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Standard Enthalpies of Formation of Gaseous Thorium, Uranium, and Plutonium Oxides

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Recently available spectroscopic data have been used to derive new values for the enthalpies of formation at 298.15 K of gaseous ThO, ThO_2, UO, UO_2, UO_3, and PuO, and PuO_2. These new values of $\Delta H_f^0(298.15\ \mathrm{K})$ are in good agreement with previously recommended values for UO, UO_2, and PuO, but not for the other molecules. Inconsistencies among evaluated thermodynamic data have been resolved for ThO (g) and UO_3 (g). Recommended values are derived for PuO_2 (g) and ThO_2 (g); however, additional experimental work on these molecules is needed.

KEY WORDS: thorium; uranium; plutonium; enthalpies of formation.

1. INTRODUCTION

A common application of tabulated thermodynamic data is the calculation of vapor pressure as a function of temperature. Such calculations require data for both the gaseous and the condensed-phase species. Generally, vapor pressures are calculated from tabulations of free energies of formation, ΔG_f^0, or from tabulations of free-energy functions, $\mathrm{fef}' \equiv -[G^0(T) - H^0(298.15\ \mathrm{K})]/T$, and the enthalpies of formation, $\Delta H_f^0(298.15\ \mathrm{K})$. The approach that uses the free-energy function is particularly useful in case for which spectroscopic data ware available, because the partition function, Q, is directly related to the molecular energy levels, and the fef $\{\equiv -[G^0(T) - H^0(0\ \mathrm{K})]/T\}$ is calculated from Q:

$$\mathrm{fef} = R \ln Q \tag{1}$$

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Table I. Summary of the Results of Consistency Checks between Spectroscopic and Thermodynamic Data for Gaseous Thorium, Uranium, and Plutonium Oxides[a]

Consistent			
UO_2:	$p\,(UO_2)$	and	$\Delta H^\circ_{sub}\,(UO_2, 298.15\ K)$
UO:	$\Delta G^\circ_f\,(UO, g)$	and	$\Delta H^\circ_f\,(UO, g, 298.15\ K)$
UO_3:	$\Delta G^\circ_f\,(UO_3, g)$	and	new $\Delta H^\circ_f\,(UO_3, g, 0\ K)$
PuO:	$\Delta G^\circ_f\,(PuO, g)$	and	$\Delta H^\circ_f\,(PuO, g, 298.15\ K)$
Inconsistent			
UO_3:	$\Delta G^\circ_f\,(UO_3, g)$	and	$D^\circ\,(UO_3, g, 0\ K)$[b]
PuO_2:	$\Delta G^\circ_f\,(PuO_2, g)$	and	$\Delta H^\circ_f\,(PuO_2, g, 298.15\ K)$
ThO:	$\Delta G^\circ_f\,(ThO, g)$	and	$\Delta H^\circ_f\,(ThO, g, 298.15\ K)$
ThO_2:	$p\,(ThO_2)$	and	$\Delta H^\circ_{sub}\,(ThO_2, 298.15\ K)$

[a] Results from [1].

[b] Atomization energy at 0 K.

The user of tabulated thermodynamic data would like to be sure that these data are internally consistent and include evaluations of all relevant data. For the vapor species of thorium, uranium, and plutonium oxide, several high-temperature techniques have been applied (e.g., mass spectrometry, mass effusion, transpiration) to measure the quantities (e.g., ion current or mass deposited) that are proportional to pressure. Assessments of the results from different studies have been done for the oxide molecules of interest and "recommended" values of the thermodynamic functions are available.

Spectroscopic information has become available recently for several of the oxides of thorium, uranium, and plutonium; these spectroscopic data have been used [1] with the methods of statistical mechanics to provide an independent check on the consistency of thermodynamic functions. The results of checks for consistency for gaseous ThO, ThO_2, UO, UO_2, UO_3, PuO, and PuO_2 are summarized in Table I. The classification of the data as "consistent" or "inconsistent" depends on the uncertainties in the data, which differ greatly for these molecules. For molecules with inconsistent data, the recommended values of $\Delta H_f^0(298.15\ K)$ are not reliable and new values are required. Even for those molecules with "consistent" data, a uniform approach to the derivation of $\Delta H_f^0(298.15\ K)$ seems desirable; we therefore derive here a new value of $\Delta H_f^0(298.15\ K)$ for each of the oxides of thorium, uranium, and plutonium.

2. METHODS

2.1. Free Energy of Formation

For each of the thorium, uranium, and plutonium oxide vapor species, the values of ΔG_f^0 as a function of T have been to an equation of the form

$$\Delta G_f^0 = a + bT \tag{2}$$

where $a \approx \Delta H_f^0$ and $b \approx -\Delta S_f^0$ over a small temperature range. In all cases, the ΔG_f^0 is from data that have been evaluated, i.e., more than one set of measured data is available and weighted averages have been taken, and no attempt has been made to reevaluate data (except for the case of gaseous ThO, which is discussed later). The values of ΔH_f^0(298.15 K) that are derived here cannot be accurate unless reliable expressions for ΔG_f^0 are available, and for some molecules, the calculations strongly suggest inaccuracies in ΔG_f^0.

If we use the sublimation of UO_2 as an example, the physically measured quantity (i.e., ion current or mass deposited) is converted to a $p(UO_2)$ value by an appropriate method, which may involve calibration of the experimental apparatus as well as making certain assumptions (e.g., that UO_2 is the predominant vapor species). In determining pressures by any process, there are potential sources of error that may depend on the temperature range, the particular apparatus, the experimental method, and the particular chemical system under study. Comparison of the results obtained by different methods is very useful, but does not always ensure a reliable pressure equation.

If we have determined $p(UO_2)$ at a measured T, then we can obtain $\Delta G_f^0(UO_2, g)$ from

$$\begin{aligned}\Delta G_{\text{sub}}{}^0 &= -RT \ln p(UO_2) \\ &= \Delta G_f^0(UO_2, g) - \Delta G_f^0(UO_2, s)\end{aligned} \tag{3}$$

Cumulative sources of error in $\Delta G_f^0(UO_2, g)$, or any other ΔG_f^0, can often be assessed by comparing the data from different experimental methods. The evaluated ΔG_f^0 equations are given in the second column of Table II. The ΔG_f^0(ThO, g) equation recommended by Rand [8] is inconsistent with a more recent equation by Ackermann and Tetenbaum [7]. The reason for this difference has not been determined, but the Ackermann and Tetenbaum equation for ΔG_f^0(ThO, g) is consistent with spectroscopic data, whereas the Rand equation is not [1]. Thus we prefer the ΔG_f^0(ThO, g) equation of Ackermann and Tetenbaum.

The primary purpose of this paper is not to evaluate ΔG_f^0 equations, and we shall generally accept the recommended equations. However, in the process of calculating ΔH_f^0(298.15 K) values, the accuracy of the recommended ΔG_f^0 equations is tested. Thus, some conclusions can be reached about the reliability of ΔG_f for gaseous ThO, ThO_2, UO, UO_2, UO_3, PuO, and PuO_2.

Table II. Free Energies of Formation and Enthalpies of Formation at 298.15 K for Gaseous Uranium, Plutoniun, and Thorium Oxide Species

		ΔH_f° (kJ mol^{-1})							
		Old values	Third-law values at 298 K				Second-law values		New values T = 298
Molecule	ΔG_f° (J mol^{-1})					avg of 3	T = 298	T = 0	
UO	$21{,}670 - 51.09T$[a] $1500 < T < 2600$	19.2[b] $T = 0$	16.7 $T = 1500$	17.1 $T = 2100$	14.6 $T = 2600$	16.1	23.6	25.3	19.9 ± 10
UO_2	$-500{,}030 + 14.88T$[c] $1800 < T < 2400$	−468[d] $T = 298$	−465.3 $T = 1800$	−464.2 $T = 2100$	−464.3 $T = 2400$	−464.6	−468.3	−466.5	−466.3 ± 5
UO_3	$-829{,}350 + 78.3T$[a] $1200 < T < 1800$	−841[e] $T = 0$	−799.8 $T = 1200$	−796.5 $T = 1500$	−795.7 $T = 1800$	−797.3	−802.8	−807.2	−800.1 ± 20
PuO	$-121{,}000 - 50.6T$[f] $1600 < T < 2100$	−92.0[f] $T = 298$	−91.0 $T = 1600$	−90.3 $T = 1900$	−90.1 $T = 2100$	−90.3	−91.6	−90.2	−91.0 ± 5
PuO_2	$-473{,}200 + 18.2T$[f] $1600 < T < 2100$	−422[f] $T = 298$	−471.6 $T = 1600$	−475.2 $T = 1900$	−477.6 $T = 2100$	−474.8	−452.5	−448.8	−469.2 ± 20
ThO	$-70{,}790 - 51.09T$[g] $2200 < T < 2800$	−28.5[h] $T = 298$	−1.3 $T = 2200$	0.0 $T = 2600$	0.5 $T = 2800$	−1.0	−29.9	−28.6	−21.8 ± 20
ThO_2	$-546{,}800 + 27.87T$[h] $2023 < T < 3000$	−504.2[h] $T = 298$	−462.4 $T = 2100$	−451.6 $T = 2600$	−444.2 $T = 3000$	−452.7	−502.9	−499.4	−453 + 60

[a] From [2].
[b] Derived in [1].
[c] From [3]; consistent with $p(UO_2)$ in [4].
[d] From [4].
[e] Derived in [1] from D^0 in [5].
[f] Recommended value in [6].
[g] From [7]; inconsistent with [8].
[h] Recommended value in [8].

2.2. Third-Law Method

After ΔG_f^0 is determined as a function of T for a temperature range $T_l \leq T \leq T_u$ (subscript l for lower and u for upper), the value of $\Delta H_f^0(298.15\text{ K})$ can be determined for each T provided that the appropriate free-energy function data, $\text{fef}' \equiv -[G^0(T) - H^0(298.15\text{ K})]/T$, are available. For example,

$$\begin{aligned}\Delta H_f^0(\text{UO}_2, \text{g}, 298.15\text{ K}) &= \Delta G_f^0(\text{UO}_2, \text{g}) + T[\text{fef}'(\text{UO}_2, \text{g}) \\ &= -\text{fef}'\,(\text{U}, \text{ref}) - \text{fef}'(\text{O}_2, \text{g})]\end{aligned} \tag{4}$$

Data for gaseous O_2 [9] and for Th, U, and Pu in their reference states [10] are available. For the gaseous oxide molecules, the fef′ values have been calculated by means of Eq. (1). Spectroscopic data for gaseous thorium, uranium, and plutonium oxides are summarized in Table III. The electronic contribution to the free-energy function has been calculated from a density-of-states model [15] with three parameters, of which one, the ionization potential (A), is empirical. The spacing of electronic states, x in Table III, is constant, and the degeneracy, g in Table III, increases with energy:

$$g_n = g/(1 - \epsilon_n/A) \tag{5}$$

where $\epsilon_n = n \cdot x$. The model parameters for UO_2 (g) were determined [15] from the empirical vapor pressure equation [4]; for other molecules, empirical data on atomic energy levels were used with the atomic states approximation [18] in cases for which consistent data [1] are available. The atomic states approximation has been used to provide an upper limit [1,16,17] to the electronic contribution, except for UO_3(g) and ThO_2(g), because this model must underestimate the electronic contribution for these molecules (U^{6+} and Th^{4+} are both isoelectronic with Rn and have no low-lying electronic states). No corrections for vibrational anharmonicity, vibration-rotation interaction, or centrifugal stretching have been included in the calculated fef′ for the metal oxides because these corrections are expected to be small relative to the uncertainties [15].

2.3. Second-Law Method

In general, the ΔG_f^0 equation, Eq. (2), for the temperature range $T_l \leq T \leq T_u$ can be treated by an alternative method to derive $\Delta H_f^0(298.15\text{ K})$. For one set of experimental data that give ΔG_f^0 as a function of T, the second-law method differs from the third-law method in the way in which these data are averaged, as well as in the requirements for additional data to calculate $\Delta H_f^0(298.15\text{ K})$. If evaluated or averaged ΔG_f^0 values are available,

Table III. Summary of Data for Uranium, Plutonium, and Thorium Oxide Vapor Species

	UO	UO_2	UO_3	PuO	PuO_2	ThO	ThO_2
Rotation[a]	$r = 0.184$ nm	Linear $r = 0.179$ nm	*T*-shaped $2r = 0.176$ nm $r = 0.179$ nm	$r = 0.183$ nm	Linear $r = 0.179$ nm	$r = 0.184$ nm	$\theta = 122.5°$, $r = 0.180$ nm
Vibration[b] (frequency in cm^{-1})	820.0	765.4, 225.2 (2) 776.1	843.5, 745.6 852.6, 186.2 211.6, 151.5	822.3	746.50 230 (2) 794.25	890.99	787.40 250 735.35
Electronic[c]	$g = 1$, $x = 920\ cm^{-1}$ $IP = 45{,}000\ cm^{-1}$	$g = 1$, $x = 230$ $IP = 45{,}000$	$g = 1$, $x = 2300$ $IP = 85{,}000$	$g = 1$ $x = 260$ $IP = 47{,}000$	$g = 1$ $x = 670$ $IP = 75{,}000$	$g = 1$ $x = 430$ $IP = 49{,}000$	$g = 1$ $x = 2300$ $IP = 70{,}000$

[a] Bond angles are measured [11–14]; bond distances are estimated [13], except for ThO [16].

[b] All frequencies above 300 cm^{-1} are measured [11–14, 16]; those below are estimated [15] except for UO_2 and UO_3 [21].

[c] Model parameters [15] are derived; ionization potentials are measured [17].

which is the case for all the molecules of interest, the two methods differ in their requirements for data to obtain $\Delta H_f^0(298.15\text{ K})$.

If we again consider the sublimation of UO_2 as an example, we may consider the first term on the right-hand side of Eq. (2) to be $\Delta H_f^0(T_a)$, where $T_a = (T_u + T_l)/2$; then

$$\begin{aligned}\Delta H_f^0(UO_2, g, 298.15\text{ K}) = {} & \Delta H_f^0(UO_2, g, T_a) + [H^0(U, \text{ref}, T_a) \\ & -H^0(U, s, 298.15\text{ K})] + [H^0(O_2, g, T_a) \\ & -H^0(O_2, g, 298.15\text{ K})] - [H^0(UO_2, g, T_a) \\ & -H^0(UO_2, g, 298.15\text{ K})]\end{aligned} \qquad (6)$$

If, on the other hand, we have an evaluated equation for the sublimation pressure of UO_2 for $T_l \leqslant T \leqslant T_u$ (which is the case for UO_2 [4]), then the slope of $R \ln p$ vs T^{-1} gives $-\Delta H_{\text{sub}}^0$ (UO_2, T_a) and

$$\begin{aligned}\Delta H_{\text{sub}}^0(UO_2, 298.15\text{ K}) = {} & \Delta H_{\text{sub}}^0(UO_2, T_a) + \int_{298}^{T_a} C_p^0(UO_2, s)\, dT \\ & - \int_{298}^{T_a} C_p^0(UO_2, g)\, dT\end{aligned} \qquad (7)$$

$$\begin{aligned}\Delta H_f^0(UO_2, g, 298.15\text{ K}) = {} & \Delta H_{\text{sub}}^0(UO_2, 298.15\text{ K}) \\ & +\Delta H_f^0(UO_2, s, 298.15\text{ K})\end{aligned} \qquad (8)$$

The use of the second-law method to obtain $\Delta H_f^0(298.15\text{ K})$ for each of the molecules of interest requires knowledge of enthalpy differences between some high temperature, T_a, and 298.15 K. If the appropriate heat capacities are known in this temperature range, then the enthalpy increments can be obtained from the integrals.

Enthalpy increments are available for gaseous O_2 [9] and for Th, U, and Pu in their reference states [10]. For the oxide molecules of interest, the enthalpy increments can be calculated from spectroscopic data. It is important to note that the heat capacity depends on first and second derivatives of the partition function, as well as on the partition function itself. Thus the free-energy function that is required for a third-law analysis, Eq. (1), is sensitive to the spectroscopic data in a different way than is the heat capacity.

3. RESULTS

Third-law values of $\Delta H_f^0(298.15\text{ K})$ were calculated at the lower and upper temperature limits of the ΔG_f^0 equation and at a middle temperature for each of the seven molecules. These values, which are given in columns 4 to 6 in Table II, were averaged to give the value in column 7, which is considered to be the "best" third-law ΔH_f^0 value. The three values averaged for ThO_2 show a temperature dependence; thus the average $\Delta H_f^0(298.15\ K)$ value should be viewed with caution.

A second-law value of $\Delta H_f^0(298.15\ K)$ was calculated from the constant term in the ΔG_f^0 equation, assuming that this term is $\Delta H_f^0(T_a)$, where T_a is approximately the average temperature for the range in which the ΔG_f^0 equation is valid. These second-law values of ΔH_f^0 at 298.15 K and 0 K are given in columns 8 and 9 of Table II. The new second- and third-law values of $\Delta H_f^0(298.15\ K)$, columns 7 and 8 of Table II, agree within 10 kJ for gaseous UO, UO_2, UO_3, and PuO, For these molecules, we give the two values equal weight in averaging to obtain the recommended values given in the last column in Table II. The estimated uncertainty that is given is no less than the difference between the second- and third-law values. The new $\Delta H_f^0(298.15\ K)$ values for gaseous UO, UO_2, and PuO are in good agreement with the old recommended values, but the new value for $UO_3(g)$ is substantially different from the old.

For PuO_2, the three third-law values are in reasonably good agreement; therefore we give triple weight to the averaged third-law value in averaging it with the second-law value to obtain the recommended value in the last column in Table II. The recommended $\Delta H_f^0(PuO_2,\ g,\ 298.15\ K)$ value is more than 40 kJ smaller than the previously recommended [6] value.

The derivation of both the second-law and third-law $\Delta H_f^0(ThO,\ g,\ 298.15\ K)$ values depends upon the electronic contribution to the thermodynamic functions of ThO(g). Table IV shows the differences among the calculated electronic contributions to the free-energy function of ThO(g) at 2000 at 298.15 K. The atomic states model [18,19], which should yield the upper limit, gives a much larger contribution than that from known states [16]. The parameters for the density-of-states model [15], which were used to calculate column 5 in Table IV, were based on the assumption that the atomic states model overestimates the electronic contribution to the free-energy function by a factor of 2 at 2000 K. The choice of these values for the model parameters was made to minimize the uncertainty in the thermodynamic functions at higher temperatures (to 6000 K) and may not be the optimum choice for the present application. However, the values of the model parameters were chosen in a consistent manner for all the oxides of Th, U, and Pu; and, as shown in Table II, these choices do give good agreement between second- and third-law values of $\Delta H_f^0(298.15\ K)$ for gaseous (UO, UO_2, UO_3, and PuO.

Rand [8] has tabulated thermodynamic functions for ThO(g) that include an electronic contribution that is larger than that from observed

Table IV. Calculated Electronic Contributions to the Free-Energy Function of ThO(g)[a]

T (K)	Known states[b]	Th^{2+} states[c]	Rand[d]	Model[e]
298.15	0.0	13.7	0.0	1.1
2000	0.4	22.4	0.8	11.2

[a] In J K^{-1} mol^{-1}.
[b] From [16].
[c] From [20].
[d] From [8].
[e] For $g = 1, x = 430$ cm^{-1}, IP = 49,000 cm^{-1} [15].

states [16], but is smaller than that from the increasing density-of-states model. The second- and third-law values of ΔH_f^0 (ThO, g, 298.15 K) that are derived from the ΔG_f^0 equation of Ackermann and Tetenbaum [7] with Rand's free-energy functions for ThO are in reasonably good agreement with each other.

To obtain a value of ΔH_f^0(ThO, g, 298.15 K) that is consistent with all uncertainties, including those of the electronic contribution, we attach equal weight to each of the second- and third-law (averaged) values in Table II and to the corresponding values that were derived with Rand's [8] fef′(ThO, g) and average these four values to obtain ΔH_f^0 (ThO, g, 298.15 K) = −21.8 kJ mol^{-1} (−5.2 kcal mol^{-1}).

For ThO_2(g) the second- and third-law values of ΔH_f^0 (298.15 K) are in poor agreement with each other. In addition, the third-law values that are derived at different temperatures have an unacceptable temperature dependence. These inconsistencies, which suggest an error in the ΔG_f^0 equation, must be resolved before a value of ΔH_f^0 can be recommended with confidence. For ThO_2(g) we recommend the average third-law value, ΔH_f^0(ThO_2, g, 298.15 K) = −453.0 kJ mol^{-1}, until the lack of agreement between second- and third-law values can be resolved.

It is difficult to assign a quantitative value to the uncertainty of these ΔH_f^0 values, but some quantitative indication of the expected accuracy seems warranted. Account must be taken of the agreement between second- and third-law values, as well as the sum of all uncertainties in data for the elements.

Uncertainties in spectroscopic parameters, particularly estimated parameters, have been considered in detail [1,15]. For example, the U—O bond distance in UO is estimated to be 0.184 nm by correlation with known distances in monoxides and is very unlikely to be outside the range 0.180 to 0.190 nm. This uncertainty in the bond distance gives an uncertainty of ±0.5

J K^{-1} mol^{-1} in the calculated fef′(UO, g) at 2000 K, which in turn results in an uncertainty in ΔH_f^0(UO, g, 298.15 K) of 1.0 kJ mol^{-1}. For UO_3, in contrast, significant uncertainty arises from the need to estimate three vibrational frequencies. The estimated uncertainty in the vibrational contribution to fef′(UO_3, g) is less than 10 J K^{-1} mol^{-1} at 2000 K (probably much less), and this uncertainty is a major component of the uncertainty given in Table II for ΔH_f^0(298.15 K). The uncertainties in ΔH_f^0(298.15 K) values include those that result from uncertainties in spectroscopic data. A judgment of uncertainty (loosely defined as the limits of the 90% confidence level), together with the recommended ΔH_f^0 values, is given in the last column of Table II.

4. CONCLUSIONS

Spectroscopic data for ThO, ThO_2, UO, UO_2, UO_3, PuO, and PuO_2 have been used to derive new values of ΔH_f^0(298.15 K) for each gaseous molecule by both the second- and third-law methods. Evaluated ΔH_f^0 equations of the form of Eq. (2) were used as the starting point for each calculation, except that the ΔH_f^0(ThO, g) equation of Ackermann and Tetenbaum [7] is to be preferred over that of Rand [8]. The ΔH_f^0(298.15 K) values obtained by the second- and third-law methods are in good agreement with each other for UO, UO_2, UO_3, and PuO, and with previously recommended values, except for UO_3. The agreement between second- and third-law values is not as good for the cases of ThO and PuO_2 and is poor for ThO_2. Furthermore, the third-law values for ThO_2 that were derived from ΔH_f^0 at different temperatures show an unacceptable temperature dependence.

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NOTE ADDED IN PROOF

The three low-frequency vibrational modes of UO_3 and the bending mode of UO_2 have been measured [21]: for UO_3 $\bar{\nu}_3$ = 186.2, $\bar{\nu}_5$ = 211.6, and $\bar{\nu}_6$ = 151.5 cm^{-1}. These measured data replace some of the estimated parameters that are given in Table III and lead to revised estimates of other parameters; Table III has been revised accordingly. The resulting changes in calculated values in Table II have also been made. The largest change in a calculated ΔH_f^0 (298.15 K) value is for UO_3 and that change of 4.2 kJ mol^{-1} is well within the estimated uncertainty that is given in the last column in Table II.

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