

Technical Issues Associated with Air Ingression During Core Degradation

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INTRODUCTION

The sophistication with which reactor accident progression can be predicted today is really quite astounding. In the early days of the commercial development of nuclear power, detailed analyses of core degradation under reactor accident conditions were not attempted. It was simply assumed that the combination of decay heat and the heat of steam reaction with the zircaloy cladding on the fuel would lead to relatively uniform degradation of the core, fuel melting and, eventually, relocation of the core materials to the lower plenum of the reactor vessel. The molten core debris would penetrate the vessel, cascade into the reactor cavity and engage in interactions with water or with concrete. The assumption that all the zircaloy cladding was oxidized by steam provided a convenient bound on the amount of hydrogen that would eventually reach the containment and pose there a threat to the integrity of the containment.

This simple bounding view of the core degradation process was changed significantly by the accident at Three Mile Island. The post-accident examinations of the core at this damaged reactor showed that indeed the central regions of the core had heated to the point of melting. Much of the cladding on the fuel in the central regions had been oxidized by steam to form hydrogen. Molten core debris had relocated to the lower plenum of the reactor vessel even though there were significant flows of coolant at the time of the relocation. Various analyses have shown that had the coolant flows not been promptly established the core debris would have soon penetrated the reactor vessel.

The unexpected feature of the damaged core at Three Mile Island was that nearly half of the core was still in place as fuel rods. Some rods were damaged significantly, but some were full rods only slightly more oxidized than would have been the case were there no accident. It was obvious that heat losses by both convection and radiation kept the peripheral regions of the core cool so that core degradation was very nonuniform. Only the central portions of the core were involved in the extensive, early degradation of the accident. Very soon after these findings of the post-accident analysis were made known a variety of computer models of core degradation were developed to account for the non-uniform core degradation. It was soon found that the highly nonuniform degradation was the norm for reactor accidents. Such degradation involving only the central regions of the core early could be expected for nearly all reactor accidents and not just the unique accident at Three Mile Island. Much of the analytic attention was then focused on how extensively fuel would be involved in the degradation before molten core debris drained from the

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core region into the reactor vessel plenum. Crust formation on the lower core support plate and the dissolution of fuel rods in molten Zr-U-O mixtures have been much investigated and debated.

Much less attention has been devoted to the issue of what happens to the fuel that remains largely intact within the reactor core region. Of particular interest is the behavior of this residual fuel, which could amount to 50-80% of the core, after the vessel ruptures. In this presentation, some of the technical issues that arise in connection with the prediction of the behavior of the residual fuel are discussed. The attention is needed because simple geometry means so much of the fuel and the fission products contained in this fuel are involved in this later stage of the core degradation.

AIR INGRESSION

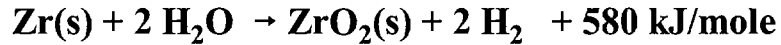
The immediate consequences of vessel penetration by core debris and expulsion of the core debris into the reactor cavity can be quite dramatic especially if the reactor coolant system has remained pressurized as it did during the accident at Three Mile Island. Even if pressurization is not maintained, as some have argued despite the evidence from the Three Mile Island accident, the processes following core debris expulsion from the vessel can be quite impressive. Expulsion of core debris into a reactor cavity filled with water could cause steam explosions or certainly sufficient steam generation that core debris could be entrained and expelled from the cavity. Expulsion of core debris into a dry reactor cavity would lead to a very vigorous initial interaction with the structural concrete. This initial interaction would be driven by decay heat and the chemical reactions of whatever zirconium in the core debris that had not been oxidized during the core degradation process.

The initial, vigorous interactions of expelled core debris will not last long. Even core debris interactions with concrete are intense only so long as there is metallic zirconium to augment decay heat with chemical reaction heat to drive the interaction. Once this inventory of metallic zirconium is depleted, the interactions progress at a quasi-steady state rate dictated by decay heat. Dissolution of core debris into the molten products of concrete decomposition drops temperature to the melting range of concrete and steel.

The question remains, then, what happens to the residual fuel in the reactor vessel that was not part of the early core degradation and was not expelled into the reactor cavity? One might suppose, at first, that this residual fuel would slowly heat and drain out of the vessel into the cavity. Certainly, this is possible because much of the convective cooling that protected this fuel during the earlier stages of the reactor accident has been lost and radiation cooling of the residual fuel becomes progressively less effective as the surroundings heat. But, on closer inspection, the continued degradation of this fuel might not be so benign. The configuration of fuel within a ruptured vessel and with leakage pathways in the rest of the reactor coolant system is reminiscent of nothing so much as a furnace box. Certainly, in the case of very open reactor cavities typical of large dry containment PWRs in the USA, residual fuel will be exposed to air drawn from the reactor containment into the "furnace box" that is the damaged reactor pressure

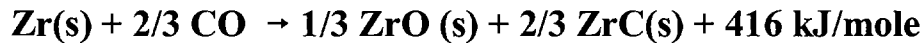
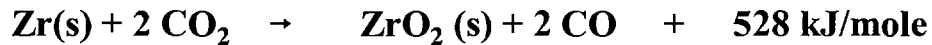
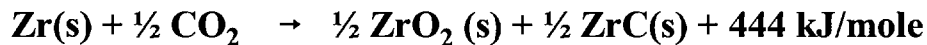
vessel.

Air interactions with fuel must be, surely, very dramatic processes. The heat of reaction of air with hot zircaloy cladding is about twice that of the heat of reaction of steam with zircaloy cladding:



Furthermore, the chemical kinetics of air reaction with zircaloy are quite similar to those of steam reactions with zircaloy as would be expected since rate limitations are predominantly the result of transport through the product oxide. Actually, air reactions with zircaloy may be even faster because the oxide product of air oxidation is, apparently, more susceptible to decrepitation and exfoliation and this leads to breakaway oxidation that progresses more rapidly than conventional parabolic kinetics.

The rate of air oxidation of zircaloy, like the rate of steam oxidation of zircaloy, can become so rapid that it is limited by the availability of gaseous reactant rather than chemical processes. There is, then, a need to have at least an order-of-magnitude estimate of the rate at which air can be drawn into the reactor vessel from the reactor cavity. The need for this estimate is especially acute in the case of reactor cavities that are more closed than those found in many of the large, dry containment PWRs. In these more confined environments, even low intensity interactions of expelled core debris with concrete could produce enough gas to prevent air from intruding into the reactor cavity. Instead, gases that are drawn into the reactor vessel would be the products of concrete decomposition including CO₂, CO, H₂ and H₂O. Reactions zircaloy with CO and CO₂ are not as well known as reactions of steam and air with the metal. Some possible reactions are:



Heats of reaction are about the same as the heats of reaction of steam with zircaloy. The complication of the additional refractory product, zirconium carbide, are difficult to predict.

A SIMPLE MODEL OF AIR INGRESSION

To gain at least a qualitative impression of the nature of air ingress into the reactor vessel following vessel rupture, a simple, two-volume model is derived here. The essential features of the model are shown in the schematic diagram in Figure 1. One volume is the reactor cavity. Reactor cavities in the world population of nuclear power plants are quite variable in design. For the purposes of this simplified, order-of-magnitude analysis, a cavity design that is quite restricted has been selected. The second volume is the reactor vessel and the associated coolant system.

Core debris expelled from the reactor vessel in the earlier stages of an accident will interact with the structural concrete of the reactor cavity. The concrete is assumed to have a density of 2.4 g/cm^3 and to be made with limestone aggregate so upon ablation by core debris, 22 weight percent of the concrete is released as carbon dioxide. It is assumed that the gaseous products of concrete decomposition emerge into the reactor cavity atmosphere at a selected temperature, T^* . Concrete erosion is assumed to take place at a selected rate and to produce gaseous products at the rate of Q moles/s. This highly simplified depiction of core debris interactions with concrete during a reactor accident is not especially realistic. The simplified description has been adopted to illustrate the effects of core debris interaction on the natural convection of air into the reactor cavity. Some scoping analyses were done assuming that a water pool overlay the core debris. The water pool could either quench the debris so only steam emerged from the pool into the reactor cavity, or, as observed in experiments [1], core debris interactions with concrete continued despite the presence of the water so both carbon dioxide and steam emerged into the reactor cavity. Results for these more complicated situations did not differ qualitatively from those of the more simplified situation discussed here.

Hot gases can flow out of the reactor cavity through the annular space around the reactor vessel. This space will not be gas tight, but it will be congested with shielding blocks and vessel insulation. There may be, then, a substantial flow resistance along this path. For the purposes of the simplified model, this flow path is characterized as a circular orifice of diameter D_{out} and a friction factor f . It is also assumed that there are other leak paths into the reactor cavity that collectively can be characterized by a circular orifice of diameter D_{in} and a discharge coefficient K .

The reactor vessel is taken to have been penetrated by core debris at the pole of the lower plenum. The penetration is taken to be circular with diameter D_{ves} . Penetration of the reactor vessel by core debris has received some attention in recent years in connection with the issues of direct containment heating [2]. Several scenarios leading to vessel penetration have been examined. Because of the many uncertainties associated with predicting the size of the hole in the vessel, distributions rather than point values of the hole size have been estimated. Examples of two of these distributions are shown in Figure 2. Median values (50% quantile values) varied among the various scenarios from about 23 to 50 cm. These same studies showed that substantial amounts of clad reactor fuel remained in the vessel following vessel penetration. Typically,

median values of the retained fuel amounted to 60 to 80% of the initial core loading of fuel.

Leak paths will exist in the reactor coolant system. Leak paths may be a rupture of the coolant system that was one of the accident initiators. Or, the rupture may have occurred during core degradation as a result of natural convection heating and creep rupture of the reactor nozzles, the surge line, or even the steam generator tubes [3]. These leak paths are assumed for the purposes of the simplified model to be characterized by a circular orifice of diameter D_{rcs} and friction factor f_{rcs} .

The ambient atmosphere of the reactor containment is taken to be normal air (mole fraction oxygen in air, x_{air} , is taken to be 0.2) at temperature T_{amb} and pressure P_{amb} . The gas temperature in the well-mixed, reactor cavity, T , is found from:

$$Q \int_{T_r}^T C_p(\text{conc}) dT + M_{in} \int_{T_r}^T C_p(\text{air}) dT = Q \int_{T_r}^{T^*} C_p(\text{conc}) dT + M_{in} \int_{T_r}^{T_{amb}} C_p(\text{air}) dT$$

where

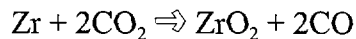
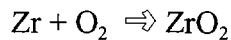
- M_{in} = molar rate of air flow into the cavity, moles/s,
- $C_p(\text{conc})$ = heat capacity of the gaseous products of concrete decomposition, J/mole-K,
- $C_p(\text{air})$ = heat capacity of air, J/mole-K,
- T_r = reference temperature, 298.15 K.

As additional simplifications, the gaseous products of concrete decomposition are taken to be just carbon dioxide and the heat capacity of air is taken to be that of nitrogen [4]:

$$C_p(\text{conc}) = 45.367 + 8.686(T/1000) - 961900/T^2 \quad \text{J/mole-K}$$

$$C_p(\text{air}) = 27.267 + 4.929(T/1000) - 33054/T^2 \quad \text{J/mole-K}$$

Gases in the reactor vessel are taken to react with the zircaloy cladding on the fuel according to the reactions:



The first of these reactions removes gas from the atmosphere thus creating additional driving force for flow into the vessel. The rate of gas removal is taken to be M_{rxn} moles/s which is found by assuming the oxidations go to completion rapidly. The second reaction is isomolar, but does result in a reduction in the gas density which also augments the driving force for gas flow into the vessel. The exothermic nature of the reactions is taken to produce a gas temperature within the vessel of T_{ves} . The heated gases flow out of the vessel and into the piping of the reactor coolant

system. The gases lose heat to the piping so that they are at the temperature T_{pipe} at the point that they emerge from the reactor coolant system.

The mass balance equations for the two-volume model are:

$$M_{\text{out}} + M_{\text{ves}} = Q + M_{\text{in}}$$

$$M_{\text{ves}} = M_{\text{rxn}} + M_{\text{res}}$$

$$M_{\text{rxn}} = 0.2 x_{\text{air}} M_{\text{ves}}$$

A complete model of the flow into the reactor vessel would consider pressure drops at the vessel penetration, pressure drops for flow through the damaged core as well as by-pass flow, kinetics of gas reaction with the cladding and the fuel, and pressure drops associated with flow into the piping system. Such a more detailed model has been used to examine the flows for a specific reactor. A detailed model of general applicability would require quite a few assumptions about geometry and core damage. A much simpler model is used for the illustrative purposes of this paper. This model assumes flows are driven by buoyancy which probably leads to lower bounds on the actual flows.

The Bernoulli equations for the model describe flow into the reactor cavity and into the reactor vessel:

$$P_{\text{amb}} - P = 0.5 \rho_{\text{amb}} K s^2 / 1.01325 \times 10^6, \quad \text{atms}$$

$$P - P_{\text{ves}} = 0.5 \rho K_{\text{ves}} v^2 / 1.01325 \times 10^6, \quad \text{atms,}$$

$$s = \frac{4 M_{\text{in}} R T_{\text{amb}}}{\pi D_{\text{in}}^2 P_{\text{amb}}}, \quad \text{cm/s}$$

$$v = \frac{4 M_{\text{ves}} R T}{\pi D_{\text{ves}}^2 P}, \quad \text{cm/s}$$

where R is the Universal gas constant = 82.06 cm³-atm/mole-K.

Flows of gases from the cavity into the containment and out of the reactor coolant system are driven by buoyancy forces and retarded by drag. Equating these forces yields:

$$M_{\text{out}} = \pi D_{\text{out}}^2 \frac{P}{RT} \sqrt{\frac{D_{\text{out}} (\rho_{\text{amb}} - \rho) g}{8 \rho f}}$$

$$M_{\text{rcs}} = \pi D_{\text{rcs}}^2 \frac{P_{\text{ves}}}{RT_{\text{pipe}}} \sqrt{\frac{D_{\text{rcs}} (\rho_{\text{amb}} - \rho_{\text{rcs}}) g}{8 \rho_{\text{rcs}} f_{\text{rcs}}}}$$

$$\rho_{\text{rcs}} = \frac{MW' P_{\text{rcs}}}{RT_{\text{pipe}}}$$

$$MW' = \frac{28.00055(1 - x_{\text{air}}) + 28.0134x_{\text{air}}0.8}{(1 - 0.2x_{\text{air}})}$$

where g is the gravitational acceleration = 980 cm/s².

Parametric values used for the example calculation with the two-volume model are shown in Table 1. Results of the calculation are shown in Figures 3 to 7 as functions of the assumed rate of concrete erosion. The detailed results are indeed sensitive to the assumed geometry and to the assumed conditions. In fact, the model is probably too crude to ascribe great significance to the quantitative results. The qualitative results, which are consistently obtained for wide variations in the parametric quantities, are thought to be more significant.

The air flow into the reactor cavity is shown in Figure 3. There is, indeed, a rate of concrete erosion that is sufficient to produce gas at a rate that pressurizes the cavity so there is no air flow into the cavity. For the conditions chosen for the example calculation, this limiting concrete erosion rate is about 30 cm/hr. Such a high concrete erosion rate will occur when high temperature core debris first contacts concrete, especially if metallic zirconium is present in the core debris. Such high concrete erosion rates are difficult to sustain once metallic zirconium has been consumed by oxidation and the core debris has been diluted by the condensed products of concrete decomposition.

With decreasing gas generation within the cavity, more air is drawn into the cavity until a maximum in the air flow rate is reached. For the example calculation, this maximum occurs at an erosion rate of about 3 cm/hr. Further reductions in the concrete erosion rate, leads to lower flows into the cavity. Were the vessel volume not included in the model, air flow into the cavity would go to zero as the concrete erosion rate went to zero. With the vessel model included, air flow into the cavity remains substantial even if the gas production within the cavity is zero.

The temperature of the cavity atmosphere is shown as a function of concrete erosion rate

in Figure 4. The variation of the cavity atmosphere temperature with concrete erosion rate is not especially sensitive to the selected value of T^* . Rapid cooling of the atmosphere by the air drawn into the cavity is consistent with observations during tests of melt-concrete interactions which showed there to be a significant temperature gradient above the melts.

An important result is the predicted composition of the cavity atmosphere shown in Figure 5. The mole fraction of air in the atmosphere rises rapidly with decreasing concrete erosion rate. That is, over most of the range of erosion rates examined here there is a substantial mole fraction of air in the cavity available to go into the reactor vessel.

The flow of gas into the reactor vessel from the cavity is shown in Figure 6. The chemical energy imparted to the residual fuel in the vessel by reaction with the gas is shown in Figure 7. This energy from chemical reaction will, of course, be concentrated in a reaction zone rather than being distributed over the entire inventory of residual fuel as is the case with decay power. This leads to one of the substantial questions associated with air ingress into the reactor vessel. How will the highly energetic chemical reactions of the gases affect degradation of the residual fuel? In the absence of experimental data, it really is not possible to predict how the mixture of air and other oxidants will affect the degradation. Two limiting cases can be defined:

- high temperatures produced by the reaction of clad with air will cause the clad to melt and flow away from the fuel, leaving fuel exposed to the oxidants, and
- high temperatures produced by the reaction of clad with air will cause the cladding to melt and dissolve fuel so that fuel and clad relocates from the core as a mixture.

In the first of these limiting cases, fuel is exposed to oxidant and the consequences of this are discussed briefly in the next section of this paper and in more detail in a subsequent presentation by Kupferschmidt and Dickson. In the second case, residual metallic zirconium will “getter” oxidants and protect the fuel from reaction throughout the relocation. The effects of air and other oxidants on fission product behavior in the reactor coolant system are still considerable and are discussed in the final section of this paper.

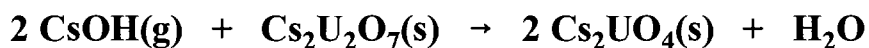
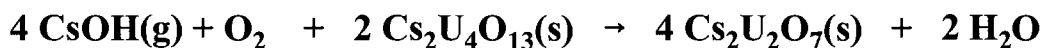
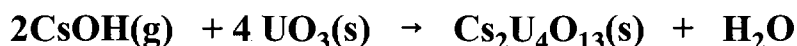
EFFECTS OF AIR ON RELEASE AND AEROSOL GENERATION

Air ingress greatly raises the oxygen potential of the vessel atmosphere [5]. When fuel is exposed to this higher oxygen potential the vaporization of both the fuel and the fission products it contains is greatly increased. Experimental studies to be discussed in a subsequent paper will describe the tremendous effect of air on the vaporization of ruthenium and molybdenum from the fuel. In the atmosphere of steam and hydrogen produced during degradation of fuel prior to vessel rupture, minimal releases of ruthenium are expected. Greatly increase releases of the radiologically significant fission product in air would alter substantially the assumed radionuclide releases during a reactor accident [6]. Accentuation of the releases of tellurium, too, might be expected once air intrusion occurs.

A more subtle effect is the substantial releases of hexavalent uranium largely in the form of $\text{UO}_3(\text{g})$. Once this vapor emerges from the core and cools, it will condense to form aerosol. This additional aerosol will affect the transport of radioactive aerosols out of the reactor coolant system and into the containment. The effects will be to greatly increase the agglomeration of radioactive aerosols, perhaps, to sizes that more readily deposit in the reactor coolant system. The hexavalent uranium will also affect the chemistry of the fission products deposited in the reactor coolant system and these effects are discussed in the next section of this paper.

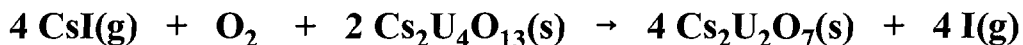
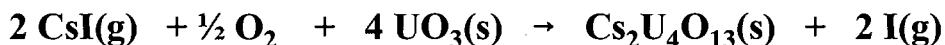
EFFECTS ON FISSION PRODUCT CHEMISTRY

The models of fission product transport in the reactor coolant system and the results of the PHEBUS-FP tests have established that significant fractions of the volatile fission products (primarily Cs and I) released from degrading reactor fuel under accident conditions can deposit in the reactor coolant system. These deposited radionuclides will continue to heat as a result of radioactive decay and can reach temperatures sufficient that they begin to revaporize. This creates a late release of radioactivity from the reactor coolant system to the reactor containment at a time when it is more likely that the reactor containment has been breached. The extent to which deposited fission products can revaporize does depend critically on the chemical forms they adopt on surfaces in the reactor coolant system. The availability of substantial amounts of hexavalent uranium could alter the chemical forms of deposited fission products. Consider the formation of cesium uranates:



A map of the partial pressure of $\text{CsOH}(\text{g})$ in equilibrium with these uranate phases as a function of temperature is shown in Figure 8. It is apparent from these results that the higher uranates especially can keep the partial pressure of cesium low and inhibit revaporization of cesium from the reactor coolant system. It would be expected, then, that abundant hexavalent uranium available in the reactor coolant system following air intrusion would suppress cesium revaporization in the later stages of an accident.

Another set of reactions of interest involve the chemical form of iodine. Some pertinent reactions are:





A plot of the ratio of atomic iodine partial pressure to the partial pressure of CsI is shown in Figure 9. Ratios shown in this figure indicate that the higher cesium uranates are better able to react with CsI to liberate iodine in a form that will remain gaseous if it reaches the containment. Whether abundant hexavalent urania leads to greater releases of gaseous iodine to the containment or not depends, of course, on the opportunities the iodine species have to react with other things along the flow path to the containment.

CONCLUSION

This paper has shown that it is possible to get significant air intrusion into a ruptured reactor vessel even from a reactor cavity with restricted access. This suggests that there is some importance to considering the consequences of air intrusion following vessel penetration by core debris. The consequences will depend on the nature of core degradation in air and other oxidizing gases. If, indeed, fuel becomes exposed to strongly oxidizing gases, significant releases of ruthenium and hexavalent urania can be expected. Hexavalent urania could alter the nature of cesium release and cesium revaporization from the reactor coolant system. Hexavalent urania could destabilize CsI and enhance the formation of gaseous iodine unless there are other materials that will react readily with atomic iodine along the flow path to the reactor containment.

Table 1. Parametric Values Used in the Example Calculation

Parameter	Value
D_{in}	30 cm
D_{out}	100 cm
D_{ves}	50 cm
D_{rcs}	35 cm
T_{amb}	300 K
T^*	1500 K
T_{pipe}	600 K
T_{ves}	2000 K
P_{amb}	1 atms
K	0.5
K_{ves}	0.5
f	0.2
f_{rcs}	0.02

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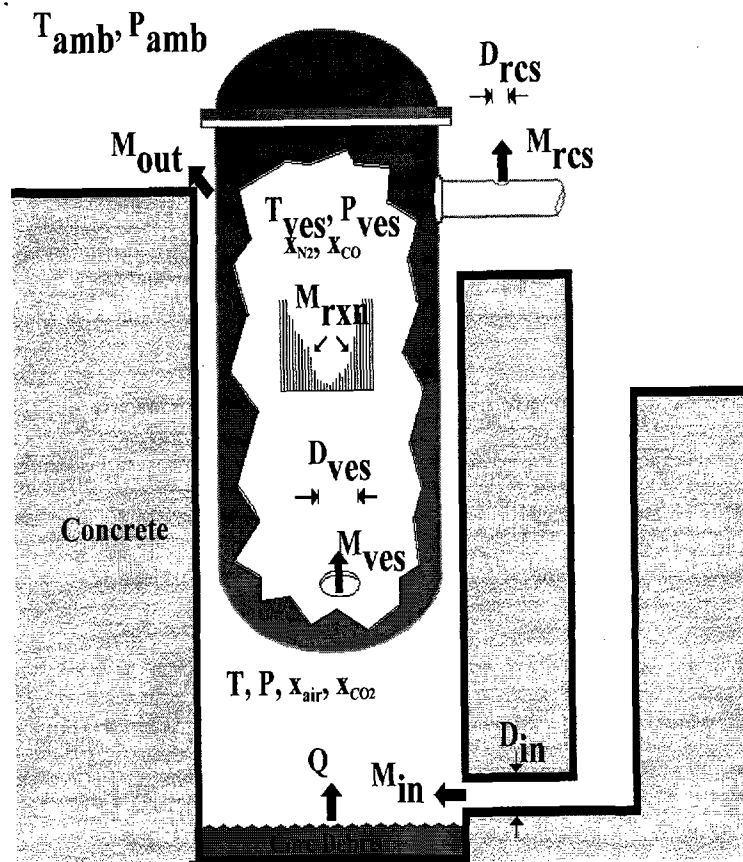


Figure 1. Schematic depiction of the two-volume model.

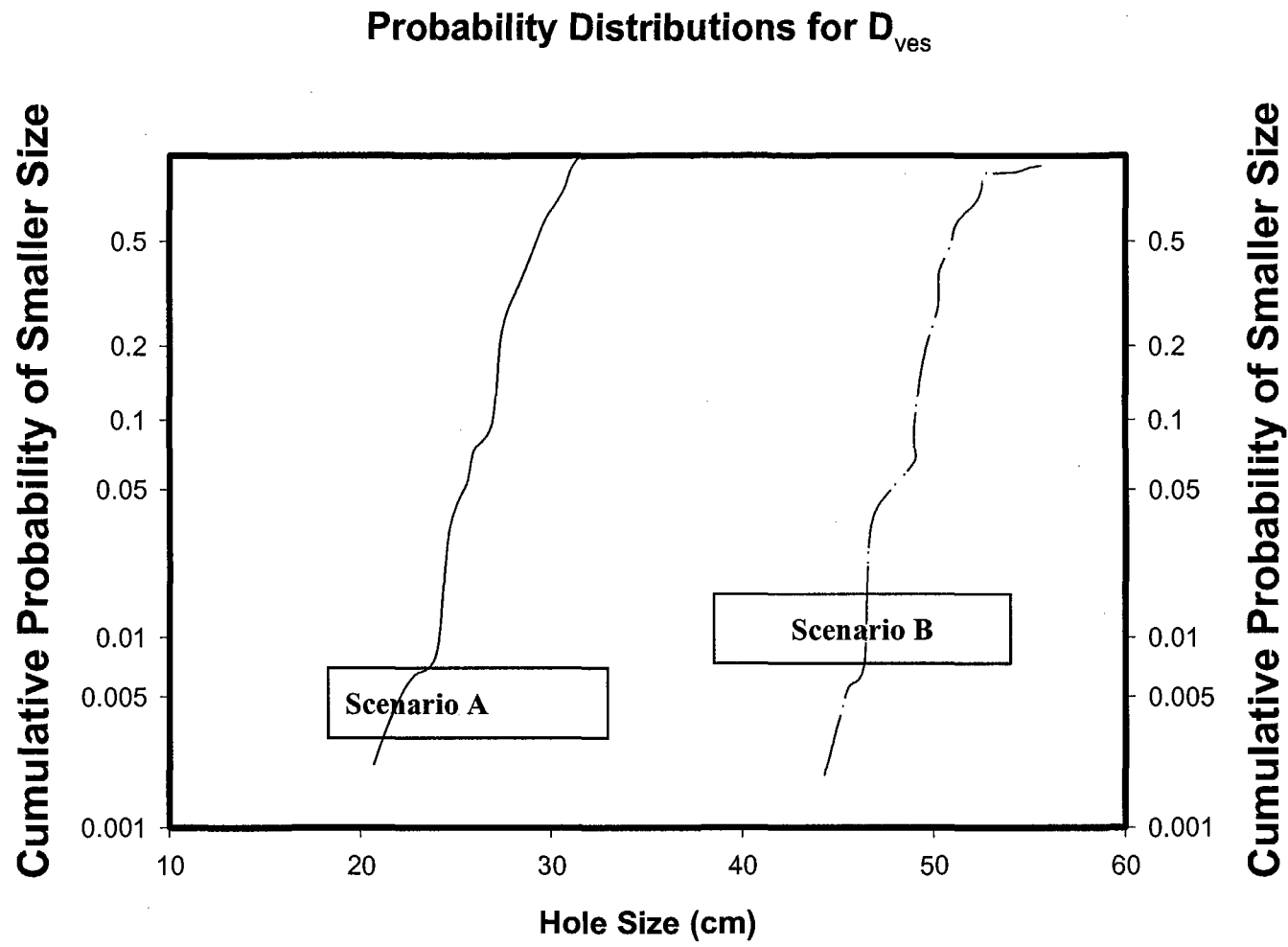


Figure 2. Cumulative probability distributions for the vessel hole size [2].

Example Calculation

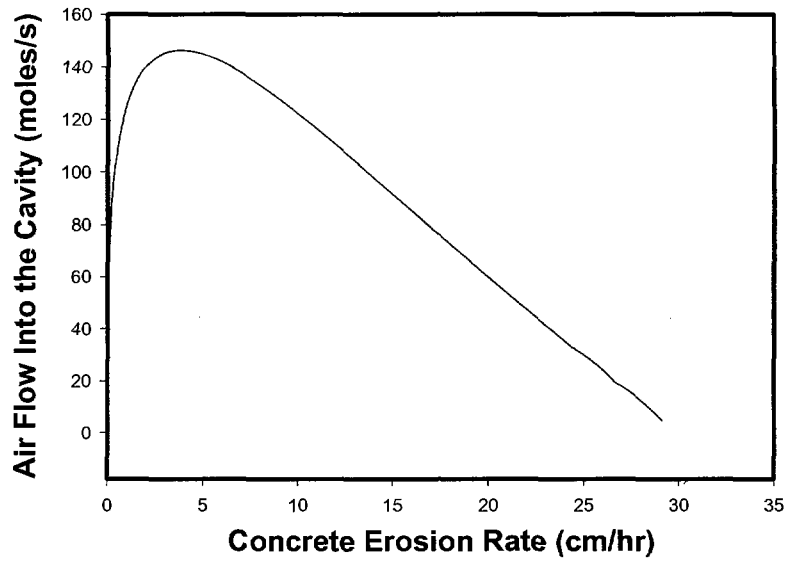


Figure 3. Air flow into the reactor cavity as a function of the effective concrete erosion rate.

Example Calculation

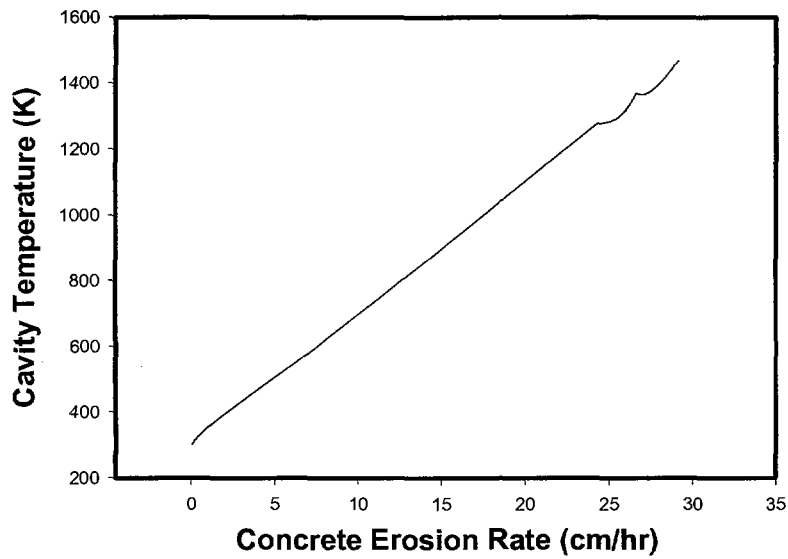


Figure 4. Cavity atmosphere temperature as a function of concrete erosion rate.

Example Calculation

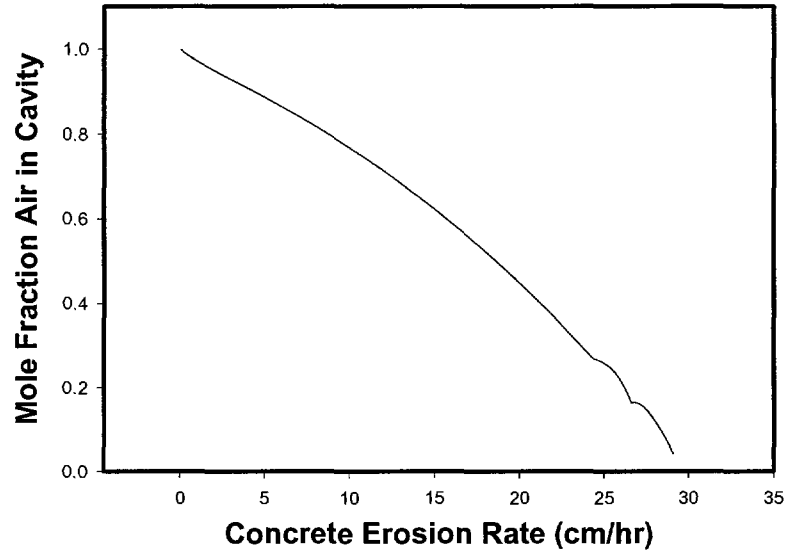


Figure 5. Composition of the reactor cavity atmosphere as a function of the effective concrete erosion rate.

Example Calculation

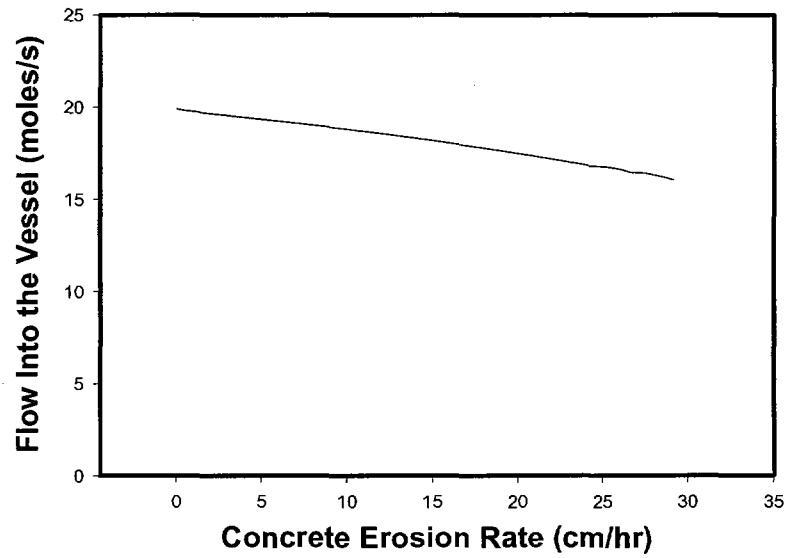


Figure 6. Gas flow into the reactor vessel as a function of the effective concrete erosion rate.

Example Calculation

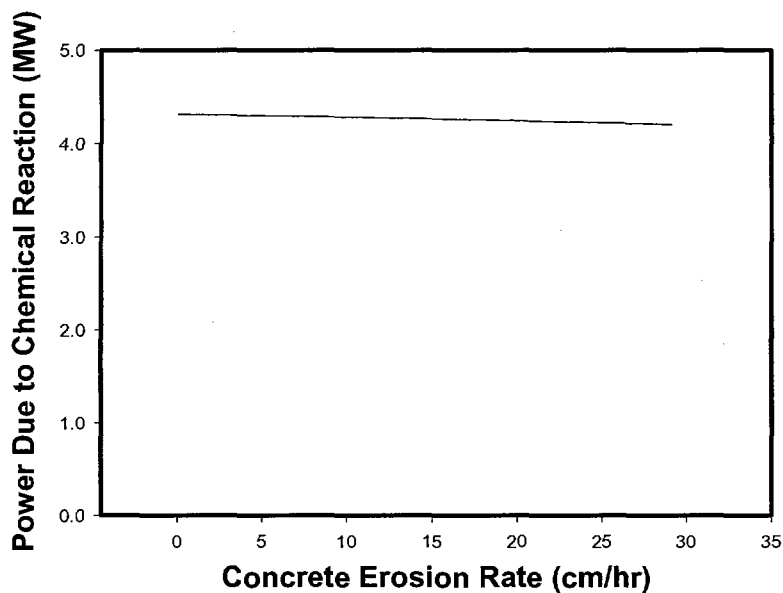


Figure 7. Power deposited in residual fuel due to chemical reactions of gas with the cladding.

Cesium Holdup by Hexavalent Urania

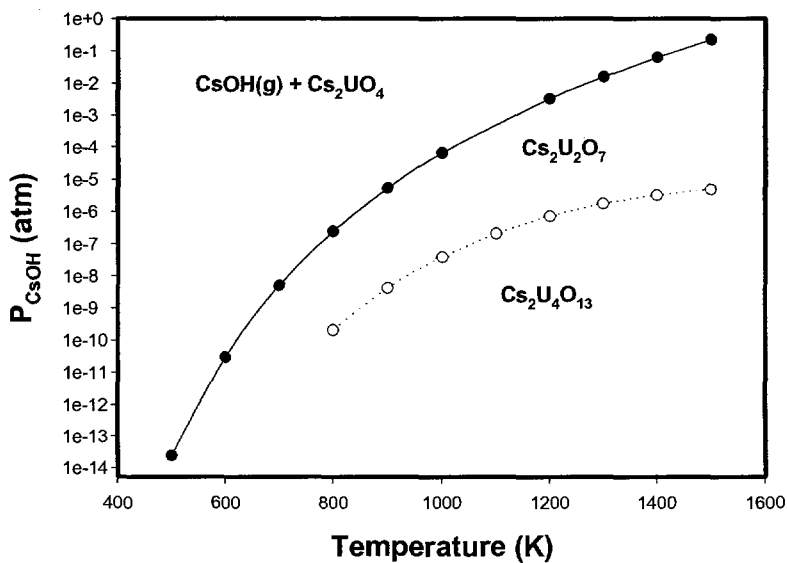


Figure 8. Effects of various cesium uranates on the equilibrium CsOH partial pressure in the reactor coolant system as a function of temperature.

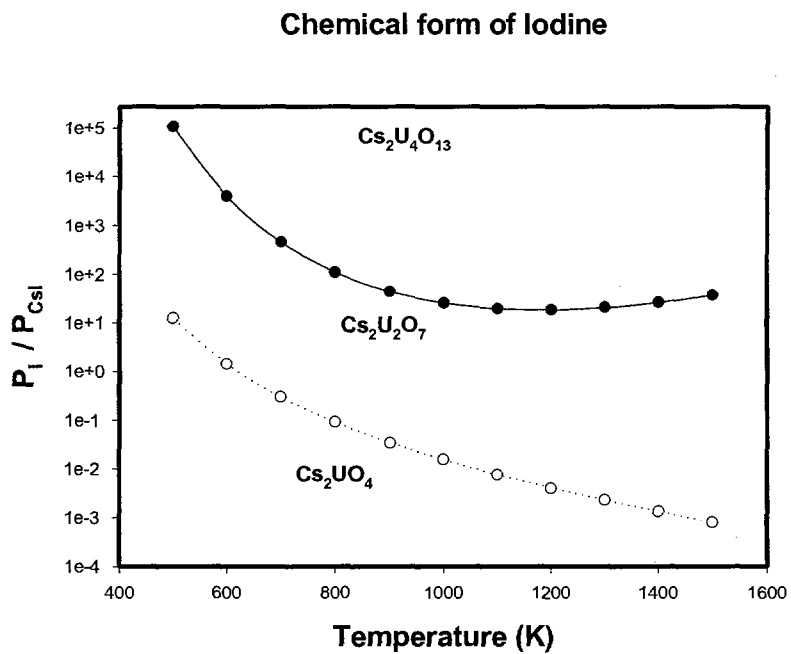


Figure 9. Effects of various cesium uranates on the ratio of the partial pressures of I(g) and CsI(g). The partial pressure of air was taken to be 0.2 atm.