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R. A. Lorenz  
Chemical Technology Division  
Oak Ridge National Laboratory  
Oak Ridge, Tennessee 37830

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# THE VAPORIZATION OF STRUCTURAL MATERIALS IN SEVERE ACCIDENTS

R. A. Lorenz  
Chemical Technology Division  
Oak Ridge National Laboratory  
Oak Ridge, Tennessee 37830

## ABSTRACT

Vaporized structural materials form the bulk of aerosol particles that can transport fission products in severe LWR accidents. As part of the Severe Accident Sequence Analysis (SASA) program at Oak Ridge National Laboratory, we have developed a model based on a mass transport coefficient to describe the transport of materials from the surface of a molten pool. In many accident scenarios, the coefficient can be calculated from existing correlations for mass transfer by natural convection. Data from SASCHA fuel melting tests (Karlsruhe, Germany) show that the partial pressures of many of the melt components (Fe, Cr, Co, Mn, Sn) required for the model can be calculated from the vapor pressures of the pure species and Raoult's law. These calculations indicate much lower aerosol concentrations than reported in previous studies.

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SUMMARY

Vaporized structural materials form the bulk of aerosol particles that can transport fission products in severe light water reactor (LWR) accidents. As part of the Severe Accident Sequence Analysis (SASA) program at Oak Ridge National Laboratory,<sup>1,2</sup> the vapor pressures of components of molten mixed structural and core materials were calculated from published experimental data.<sup>3,4</sup> These vapor pressures were then used to calculate the rate at which melted material vaporized in two geometries: exposed molten surfaces with gas (steam) flow past the surfaces, and a molten pool with no forced gas flow. This analysis was undertaken because it was believed that the direct extrapolation of fractional release rates obtained in small experiments to full-size reactors would result in overly conservative (high) releases.

The vapor pressures of the components of the structural material were obtained from the analysis of SASCHA fuel melting tests performed at Karlsruhe, Germany. The analysis was concentrated on data obtained at 2400°C, a temperature at which the mixtures of UO<sub>2</sub>, Zircaloy, and stainless steel were fully molten. This is also the maximum temperature expected before the melted materials would flow to the bottom of a reactor vessel. We assumed that the controlling mass transfer mechanism was gas-phase convection, probably natural convection, both for the SASCHA tests and for large core melts settled into a pool configuration. The rate of mass transport is determined from:

$$q = k_G L p \approx k_{GP} , \quad (1)$$

where

$q$  = the mass transport flux, mol/s·cm<sup>2</sup> surface;

$k_G$  = mass transfer coefficient, mol/s·cm<sup>2</sup>·atm;

$\Delta p$  = difference in pressure of transported species from pool surface  
to bulk gas, atm;

$p$  = partial pressure at the pool surface, atm.

For the case of vaporization from a very hot surface, the partial pressure in the gas leaving the system is very much lower than that at the pool surface; therefore, we will assume that  $\Delta p$  is the same as  $p$ , the partial pressure at the pool surface.

It was not possible to evaluate both  $k_G$  and  $p$  from SASCHA tests alone. We found that by assuming a mass transfer coefficient,  $k_G$ , of  $9 \times 10^{-6}$  mol/s·cm<sup>2</sup>·atm, we could obtain vapor pressures for structural materials released in SASCHA that were in reasonable agreement with literature values. Equation (1) was used to calculate the vapor pressures after evaluating the mass transport flux,  $q$ , for each component from

$$q = \frac{Nk_r}{A}, \quad (2)$$

where

$N$  = amount of the component in the melt, mol;

$k_r$  = release rate coefficient, fraction/s;

$A$  = pool surface area = 19.6 cm<sup>2</sup> for SASCHA.

The results of these calculations are shown in Table 1. Data for silver, cadmium, and indium were not included since these elements are not present in significant amounts in the Browns Ferry reactors under study. The fractional release rates chosen were based on SASCHA tests conducted in

both air and steam at 2400 and 2800°C. The  $\text{UO}_2$  release rate is lower than that found in recent SASCHA tests; our choice was based partly on relative release rates found in other fuel melting tests.<sup>5-8</sup>

If the mass transfer model is valid, the partial pressures of the individual species calculated above should be in reasonable agreement with pressures calculated from published vapor pressures of pure materials. Some uncertainty is involved in identifying the chemical form of each material at the melt surface. In our calculations, we assumed that each component (except  $\text{UO}_2$ ) existed in the elemental form since only partial oxidation of the SASCHA melts occurred. We also assumed that Raoult's law could be used to calculate the partial pressure above the melt. The results of these calculations, which are summarized in Table 2, are in generally good agreement with experimental values determined from the mass transfer coefficient  $k_G = 9 \times 10^{-6} \text{ mol/s}\cdot\text{cm}^2\cdot\text{atm}$ . However, the values for zirconium and tin show significant differences.

A calculation was made to determine whether natural convection might be the controlling mass transfer mechanism in SASCHA tests. From a correlation for heat transfer by natural convection above a heated flat surface under laminar flow conditions<sup>9</sup> and the heat transfer-mass transfer analogy,<sup>10</sup> we calculated a  $k_G$  value of  $6 \times 10^{-6} \text{ mol/s}\cdot\text{cm}^2\cdot\text{atm}$  for SASCHA tests in 2-bar air atmosphere at 2400°C. This is better agreement with the assumed value of  $k_G$  than would be expected, considering that SASCHA is not an ideal geometry for calculating natural convection, and that the correlation has not been verified as applicable to the small-size, high-temperature conditions of SASCHA.

Our analysis of the corium mixture, Table 1, indicates a total vapor pressure for this material of  $1.5 \times 10^4$  Pa (0.15 atm) at 2400°C. This is equivalent to a mass concentration of 39 g/m<sup>3</sup> at 2400°C, well below aerosol concentration levels predicted or assumed in other studies. For those PWRs that use a control rod fabricated of Ag-In-Cd alloy, the vapor pressures of these materials can be expected to more than double the total concentration of vaporized material shown above.<sup>4</sup>

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Table 1. Release of structural materials in typical SASCHA test at 2400°C

Material	Mass in SASCHA (g)	Release rate			Aerosol composition (mass %)	Effective vapor pressure <sup>a</sup> (atm)
		(fraction/min)	(g/min)	(mol/min)		
Fe	54.4	$6.2 \times 10^{-4}$	0.0336	$6.05 \times 10^{-4}$	36.80	0.0569
Mn	1.6	$2.1 \times 10^{-2}$	0.0336	$6.11 \times 10^{-4}$	36.80	0.0576
Cr	15.4	$1.0 \times 10^{-3}$	0.0150	$2.95 \times 10^{-4}$	16.43	0.0279
Sn	0.45	$1.1 \times 10^{-2}$	0.0050	$4.20 \times 10^{-5}$	5.48	0.0039
Ni	7.4	$3.0 \times 10^{-4}$ <sup>b</sup>	0.0022	$3.78 \times 10^{-5}$	2.41	0.0036
UO <sub>2</sub>	90.0	$1.5 \times 10^{-5}$	0.0014	$5.00 \times 10^{-6}$	1.53	$4.0 \times 10^{-4}$
Zr	29.55	$1.3 \times 10^{-5}$	$3.8 \times 10^{-4}$	$4.22 \times 10^{-6}$	0.42	$4.0 \times 10^{-4}$
Co	0.6	$4.0 \times 10^{-4}$	$1.3 \times 10^{-4}$	$4.35 \times 10^{-6}$	0.14	$3.8 \times 10^{-4}$
Si	0.6					
Total	200.0		0.0913	$1.60 \times 10^{-3}$ <sup>c</sup>	100.0	0.151

<sup>a</sup>Calculated assuming  $k_G = 9 \times 10^{-6}$  mol/s·cm<sup>2</sup>·atm.

<sup>b</sup>This release rate was assumed since experimental results were not available.

<sup>c</sup>Average molecular weight =  $0.0913/1.60 \times 10^{-3} = 57.1$ .

Table 2. Comparison of calculated and experimental partial pressures in SASCHA at 2400°C

Material	Amount in SASCHA		Vapor pressure of element (atm)	Partial pressure by Raoult's law <sup>a</sup> (atm)	Experimental P if $k_G = 9 \times 10^{-6} b$ mol/s·cm <sup>2</sup> ·atm (atm)
	Mol	Mol fraction in liquid			
Fe	0.974	0.460	0.096	0.044	0.0569
Mn	0.0291	0.0137	5.0	0.0685	0.0576
Cr	0.296	0.140	0.24	0.0336	0.0279
Sn	0.00379	0.00178	0.39	0.0007	0.0039
UO <sub>2</sub>	0.333	0.157	0.00117 <sup>c</sup>	$1.84 \times 10^{-4}$	$4.7 \times 10^{-4}$
Ni	0.126	0.0595	0.066	0.0039	0.0036
Zr	0.324	0.153	$1.5 \times 10^{-5}$	$2.3 \times 10^{-6}$	$4.0 \times 10^{-4}$
Co	0.0102	0.00482	0.06	$2.9 \times 10^{-4}$	$3.8 \times 10^{-4}$
Si	0.0214	0.0101			
Total	2.117			0.151	0.151

<sup>a</sup>Partial pressure = vapor pressure of pure substance × mol fraction in liquid.

$$b_P = 94.2 \frac{R \text{ mass}}{MW}$$

<sup>c</sup>Vapor pressure of pure UO<sub>2</sub>.