach that does not appear in the other two calculational methods. In all three calculational approaches, the diffusion coefficient values that were required were derived from a rather sparse amount of annealing data.<sup>3-5</sup>

For long-lived isotopes, the gap fraction is directly proportional to the net rate of transport of the isotope through the  $UO_2$  crystal lattice. All three groups whose values are listed in Table I thus imply that the differences in size and mass between krypton and xenon have no significant effect on their rates of release from the  $UO_2$  matrix. In contrast, whereas the ANC and BCL results for the halogens suggest diffusion as neutral species through the  $UO_2$  lattice (like the noble gases), the ORNL value indicates enhanced transport relative to that of the noble gases. In like manner, all three values for the alkali metals indicate transport either as charged species or chemical interaction with the  $UO_2$  (or both), but there is disagreement concerning whether or not this results in enhanced transport or delays the diffusion process. On the other hand, there is agreement regarding a marked decrease in the release of the alkaline earths from the fuel.

The fraction of rod inventory that would be released in the event of loss of cladding integrity is stated in WASH-1400 as the product of the gap fraction and an escape fraction. The escape fractions, which are presented in Table II, were developed in part from chemical considerations. For example, although the boiling points of both elemental iodine and elemental cesium would be expected to be exceeded prior to cladding rupture, the escape fractions for these fission products were taken to be one-third, rather than unity as for the noble gases. Retention of a large fraction of the iodine gap inventory was believed to result from chemical reaction with the Zircaloy cladding, whereas the cesium was believed to be retained by interaction with both the cladding and the fuel. Although thermodynamics calculations were made which indicated the formation of  $CsI$ , this result was discounted for lack of experimental support. Since cesium is present in about a tenfold excess over that of iodine, the fact that the Reactor Safety Study yielded cesium releases from the gap region that were 5 to 30 times higher than the corresponding releases of iodine did not appear contradictory to the Reactor Safety Study researchers to the position they had taken with regard to  $CsI$  formation.

For strontium and barium, neither the elements nor the oxides have appreciable volatility at projected cladding rupture temperatures. An escape fraction of  $10^{-4}$  was therefore selected; this value was based upon crude in-pile experimental data.<sup>6</sup> Tellurium, on the other hand, would have a significant vapor pressure under cladding rupture conditions, but it was presumed to be released from the fuel in elemental form and to react subsequently with the Zircaloy.

## 2. Meltdown Release

Cladding rupture temperatures were estimated to range between 760° and 1100°C. In a core meltdown accident, initial melting was estimated to occur within minutes of cladding rupture, and the entire core was assumed to become molten within 30 min to 2 h. As a consequence, fission product release between the period of cladding rupture and fuel melting was considered to be insignificant. Release during the meltdown phase was described in terms of the fraction of the core that became molten and the release fraction from molten fuel. These release fractions are presented in Table III.

Although almost complete release of the noble gases and halogens from molten fuel was estimated, some solubility of the alkali metals (and therefore less than complete release) was postulated, based upon experimental observation and thermodynamics considerations. (It is of some interest to note that this result has largely been ignored in subsequent studies, but may have been verified by data obtained from analyses of core debris from the Three Mile Island Reactor<sup>7</sup>.)

High retention of the tellurium in the molten phase, like its low release from the gap region, is predicated on its reaction with unoxidized Zircaloy in the melt. For the remaining fission products, the release fractions reflect volatility considerations under a chemically reducing environment that is presumed to prevail due to the presence of metallic zirconium and hydrogen.

## 3. Vaporization Release

When the molten mass penetrates the reactor vessel, a two-phase mixture, metal and oxide, is postulated to fall (or run) into the reactor cavity. The heavier oxide phase would first contact the concrete, and the gases resulting from the decomposition of the concrete, H<sub>2</sub>O and CO<sub>2</sub>, would sparge the molten material. At the point of reactor vessel failure, however, the molten material becomes exposed to chemically oxidizing conditions. As a consequence, oxides from the concrete and from oxidation of the initially lighter iron phase would dissolve in the oxide phase, thus reducing its density so that, eventually, the ordering of the two layers would reverse.

Because a complete mechanistic description appeared to pose an intractable problem, an empirical approach to fission product release during this period was developed. The key factors involved the recognition that the environment was now chemically oxidizing and that a metallic iron phase was present. Release fractions for this phase of the accident are presented in Table IV.

Note that, at this point, all of the zirconium is presumed to have been oxidized, thus releasing the chalcogens, but the noble metals, Mo and Tc, were assumed to distribute to the molten iron phase.

#### 4. Oxidation Release

In the event a steam explosion was to occur as the molten material escaped from the reactor, finely divided fuel particulates would be dispersed within the reactor containment. In a pressurized water reactor (PWR), these particulates would thus be ejected into a steam-air atmosphere, where oxidation of the fuel and some of the fission products contained within the particulates would result. For this release component, the oxidation release fractions presented in Table V were developed.

#### B. TRANSPORT THROUGH THE PRIMARY SYSTEM

An accurate description of the various mechanisms and the thermal hydraulic conditions affecting fission product transport through the primary system appeared to present too formidable a task to be undertaken at the time of the Reactor Safety Study, hence the study considered this aspect of the problem primarily in a qualitative manner. No attenuation of the fission product concentrations upon transport through the primary system of a PWR was allowed, as it was argued that such an attenuation was already taken into account in the release fractions, since those values were based upon experimental data in which the release was indicative of a high temperature region of the apparatus rather than from the peak temperature fission product source.

Two exceptions, both involving boiling water reactors (BWRs), were noted, however. In one of these, the fission product escape pathway involved transport through a large column of water. In this case, only 10% of all of the fission products released from the core (except the noble gases) was estimated to escape from the primary system. In the second case, dry heatup of the fuel was assumed to occur; in this instance only two-thirds of the fission products are assumed to escape from the primary system owing to the absence of a driving force.

Although some consideration was given to chemical effects in this aspect of the study (iodine as  $I_2$  or HI was considered), because of the global nature of the treatment, the results were independent of chemical form.

#### C. BEHAVIOR IN THE CONTAINMENT

In the Reactor Safety Study, two models were employed to describe fission product behavior in the containment. In one of these, the containment was considered to be a single, well-mixed volume; in the other, a multicompartiment description, consisting of four internal compartments for a PWR, and as many as six compartments for a BWR, was employed. For each compartment, a mass balance expression was derived which consisted of three groups of terms: (1) source terms for the compartment; (2) terms accounting for airborne depletion within the compartment; and (3) compartment leakage terms.

Only three classes of fission products were considered; these were the noble gases, iodine (as elemental iodine and organic iodine), and particulates (all other fission products).

No depletion of noble gases was considered, nor were depletion terms developed for the organic iodides, although such terms were implicit in considerations of their formation from elemental iodine. Elemental iodine depletion mechanisms included deposition on surfaces, removal by sprays, equilibria with aqueous media, and, for BWRs, pool scrubbing and filtration. The same depletion mechanisms (except equilibria with aqueous media) were also considered to describe attenuation of the concentrations of particulates. Also, for particulates, the particle size distribution was assumed to be monodisperse but time dependent, decreasing linearly with time from 15  $\mu\text{m}$  to 5  $\mu\text{m}$  during the first 4 hours of the accident, and remaining at 5  $\mu\text{m}$  thereafter.

The organic iodides were assumed to be chemical analogues of methyl iodide. Their formation was expressed as percent of (elemental) iodine converted. Estimates, which were based upon data presented by Postma and Zavadoski,<sup>8</sup> were developed for two cases for PWRs: one which applied to accidents in which containment sprays were operable; and a second which applied to compartments in which sprays would not be expected to deplete the airborne molecular iodine concentration. For the case in which sprays were not operable, 0.7% of the iodine was estimated to appear as methyl iodide, whereas only 0.4% conversion was estimated for situations involving sprays. For BWR accidents, 0.7% conversion was assumed for all accidents.

Interestingly, a distinction was made between the formation of methyl iodide by radiolytic reactions and by nonradiolytic reactions in developing the conversion estimates. This was the only situation in which radiolytic effects were considered.

Attenuation of the fission product concentrations upon transport through the leaks in the containment and particularly upon failure of the basemat were also considered in the Reactor Safety Study, but this aspect is regarded as outside the scope of the present report.

### III. CURRENT STATUS

The Reactor Safety Study was not favorably received by the technical community.<sup>9</sup> Most of the criticisms concerned the methodologies that were employed to classify the various types of severe core damage accidents, the completeness of the study, and the use of probabilistic methods.<sup>10</sup> Surprisingly little criticism was made of the treatment of the physicochemical aspects of fission product transport in the study, even though these establish the consequences in the risk formula and are more readily amenable to experimental verification than the probability values that constitute the other part of the risk equation.

In spite of its deficiencies, WASH-1400 has proven to be a truly seminal document; directly or indirectly, it has influenced the course of severe accident research for the past decade. It is thus appropriate to describe the current status of our understanding of fission product behavior using WASH-1400 as the benchmark.

## A. FISSION PRODUCT RELEASE MECHANISMS

The four major conditions under which significant fission product releases can occur that were identified in the Reactor Safety Study remain essentially unchanged, except that vaporization release is now more usually described as release due to core-concrete interactions, and oxidation release is now associated with reactor vessel failure at elevated pressure and/or hydrogen combustion in containment rather than with steam explosions. In addition, due partly to the realization that the timing of fission product release can be important and that severe core damage accidents need not necessarily involve extensive core melt (as was initially believed to have been the case at the Three Mile Island Reactor), considerably more attention has been given to fission product releases over the period just after cladding failure and prior to fuel melting.

### 1. Gap Release

Changes in fuel design and operation have resulted in considerable reduction in the gap inventories of the current generation of reactor fuel rods (this may change, however, as the fuel is utilized to higher burnups). As reported previously,<sup>11</sup> a significant data base has been developed and considerable progress has been made in describing the development of the gap inventories of the noble gas fission products. In addition, although measurements of releases of Te, I, Xe, and Cs isotopes from small samples of unclad fuel indicate decidedly different rates of migration of these nuclides through the  $UO_2$  fuel matrix,<sup>12-14</sup> the results are not consistent and are at variance with direct measurements of gap inventories of commercial irradiated fuel rods<sup>15-17</sup> and with observed rates of release of I, Kr, and Cs from such rods at elevated temperatures.<sup>18,19</sup> Although this facet is relatively inconsequential, the cause for the discrepancy between the results derived from experiments with small, unclad fuel fragments and those obtained from tests with relatively large, clad, fuel rod segments should be elucidated.

Direct identification of the chemical form of iodine within the gap region is lacking, but considerations of its migration behavior within intact fuel rods<sup>20,21</sup> and its release characteristics from fuel with defected claddings<sup>22</sup> clearly suggest that the bulk of the iodine within the gap region resides there as CsI. Moreover, since cesium is present in ten-fold excess relative to that of iodine, elemental cesium appears to be the dominant form of cesium in the gap region, but its partial pressure may be determined by equilibrium with cesium uranate solid-phase compounds.

The simple concept of specifying invariant gap escape fractions to account for fission product escape at the time of cladding failure (and including the period up to melting of the fuel) has been replaced by more mechanistic models. For the noble gases, complete release of the gap inventories is still postulated, whereas for cesium and iodine, the release is described<sup>23</sup> as the result of two processes, the sweeping action of the fission gases at the time of cladding rupture ("burst release"), and diffusion of the gas-phase cesium and iodine species through the gap region and out of the rupture location afterwards ("diffusion release").

(The functional dependences of these types of release on fission product and fuel rod geometry are presented elsewhere.<sup>23</sup>) In contrast, all other fission products are assumed to escape only as fuel fines components during rod rupture, and the amount of fuel fines ejected at the time of cladding rupture is taken to be 0.003% of the total amount of fuel in the rod, and independent of rupture temperature.<sup>24</sup> (Although the data base for the fuel fines release value is sparse, the result is more or less consistent with the gap escape fraction for the alkaline earths that was employed in the Reactor Safety Study.)

Burst release, which is assumed to occur instantaneously, is the dominant mode for fission product release at temperatures up to about 1200°C, although diffusion release can be significant for cesium and iodine nuclides if the duration of the release is long.

## 2. Meltdown Release

At temperatures in excess of 1200°C, transport through the  $UO_2$  is considered to be the process that limits the rates of escape of the fission products from the fuel. For this situation, three major computational programs are currently in use. One of these, CORSAR,<sup>25</sup> represents an empirical correlation of experimental data, whereas the others, FASTGRASS<sup>26</sup> and VICTORIA,<sup>27</sup> have mechanistic bases. FASTGRASS, for example, includes intra- and intergranular migration of fission products via diffusion to noble gas bubbles and subsequent migration of the bubbles along interlinked pores and micro-cracks. Local chemical equilibria are also included to establish the chemical forms of specific fission product nuclides.

The VICTORIA code also contains a mathematical description of diffusion within  $UO_2$  grains, and includes other transport limitations (vaporization and liquid- and gas-phase transport) as well. Moreover, VICTORIA contains an extensive treatment of chemical equilibria associated with the fuel-fission product system. These models all appear to predict the complete loss of cesium, iodine, and the noble gases even prior to fuel melting, and significant loss of less volatile fission product species, such as the isotopes of Sb, Ba, and Sr. Thus, for severe core damage accidents which develop in a gradual manner, the newer approaches predict considerably greater releases of most of the fission products earlier into the accident than would be predicted using the WASH-1400 methodology.

## 3. Vaporization Release

Considerable research has been conducted on core-concrete interactions and the concomitant releases of fission products since the issuance of the Reactor Safety Study. As a consequence, the use of invariant release fraction values for pertinent fission product groups has been replaced by the utilization of two computer programs, CORCON<sup>28</sup> and VANESA,<sup>29</sup> or by an integral code, MAAP.<sup>30</sup> The CORCON code was developed primarily to describe ablation of concrete structures by molten core components that have exited from the reactor vessel; it is therefore employed to calculate the temperature history of the melt and the rate of generation of gases released due to decomposition of the concrete. In the current practice, these parameters are input into the VANESA code, which is used to calculate the



time-dependent release of material from the molten masses as the result of four processes: (1) evaporation, (2) sparging by the concrete decomposition gases, (3) chemical reaction, and (4) mechanical formation of aerosols by the sparge gases.

The chemistry of approximately 150 vapor species representing 27 selected elements is described by the current version of the VANESA code, and efforts are presently under way to integrate CORCON and VANESA into a single computational program. Comparison of results using the WASH-1400 methodology with the results of CORCON/VANESA computations is complicated by changes in chemical conditions that are postulated to occur over the period in which vaporization (or core-concrete interaction) release is significant. As an example, WASH-1400 assumes complete releases of tellurium and antimony nuclides during this period, owing to the assumption that all of the zirconium in the melt is oxidized. In the CORCON/VANESA approach, however, a significant amount of metallic zirconium is postulated to be present, and the releases of tellurium and antimony occur in a more gradual manner until oxidation of zirconium is complete.

Thermal hydraulics and chemistry considerations have already been integrated in the MAAP code, and chemical equilibria among 34 species are determined to ascertain the corresponding partial pressures contained within the sparge gas hubbles.

#### 4. Oxidation Release

Relatively little research has been conducted since the Reactor Safety Study on fission product chemical behavior as finely divided molten fuel is dispersed throughout the reactor containment as a result of steam explosions or the ejection of the molten core through a breach in the reactor vessel at high primary system pressure. However, experiments have been performed<sup>31</sup> to investigate the dispersion of molten material from vessels under high pressure, and these experiments are to be extended to include studies of fission product behavior during the dispersion process.<sup>32</sup> Also, research has been conducted on the possible formation of volatile iodine chemical forms as the result of hydrogen burns within containment; these studies have demonstrated thermal decomposition of CsI in the presence of alumina aerosols.<sup>33</sup>

#### B. TRANSPORT THROUGH THE PRIMARY SYSTEM

Although the chemistry of the fission products within the primary system is determined in large measure by the  $H_2/H_2O$  ratio, additional complexities are introduced by the presence of oxides of structural components, and materials for control of the nuclear reaction (boron carbide in BWRs and borates, silver, indium, and cadmium in PWRs). Much of the current focus is on the possible formation of volatile iodine-containing species; such studies have demonstrated the decomposition of CsI as a result of the formation of thermodynamically preferred cesium metaborate compounds.<sup>34</sup> However, some of the research has been directed to an examination of enhanced volatility of fission product oxides in the presence of steam<sup>35, 36</sup> and, perhaps more importantly, to interactions of fission products with aerosol and structure surfaces.<sup>37</sup>

A major current concern<sup>38</sup> involves resuspension of fission products deposited on primary system surfaces, since this phenomenon can lead to more severe consequences in certain accident scenarios (late containment failure) than the consequences that would result under the apparently conservative assumption of no fission product deposition in the primary circuit as was made in the Reactor Safety Study.

Although a computational model (TRAP-MELT)<sup>39</sup> has been formulated to describe fission product transport in the primary system, the chemical aspects of the phenomena involved are lacking. Indeed, a unified treatment of the chemistry in the primary circuit is lacking, although VICTORIA is intended to provide this treatment.

#### C. BEHAVIOR IN THE CONTAINMENT

Although the methodology developed in WASH-1400 to describe fission product behavior in the reactor containment and associated volumes has not been altered appreciably in current approaches, considerable refinement has nonetheless resulted. Now, for example, multicompartment descriptions of the pertinent volumes are employed in the computational models for heat and mass transfer,<sup>40</sup> and rather sophisticated computational programs are being utilized to describe aerosol behavior.<sup>41,42</sup> Moreover, the aerosol computer codes use particle size distribution functions that vary with time as dictated by the physics.

Considerable attention is being given to fission product iodine behavior in the containment,<sup>11</sup> and descriptions of the chemical kinetics and equilibria of recognized processes have been unified in the computational program TRENDS.<sup>43</sup> Although the computer program currently employs a number of empirical relationships that have been developed to correlate experimental observation, these relationships will unquestionably be replaced eventually with more mechanistic descriptions as ongoing research reaches fruition. Such research includes studies of radiolysis effects on iodine aqueous chemistry,<sup>44</sup> investigations of iodine interactions with surfaces,<sup>45</sup> and studies of the mechanisms for the formation of organic iodides.<sup>45-48</sup>

#### IV. CONCLUDING REMARKS

There is a common misperception that, after over 10 years of extensive research of phenomena associated with severe core damage accidents, including detailed considerations of the chemistry involved, the uncertainties in the results are even larger than what was perceived to be the uncertainties attending the estimates provided by the Reactor Safety Study. The actual situation, of course, is to the contrary; we have simply become aware of a larger number of processes that can provide alternate modes of behavior than those previously postulated.

What is lacking, however, is a methodology to ascertain the significance a single process may have on overall behavior. Moreover, no amount of modelling and calculation, however sophisticated, can ever be so convincingly complete as direct observation. Yet, ever greater emphasis is being given to computer code development, partly in response to financial

constraints, and less to experimental investigations. And, disturbingly, the notion of the "critical experiment" appears to be an endangered concept.

TABLE I. Gap fractions of selected long-lived fission products<sup>a</sup>

Fission product	Gap fraction		
	ANC	BCL	ORNL
Xe, Kr	0.06	0.10	0.08,
I, Br	0.06	0.10	0.14
Cs, Rb	0.20	0.05	0.21
Sr, Ba	0.000004	0.02	0.02

<sup>a</sup>From WASH-1400, App. VII (ref. 1).

TABLE II. Gap escape fractions<sup>a</sup>

Fission product	Escape fraction
Xe, Kr	1
I, Br	1/3
Cs, Rb	1/3
Sr, Ba	10 <sup>-4</sup>
Te, Se, Sb	10 <sup>-3</sup>

<sup>a</sup>From WASH-1400, App. VII (ref. 1).

TABLE III. Meltdown release fractions<sup>a</sup>

<u>Fission product</u>	<u>Release fraction</u>
Xe, Kr	0.9
I, Br	0.9
Cs, Rb	0.8
Te	0.15
Ba, Sr	0.1
Noble Metals	0.03
Rare Earths	0.003
Zr, Nb	0.003

<sup>a</sup>From WASH-1400, App. VII (ref. 1).

TABLE IV. Vaporization release fractions

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<u>Fission product</u>	<u>Release fraction</u>
Xe, Kr	1
I, Br	1
Cs, Rb	1
Te, Se, Sb	1
Ru, Rh, Pd, Mo, Tc	0.05
Refractory Oxides	0.01

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<sup>a</sup>From WASH-1400, App. VII (ref. 1).

TABLE V. Oxidation release fractions

<u>Fission product</u>	<u>Release fraction</u>
Xe, Kr	0.9
I, Br	0.9
Te, Se (Sb)	0.6
Ru (Mo, Tc, Pd, Rh)	0.9

<sup>a</sup>From WASH-1400, App. VII (ref. 1)



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