

Equation of State of UO_2 ¹

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An international project supported by INTAS (International Association for Promotion of Cooperation with Scientists from the New Independent States of the former Soviet Union) was started in 1994 with the intent of constructing an equation of state (EOS) for liquid and gaseous UO_2 , which fully reproduces the comprehensive thermodynamic database for this compound. The new equation of state was devised for applications encompassing hypo- and hyper-stoichiometric compositions. A so-called "chemical model" was used for the theoretical description of liquid urania. This model is based on the thermodynamic perturbation theory (TPT) modified in order to account for the specific properties of the system investigated. It describes, in a unified formalism, a *multicomponent mixture* of chemically reactive, strongly interacting neutral and charged molecules and atoms. Comparisons of the predicted equilibrium vapor pressures with literature data provided an initial validation of the model up to temperatures of the order of 5500 K. A further, positive result is the fairly good agreement of the predicted heat capacity with experimental values, which extend up to 8000 K. A characteristic feature of non-congruent vaporization in $\text{UO}_{2\pm x}$ is the production of a very high maximum vapor pressure ($P_{\text{max}} \sim 1$ GPa) as well as a substantial oxygen enrichment of the vapor phase over boiling UO_2 ($(\text{O}/\text{U})_{\text{max}} \sim 7$). The critical point of a *truly non-congruent* phase transition in UO_2 was also calculated. This point *essentially differs* from that defined for a gas-liquid phase transition in *simple liquids*; in particular, the equation $(\partial P/\partial V)_c \sim (P/V) \neq 0$ applies here. The predicted critical parameters are: $T_c \approx 10120$ K, $P_c \approx 965$ MPa, $\rho_c \approx 2.61$ g · cm⁻³.

KEY WORDS: equation of state; heat capacity; high temperatures; non-congruent evaporation; thermodynamic perturbation theory; vapor pressure, uranium dioxide.

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1. INTRODUCTION

The equation of state EOS of fluid UO_2 was first needed in the analysis of hypothetical accidents in nuclear fast reactors, where temperatures are expected at which the vapor pressure of the fuel is sufficiently high to produce liquid mass displacements. In the last decade, however, increasing attention was directed on the effects of the atmospheric environment on the vaporization rate of the molten fuel. On the other hand, empirical thermodynamic properties of uranium at high temperatures are presently restricted to the near-stoichiometric or congruent-vaporization compositions. A new EOS was, therefore, devised for applications encompassing hypo- and hyperstoichiometric compositions.

The main problem in the construction of a physically plausible EOS of UO_2 is the complexity of both its liquid and vapor structure, i.e., a presumably highly ionized ionic character of the condensed state, and a weakly ionized molecular character of the gaseous state. Solid UO_2 is usually described as a pure ionic mixture of U^{+n} and O^{-2} , and the vapor as a neutral molecular mixture; the structure of the liquid is probably somewhere in between.

2. GENERAL APPROACH AND MODEL

A so-called "chemical" (quasi-molecular) description of *dense liquid* UO_2 was applied, based on the demonstrated formal correspondence of the partition function of ionic liquids to that of liquids formed of virtual molecular clusters (Stillinger-Reiss theorem [1-3]). In such a model, detailed information is needed to describe the contributions of the internal degrees of freedom of complex (bounded) particles, e.g., energies of ionization, dissociation, excitation energy levels, etc. On the other hand, however, the advantage is that the renormalized "effective" interaction of free (unbounded) particles is relatively weak, and, furthermore, arbitrary stages of ionization and different structures of complex particles can be composed in the frame of a unified calculation procedure. In principle, once a proper definition of the effective interaction of free particles is achieved, and the bound states are properly restricted, the model can continually describe the wide variety of states ranging from the presumably highly ionized liquid at very high temperatures, to the "cold" vapor represented by a neutral molecular mixture.

Along with "free" UO_2 a number of "clusters," like U_2O_4 , U_2O_2 , U_2O_3 , U_2O_5 , etc. as well as free atoms and molecules of uranium and oxygen (U , U_2 , O , O_2), and ions (UO_2^+ , UO^+ , U^+ , UO_3^- , UO_2^- , O^-) were included in the picture as distinct species with their own partition

functions, and parameters of repulsion (intrinsic volumes) and short-range attraction.

This model can be considered as sufficiently realistic for dense vapors and dense, expanded liquids in the high-temperature region of phase coexistence. The liquid phase is essentially described as a set of conventionally distinguishable clusters (mainly with one uranium in the center and several oxygen constituents), which are allowed to freely exchange oxygens and/or electrons.

In a chemical model, the calculation of the equilibrium composition and of the thermodynamic functions starts from the definition of the Helmholtz free energy $F = F(V, T, N_j)$, and a set of arbitrary (non-equilibrium) numbers of particles of all species, $\{N_j\}$. The free energy $F(T, V, \{N_j\})$ is the sum of three contributions: the ideal-gas part, $\Delta F^{(\text{id})}$, the interactions of neutral and charged-neutral particles, ΔF_n , and of charged-charged particles, ΔF_c , i.e.,

$$F(T, V, \{N_j\}) = \Delta F^{(\text{id})} + \Delta F_n + \Delta F_c \quad (1)$$

The set of equilibrium concentrations $n_i = N_i/V$ ($n = \sum n_i$) corresponds to the conditioned minimum of the free energy, Eq. (1), with respect to the variation of N_i at constant temperature, T , and volume, V , under the constraint of:

- (a) Conservation of mass: $\sum n_i m_i = \text{Const} \equiv \rho$
- (b) Chemical proportion: $(\sum n_i \nu_{ai}) / (\sum n_j \nu_{bj}) = \text{Const}$
- (c) Electro-neutrality: $\sum n_i Z_i^{(+)} = \sum n_i Z_i^{(-)}$

The general conditions for the free-energy minimum are equivalent to a system of equations for the chemical potentials, or may also be written in the form of a mass-action law. When explicit expressions for the free energy, Eq. (1), are known, the minimization becomes a mere numerical problem.

After the equilibrium composition $\{n_j\}$ is evaluated, all thermodynamic functions can also be deduced from the thermal and caloric equations of state, $P = P(T, \{n_i\})$ and $U = U(T, \{n_i\})$, which are self-consistently defined as the first derivatives of the free energy. Actually, it is well known that, in spite of the additional, implicit dependence of $F(T, V, \{N_j\})$ on T and V through the *variable* equilibrium composition, its first *full* derivatives, i.e., pressure, entropy and chemical potentials, are equal to the corresponding *partial* derivatives of the free energy at fixed composition $\{n_i\}$ at the minimum of $F(T, V, \{N_j\})$. However, where the *second* derivatives are concerned (e.g., for heat capacities, compressibility, etc.), this additional dependence of the free energy $F(T, V, \{N_j\})$ must be taken into account *explicitly*.

Therefore, these quantities were calculated via a direct numerical differentiation using a program package, developed by Gryaznov [4].

The contribution of neutral and charged-neutral interactions to the free energy, ΔF_n , was formulated with the thermodynamic perturbation theory (TPT). Application of TPT to a particular system, first necessitates splitting of the microscopic energy of the system into the energy of some "unperturbed," or "reference" system (usually consisting of fully repulsive interactions) and an additional, attractive contribution—in some sense small—representing the "perturbation" energy. The structure and the thermodynamic properties of the "unperturbed" system must be completely defined.

The original approach of Barker and Henderson [5] and Verlet and Weiss [6] was adopted in a modified form, which accounts for the specific properties of the system studied; this model describes, in a unified formalism, a *multicomponent mixture* of chemically reactive, strongly interacting neutral and charged molecules and atoms.

The "hard-spheres mixture" approximation [7] was used for the contributions of the "reference" repulsive interactions of all (charged and neutral) species to the free energy. A simplified version of Verlet and Weiss [6] expressions was then assumed for the first-order TPT short-range attractive contributions to ΔF_n , within the so-called "one-fluid approximation" for mixtures:

$$\Delta F_n = \Delta F_{\text{HS}}^{(0)}(\eta) - \frac{16}{9} \pi n \varepsilon_{\text{MIX}} \sigma_{\text{MIX}}^3 \left[\left(\frac{\sigma_{\text{MIX}}}{d(T)} \right)^{12} M_{12}(\eta, T) - \left(\frac{\sigma_{\text{MIX}}}{d(T)} \right)^6 M_6(\eta, T^*) \right] \quad (2)$$

Here M_{12} and M_6 are moments of the hard-sphere radial distribution function; $T^* = kT/\varepsilon_{\text{MIX}}$ is the reduced temperature; and $\eta = 1/6\pi N/V[d(T)]^3$ is the packing fraction. Approximate expressions for M_{12} and M_6 were used as proposed in Ref. 6 for a Lennard–Jones (12–6) potential and conventional combination and mixing rules [3].

Using a central-symmetrical Lennard–Jones (12–6) potential for the description of interactions between uranium dioxide molecules (as well as for other components of the liquid mixture) might appear as a too drastic simplification, since most molecules (or ion clusters) do not have a centrosymmetrical structure; hence, a significant anisotropy in the repulsive interactions must be expected [3]. Consequently, the parameters ε and σ of the Lennard–Jones (12–6) potential should be considered as effective quantities, i.e., as appropriately averaged over all orientations. In this respect, also the definition of the effective molecular diameter $d(T)$ in Eq. (2) needs a more detailed verification. Therefore, all the used parameters of the Lennard–Jones

(12-6) potential, $\langle \sigma \rangle$, $\langle \varepsilon \rangle$, and $d(T)$, were analyzed and expressed through "central components," σ_0 , ε_0 , and $d_0(T)$ and a dimensionless anisotropy parameter Q equal to the ratio of the mean-squared value of the anisotropic contribution to the corresponding isotropic repulsive contribution:

$$d(T) = \frac{d_0(T)}{1 + (\sigma_0/d_0)(\varepsilon_0 Q^2/3kT)} \quad (3)$$

$$\langle \sigma \rangle = \sigma_0 \left(1 - \frac{nQ^2}{36} \right); \quad \langle \varepsilon \rangle = \varepsilon_0 \left(\frac{\langle \sigma \rangle}{\sigma_0} \right)^6$$

Analysis of Eq. (3) shows that, compared to a pre-averaged isotropic potential, the anisotropy of the molecular interaction causes a notable *decrease* of the effective molecular diameter $\langle \sigma \rangle$. The effective diameter, $d(T)$, decreases even more markedly than $\langle \sigma \rangle$. Finally, when anisotropy of the repulsive interactions is taken into account, the ratio $\langle \sigma \rangle/d(T)$ *increases* with respect to the case of central symmetry. Consequently, the effective potential well depth $\langle \varepsilon \rangle$, proportional to the sixth power of $\langle \sigma \rangle/\sigma_0$, becomes much more pronounced.

We calculated the values of the parameters, ε_0 , σ_0 , and Q , from the known bond lengths and atomic radii of the UO_2 molecule and its heat of vaporization [3]. A value of $Q \sim 0.66$ was obtained, close to those deduced from experimental data which reproduce the density, Gibbs energy, and compressibility at the melting point, i.e., the three most sensitive properties of liquid $\text{UO}_{2.00}$, when Coulombic contributions are not taken into account.

The non-ideality correction, ΔF_c , due to Coulomb long-range ionic interactions has a one-parametric form, and consists of a superposition of MSA (mean spherical approximation) + DHSA (Debye-Hückel for charged sphere approximation) [8, 9] combined with the "one-fluid" approximation for the case of an ionic mixture with different diameters [10]. In the reported EOS calculations the diameters of all ionic species were assumed to be equal to the diameters of their neutral counterparts.

$$\frac{\Delta F_c}{VkT} \equiv \left(\frac{\Delta F_c}{VkT} \right)_{\text{DHLL}} \theta(x, v) \cong \left(\frac{\Delta F_c}{VkT} \right)_{\text{DHLL}} \theta(x)$$

$$x \equiv \langle \sigma \rangle / r_D, \quad r_D^2 \equiv \frac{4\pi e^2}{VkT} \sum_{\alpha=1}^2 N_\alpha Z_\alpha^2 \quad (4)$$

$$\theta(x) = (3\alpha/x^3) [\text{Ln}(1+x) - x + x^2] + (1-a) \{ [2(1+2x)^{3/2} - 3x^2 - 6x - 2] / x^3 \} \quad (5)$$

$$\langle \sigma \rangle \equiv \left[\left(\sum n_i \sigma_i^3 \right) / \left(\sum n_i \right) \right]^{1/3} \quad (6)$$

Table I. Selected Properties of the Coexisting Phases of $\text{UO}_{2,00}$ along the Boiling Line

| | T (K) | 3120 | 4000 | 5000 | 6000 | 7000 | 8000 |
|--|---------|---------|---------|---------|---------|---------|---------|
| Pressure (MPa) | | 0.00398 | 0.147 | 2.33 | 20.2 | 112 | 388 |
| Density ($\text{g} \cdot \text{cm}^{-3}$) | Liquid | 8.8710 | 8.1257 | 7.3536 | 6.5333 | 5.6797 | 4.8761 |
| | Vapor | 0.00005 | 0.00105 | 0.00956 | 0.0439 | 0.1535 | 0.4718 |
| H ($\text{kJ} \cdot \text{kg}^{-1}$) | Liquid | -6406.1 | -6005.0 | -5628.8 | -5238.0 | -4779.4 | -4158.0 |
| | Vapor | -5394.6 | -4962.9 | -4339.0 | -3454.6 | -2519.8 | -1935.9 |
| S ($\text{kJ} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$) | Liquid | 1.1620 | 1.2759 | 1.3599 | 1.4305 | 1.4987 | 1.5743 |
| | Vapor | 1.7776 | 1.8288 | 2.1460 | 2.7825 | 3.3774 | 3.2838 |
| C_p ($\text{kJ} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$) | Liquid | 0.5112 | 0.3992 | 0.3718 | 0.4161 | 0.5242 | 0.8517 |
| | Vapor | 0.8295 | 1.5394 | 2.2706 | 2.5293 | 2.5331 | 2.2507 |
| β_T (10^{-10} Pa^{-1}) | Liquid | 0.395 | 0.529 | 0.757 | 1.17 | 1.97 | 3.30 |
| | Vapor | 25.95 | 0.7468 | 0.0503 | 0.0059 | 0.0011 | 0.0003 |
| α_T (10^{-4} K^{-1}) | Liquid | 1.09 | 0.95 | 1.08 | 1.34 | 1.81 | 2.83 |
| | Vapor | 5.58 | 7.10 | 7.01 | 5.24 | 4.05 | 3.50 |
| O/U | Liquid | 2.0 | 2.0 | 2.0 | 2.0 | 2.0 | 2.0 |
| | Vapor | 2.42 | 2.50 | 3.07 | 4.57 | 6.52 | 6.36 |
| Ionization (%) | Liquid | 9.5 | 13.0 | 17.2 | 21.7 | 26.3 | 29.2 |
| | Vapor | 3.8 | 6.7 | 9.7 | 5.1 | 8.6 | 13.5 |

Here $(\Delta F_c/VkT)_{\text{DHLL}} \equiv -(12\pi r_D^3)^{-1}$ is the reduced Coulomb free energy within Debye–Hückel limiting law (DHLL) [10].

It was assumed that charged species of the *same* sign are *not* submitted to the short-range Van der Waals attraction, on account of the strong ordering of the positive and negative charges produced by Coulomb's repulsion. Actually, significant ionization takes place both in the vapor and in the liquid phases of UO_2 due to this strong interaction.

3. MODEL VALIDATION

The model was calibrated with a two-stage procedure. The interaction parameters were first evaluated theoretically, and then adjusted to closely reproduce selected properties of molten UO_2 at the melting point. Properties were chosen, which are sufficiently sensitive to variation of the model parameters and for which experimental measurements are adequately accurate.

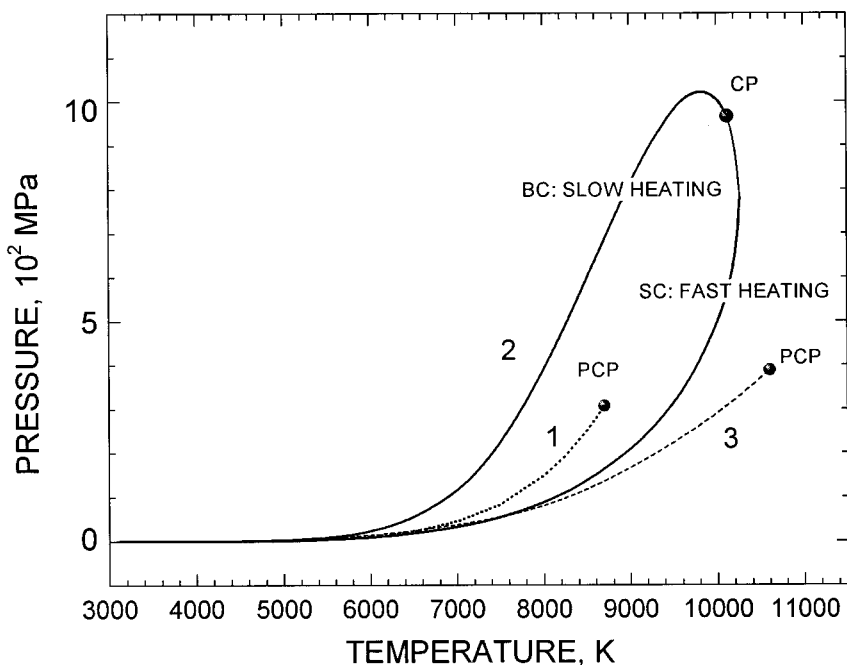


Fig. 1. Vapor–liquid phase equilibrium in stoichiometric uranium dioxide. (1) Gas–liquid coexistence in PCE-mode (BC=SC) with pseudocritical point $\{“\text{PCP}”-(\partial P/\partial V)_T=0; (\partial^2 P/\partial V^2)_T=0; (\partial^3 P/\partial V^3)_T=0\}$. (2) Boiling (BC) and saturation (SC) curves in total equilibrium (EOS “INTAS-99”) [14]. (3) Total vapor pressure calculated by Fischer [13] with pseudocritical point (PCP).

The microscopic parameters for each species were first deduced from independent data sources [3]. A sensitivity study made it possible to select those species whose parameters are dominant (UO_2) or subdominant (UO , UO_3) in defining the properties of liquid UO_2 . The relative values of all other parameters were then fixed with respect to those of the UO_2 free molecule. The adjustment of the dominant and subdominant parameters was made on the basis of properties of stoichiometric liquid $\text{UO}_{2.00}$, i.e., density, Gibbs free energy, heat capacity, compressibility, and oxygen potential at $T_m = 3120$ K. No other properties have been fitted at higher temperatures. Hence, the behavior of the EOS in the broad temperature range investigated is a property inherent in the physical fundamentals of the model. The calculated values of the thermodynamic functions are presented in Table I and compared with other predictions and measurements in Figs. 1 to 3.

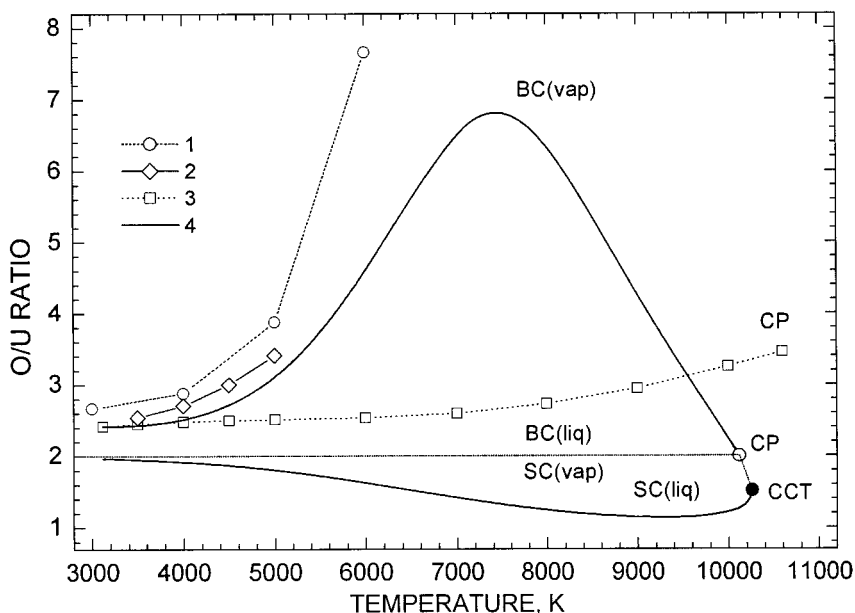


Fig. 2. U/O ratio of coexisting phases on the boiling and saturation curves. (1) Open circles: calculations of Green and Leibovitz [14]. (2) Open diamonds: O/U ratio from the calculated composition of Ohse et al. [15]. (3) Squares: O/U ratio (total) reconstructed from calculations of Fischer [13]. (4) Solid line: present work. BC, boiling curve; SC, saturation curve; CP, true critical point; PCP, pseudo-critical point in Fischer's calculations; CCT, point of maximum temperature on SC (*cricondentherm*).

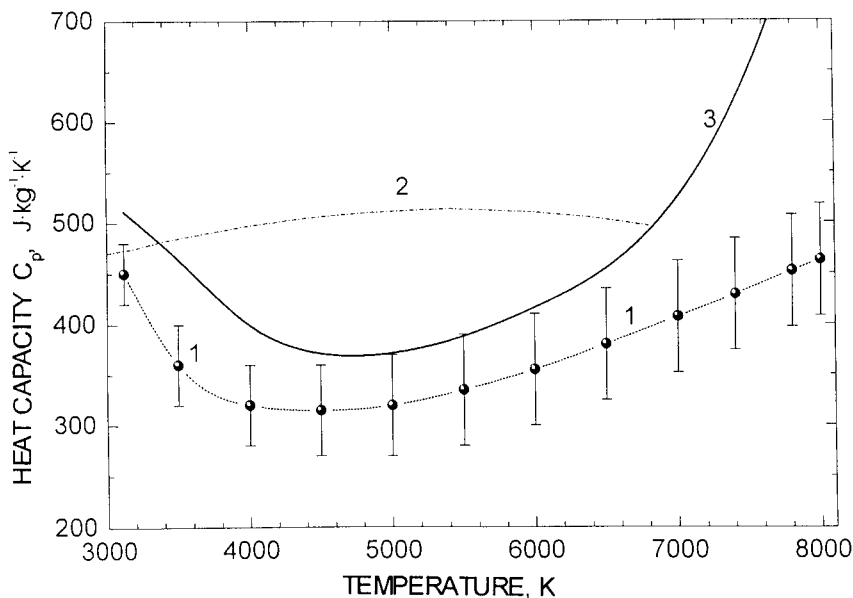


Fig. 3. Heat capacity of liquid uranium dioxide. Comparison of predictions and measurements [16]. (1) Measurements of Ronchi et al. [16]. (2) C_p calculated by Fischer [13]. (3) Present calculation.

4. RESULTS AND DISCUSSION

Comparisons of the predicted equilibrium vapor pressure with literature data provided a first validation of the model up to temperatures of the order of 5500 K. A relevant result is the unusual structure of the gas-liquid phase transition boundary, which exhibits a *striking difference* with respect to that of ordinary liquids. This feature is caused by the *non-congruency* of the coexisting phases in $\text{UO}_{2\pm x}$. Both the total vapor pressure and the degree of oxygen enrichment of the vapor phase strongly depend on the evaporation regime. Two distinct pressure-temperature functions $P=P(T)$ were, therefore, identified and calculated corresponding to the two limiting regimes of: (a) *slow* evaporation under “global equilibrium conditions,” corresponding to the *boiling curve* and (b) *fast*, nonequilibrium evaporation, called “forced-congruent mode” (FCM), corresponding to *saturation conditions* (SC).

In Fig. 1 curve (1) obtained by enforcing a partially-congruent phase equilibrium—PCE mode, with $(\text{U}/\text{O})_{\text{liq}} = (\text{U}/\text{O})_{\text{vap}}$ —is compared with those where noncongruency is governing evaporation (BC and SC curves 2). The predictions of Fisher [13] (curve 3) are also plotted in the same graph. In

contrast with nonreacting one-component fluids, uranium dioxide exhibits a coexistence curve and critical region of essentially different forms. The heat of vaporization is a nonmonotonic function of temperature, and the U/O ratio in the vapor phase increases markedly with temperature (Fig. 2).

The reliability of the model is corroborated by the fairly good agreement of the predicted heat capacity with the experimental values of Ronchi et al. [16], which extend up to 8000 K (Fig. 3). In fact, it should be noted, so far, none of the existing liquid UO_2 models was able to reproduce correctly the observed temperature dependence of C_p .

A characteristic feature of noncongruent vaporization in $\text{UO}_{2\pm x}$ is the production of a comparatively high maximal vapor pressure ($P_{\max} \sim 1$ GPa), as well as a substantial oxygen enrichment, $(\text{O/U})_{\max} \sim 7$, of the vapor phase over the boiling liquid, as shown in Fig. 1.

The critical point of the *truly noncongruent* phase transition in UO_2 was also calculated. This point *essentially differs* from that defined for a gas-liquid phase transition in *simple liquids*; in particular, we have here: $(\partial P/\partial V)_c \sim (P/V) \neq 0$.

The predicted critical parameters are: $T_c \approx 10120$ K, $P_c \approx 965$ MPa, and $\rho_c \approx 2.61$ g · cm⁻³ [11].

Details on the results of calculations performed in the high-pressure and high-temperature range $\{P \leq 10$ GPa, $T \leq 20000$ K $\}$ will be published separately.

5. CONCLUSIONS

The new EOS essentially differs from all previously developed models. The structure of the equation, as well as the broad physical and chemical conditions covered, makes it particularly suitable for applications in nuclear reactor safety analysis. Effective computational algorithms enable numerical calculation of combined phases, as well as chemical and ionization equilibrium in the liquid, described as a strongly interacting uranium-oxygen system. These can be successfully applied to analyze situations involving strongly noncongruent evaporation.

The complete phase diagram of condensed and gaseous $\text{UO}_{2\pm x}$ up to the *true critical point* has been constructed. For the first time this diagram correctly reproduces the *noncongruent bivariant* nature of the evaporation in the uranium-oxygen system.

The present calculations have the following limitations:

- The excitation partition functions of the complex particles were assumed to be equal to their *ideal-gas* counterparts taken from Glushko's Handbook [12] and from the recommendations of

IVTAN-1996 [3], whereby the value of the reduced thermodynamic potential, $\Phi^0(T) \equiv G(T)/T$ was used for this extraction. No cut-off procedure and no perturbation of internally bound excited states of complex particles under dense-liquid surrounding were taken into account.

- Only single-stage ionization is included. Therefore, the main charged species in the present model are molecular ions, first of all UO_3^- , UO_2^+ , and UO^+ . However, sensitivity studies show that the presence of other ionic species has little influence on the results.
- Finally, it should be emphasized that first-order TPT does not allow a detailed description of the near-critical region, on account of the neglect of fluctuations, which become important in the closest vicinity of the critical point.

Despite these limitations, the present EOS provides the means for a more realistic description of the interplay between thermodynamic aspects and hydrodynamic and diffusive processes occurring during severe reactor accidents.

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