
Submission for the CSNI/GREST Benchmark Exercise on Chemical Thermodynamic Modeling in Core-Concrete Interaction Releases of Radionuclides

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ABSTRACT

A submission for the CSNI/PWG-4/GREST standard problem on chemical thermodynamic modeling in core-concrete interaction releases of radionuclides is described. Part A of the exercise is a highly defined benchmark calculation in which data and speciation are specified. The problem is, however, ambiguous concerning the definition of an ideal solution. Consequently, two solutions are provided. In one solution, specified species are treated as molecular entities to define the ideal solution. In the second, mixing is assumed to occur ideally on cationic and anionic lattices. The different results obtained in these calculations illustrate the importance of condensed phase modeling in the analyses of high temperature melt interactions with concrete.

Part B of the exercise consists of six problems in which the temperatures, pressures and bulk compositions of the melts are specified. Data and speciation are to be supplied as parts of the solutions to the problems. Results of calculations for these six problems are presented. Additional solutions are provided to illustrate the effects of speciation in the condensed oxide phase, non-ideality in the condensed metal phase and uncertainty in the thermodynamic properties of gas phase species.

It is demonstrated by the calculations that a variety of challenges arise in the accurate modeling of thermochemistry of high temperature melts. A systematic exploration of approximations is clearly needed. Experimental data on the thermochemistry of such high temperature melts will not be useful if there is not an adequate theoretical framework to interpolate and extrapolate the data.

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1. INTRODUCTION

The release of radionuclides during core debris interactions with concrete in a severe nuclear reactor accident has been recognized since the early 1970's [1]. The relative importance of the melt/concrete release process has increased in reactor accident analyses as new methods of source term analysis have been introduced [2,3] and more mechanistic models of the melt/concrete interaction process have been devised [4]. The newer models and experiments [5,6,7,8] have shown that during melt interactions with concrete, tremendous quantities of aerosol material are produced which can drastically affect the longevity of radioactive material suspended in the reactor containment. Radionuclide releases predicted with the modern models are highly dependent on the peculiarities of the accidents and power plants in question [9]. In some situations the releases are less than those predicted in the older models such as that developed for the Reactor Safety Study [1]. In other cases, however, significantly higher releases of particularly the more refractory alkaline earths and the rare earths are predicted.

The chemical system created by the interaction of core debris with concrete is, obviously, quite complex. Any model of the release of radionuclides and generation of aerosols during core debris interactions with concrete will, necessarily, be approximate. Data for evaluating the accuracy of modern models of radionuclide release and aerosol generation are scarce. Where comparisons have been possible, the predictions of the VANESA model [4] of aerosol generation during core debris interactions with concrete have agreed with experiment results to within about a factor of 2 to 3. Available data do not, however, extend into regimes where the highest releases of radionuclides are predicted to occur. Until more data are available, it is useful to examine the assumptions and approximations made in the model that lead to predictions of high (and low) generation rates of radioactive and non-radioactive aerosols.

A key element of models of aerosol generation during core debris interactions with concrete is the treatment of thermochemistry. It is, in fact, the analyses of melt thermochemistry that distinguishes modern models from those used in the past and even release models developed for other accident phenomena. The thermochemistry defines the driving force for vaporization of volatile constituents while core debris interacts with concrete. It also defines the maximum extent of vapor formation and consequently aerosol production. Approximations made in the description of the thermochemistry of the complex melts have a direct influence on the quality of predictions of vaporization during melt interactions with concrete.

The Group of Experts on the Source Term (GREST) of the Committee for the Safety of Nuclear Installations (CSNI) has recently turned its attentions to the issues of core debris interactions with concrete and the attendant release of radionuclides. Dr. P. Clough, chairman of GREST, has defined a set of "benchmark problems" to assess the current capabilities for predicting the thermochemistry of melts interacting with concrete. That is, the benchmark problems do not test the adequacy of models used to predict radionuclide release during core debris interactions with concrete. They are, instead, tests of one part, albeit an important part, of the analyses necessary to predict these releases. The exercise is then of value for the modeling of other chemical processes in reactor accidents as well as for the modeling of core debris interactions with concrete.

The benchmark exercise is divided into two types of standard problems to be confronted by the analysts. Part A of the exercise consists of a highly specified standard problem in which the temperature, pressure, bulk composition, and speciation of the melt are defined along with thermochemical data for the melt and gas phase species. The standard problem in part A is "...intended to test the numerical solution techniques involved in defining chemical equilibrium...". Part B of the benchmark exercise consists of a set of six standard problems in which only the bulk compositions, temperatures and pressures of the melts are defined. Speciation of the gas phase and the melt phases is deliberately left to the discretion of the analyst as are the thermochemical data for the species. The purpose of the part B standard problems is to illustrate the variability of results obtained when various treatments are employed to determine the equilibrium chemistry.

The balance of this document consists of a submission for the GREST benchmark exercise for chemical thermodynamic modeling. The submission was prepared at the request of J. Mitchell of the U. S. Nuclear Regulatory Commission as part of the work on modeling core debris-concrete interactions sponsored at Sandia National Laboratories. Solutions to both the part A and part B problems are discussed below. The document concludes with some additional calculations to illustrate the sensitivities of the equilibrium analyses to factors not considered in the benchmark exercise.

2. MODEL USED IN THE EXERCISE

The benchmark exercise does not test computer codes used in accident analyses for predicting the release of radionuclides during core debris interactions with concrete. It tests only the treatment of chemical equilibrium that is a part of such computer models. Computer codes that model the

the dynamic release process are optimized to treat release as it is hypothesized to occur in reactor accidents. Such codes are ill-suited to treat the generalized, static problems defined in the benchmark exercise. No attempt was made then to use one of these accident analysis models for the benchmark exercise. Instead, the equilibrium solver in the VANESA model [4] was extracted and modified to serve as a "stand-alone" chemical equilibrium computer code.

Modern chemical equilibrium solvers fall typically in one of two classes--those that attempt to find the minimum free-energy and those using the so-called Brinkley equilibrium constant method [10]. Free-energy minimization has achieved a broad popularity for the solution of equilibrium problems. Free-energy minimization suffers, however, from the difficulties of mathematical scaling, finite convergence neighborhoods, and truncation caused by convergence criteria. The technique is then poorly suited for the repetitive solution of essentially similar problems. The equilibrium constant method, while it is entirely equivalent in a mathematical sense to free-energy minimization, is more difficult to implement as a generalized solver capable of treating a wide range of problems.

The equilibrium solver in the VANESA model uses a slightly modified equilibrium constant method. The equilibrium constant method converts the problem into a set of non-linear equations. The equations are solved in the code used here by a cyclic Newton technique. Convergence is declared when the mass balance constraints are achieved to 1 part in 10^{10} . This convergence criterion was dictated by the internal precision of the computing machine. More demanding convergence criteria could be imposed in other systems.

The adequacy of the "stand-alone" solver was tested by comparison of calculated results to published results for the so-called "blast furnace problem" [11]. This comparison is shown in Table 1.

3. BENCHMARK PROBLEM A

3.1 Definition of the Problem

The specifications of the problem in part A of the benchmark exercise are shown in Table 2. The problem is highly constrained except where it specifies that the condensed phases are to be treated as ideal solutions. There is no difficulty with the treatment of the metal phase. (It is readily apparent, however, that in reality the prescribed

TABLE 1 Calculated Results for the "Blast
Furnace Problem"

Species	Moles	
	lit. [11]	This work
Fe(solid)	42.83	42.8270
CaO(solid)	0.756	0.75620
CO(gas)	81.62	81.624
CO ₂ (gas)	6.66	6.6637
H ₂ (gas)	6.43	6.4262
H ₂ O(gas)	0.444	0.4436
CH ₄ (gas)	6.55 x 10 ⁻³	6.550 x 10 ⁻³
CH ₂ O(gas)	2.36 x 10 ⁻⁶	2.357 x 10 ⁻⁶
CHO(gas)	1.05 x 10 ⁻⁶	1.052 x 10 ⁻⁶
OH(gas)	7.93 x 10 ⁻¹¹	7.923 x 10 ⁻¹¹
O ₂ (gas)	1.73 x 10 ⁻¹⁹	1.729 x 10 ⁻¹⁹

TABLE 2. Specifications for Benchmark Problem A

Input

- (1) Specification of the system. The system consists of a gas phase, a liquid oxide solution phase, and a liquid metal solution phase, all in equilibrium. The components in each phase are as follows:

Gas phase: O_2 , UO_2 , UO_3 , UOH , H_2 , H_2O , Zr , Cr , La ,
 LaO , K , KOH

Oxide solution: UO_2 , $U_2O_{4.5}$, ZrO_2 , Cr_2O_3 , La_2O_3 , K_2O

Metal solution: U , Zr , Cr , La , K

- (2) Thermodynamic data. Expressions for the Gibbs energy of formation are in the form:

$$\Delta G_f^0 = A + B \cdot T \quad \text{J-mol}^{-1}$$

where T is in Kelvin. The Gibbs energies of formation of all elements in their normal physical form at 298.15 K are taken to be zero. Author's note: This reference state applies to all elements except potassium. Liquid potassium is the reference state for the calculation.)

The values for the coefficients A and B for the species are:

Species	A	B
O_2 (g)	0.0	0.0
UO_2 (g)	-5.083560E+5	2.280E+1
UO_3 (g)	-8.368000E+5	8.117E+1
UOH (g)	2.493900E+4	-2.619E+0
H_2 (g)	0.0	0.0
H_2O (g)	-2.464376E+3	5.481E+1
Zr (g)	6.095980E+5	-1.320E+2
Cr (g)	3.794340E+5	-1.318E+2
La (g)	4.208690E+5	-1.142E+2
LaO (g)	-1.470880E+5	-5.272E+1
K (g)	6.886400E+4	-6.976E+1
KOH (g)	-2.402840E+5	1.074E+1

TABLE 2 (continued)

Species	A	B
UO ₂ (o)	-1.080000E+6	1.690E+2
U ₂ O _{4.5} (o)	-2.250000E+6	3.914E+2
ZrO ₂ (o)	-1.098000E+6	1.940E+2
Cr ₂ O ₃ (o)	-1.120776E+6	2.534E+2
La ₂ O ₃ (o)	-1.790049E+6	2.820E+2
K ₂ O (o)	-4.547004E+5	2.254E+2
U (m)	0.0	0.0
Zr (m)	0.0	0.0
Cr (m)	0.0	0.0
La (m)	0.0	0.0
K (m)	0.0	0.0

(3) Input amounts in moles

H ₂ O (g)	5.0E+4
UC ₂ (o)	3.0E+5
La ₂ O ₃ (o)	1.0E+2
K ₂ O (o)	1.5E+4
Zr (m)	1.0E+5
Cr (m)	5.0E+5

(4) Conditions and representation of the melt. The temperature of the system is constant at 2500 K and the gas pressure is 1 atmosphere. The oxide and metal solutions are treated as ideal solutions. The final equilibrium composition that is calculated is such that the gas phase is in equilibrium with the oxide and metal solutions.

Output

The output required is the number of moles of each input vapor species in equilibrium with the melt.

metal solution would not be ideal in its behavior.) It is clear that the architect of the problem intends, as is common in the literature, that the specified metal phase species mix randomly so that the free-energy of mixing consists of only configurational entropy. Ambiguity arises in connection with the treatment of the oxide phase. It may be that the problem architect intends that the specified oxide species be treated as molecular entities and that they mix randomly. But, the specified species are not molecular entities. In the solid-state, they are all extended lattice oxides. An alternate definition of an ideal solution for these oxides, one that is commonly used in the literature, is to hypothesize the existence of two lattices in the liquid phase--a lattice for cations and a lattice for anions. Since the only anion is O^{2-} there is no entropy of mixing on the anionic lattice. An ideal solution is created when the cations mix randomly on the cationic lattice.

Because it is unclear which definition of ideal solution is intended--an *ad hoc* solution treating the species as molecular entities or a two lattice solution model--part A of the benchmark exercise was solved using both models. For the molecular solution model, the free energy of mixing in the oxide phase was taken to be:

$$G_{mix} = -RT \sum_i Y_i \ln(Y_i)$$

where G_{mix} = free-energy of mixing
 R = gas constant
 T = absolute temperature
 Y_i = mole fraction of the i th specified species

For the two-lattice model, the free-energy of mixing was taken to be

$$G_{mix} = -RT \sum_i z_i \ln(z_i)$$

where $z_i = n_i m_i / \sum_j n_j m_j$

n_i = number of cations in the i th species

m_i = number of moles of the i th species in the melt.

This definition of the free-energy of mixing in the two-lattice model amounts to considering the species $U_2O_{4.5}$, La_2O_3 , Cr_2O_3 and K_2O to actually be $UO_{2.25}$, $LaO_{1.5}$, $CrO_{1.5}$, and $KO_{0.5}$.

3.2 Results of the Calculations for Part A

Results of the calculations for part A using the molecular solution model and the two-lattice model are shown in Tables 3 and 4, respectively. The two calculations yield rather similar oxygen partial pressures--about 7×10^{-14} atmospheres or an oxygen potential of about -150 kcal. Contributions of other vapor species to the equilibrium atmosphere above the melt are also quite similar except in the case of lanthanum-bearing species. The partial pressure of La(g) is 7.16×10^{-7} atmospheres in the case of the molecular model and 2.97×10^{-8} atmospheres in the case of the two-lattice model. The partial pressure of LaO(g) is 8.9×10^{-5} atmospheres for the molecular solution model and 3.7×10^{-6} atmospheres in the case of the two lattice model. This order-of-magnitude difference is directly the result of the solution models since the oxygen potentials are nearly the same in the two calculations. The result illustrates that the vapor pressure of lanthanum-bearing species is not dependent on just temperature and oxygen potential. Further discussions of this issue with respect to lanthanum vaporization have been reported recently [17].

The most substantive differences in the results for the condensed phase have to do with species specified to have two cations in the problem definition. More potassium, chromium and higher valent uranium are predicted to be present in the oxide phase with the two-lattice model than with the molecular solution model.

These results point out the importance of solution phase modeling for the prediction of radionuclide release, which has been noted elsewhere [12]. To date, this aspect of the thermochemistry of core debris has received less attention than has speciation of the gas phase. A much smaller data base is available to guide modeling of the liquid phase.

4. BENCHMARK PROBLEM B

4.1 Definitions of the Problems

Part B of the benchmark exercise presents a set of problems whose specifications are defined in Table 5. Six problems are defined involving three composition cases each analyzed at two temperatures. These problems are substantially more complicated and less constrained than that defined in part A. To address these problems, three distinct tasks are required:

1. Select vapor species.
2. Formulate a model of the oxide phase
3. Decide whether non-idealities in the condensed phases are to be recognized.

TABLE 3. Results Benchmark Problem A - Molecular Solution Model

Species	Moles	Mole Fraction	Partial Pressure (atms.)
<u>Gas Phase</u>			
O ₂	5.888 x 10 ⁻⁹		7.11 x 10 ⁻¹⁴
UO ₂	3.383		4.08 x 10 ⁻⁵
UO ₃	5.871 x 10 ⁻³		7.09 x 10 ⁻⁸
UOH	1.510 x 10 ⁻³		1.82 x 10 ⁻⁸
H ₂	49996.9		0.604
H ₂ O	2.576		3.11 x 10 ⁻⁵
Zr	0.0447		5.40 x 10 ⁻⁷
Cr	3067.0		0.0370
La	0.0593		7.16 x 10 ⁻⁷
LaO	7.364		8.89 x 10 ⁻⁵
K	29742.0		0.3591
KOH	1.107		1.34 x 10 ⁻⁵

Condensed Phases

UO ₂ (c)	275520	0.8284	
U ₂ O _{4.5} (c)	14.538	4.371 x 10 ⁻⁵	
ZrO ₂ (c)	56985.2	0.17133	
Cr ₂ O ₃ (c)	1.602 x 10 ⁻⁵	4.216 x 10 ⁻¹¹	
La ₂ O ₃ (c)	68.647	2.064 x 10 ⁻⁴	
K ₂ O (c)	2.371 x 10 ⁻⁹	7.130 x 10 ⁻¹⁵	
U (c)	24447.3	0.21313	
Zr (c)	43014.7	0.3750	
Cr (c)	46933.0	0.4092	
La (c)	55.284	4.820 x 10 ⁻⁴	
K (c)	256.90	2.240 x 10 ⁻³	

TABLE 4. Results Benchmark Problem A - Two Lattice Solution Model

Species	Moles	Mole Fraction	Partial Pressure (atms.)
<u>Gas Phase</u>			
O ₂	5.8433 x 10 ⁻⁹		7.057 x 10 ⁻¹⁴
UO ₂	3.35648		4.053 x 10 ⁻⁵
UO ₃	5.80378 x 10 ⁻³		7.009 x 10 ⁻⁸
UOH	1.50379 x 10 ⁻³		1.816 x 10 ⁻⁸
H ₂	49996.9		0.60379
H ₂ O	2.56622		3.099 x 10 ⁻⁵
Zr	4.48504 x 10 ⁻²		5.4164 x 10 ⁻⁷
Cr	3059.69		3.695 x 10 ⁻²
La	2.4628 x 10 ⁻³		2.9742 x 10 ⁻⁸
LaO	0.304868		3.6817 x 10 ⁻⁶
K	29741.3		0.35917
KOH	1.10283		1.3318 x 10 ⁻⁵

Condensed Phases

UO ₂ (c)	273313	0.82219
UO _{2.25} (c)	2179.18	0.006555
ZrO ₂ (c)	56730.5	0.17066
CrO _{1.5} (c)	2.28882	6.885 x 10 ⁻⁶
LaO _{1.5} (c)	197.389	5.938 x 10 ⁻⁴
KO _{0.5} (c)	0.02802	8.429 x 10 ⁻⁸
U (c)	24504.6	0.21314
Zr (c)	43269.5	0.37635
Cr (c)	46938.0	0.40826
La (c)	2.30327	2.003 x 10 ⁻⁵
K (c)	257.537	2.24 x 10 ⁻³

TABLE 5. Specifications of Benchmark Problem B

This is based on three test cases A, B and C of more realistic composition. Participants should use their own chemical thermodynamic data and choice of species in all phases.

Input

- (1) In each Case, the system consists of a gas phase, a liquid oxide solution phase, and a liquid metal solution phase, all in equilibrium. The initial components in each phase are as follows:

Gas phase: H_2O , CO_2

Oxide solution: UO_2 , SiO_2 , SrO , CaO , La_2O_3 , CeO_2

Metal solution: Zr , Fe , Mo

The methods adopted to define the final equilibrium in terms of permissible species in each phase, activity coefficients, etc. are entirely at the choice of the participant.

- (2) Input amounts. The initial compositions in the three Cases are defined in the following table.

Component	Amount (moles)		
	Case A 'reducing'	Case B 'not as reducing'	Case C 'CaO/SiO ₂ =10'
UO_2	10^5	10^5	10^5
Zr	10^5	10^4	10^4
SiO_2	10^4	10^5	10^4
Fe	10^5	10^5	10^5
SrO	10^2	10^2	10^2
CaO	10^4	10^5	10^5
La_2O_3	10^2	10^2	10^2
CeO_2	10^3	10^3	10^3
Mo	10^2	10^2	10^2
H_2O	10^2	10^2	10^2
CO_2	10^2	10^2	10^2

The objective of part B of the benchmark exercise is to illustrate how results vary with the choice of treatment of the equilibrium analyses. It would be of interest, then, to explore how approximations made in each of these three tasks affect the results. Time constraints prevent any such extensive or systematic analysis. Instead, a fairly conventional analysis is done as described below. A few perturbations of this analysis are presented later in this section of the submission to provide some indications of how various treatments could affect the results.

Vapor species considered for the analysis were in the main well-known species listed in the data base for the VANESA model [4]. A few additional species recently suggested to be important [13] were added. The condensible vapor species considered are constituents of the M-O-H systems where M is an element in the bulk composition of the melt. That is, intermetallic vapor species and mixed oxide vapor species such as $\text{CaMoO}_4(\text{g})$ were not represented in the species list though there is evidence they could contribute to vaporization. Few vapor phase hydrides are included.

Omissions of some vapor species as cited above is not the only vapor speciation concern arising in the analysis of core debris interactions with concrete. Concretes, when intensely heated by melts, also evolve HCl, HF and sulfur or sulfur oxides. Reactions of these vapors could create volatile species. Certainly it is observed that aerosols collected during tests of core debris interactions with concrete contain halides and sulphides [4].

The oxide melt phase presented in part B is far more complex than that in part A of the benchmark exercise. In part A, no mixing could occur on the anion lattice. In part B, anions other than O^{2-} such as SiO_4^{4-} and MoO_4^{2-} can be hypothesized. Mixing models which involve random substitution on both the cationic and anionic lattices could be formulated. Some anions are sufficiently large that the concept of a lattice model becomes difficult to implement. Instead, polymerization models such as



have been used to describe mixing in such systems [14,15]. Some work is underway to develop mixing models of this type for the VANESA model of release during core debris interactions with concrete. These models could not, however, be made available for the benchmark exercise.

A molecular model of the oxide phase was selected. The model was chosen deliberately to be more complex than that currently being used in released versions of the VANESA code. Notable features of the oxide phase model include:

- alkaline earths in the oxide phase are allowed to form oxides, orthosilicates, metasilicates, zirconates and molybdates.
- cerium can be present in both the tri-valent and the tetra-valent states.
- molybdenum can be present in both the tetra-valent and the hexa-valent states.

The many condensed species of the oxide phase are treated as molecular entities, that is distinct anion and cationic lattices are not invoked, and are assumed to form an ideal solution.

The metallic phase was assumed to be an ideal solution. The substantial data base for ferrous melts shows that metal phases such as those arising in the part B problems are not ideal. Consequently, one of the perturbed analyses discussed below examines the effects of non-ideality in the metal phase.

4.2 Results Obtained for the Problems in Part B

Results obtained for the six problems in part B of the exercise are presented in Tables 6-11. Each problem produces a forbiddingly large quantity of numbers. Each element varies in its behavior. Only a limited discussion of the behaviors is presented below.

1. Oxygen Potentials: Oxygen partial pressures calculated for the six problems are

	Oxygen Partial Pressures (atms)	
	2000 K	2500K
Case A	2.3×10^{-18}	2.3×10^{-13}
Case B	4.7×10^{-15}	8.1×10^{-10}
Case C	1.7×10^{-17}	1.9×10^{-12}

Case A is clearly the situation with the lowest oxygen potential. Interestingly, though cases B and C differ only in the amount of silicon dioxide assumed initially to be present, Case C is substantially more reducing than Case B.

2. Uranium Behavior: Substantial reduction of UO_2 to uranium metal occurs in Case A. Much less reduction occurs in Case C and in Case B there is little reduction. In all cases $UO_2(g)$ is an important uranium-bearing vapor species. In the most reducing cases, $UO(g)$ contributes to the vapor pressure and at 2500 K in Case B, $UO_3(g)$ is an important uranium-bearing species.

TABLE 6. Results Obtained in the Case A Problem
with T = 2000 K

SPECIES	MOLES	MOLE FRACTION	PRESSURE (ATMS)	FREE-ENERGY (CAL/MOLE)
METALS				
U metal	6.524e-04	3.265e-01		-44487
Zr metal	2.437e-04	1.228e-01		-39882
Fe metal	1.888e-05	5.885e-01		-98587
Mo metal	1.888e-02	5.885e-04		-24151
Sr metal	7.448e-02	3.723e-07		-43187
Ca metal	1.484e-01	7.827e-05		-36758
Si metal	9.988e-03	4.995e-02		-23348
C graphite	1.728e-02	5.885e-04		-18998
OXIDES				
UO ₂ (s)	3.478e-04	3.187e-01		-319892
SiO ₂ (s)	3.254e-08	2.989e-05		-269347
CoO(s)	2.379e-02	2.127e-03		-188111
LaO _{1.5} (s)	2.888e-02	1.788e-03		-272569
CoO ₂ (s)	2.988e-08	2.672e-05		-321816
MoO ₂ (s)	3.453e-18	3.887e-15		-197188
FeO(s)	6.514e-01	5.823e-06		-123154
ZrO ₂ (s)	6.581e-04	5.883e-01		-315457
CaO _{1.5} (s)	9.978e-02	8.913e-03		-278888
CaSiO ₃	5.469e-01	4.889e-06		-474825
CaSiO _{1.5} O ₂	3.875e-01	2.749e-04		-335418
SrSiO ₃	3.398e-02	2.838e-07		-491833
SrO(s)	2.741e-01	2.458e-06		-188475
SrSi ₂ O ₇	7.445e-01	6.658e-06		-347876
CaMoO ₄	9.872e-09	8.828e-14		-584532
SrMoO ₄	1.535e-12	1.373e-17		-498937
CaZrO ₃	9.717e-03	8.686e-02		-528428
SrZrO ₃	9.887e-01	8.839e-04		-529442
MoO ₃ (s)	1.119e-21	1.888e-26		-173498
GASES				
H ₂	3.992e-01		9.975e-01	-77358
H	1.618e-01		1.618e-03	-13137
OH	8.815e-08		8.888e-18	-93815
O	1.828e-18		1.818e-12	-28857
H ₂ O	5.318e-04		5.387e-06	-168894
U(g)	5.718e-08		5.789e-08	17358
UO	2.928e-04		2.923e-06	-138892
UO ₂ (g)	1.881e-04		1.658e-06	-271648
UO ₃	5.671e-08		5.662e-10	-377716
UO ₄	1.559e-08		1.558e-10	-135665
U(OH) ₂	9.677e-12		9.681e-14	-282881
UO ₂ (OH) ₂	1.444e-13		1.442e-15	-541882
Zr(g)	1.428e-08		1.424e-18	48145
ZrO	5.261e-06		5.252e-20	-113146
ZrO ₂ (g)	9.119e-09		9.183e-11	-225679
ZrOH	1.264e-09		1.262e-11	-118788
Zr(OH) ₂	1.976e-18		1.973e-12	-287818

Note in these and other tables A.AAAe-BB equals A.AAA x 10^{-BB}

TABLE 6. (continued)

SPECIES	MOLES	MOLE FRACTION	PRESSURE (ATMS)	FREE-ENERGY (CAL/MOLE)
Fe(g)	2.852e-02		2.849e-04	497
FeO(g)	1.988e-09		1.985e-11	-73879
FeOH	2.873e-09		2.869e-11	-113267
Fe(OH) ₂	1.281e-12		1.199e-14	-258838
Mo(g)	3.183e-11		3.898e-13	68117
MoO	1.636e-15		1.633e-17	-38535
MoO ₂ (g)	4.125e-17		4.118e-19	-181789
MoO ₃ (g)	8.357e-22		8.343e-24	-256559
MoO ₂ (OH) ₂	1.492e-25		1.498e-27	437426
Ce(g)	2.529e-02		2.524e-04	-41848
CeO(g)	1.157e-09		1.155e-11	-112476
Ce(OH)	5.882e-08		5.853e-08	-184479
Ce(OH) ₂	8.867e-18		8.855e-12	-325578
CeH	1.824e-04		1.823e-06	-58552
Ce ₂	7.511e-09		7.498e-11	-56874
Sr(g)	2.821e-04		2.818e-06	-43903
SrO(g)	1.374e-18		1.371e-12	-131262
SrOH	4.358e-08		4.351e-18	-192837
Sr(OH) ₂	1.417e-12		1.415e-14	-328251
SrH	7.869e-07		7.858e-09	-66535
La(g)	8.726e-09		8.711e-11	1849
LaO(g)	4.179e-06		4.172e-08	-161279
LaOH	2.774e-18		2.769e-12	-161738
La(OH) ₂	4.131e-18		4.124e-12	-339797
La ₂ O	1.842e-13		1.848e-15	-182712
(LaO) ₂	5.581e-12		5.552e-14	-336321
Ce(g)	9.715e-08		9.699e-18	-5583
CeO	5.113e-05		5.185e-07	-168286
CeO ₂ (g)	1.617e-07		1.614e-09	-283288
CeOH	2.586e-09		2.581e-11	-167624
Ce(OH) ₂	1.433e-11		1.431e-13	-323492
(CeO) ₂	2.348e-09		2.342e-11	-354451
CO	2.254e-03		2.251e-05	-136472
CO ₂	2.647e-09		2.642e-11	-228823
Si(g)	2.895e-04		2.891e-06	16718
SiO	3.871e-02		3.864e-04	-141826
SiO ₂ (g)	7.359e-18		7.347e-12	-288978
SiOH	1.827e-08		1.824e-18	-122619
Si(OH) ₂	2.278e-11		2.274e-13	-272524
SiH	1.495e-04		1.493e-06	-28827
SiH ₄	4.985e-06		4.897e-08	-12389
Si ₂	1.144e-07		1.142e-09	1137
O ₂	2.351e-16		2.347e-18	-114214
Ce ₂ O	2.726e-18		2.722e-18	-153189
La ₂ O	1.832e-24		1.838e-26	-128802
Sr ₂ O	5.142e-17		5.133e-19	-174489
(SiO) ₂	1.186e-12		1.184e-14	-218392

TABLE 7. Results in the Case A Problem
with T = 2500 K

SPECIES	MOLES	MOLE FRACTION	PRESSURE (ATMS)	FREE-ENERGY (CAL/MOLE)
METALS				
U metal	4.833e 04	2.418e-01		-61417
Zr metal	4.118e 04	2.060e-01		-47041
Fe metal	9.999e 04	5.001e-01		-43668
Mo metal	1.000e 02	5.002e-04		-36205
Sr metal	1.536e 00	7.683e-06		-58511
Cu metal	2.531e 02	1.266e-03		-50079
Si metal	9.959e 03	4.982e-02		-35223
C graphite	9.002e 01	4.993e-04		-16221
OXIDES				
UO ₂ (o)	5.166e 04	4.552e-01		-355953
SiO ₂ (o)	4.494e 00	3.960e-05		-291158
CaO (o)	1.735e 03	1.528e-02		-208150
LaO _{1.5} (o)	2.000e 02	1.762e-03		-298779
CaO ₂ (o)	8.864e 00	7.810e-05		-351193
MoO ₂ (o)	7.498e-08	6.606e-12		-226029
FeO (o)	9.630e 00	8.484e-05		-146234
ZrO ₂ (o)	5.078e 04	4.474e-01		-342287
CaO _{1.5} (o)	9.911e 02	8.732e-03		-301778
CaSiO ₃	6.724e-01	5.924e-06		-310642
CaSi _{0.5} O ₂	6.525e 01	5.749e-04		-362612
SrSiO ₃	7.048e-02	6.209e-07		-533227
S-O (o)	3.681e 00	3.243e-05		-211362
SrSi _{0.5} O ₂	2.423e 00	2.134e-05		-380042
CaMoO ₄	1.339e-11	1.180e-16		-557770
SrMoO ₄	2.173e-15	1.914e-20		-548211
CaZrO ₃	7.043e 03	6.998e-02		-561992
SrZrO ₃	9.227e 01	8.129e-04		-573649
MoO ₃ (o)	1.471e-20	1.296e-25		-226365
GASES				
H ₂	9.060e 01		9.090e-01	-100342
H	2.589e 00		2.385e-02	-31848
OH	4.457e-05		4.105e-07	-123047
O	7.552e-07		6.957e-09	-52381
H ₂ O	7.986e-03		7.356e-05	-199231
U (g)	1.466e-03		1.350e-05	-12767
UO	4.810e-02		4.431e-04	-175805
UO ₂ (g)	3.278e-02		3.020e-04	-319597
UO ₃	6.071e-05		5.592e-07	-434038
UOH	5.402e-06		4.976e-08	-181033
U(OH) ₂	5.866e-09		5.403e-11	-343230
UO ₂ (OH) ₂	6.125e-11		5.642e-13	-611960
Zr (g)	3.120e-05		2.874e-07	19939
ZrO	4.192e-03		3.862e-05	-150103
ZrO ₂ (g)	3.111e-05		2.866e-07	-271440
ZrOH	1.479e-06		1.363e-08	-161019
Zr(OH) ₂	1.208e-07		1.112e-09	-344676

TABLE 7. (continued)

SPECIES	MOLES	MOLE FRACTION	PRESSURE (ATMS)	FREE-ENERGY (CAL/MOLE)
Fe(g)	2.173e-08		2.002e-02	-27680
FeO(g)	7.039e-06		6.483e-08	-110581
FeOH	2.502e-06		2.305e-08	-155850
Fe(OH) ₂	1.528e-09		1.407e-11	-315187
Mo(g)	4.516e-08		4.160e-10	33345
MoO	2.702e-11		2.487e-13	-75479
MoO ₂ (g)	1.800e-12		1.658e-14	-207723
MoO ₃ (g)	2.012e-16		1.854e-18	-308216
MoO ₂ (OH) ₂	3.888e-20		3.582e-22	-512245
Ca(g)	2.977e-08		2.742e-02	-65358
CaO(g)	8.969e-06		8.261e-08	-147899
Ca(OH)	2.077e-03		1.913e-05	-225357
Ca(OH) ₂	6.349e-07		5.848e-09	-381256
CaH	2.055e-02		1.893e-04	-91046
Ca ₂	8.813e-05		6.091e-07	-95353
Sr(g)	2.123e-02		1.955e-04	-74591
SrO(g)	5.600e-07		5.166e-09	-167918
SrOH	1.655e-05		1.524e-07	-235141
Sr(OH) ₂	2.013e-09		1.854e-11	-386463
SrH	1.502e-04		1.383e-06	-100401
La(g)	4.797e-06		4.419e-08	-27606
LaO(g)	8.088e-04		7.450e-06	-198775
LaOH	1.186e-07		1.092e-09	-205327
La(OH) ₂	5.741e-08		5.208e-10	-397029
La ₂ O	1.898e-10		1.748e-12	-234677
(LaO) ₂	3.935e-09		3.624e-11	-395434
Ce(g)	7.386e-05		6.803e-07	-36237
CeO	1.048e-02		9.657e-05	-206551
CeO ₂ (g)	1.262e-04		1.162e-06	-330291
CeOH	1.295e-06		1.193e-08	-212252
Ce(OH) ₂	9.023e-09		8.311e-11	-393684
(CeO) ₂	1.161e-06		1.070e-08	-413784
CO	1.872e-01		1.658e-03	-167880
CO ₂	2.392e-06		2.204e-08	-257794
Si(g)	2.493e-02		2.297e-04	-8498
SiO	1.779e-08		1.639e-02	-175395
SiO ₂ (g)	1.009e-06		9.295e-09	-249641
SiOH	4.630e-06		4.265e-08	-161921
Si(OH) ₂	3.572e-09		3.291e-11	-322420
SiH	6.567e-03		6.049e-05	-52278
SiH ₄	3.312e-06		3.051e-08	-165785
Si ₂	3.483e-05		3.209e-07	-25966
O ₂	2.536e-11		2.336e-13	-146901
Ce ₂ O	1.730e-13		1.594e-15	-189996
La ₂ O	7.392e-20		6.809e-22	-161000
Sr ₂ O	6.341e-14		5.841e-16	-205471
(SiO) ₂	6.114e-11		5.632e-13	-201520

TABLE 8. Results Obtained in the Case B Problem
with T = 2000 K

SPECIES	MOLES	MOLE FRACTION	PRESSURE (ATMS)	FREE-ENERGY (CAL/MOLE)
METALS				
U metal	2.583e-01	2.288e-04		-44487
Zr metal	1.835e-01	1.672e-06		-33622
Fe metal	9.989e-04	9.182e-01		-38527
Mo metal	1.888e-02	9.112e-04		-24151
Sr metal	2.659e-04	2.423e-09		-43187
Ca metal	1.967e-02	1.793e-05		-36758
Si metal	9.632e-03	8.776e-02		-23348
C graphite	9.981e-01	9.894e-04		-18998
OXIDES				
UO ₂ (c)	9.997e-04	4.372e-01		-319892
SiO ₂ (c)	2.355e-04	1.838e-01		-269347
CaO(c)	5.568e-03	2.435e-02		-188111
LaO _{1.5} (c)	2.888e-02	8.747e-04		-272569
La ₂ O ₃ (c)	1.969e-01	8.611e-05		-321816
MoO ₂ (c)	2.589e-06	1.132e-11		-197168
FeO(c)	1.887e-02	4.754e-04		-123154
ZrO ₂ (c)	3.716e-03	1.825e-02		-315457
CaO _{1.5} (c)	9.883e-02	4.287e-03		-276886
CaSiO ₃	4.532e-04	1.962e-01		-474825
CaSi ₂ O ₇	4.283e-04	1.873e-01		-335418
SrSiO ₃	7.176e-01	3.139e-04		-491835
SrO(c)	1.638e-01	7.156e-07		-188475
SrSi ₂ O ₇	2.644e-01	1.157e-04		-347876
CaMoO ₄	2.661e-08	1.164e-13		-584532
SrMoO ₄	1.855e-13	4.816e-19		-496937
CaZrO ₃	6.282e-03	2.748e-02		-528428
SrZrO ₃	1.638e-02	7.131e-06		-529442
MoO ₃ (c)	2.633e-22	1.151e-27		-173496
GASES				
H ₂	9.989e-01	9.654e-01		-77358
H	1.645e-01	1.589e-03		-13137
OH	4.821e-06	3.886e-08		-93615
O	4.728e-09	4.569e-11		-28857
H ₂ O	2.385e-02	2.385e-04		-166894
U(g)	4.125e-09	3.987e-11		17358
UC	9.481e-06	9.163e-08		-136892
UO ₂ (g)	2.414e-04	2.333e-06		-271648
UO ₃	3.788e-06	3.576e-08		-377716
UOH	4.966e-10	4.799e-12		-135665
U(OH) ₂	1.361e-11	1.316e-13		-282821
UO ₂ (OH) ₂	4.895e-18	3.957e-12		-541692
Zr(g)	2.819e-13	1.952e-15		48145
ZrO	3.344e-09	3.232e-11		-113146
ZrO ₂ (g)	2.682e-10	2.515e-12		-225679
ZrOH	7.983e-13	7.637e-15		-118788
Zr(OH) ₂	5.457e-12	5.274e-14		-287818

TABLE 8. (continued)

SPECIES	MOLES	MOLE FRACTION	PRESSURE (ATMS)	FREE-ENERGY (CAL/MOLE)
Fe(g)	3.855e-02		3.726e-04	497
FeO(g)	1.668e-07		1.684e-09	-73079
FeOH	2.384e-07		2.384e-09	-113267
Fe(OH) ₂	4.399e-09		4.251e-11	-258830
Mo(g)	5.835e-11		5.639e-13	60117
MoO	1.381e-13		1.335e-15	-38535
MoO ₂ (g)	1.563e-13		1.511e-15	-161709
MoO ₃ (g)	1.421e-16		1.374e-18	-256559
MoO ₂ (OH) ₂	1.103e-18		1.066e-20	-437426
Co(g)	6.663e-03		6.440e-05	-41840
CoO(g)	1.369e-08		1.323e-10	-112476
Co(OH)	5.890e-05		5.693e-07	-184478
Co(OH) ₂	3.529e-07		3.410e-09	-325570
CoH	2.604e-05		2.516e-07	-58552
Co ₂	5.049e-10		4.880e-12	-56874
Sr(g)	1.359e-06		1.313e-08	-49903
SrO(g)	4.144e-11		4.005e-13	-131262
SrOH	1.294e-08		1.250e-10	-192637
Sr(OH) ₂	1.857e-11		1.795e-13	-328251
SrH	5.203e-09		5.029e-11	-66535
La(g)	1.466e-11		1.417e-13	1049
LaO(g)	3.152e-07		3.046e-09	-161279
LaOH	2.058e-11		1.989e-13	-161730
La(OH) ₂	1.354e-09		1.308e-11	-339797
La ₂ O	1.278e-17		1.235e-19	-182712
(LaO) ₂	3.063e-14		2.960e-16	-336321
Ce(g)	1.605e-10		1.551e-12	-5583
CeO	3.792e-06		3.665e-08	-168286
CeO ₂ (g)	5.382e-07		5.201e-09	-283209
CeOH	1.872e-10		1.809e-12	-167624
Ce(OH) ₂	4.619e-11		4.464e-13	-323492
(CeO) ₂	1.249e-11		1.208e-13	-354451
CO	1.900e-01		1.833e-03	-136472
CO ₂	1.001e-05		9.675e-08	-220003
Si(g)	3.001e-04		3.674e-06	16718
SiO	3.153e-00		3.048e-02	-141826
SiO ₂ (g)	2.691e-06		2.601e-08	-208970
SiOH	1.464e-06		1.415e-08	-122619
Si(OH) ₂	8.064e-08		7.793e-10	-272526
SiH	2.671e-04		2.581e-06	-20627
SiH ₄	8.339e-06		8.059e-08	-123097
Si ₂	3.647e-07		3.525e-09	11318
O ₂	4.893e-13		4.720e-15	-114270
Ce ₂ O	3.234e-20		3.126e-22	-153188
La ₂ O ₂	3.494e-24		3.377e-26	-120000
Sr ₂ O	1.009e-19		9.754e-22	-174469
(SiO) ₂	7.103e-09		6.865e-11	-210392

TABLE 9. Results Obtained in the Case B Problem
with T = 2500 K

SPECIES	MOLES	MOLE FRACTION	PRESSURE (ATMS)	FREE-ENERGY (CAL/MOLE)
METALS				
U metal	6.472e-02	6.325e-05		-61417
Zr metal	2.958e-01	2.890e-06		-47041
Fe metal	9.724e-04	9.523e-01		-43668
Ko metal	1.800e-02	8.773e-01		-36205
Sr metal	1.615e-03	1.578e-08		-58511
Ca metal	1.259e-01	1.230e-04		-50279
Si metal	4.966e-03	4.853e-02		-35223
C graphite	4.856e-02	3.964e-05		-16221
OXIDES				
UO ₂ (c)	9.999e-04	4.148e-01		-355953
SiO ₂ (c)	3.239e-04	1.343e-01		-291158
CaO(c)	2.114e-04	8.764e-02		-208150
LaC _{1.5} (c)	2.800e-02	8.292e-04		-298779
CoO ₂ (c)	6.427e-01	2.665e-04		-351193
MoO ₂ (c)	1.884e-03	4.493e-09		-226029
FeO(c)	2.294e-03	9.511e-03		-146234
ZrO ₂ (c)	5.271e-03	2.185e-02		-342287
Co _{1.5} (c)	9.356e-02	3.879e-03		-301778
CoSiO ₃	2.779e-04	1.152e-01		-518642
CoSi _{0.5} O ₂	4.630e-04	1.920e-01		-362612
SrSiO ₃	6.155e-01	2.552e-04		-533227
SrO(c)	9.480e-01	3.930e-06		-211362
SrSi _{0.5} O ₂	3.633e-01	1.506e-04		-380042
CaMoO ₄	6.546e-05	2.714e-10		-557770
SrMoO ₄	2.245e-10	9.309e-16		-548211
CaZrO ₃	4.728e-03	1.960e-02		-561992
SrZrO ₃	1.161e-02	4.812e-06		-573649
MoO ₃ (c)	1.254e-14	5.200e-20		-226365
GASES				
H ₂	8.854e-01		7.010e-03	-100342
H	2.586e-01		2.894e-03	-31848
OH	2.626e-02		2.127e-06	-123047
O	5.067e-03		4.104e-07	-52381
H ₂ O	4.132e-01		3.347e-05	-199231
U(g)	4.361e-05		3.533e-09	-12767
UO	8.443e-02		6.839e-06	-175825
UO ₂ (g)	3.395e-02		2.750e-04	-319507
UO ₃	3.710e-01		3.805e-05	-434038
UOH	8.327e-07		6.745e-11	-181033
U(OH) ₂	4.685e-09		3.795e-13	-342230
UO ₂ (OH) ₂	1.783e-07		1.380e-11	-611960
Zr(g)	4.978e-08		4.832e-12	19939
ZrO	3.947e-04		3.197e-08	-150103
ZrO ₂ (g)	1.728e-04		1.400e-08	-271440
ZrOH	1.223e-08		9.906e-13	-161019
Zr(OH) ₂	5.173e-09		4.190e-13	-344676

TABLE 9. (continued)

SPECIES	MOLES	MOLE FRACTION	PRESSURE (ATMS)	FREE-ENERGY (CAL/MOLE)
Fe(g)	4.695e-02		3.003e-02	-27680
FeO(g)	8.073e-02		7.268e-06	-110581
FeOH	2.001e-03		2.269e-07	-155850
Fe(OH) ₂	8.861e-06		7.178e-10	-315187
Mo(g)	1.003e-05		8.127e-10	33345
MoO	3.539e-07		2.867e-11	-75479
MoO ₂ (g)	1.392e-06		1.128e-10	-207723
MoO ₃ (g)	9.183e-09		7.438e-13	-300216
MoO ₂ (OH) ₂	8.073e-13		6.539e-17	-512245
Co(g)	3.290e-01		2.665e-03	-65358
CoO(g)	5.848e-03		4.737e-07	-147899
Co(OH)	1.189e-01		9.634e-06	-225357
Co(OH) ₂	1.884e-04		1.526e-08	-381256
CoH	1.004e-02		1.615e-06	-91046
Co ₂	7.103e-05		5.754e-09	-95353
Sr(g)	4.958e-03		4.016e-07	-74591
SrO(g)	7.729e-06		6.260e-10	-167910
SrOH	2.003e-05		1.622e-09	-235141
Sr(OH) ₂	1.262e-08		1.023e-12	-386463
SrH	3.080e-06		2.495e-10	-100401
La(g)	5.664e-07		4.588e-11	-27606
LaO(g)	5.634e-03		4.564e-07	-198775
LaOH	7.253e-08		5.875e-12	-205327
La(OH) ₂	1.820e-07		1.474e-11	-397829
La ₂ O	1.373e-12		1.112e-16	-234677
(LaO) ₂	1.679e-09		1.360e-13	-395434
Ce(g)	8.233e-06		6.669e-10	-36237
CeO	6.895e-02		5.585e-06	-206551
CeO ₂ (g)	4.897e-02		3.967e-06	-330291
CeOH	7.478e-07		6.057e-11	-212252
Ce(OH) ₂	2.700e-08		2.187e-12	-383684
(CeO) ₂	4.416e-07		3.577e-11	-413784
CO	9.587e-01		7.766e-03	-167880
CO ₂	7.518e-02		6.090e-06	-257794
Si(g)	2.762e-00		2.237e-04	-8498
SiO	1.163e-04		9.418e-01	-175395
SiO ₂ (g)	3.891e-01		3.152e-05	-249641
SiOH	2.657e-03		2.153e-07	-161921
Si(OH) ₂	1.062e-05		9.606e-10	-322420
SiH	6.389e-02		5.175e-06	-52278
SiH ₄	2.182e-08		1.768e-12	-165785
Si ₂	3.759e-03		3.045e-07	-25966
O ₂	1.004e-05		8.133e-10	-146901
Ce ₂ O	1.115e-15		9.036e-21	-189996
La ₂ O	3.038e-17		2.461e-21	-161000
Sr ₂ O	1.795e-15		1.454e-19	-205471
(SiO) ₂	2.297e-05		1.061e-09	-251520

TABLE 10. Results Obtained in the Case C Problem
with T = 2000 K

SPECIES	MOLES	MOLE FRACTION	PRESSURE (ATMS)	FREE-ENERGY (CAL/MOLE)
METALS				
U metal	7.459e-03	6.778e-02		-44487
Zr metal	5.753e-02	5.227e-05		-33622
Fe metal	9.999e-04	9.886e-01		-38527
Mo metal	1.882e-02	9.886e-04		-24151
Sr metal	4.654e-01	4.229e-06		-43187
Ca metal	5.116e-02	4.649e-03		-36758
Si metal	1.882e-03	1.718e-02		-23348
C graphite	9.999e-01	9.885e-04		-18998
OXIDES				
UO ₂ (c)	9.254e-04	4.777e-01		-319892
SiO ₂ (c)	1.429e-01	7.377e-05		-269347
CoO(c)	7.417e-04	3.829e-01		-188111
LaO _{1.5} (c)	2.888e-02	1.832e-03		-272569
CoO ₂ (c)	4.921e-02	2.548e-05		-321816
MoO ₂ (c)	8.841e-09	4.151e-14		-197168
FeO(c)	5.573e-02	2.877e-05		-123154
ZrO ₂ (c)	3.617e-02	1.867e-03		-315457
CoO _{1.5} (c)	9.951e-02	5.137e-03		-276226
CaSiO ₃	4.324e-02	2.232e-03		-474825
CaSi _{0.5} O ₂	1.527e-04	7.883e-02		-335418
SrSiO ₃	4.629e-02	2.379e-05		-491835
SrO(c)	1.467e-01	7.574e-05		-188475
SrSi _{0.5} O ₂	6.346e-01	3.276e-04		-347876
CaMoO ₄	2.878e-06	1.589e-11		-584532
SrMoO ₄	8.219e-11	4.243e-16		-496937
CoZrO ₃	9.616e-03	4.964e-02		-528422
SrZrO ₃	1.682e-01	8.671e-05		-529442
MoO ₃ (c)	1.937e-21	1.882e-26		-173496
GASES				
H ₂	9.991e-01		9.827e-01	-77358
H	1.632e-01		1.622e-03	-13137
OH	2.419e-07		2.375e-09	-93615
O	2.822e-18		2.778e-12	-28257
H ₂ O	1.447e-03		1.428e-05	-166894
U(g)	1.227e-06		1.185e-08	17352
UO	1.682e-04		1.651e-06	-136292
UO ₂ (g)	2.597e-04		2.549e-06	-271642
UO ₃	2.413e-07		2.369e-09	-377716
UOH	8.882e-09		8.717e-11	-135665
U(OH) ₂	1.488e-11		1.468e-13	-282821
UO ₂ (OH) ₂	1.645e-12		1.614e-14	-541692
Zr(g)	6.215e-12		6.182e-14	48145
ZrO	6.248e-09		6.125e-11	-113146
ZrO ₂ (g)	2.943e-11		2.889e-13	-225679
ZrOH	1.486e-12		1.459e-14	-118728
Zr(OH) ₂	6.272e-13		6.156e-15	-287818

TABLE 10. (continued)

SPECIES	MOLES	MOLE FRACTION	PRESSURE (ATMS)	FREE-ENERGY (CAL/MOLE)
Fe(g)	3.789e-02		3.719e-04	497
FeO(g)	9.890e-09		9.788e-11	-73879
FeOH	1.432e-08		1.405e-10	-113267
Fe(OH) ₂	1.614e-11		1.585e-13	-258830
Mo(g)	5.729e-11		5.624e-13	60117
MoO	8.220e-15		8.069e-17	-38535
MoO ₂ (g)	5.641e-16		5.537e-18	-161709
MoO ₃ (g)	3.110e-20		3.053e-22	-256559
MoO ₂ (OH) ₂	1.486e-23		1.459e-25	-437426
Ca(g)	1.781e-08		1.678e-02	-41840
CaO(g)	2.119e-07		2.080e-09	-112476
Ca(OH)	9.191e-04		9.022e-06	-184478
Ca(OH) ₂	3.365e-07		3.303e-09	-325570
CaH	6.701e-03		6.578e-05	-58552
Ca ₂	3.344e-05		3.282e-07	-56874
Sr(g)	2.335e-03		2.292e-05	-49903
SrO(g)	4.319e-09		4.239e-11	-131262
SrOH	1.359e-06		1.334e-08	-192837
Sr(OH) ₂	1.192e-10		1.170e-12	-328251
SrH	9.013e-06		8.848e-08	-66535
La(g)	1.142e-09		1.120e-11	1049
LaO(g)	1.488e-06		1.460e-08	-161279
LaOH	9.792e-11		9.612e-13	-161730
La(OH) ₂	3.935e-10		3.862e-12	-339797
La ₂ O	4.770e-15		4.683e-17	-182712
(LaO) ₂	6.931e-13		6.803e-15	-336321
Ce(g)	1.268e-08		1.245e-10	-5583
CeO	1.817e-05		1.783e-07	-168286
CeO ₂ (g)	1.563e-07		1.534e-09	-283208
CeOH	9.040e-10		8.873e-12	-167624
Ce(OH) ₂	1.363e-11		1.338e-13	-323492
(CeO) ₂	2.913e-10		2.859e-12	-354451
CO	1.133e-02		1.112e-04	-136472
CO ₂	3.619e-08		3.553e-10	-220003
Si(g)	7.293e-05		7.159e-07	16718
SiO	3.668e-02		3.600e-04	-141826
SiO ₂ (g)	1.898e-09		1.863e-11	-200970
SiOH	1.717e-08		1.685e-10	-122619
Si(OH) ₂	5.777e-11		5.678e-13	-272526
SiH	5.164e-05		5.069e-07	-20627
SiH ₄	1.651e-06		1.621e-08	-123097
Si ₂	1.364e-08		1.338e-10	11318
O ₂	1.771e-15		1.738e-17	-114270
Ce ₂ O	1.244e-17		1.221e-19	-153188
LaO ₂	9.998e-25		9.814e-27	-128800
Sr ₂ O	1.836e-14		1.802e-16	-174469
(SiO) ₂	9.761e-13		9.581e-15	-210392

TABLE 11. Results Obtained in the Case C Problem
with T = 2500 K.

SPECIES	MOLES	MOLE FRACTION	PRESSURE (ATMS)	FREE-ENERGY (CAL/MOLE)
METALS				
U metal	3.546e 03	3.212e-02		-61417
Zr metal	6.862e 01	5.491e-04		-47041
Fe metal	9.991e 04	9.850e-01		-43668
Mo metal	1.888e 02	9.858e-04		-36205
Sr metal	1.388e 00	1.178e-05		-58511
Ca metal	1.329e 03	1.204e-02		-58079
Si metal	5.352e 03	4.848e-02		-35223
C graphite	9.868e 01	8.939e-04		-16221
OXIDES				
UO ₂ (c)	9.645e 04	4.861e-01		-355953
SiO ₂ (c)	5.147e 01	3.898e-04		-291158
CaO(c)	8.176e 04	4.128e-01		-288150
LaO _{1.5} (c)	2.888e 02	1.888e-03		-288779
CaO ₂ (c)	1.484e 01	7.476e-05		-351193
MoO ₂ (c)	1.988e-06	9.617e-12		-226029
FeO(c)	8.637e 01	4.353e-04		-146234
ZrO ₂ (c)	1.922e 03	9.587e-03		-342287
CaO _{1.5} (c)	9.852e 02	4.965e-03		-381778
CaSiO ₃	2.479e 02	1.250e-03		-518642
CaSi _{0.5} O ₂	8.683e 03	4.335e-02		-362612
SrSiO ₃	4.189e 00	2.111e-05		-533227
SrO(c)	2.796e 01	1.489e-04		-211362
SrSi _{0.5} O ₂	5.148e 01	2.594e-04		-388042
CaO _{0.4}	2.685e-08	1.313e-13		-557770
SrO _{0.4}	6.813e-13	3.434e-18		-548211
ZrO ₃	8.822e 03	4.843e-02		-561992
Zr ₂ O ₃	1.582e 01	7.569e-05		-573649
Mo ₂ O ₃ (c)	1.861e-18	5.349e-24		-226365
GASES				
H ₂	9.827e 01		6.264e-01	-180342
H	3.186e 00		1.980e-02	-31848
OH	1.516e-04		9.662e-07	-123047
O	3.894e-06		1.972e-08	-52381
H ₂ O	2.255e-02		1.437e-04	-199231
U(g)	2.814e-04		1.784e-06	-12767
UO	2.618e-02		1.669e-04	-175825
UO ₂ (g)	5.859e-02		3.225e-04	-319597
UO ₃	2.656e-04		1.693e-06	-434838
UOH	2.441e-06		1.556e-08	-181833
U(OH) ₂	6.238e-09		3.976e-11	-343238
UO ₂ (OH) ₂	5.236e-10		3.338e-12	-611860
Zr(g)	1.282e-07		7.660e-10	19939
ZrO	4.579e-05		2.918e-07	-158183
ZrO ₂ (g)	9.633e-07		6.140e-09	-271448
ZrOH	1.341e-08		8.548e-11	-161819
Zr(OH) ₂	2.577e-09		1.642e-11	-344676

TABLE 11. (continued)

SPECIES	MOLES	MOLE FRACTION	PRESSURE (ATMS)	FREE-ENERGY (CAL/MOLE)
Fe(g)	5.682e-02		3.622e-02	-27680
FeO(g)	5.218e-05		3.326e-07	-118581
FeOH	1.540e-05		9.815e-08	-155850
Fe(OH) ₂	2.213e-08		1.411e-10	-315187
Mo(g)	1.182e-07		7.533e-10	33345
MoO	2.003e-10		1.277e-12	-75479
MoO ₂ (g)	3.787e-11		2.414e-13	-207723
MoO ₃ (g)	1.200e-14		7.651e-17	-308216
MoO ₂ (OH) ₂	4.531e-18		2.888e-20	-512245
Co(g)	4.091e-01		2.607e-01	-65358
CoO(g)	3.494e-04		2.227e-06	-147899
Co(OH)	6.717e-02		4.262e-04	-225357
Co(OH) ₂	4.833e-05		3.080e-07	-381256
CoH	2.344e-01		1.494e-03	-91846
Co ₂	8.640e-03		5.507e-05	-95353
Sr(g)	4.701e-02		2.897e-04	-74591
SrO(g)	3.522e-06		2.245e-08	-167918
SrOH	8.626e-05		5.498e-07	-235141
Sr(OH) ₂	2.470e-08		1.574e-10	-386463
SrH	2.761e-04		1.760e-06	-100401
La(g)	8.305e-07		5.294e-09	-27606
LaO(g)	3.970e-04		2.531e-06	-198775
LaOH	4.831e-08		3.079e-10	-205327
La(OH) ₂	5.507e-08		3.510e-10	-397829
La ₂ O	1.116e-11		7.116e-14	-234677
(LaO) ₂	6.561e-10		4.182e-12	-395434
Ce(g)	1.271e-05		8.102e-08	-36237
CeO	5.115e-03		3.261e-05	-206551
CeO ₂ (g)	1.746e-04		1.113e-06	-330291
CeOH	5.244e-07		3.343e-09	-212252
Ce(OH) ₂	8.602e-09		5.483e-11	-383684
(CeO) ₂	1.913e-07		1.219e-09	-413784
CO	1.320e-02		8.416e-03	-167880
CO ₂	4.976e-05		3.171e-07	-257794
Si(g)	3.507e-02		2.235e-04	-8498
SiO	7.094e-02		4.521e-02	-175395
SiO ₂ (g)	1.141e-05		7.272e-08	-249641
SiOH	1.533e-05		9.760e-08	-161921
Si(OH) ₂	2.783e-08		1.774e-10	-322420
SiH	7.667e-03		4.887e-05	-52278
SiH ₄	2.212e-06		1.410e-08	-165785
Si ₂	4.768e-05		3.039e-07	-25986
O ₂	2.946e-10		1.878e-12	-146901
Ce ₂ O	1.006e-14		6.409e-17	-109996
La ₂ O ₂	.029e-19		6.558e-22	-161000
Sr ₂ O	6.103e-13		3.890e-15	-205471
(SiO) ₂	6.728e-10		4.289e-12	-251520

3. Behavior of Zirconium: Zirconium is oxidized in all cases. Metallic products of reduction do insure relatively low oxygen partial pressures are maintained.

4. Behavior of Silicon: There is a tendency, especially under strongly reducing conditions for silica to be reduced to silicon metal. Otherwise, silica tends to react with available alkaline earths to form usually orthosilicates.

5. Behavior of Carbon: In all cases, the system is reducing-enough that much of the CO₂ is reduced to carbon which is incorporated into the metallic melt.

6. Behavior of Strontium: The most significant feature of strontium behavior is its vaporization from the melt. To track this vaporization, a "figure-of-merit", F(Sr), is defined as:

$$F(\text{Sr}) = \sum_v n_{vm}(v)$$

where n_v = number of strontium atoms in the v^{th} vapor species

$m(v)$ = moles of the v^{th} vapor species.

Values of these figures-of-merit are summarized below:

	F(Sr) at	
	2000K	2500K
Case A	2.0×10^{-4}	2.1×10^{-2}
Case B	1.4×10^{-6}	5.0×10^{-3}
Case C	2.3×10^{-3}	4.7×10^{-2}

In all cases there is at least an order of magnitude increase in the figure-of-merit in going from 2000 to 2500 K. The increase in strontium release with temperature is due to both the increases in the partial pressures of strontium-bearing gases and to the increase in vapor phase volume by condensible species such as SiO(g). The increases in condensible species contributions to the vapor phase at the highest temperatures also affect release of other radionuclides.

Sr(g) is the dominant vapor species for these calculations. Of course, in other circumstances other vapor species may be important.

7. Behavior of Cerium: Cerium in the oxide melt is predicted to be predominantly in the tri-valent form. To track vaporization of cerium, a figure-of-merit, F(Ce),

which is similar in definition to that used to track strontium vaporization was defined. Summary values of this figure of merit are listed below:

	F(Ce) at	
	2000 K	2500 K
Case A	5.1×10^{-5}	1.1×10^{-2}
Case B	4.3×10^{-6}	1.2×10^{-1}
Case C	1.8×10^{-5}	5.3×10^{-3}

Ce(g), CeO(g) and CeO₂(g) are the important vapor species.

8. Behavior of Lanthanum: Again, a figure-of-merit is used to track lanthanum vaporization among the various problems:

	F(La) at	
	2000 K	2500 K
Case A	4.2×10^{-6}	8.1×10^{-4}
Case B	3.2×10^{-7}	5.6×10^{-3}
Case C	1.5×10^{-6}	4.0×10^{-4}

Little lanthanum release would be expected below 2500 K and what release does occur is predominantly the result of the vapor pressure of LaO(g).

4.3 Comparison to Results of a Simpler Model of the Oxide Phase

A fairly complex model of the condensed phase was employed in the analysis of the problems in part B of the benchmark exercise. Not all models have such complex descriptions of the condensed phase. It is of interest then to compare the results obtained above to results obtained with a simpler model. The simpler model considered here does not allow formation of silicates, zirconates or molybdates. Further, it does not allow reduction of silica to silicon, calcium oxide to calcium metal, strontium oxide to strontium metal, or uranium to uranium metal. Vapor phase speciation, however, is the same in the simpler model as in the analyses for the part B problems.

Results obtained with the simpler model for Case B at 2000 K are collected in Table 12. The most notable feature of these results is the very high partial pressure of silicon monoxide. Whereas with the more complex model, the partial pressure of SiO(g) was calculated to be only 3×10^{-2} atmospheres, with the simpler model, SiO(g) is calculated to be the dominant gas phase species.

Figures-of-merit for strontium, lanthanum and cerium vaporization for calculations with the complex and the simple models of the condensed phase are:

TABLE 12. Results Obtained for Case B at 2000 K with a Simpler Solution Model

SPECIES	MOLES	MOLE FRACTION	PRESSURE (ATMS)	FREE-ENERGY (CAL/MOLE)
METALS				
Zr metal	5.889e-01	4.997e-04		-33502
Fe metal	9.998e-04	9.975e-01		-30527
Mo metal	1.888e-02	9.977e-04		-24151
C graphite	9.687e-01	9.365e-04		-10998
OXIDES				
UO ₂ (c)	1.888e-05	5.422e-01		-319892
SiO ₂ (c)	8.114e-04	2.777e-01		-269347
CoO(c)	9.979e-04	3.415e-01		-188111
LaO _{1.5} (c)	2.888e-02	6.845e-04		-272569
CoO ₂ (c)	5.778e-02	1.977e-05		-321816
MoO ₂ (c)	2.548e-08	8.694e-14		-197168
FeO(c)	1.275e-01	4.363e-05		-123154
ZrO ₂ (c)	9.958e-03	3.485e-02		-315457
CoO _{1.5} (c)	9.942e-02	3.483e-03		-276006
SrO(c)	9.858e-01	3.374e-04		-188475
MoO ₃ (c)	2.922e-21	1.888e-26		-173496
GASES				
H ₂	9.816e-01		5.128e-03	-77358
H	2.219e-02		1.157e-04	-13137
OH	4.543e-06		2.378e-10	-93615
O	7.335e-08		3.826e-12	-28257
H ₂ O	1.963e-03		1.824e-07	-166894
U(g)	8.533e-05		4.451e-09	17350
UO	1.642e-02		8.565e-07	-136092
UO ₂ (g)	3.581e-02		1.826e-06	-271648
UO ₃	4.494e-05		2.344e-09	-377716
UOH	6.253e-08		3.267e-12	-135665
U(OH) ₂	1.847e-11		5.462e-16	-282821
UO ₂ (OH) ₂	2.288e-12		1.152e-16	-541692
Zr(g)	1.118e-08		5.832e-13	48145
ZrO	1.552e-05		8.887e-10	-113146
ZrO ₂ (g)	1.818e-07		5.269e-12	-225679
ZrOH	2.668e-10		1.392e-14	-118788
Zr(OH) ₂	1.124e-11		5.861e-16	-287818

TABLE 12. (continued)

SPECIES	MOLES	MOLE FRACTION	PRESSURE (ATMS)	FREE-ENERGY (CAL/MOLE)
Fe(g)	7.828e-08		4.883e-04	497
FeO(g)	2.822e-06		1.472e-10	-73079
FeOH	2.952e-07		1.540e-11	-113267
Fe(OH) ₂	3.322e-11		1.733e-15	-258830
Mo(g)	1.184e-08		6.175e-13	60117
MoO	2.346e-12		1.224e-16	-38535
MoO ₂ (g)	2.224e-13		1.160e-17	-161709
MoO ₃ (g)	1.693e-17		8.832e-22	-256559
MoO ₂ (OH) ₂	5.833e-23		3.843e-27	-437426
Co(g)	2.868e-02		1.879e-02	-41840
CoO(g)	3.557e-05		1.855e-09	-112476
Co(OH)	1.115e-02		5.815e-07	-184478
Co(OH) ₂	4.872e-07		2.124e-11	-325570
CoH	5.884e-02		3.869e-06	-58552
Co ₂	2.625e-03		1.369e-07	-56874
Sr(g)	1.417e-08		7.393e-25	-49903
SrO(g)	3.628e-06		1.588e-10	-131262
SrOH	8.229e-05		4.293e-09	-192837
Sr(OH) ₂	7.285e-10		3.758e-14	-328251
SrH	3.953e-04		2.862e-06	-66535
La(g)	6.774e-08		4.576e-12	1849
LaO(g)	1.579e-04		8.238e-09	-161279
LaOH	7.511e-10		3.918e-14	-161730
La(OH) ₂	3.812e-10		1.571e-14	-339797
La ₂ O	2.868e-13		1.879e-17	-182712
(LaO) ₂	4.158e-11		2.165e-15	-336321
Ce(g)	9.741e-07		5.881e-11	-5583
CeO	1.927e-03		1.805e-07	-168286
CeO ₂ (g)	2.298e-05		1.195e-09	-283208
CeOH	6.928e-09		3.613e-13	-167624
Ce(OH) ₂	1.842e-11		5.437e-16	-323492
(CeO) ₂	1.741e-08		9.883e-13	-354451
CO	3.132e-08		1.634e-04	-136472
CO ₂	1.382e-05		7.289e-10	-220003
Si(g)	2.788e-01		1.413e-03	16718
SiO	1.881e-04		9.813e-01	-141826
SiO ₂ (g)	1.344e-03		7.813e-08	-208978
SiOH	6.362e-04		5.319e-08	-122619
Si(OH) ₂	2.136e-07		1.114e-11	-272526
SiH	1.386e-08		7.728e-05	-28627
SiH ₄	1.671e-05		8.717e-10	-123097
Si ₂	9.993e-08		5.212e-14	11318
O ₂	6.357e-13		3.316e-17	-114278
Co ₂ O	5.383e-16		2.888e-20	-153188
La ₂ O ₂	1.466e-22		7.646e-27	-128820
Sr ₂ O	4.964e-11		2.589e-15	-174469
(SiO) ₂	1.365e-03		7.118e-08	-211177

	Simple Model	Complex Model
F(Sr)	1.4	1.4×10^{-6}
F(Ce)	2.0×10^{-3}	4.3×10^{-6}
F(La)	1.6×10^{-4}	3.2×10^{-7}

Vastly higher vaporization of radionuclides is predicted with the simple model largely because of the huge volume of SiO(g) created. Whether this SiO(g) forms or not depends on the activity of SiO₂ in the melt. In the complex model, this activity is kept low by reduction of SiO₂ to Si and by the formation of silicates. There is some experimental evidence that enhanced vaporization does occur when zirconium is added to siliceous melts (8) as is suggested especially by the simpler model.

4.4 Effects of Non-Ideality in the Metal Phase

Solutions to the problems posed in part B of the benchmark exercise were all calculated assuming the mixture of U, Fe, Zr, Mo, Si, Ca, Sr, and C was an ideal solution. There is substantial evidence such an alloy would not be ideal. Calcium and carbon are well-known to be non-ideal when alloyed with iron. To ascertain the effects of non-ideality in the metal phase a very approximate model drawn from the ferrous-metallurgy literature [16] was formulated and the Case B problem at 2000K was recalculated. The activity coefficients in the non-ideal metal phase model are:

$$\begin{aligned}
 \ln \gamma(\text{Zr}) &= \ln(0.037) \\
 \ln \gamma(\text{U}) &= \ln(0.027) + 9.4X(\text{U}) \\
 \ln \gamma(\text{Mo}) &= -4X(\text{C}) \\
 \ln \gamma(\text{Ca}) &= \ln(2240) - 15.8X(\text{C}) - 10.7X(\text{Si}) \\
 \ln \gamma(\text{Si}) &= \ln(0.0013) + 9.7X(\text{Si}) - 10.7X(\text{Ca}) - 10.7X(\text{Sr}) \\
 \ln \gamma(\text{C}) &= \ln(0.70) + 6.9X(\text{C}) - 15.8X(\text{Ca}) - 4X(\text{Mo}) \\
 &\quad + 9.7X(\text{Si}) + 11.6X(\text{C})^2 + 8.8X(\text{Si})^2 - 15.8X(\text{Sr})
 \end{aligned}$$

where $\gamma(i)$ = activity coefficient of the constituent i of the metal phase

$X(i)$ = mole fraction of the constituent i of the metal phase.

The activity coefficient of iron was assumed to be approximately 1. No data for the activity of Sr in iron were found so the activity coefficient was taken to be the same as that for Ca.

Results of the recalculation are shown in Table 13. Introduction of non-idealities into the model of the metal

TABLE 13. Results Obtained for Case B at 2000 K with a Non-ideal Metal Phase

SPECIES	MOLES	MOLE FRACTION	PRESSURE (ATMS)	FREE-ENERGY (CAL/MOLE)
METALS				
U metal	2.586e-08	2.388e-05		-44487
Zr metal	2.492e-02	2.287e-07		-33682
Fe metal	9.823e-04	9.818e-01		-38587
Mo metal	1.888e-02	9.188e-04		-24151
Sr metal	2.895e-08	1.823e-13		-43187
Co metal	1.543e-04	1.417e-09		-36758
Si metal	1.858e-04	9.842e-02		-23348
C graphite	9.413e-01	8.841e-04		-18998
OXIDES				
UO ₂ (c)	1.888e-05	4.353e-01		-319892
SiO ₂ (c)	2.296e-04	9.996e-02		-269347
CoO(c)	5.685e-03	2.475e-02		-188111
LaO _{1.5} (c)	2.888e-02	8.787e-04		-272569
CoO ₂ (c)	7.587e-01	3.268e-04		-321816
MoO ₂ (c)	6.966e-04	3.833e-09		-197168
FeO(c)	1.767e-03	7.692e-03		-123154
ZrO ₂ (c)	3.678e-03	1.681e-02		-315457
CoO _{1.5} (c)	9.249e-02	4.827e-03		-276886
CoSiO ₃	4.471e-04	1.955e-01		-474825
CoSi _{0.5} O ₂	4.389e-04	1.876e-01		-335418
SrSiO ₃	7.146e-01	3.111e-04		-491835
SrO(c)	1.679e-01	7.388e-07		-188475
SrSi _{0.5} O ₂	2.673e-01	1.164e-04		-347876
CoMoO ₄	1.188e-04	5.172e-18		-584532
SrMoO ₄	4.736e-18	2.862e-15		-496937
CoZrO ₃	6.328e-03	2.752e-02		-528428
SrZrO ₃	1.848e-02	7.175e-06		-529442
MoO ₃ (c)	1.157e-18	5.836e-24		-173496
GASES				
F ₂	9.953e-01		9.373e-01	-77358
H	1.663e-01		1.566e-03	-13137
OH	8.641e-05		6.253e-07	-93615
O	7.924e-08		7.463e-18	-28857
H ₂ O	3.882e-01		3.655e-03	-166894
U(g)	1.588e-11		1.488e-13	17358
UO	5.931e-07		5.586e-09	-136892
UO ₂ (g)	2.467e-04		2.323e-06	-271648
UO ₃	6.175e-05		5.815e-07	-377716
UOH	3.861e-11		2.883e-13	-135665
U(OH) ₂	1.358e-11		1.272e-13	-282821
UO ₂ (OH) ₂	1.883e-07		1.828e-09	-541692
Z ⁻ (g)	7.655e-16		7.289e-18	48145
ZrO	2.878e-18		1.958e-12	-113146
ZrO ₂ (g)	2.631e-18		2.477e-12	-225679
ZrOH	4.821e-14		4.548e-16	-118788
Zr(OH) ₂	5.357e-12		5.845e-14	-287818

TABLE 13. (continued)

SPECIES	MOLES	MOLE FRACTION	PRESSURE (ATMS)	FREE-ENERGY (CAL/MOLE)
Fe(g)	3.928e-02		3.691e-04	497
FeO(g)	2.756e-05		2.595e-08	-73079
FeOH	3.980e-00		3.673e-08	-113267
Fe(OH) ₂	1.158e-06		1.091e-08	-258830
Mo(g)	6.813e-11		5.662e-13	60117
MoO	2.324e-12		2.188e-14	-38535
MoO ₂ (g)	4.296e-11		4.045e-13	-161709
MoO ₃ (g)	6.380e-13		6.008e-15	-256559
MoO ₂ (OH) ₂	7.847e-14		7.390e-16	-437426
Co(g)	4.256e-04		4.008e-06	-41840
CoO(g)	1.428e-08		1.345e-10	-112470
Co(OH)	6.054e-05		5.701e-07	-184478
Co(OH) ₂	5.836e-06		5.496e-08	-325570
CoH	1.639e-06		1.543e-08	-58552
Co ₂	2.007e-12		1.890e-14	-56874
Sr(g)	8.718e-08		8.210e-10	-49903
SrO(g)	4.344e-11		4.090e-13	-131262
SrOH	1.336e-08		1.258e-10	-192637
Sr(OH) ₂	3.086e-10		2.906e-12	-328251
SrH	3.290e-10		3.098e-12	-66535
La(g)	2.270e-13		2.137e-15	1049
LaO(g)	7.968e-08		7.503e-10	-161279
LaOH	5.127e-12		4.828e-14	-161730
La(OH) ₂	5.425e-09		5.109e-11	-339797
La ₂ O	4.873e-20		4.589e-22	-182712
(LaO) ₂	1.907e-15		1.796e-17	-336321
Ce(g)	2.344e-12		2.208e-14	-5583
CeO	9.044e-07		8.517e-09	-168286
CeO ₂ (g)	2.096e-06		1.974e-08	-283208
CeOH	4.399e-11		4.143e-13	-167624
Ce(OH) ₂	1.747e-10		1.645e-12	-323492
(CeO) ₂	6.925e-13		6.522e-15	-354451
CO	5.868e-00		5.526e-02	-136472
CO ₂	5.051e-07		4.757e-05	-220003
Si(g)	1.420e-06		1.337e-08	16718
SiO	1.923e-01		1.811e-03	-141826
SiO ₂ (g)	2.681e-06		2.524e-06	-208370
SiOH	8.000e-08		8.287e-10	-122619
Si(OH) ₂	7.798e-08		7.343e-10	-272526
SiH	9.027e-07		9.254e-09	-20627
SiH ₄	2.935e-08		2.784e-10	-123097
Si ₂	4.956e-12		4.687e-14	11318
O ₂	1.339e-10		1.261e-12	-115270
Ce ₂ O	1.098e-22		1.034e-24	-153188
LaO ₂	1.442e-23		1.358e-25	-120000
Sr ₂ O	6.614e-21		6.229e-23	-174469
(SiO) ₂	2.575e-11		2.421e-13	-218392

phase enhanced reduction of silica to silicon and inhibited reduction of uranium and calcium. It also inhibited graphite formation in the melt. Figures-of-merit for the vaporization of strontium, cerium and lanthanum are:

	Non-ideal Metal Phase	Ideal-Metal Phase
F(Sr)	1×10^{-7}	1.4×10^{-6}
F(Ce)	3.0×10^{-6}	4.3×10^{-6}
F(La)	8.5×10^{-8}	3.2×10^{-7}

Clearly, introduction of metal phase activity coefficients different than one affected the vaporization of strontium. The depressed vaporization of Sr is the result of changes in the concentrations of SrO and Sr in the condensed phases and the increased oxygen potential - $P_{O_2} = 1.3 \times 10^{-12}$

atmospheres as opposed to $P_{O_2} = 4.7 \times 10^{-15}$ atmospheres

when activity coefficients were equal to 1. The effects of the non-ideal metal phase model on the vaporization of lanthanum and cerium are less pronounced since neither of these species contribute to the metal phase.

4.5 Effects of Uncertainty in the Properties of Gas Phase Species

The thermodynamic properties of many of the gas phase species of interest here are not well-known. To illustrate the effects of uncertainties in the thermodynamic properties on the calculations, case B at 2000 K was recalculated with properties of selected radionuclide vapor species varied toward more stable values within their recognized uncertainty ranges. To do this, the uncertainties in the free-energies of the vapor species were ascribed to the enthalpies of formation. The uncertainties in these enthalpies of formation were taken from the literature [17,18,19,20] to be:

$\Delta H_f(\text{Sr(g)})$	=	39200 ± 400
$\Delta H_f(\text{SrO(g)})$	=	-3200 ± 4000
$\Delta H_f(\text{SrOH(g)})$	=	-49120 ± 5000
$\Delta H_f(\text{Sr(OH)}_2(\text{g}))$	=	-142400 ± 10000
$\Delta H_f(\text{SrH(g)})$	=	52103 ± 5000
$\Delta H_f(\text{Sr}_2\text{O(g)})$	=	-50470 ± 5960
$\Delta H_f(\text{Ce(g)})$	=	99150 ± 600
$\Delta H_f(\text{CeO(g)})$	=	-29600 ± 4000
$\Delta H_f(\text{CeO}_2(\text{g}))$	=	-125180 ± 6000
$\Delta H_f(\text{Ce}_2\text{O(g)})$	=	-5960 ± 17900
$\Delta H_f((\text{CeO})_2(\text{g}))$	=	-153000 ± 15900

$\Delta H_f(\text{La(g)})$	=	103125 ± 994
$\Delta H_f(\text{LaO(g)})$	=	-27818 ± 2000
$\Delta H_f(\text{La}_2\text{O(g)})$	=	0 ± 13900
$\Delta H_f(\text{LaO}_2(\text{g}))$	=	-125200 ± 21900
$\Delta H_f((\text{LaO})_2(\text{g}))$	=	-133130 ± 17900

Results of the recalculation are shown in Table 14. Figures-of-merit for the vaporization of strontium, cerium and lanthanum are:

	Lower Bound Free-Energies	Best-Estimate Free-Energies
F(Sr)	1.6×10^{-6}	1.4×10^{-6}
F(Ce)	1.3×10^{-5}	4.3×10^{-6}
F(La)	5.2×10^{-7}	3.2×10^{-7}

The small uncertainties in the properties of strontium-bearing vapor species little affect the predicted vaporization of strontium. The larger uncertainties in the properties of cerium-bearing and lanthanum-bearing vapor species have more distinct effects on vaporization. The predicted vaporization of cerium may be uncertain by a factor of 3 because of uncertainties in the vapor properties.

It is unlikely that uncertainties in the thermodynamic properties of vapor species will soon be resolved. Consequently, the uncertainty in vaporization created by uncertainties in the vapor properties constitute a limit on the accuracy that can be obtained in thermodynamic calculations of core debris interacting with concrete. They also define a limit on the extent to which condensed phase models need to be refined.

5. CONCLUDING DISCUSSION

The analyses presented above have illustrated a few of the important topics that need to be considered in the thermochemical modeling of core debris interactions with concrete. An emphasis has been placed on modeling the condensed oxide phase. To obtain accurate estimates of radionuclide release and aerosol generation during core debris interactions with concrete, a far more systematic examination of this type of modeling needs to be conducted.

The need to develop models on the condensed phase of core debris cannot be abridged by experimentation alone. It is apparent from the calculations presented here that the properties of the condensed phases vary with both temperature and melt composition. Because of this, it is unlikely that so complete a data base on the thermochemical

TABLE 14. Results Obtained for Case B at 2000 K with Lower Bound Free - Energy Data for some Vapor Species.

SPECIES	MOLES	MOLE FRACTION	PRESSURE (ATMS)	FREE-ENERGY (CAL/MOLE)
METALS				
U metal	2.503e-01	2.280e-04		-44487
Zr metal	1.835e-01	1.672e-06		-33602
Fe metal	9.989e-04	9.182e-01		-30507
Mo metal	1.800e-02	9.112e-04		-24151
Sr metal	2.659e-04	2.423e-09		-43187
Co metal	1.067e-00	1.793e-05		-36758
Si metal	9.632e-03	8.776e-02		-23348
C graphite	9.981e-01	9.894e-04		-18917
OXIDES				
UO ₂ (c)	9.697e-04	4.372e-01		-319892
SiO ₂ (c)	2.355e-04	1.830e-01		-269347
CoO(c)	5.568e-03	2.435e-02		-188111
LaO _{1.5} (c)	2.800e-02	8.747e-04		-272569
CeO ₂ (c)	1.969e-01	8.611e-05		-321816
MoO ₂ (c)	2.589e-06	1.132e-11		-187168
FeO(l)	1.887e-02	4.754e-04		-123154
ZrO ₂ (c)	3.716e-03	1.629e-02		-315457
CeO _{1.5} (c)	9.823e-02	4.287e-03		-276006
CoSiO ₃	4.532e-04	1.882e-01		-474825
CoSi _{0.5} O ₂	4.283e-04	1.873e-01		-335410
Sr-SiO ₃	7.176e-01	3.139e-04		-491035
Sr-O(c)	1.636e-01	7.156e-07		-188475
Sr-Si _{0.5} O ₂	2.644e-01	1.157e-04		-347876
CoMoO ₄	2.661e-08	1.164e-13		-584532
SrMoO ₄	1.855e-13	4.616e-19		-496937
CoZrO ₃	6.282e-03	2.748e-02		-520420
SrZrO ₃	1.630e-00	7.131e-05		-529442
MoO ₃ (n)	2.633e-22	1.151e-27		-173496
GASES				
H ₂	9.989e-01		9.654e-01	-77358
H	1.645e-01		1.589e-03	-13137
OH	4.821e-06		3.886e-08	-93615
O	4.728e-09		4.569e-11	-28057
H ₂ O	1.385e-02		2.385e-04	-166894
U(g)	4.125e-09		3.987e-11	17350
UO	9.481e-06		9.163e-08	-136092
UO ₂ (g)	2.414e-04		2.333e-06	-271640
UO ₃	3.780e-06		3.576e-08	-377716
UOH	4.966e-10		4.799e-12	-135865
U(OH) ₂	1.361e-11		1.316e-13	-282801
UO ₂ (OH) ₂	4.895e-10		3.957e-12	-541692
Zr(g)	2.819e-13		1.952e-15	48145
ZrO	3.344e-09		3.232e-11	-113146
ZrO ₂ (g)	2.682e-10		2.515e-12	-225679
ZrOH	7.983e-13		7.637e-15	-118708
Zr(OH) ₂	5.457e-12		5.274e-14	-287818

TABLE 14. (continued)

SPECIES	MOLES	MOLE FRACTION	PRESSURE (ATMS)	FREE-ENERGY (CAL/MOLE)
Fe(g)	3.855e-02		3.726e-04	497
FeO(g)	1.660e-07		1.684e-09	-73879
FeOH	2.384e-07		2.384e-09	-113267
Fe(OH) ₂	4.399e-09		4.251e-11	-258832
Mo(g)	5.835e-11		5.639e-13	60117
MoO	1.381e-13		1.335e-15	-38535
MoO ₂ (g)	1.563e-13		1.511e-15	-161709
MoO ₃ (g)	1.421e-16		1.374e-18	-256559
MoO ₂ (OH) ₂	1.183e-18		1.866e-20	-437426
Co(g)	6.663e-03		6.448e-05	-41842
CoO(g)	1.369e-08		1.323e-10	-112476
Co(OH)	5.892e-05		5.693e-07	-184478
Co(OH) ₂	3.529e-07		3.418e-09	-325572
CoH	2.624e-05		2.516e-07	-58552
Co ₂	5.849e-18		4.882e-12	-56874
Sr(g)	1.522e-06		1.452e-08	-52303
SrO(g)	1.134e-10		1.096e-12	-135262
SrOH	4.552e-08		4.399e-10	-197837
Sr(OH) ₂	2.299e-10		2.222e-12	-328251
SrH	1.831e-08		1.769e-10	-71535
La(g)	1.883e-11		1.828e-13	55
LaO(g)	5.197e-07		5.022e-09	-163266
LaOH	2.858e-11		1.989e-13	-161732
La(OH) ₂	1.354e-09		1.308e-11	-339797
La ₂ O	4.232e-16		4.092e-18	-196621
(LaO) ₂	2.756e-12		2.664e-14	-354284
Ce(g)	1.865e-12		1.822e-12	-6179
CeO	1.831e-05		9.962e-08	-172268
CeO ₂ (g)	2.412e-06		2.331e-08	-289169
CeOH	1.872e-10		1.809e-12	-167624
Ce(OH) ₂	4.619e-11		4.464e-13	-323492
(CeO) ₂	6.819e-10		6.591e-12	-378347
CO	1.928e-01		1.836e-03	-136472
CO ₂	1.821e-05		9.675e-08	-228223
Si(g)	3.821e-04		3.674e-06	16718
SiO	3.153e-02		3.048e-02	-141826
SiO ₂ (g)	2.691e-06		2.621e-08	-208972
SiOH	1.464e-06		1.415e-08	-122619
Si(OH) ₂	8.864e-08		7.793e-10	-272526
SiH	2.671e-04		2.581e-06	-28627
SiH ₄	8.339e-06		8.059e-08	-123297
Si ₂	3.647e-07		3.525e-09	11318
O ₂	4.893e-13		4.729e-15	-114278
Ce ₂ O	2.918e-18		2.813e-20	-171871
La ₂ O ₂	8.546e-22		8.259e-24	-158657
Sr ₂ O	4.523e-19		4.371e-21	-188438
(SiO) ₂	7.183e-09		6.865e-11	-218392

properties of the condensed phases can be developed that analytic models will not be needed to interpolate and extrapolate the data base.

It should, however, be remembered that uncertainties in the thermodynamic properties of vapor species define a limit on the refinement of condensed phase models. It should also be remembered that modeling of the condensed oxide phase is not the only technical issue of thermochemical analyses of core debris interactions with concrete. Other topics that can be just as important include:

1. Completeness of the gas phase speciation including the importance of vapor phase hydrides, mixed oxides and intermetallics.
2. Non-ideality of the metal phase
3. Importance of halide and sulphide-bearing gases evolving from the concrete.

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13. ABSTRACT (200 words or less) <p>A submission for the CSNI/PWG-4/GREST standard problem on chemical thermodynamic modeling in core-concrete interaction releases of radionuclides is described. Part A of the exercise is a highly defined benchmark calculation in which data and speciation are specified. The problem is, however, ambiguous concerning the definition of an ideal solution. Consequently, two solutions are provided. In one solution, specified species are treated as molecular entities to define the ideal solution. In the second, mixing is assumed to occur ideally on cationic and anionic lattices. The different results obtained in these calculations illustrate the importance of condensed phase modeling in the analyses of high temperature melt interactions with concrete.</p> <p>Part B of the exercise consists of six problems in which the temperatures, pressures and bulk compositions of the melts are specified. Data and speciation are to be supplied as parts of the solutions to the problems. Results of calculations for these six problems are presented. Additional solutions are provided to illustrate the effects of speciation in the condensed oxide phase, non-ideality in the condensed metal phase and uncertainty in the thermodynamic properties of gas phase species.</p>		
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