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Estimation of the standard entropies of some Am(III) and Cm(III) compounds

R.J.M. Konings *

European Commission, Joint Research Centre, Institute for Transuranium Elements, P.O. Box 2340, D-76125 Karlsruhe, Germany

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Abstract

The standard entropies $S°(298.15\ K)$ of some actinide(III) compounds have been estimated using a semi-empirical method describing the total entropy as the sum of the lattice entropy S_{lat} and the excess entropy S_{exs}. The validity of the applied approach has been verified for the iso-electronic lanthanide(III) compounds for which a good agreement with experimental values has been obtained. The present results for the actinide(III) compounds are compared to previous estimates. Significant differences have been found, in particular for the americium compounds.

1. Introduction

Thermodynamic data for transuranium compounds are scarce, which is not surprising in view of the difficulties of handling these highly radioactive materials and the limited availability of americium and curium in pure form. Fortunately, a substantial set of experimental data has been generated for plutonium compounds between 1950 and 1985, when the expectations of plutonium reuse were still high. But for americium and curium compounds, which are currently of particular interest to the partitioning and transmutation research programmes in many countries, only few experimental determinations have been made. Almost all of them are of a thermochemical nature (enthalpies of formation, vapour pressure) as this type of measurements can be made with relatively small quantities of material (milligrams). Heat capacity measurements, which generally require samples in gram quantities when accurate adiabatic or drop calorimetric techniques are used, have not been made at all for the compounds of these elements. As a result, accurate values for the standard molar entropy are not known, which is reflected in significant uncertainties in the Gibbs energy of formation, the quantity that determines the chemical equilibrium between phases.

Estimates of the standard molar entropy of compounds of americium and curium presented in the literature are often based on qualitative methods, in which empirical correlations with the uranium and plutonium compounds are used. Also advanced Latimer schemes to resolve the entropy into contributions of the atomic components or entropy corrections for spin-only contributions of the magnetic entropy have been used [1–3]. In order to attempt to reduce the uncertainties in the standard molar entropies of some technologically relevant Am(III) and Cm(III) compounds, a semi-quantitative estimation approach is presented in this paper which considers the lanthanide(III) and actinide(III) compounds jointly.

2. Method

The entropy of the trivalent lanthanide and actinide compounds consists of a lattice component, arising mainly from the vibrations of the ions in the crystal, and an excess component [1,4–6]

$$S° = S_{lat} + S_{exs}. \quad (1)$$

The excess contribution is due to the distribution of the electrons over the energy levels, and includes the

* Tel.: +49-7247 951 391; fax: +49-7247 951 566.
E-mail address: konings@itu.fzk.de (R.J.M. Konings).

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lowering of the magnetic ordering (splitting of the ground term) by the crystalline electric field (Stark effect). It can be calculated from the partitioning function Q, which is described by the Maxwell–Boltzmann distribution law

$$Q_{\mathrm{exs}} = \sum_{i=0}^{n} g_i \mathrm{e}^{-\epsilon_i/RT}, \tag{2}$$

where ϵ_i is the energy and g_i the degeneracy of level i, R is the universal gas constant and T is the absolute temperature. The excess entropy component is then calculated from the equation

$$S_{\mathrm{exs}} = R \ln Q_{\mathrm{exs}}. \tag{3}$$

Combining Eqs. (2) and (3) gives

$$S_{\mathrm{exs}} = R \ln(g_0) + R \ln \left(\sum_{i=1}^{n} g_i \mathrm{e}^{-\epsilon_i/RT} \right). \tag{4}$$

The first term of Eq. (4) represents the temperature independent contribution of the ground state, the second term the contribution of the excited energy levels.

The lattice contribution in the series of 4f and 5f compounds is only known with sufficient accuracy when the f shell of the metal ions is empty ($4f^0$) or completely filled ($4f^{14}$). In these cases S_{exs} is zero and the experimental entropy corresponds to S_{lat}. Also in case the f-shell of the metal ion is half filled ($4f^7$), S_{lat} can be derived easily from the experimental entropy as only a correction for the temperature independent term in Eq. (4) needs to be made, in absence of significant crystal-field splitting of the ground state. For compounds containing ions with partially filled f-shell S_{lat} has been obtained empirically by interpolation of these values, as will be discussed in the following sections.

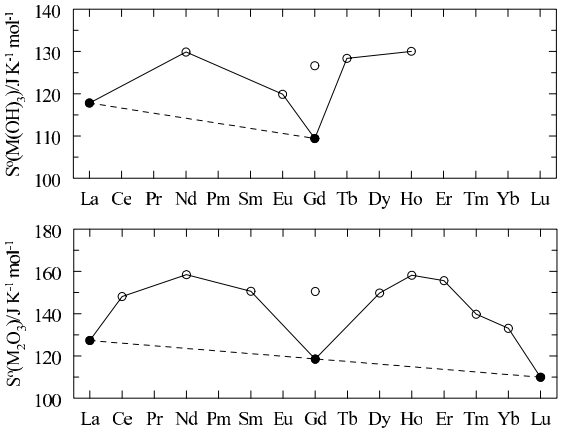

Fig. 1. The variation of the S_{exp} (○) and S_{lat} (●) in the lanthanide (hydr)oxides.

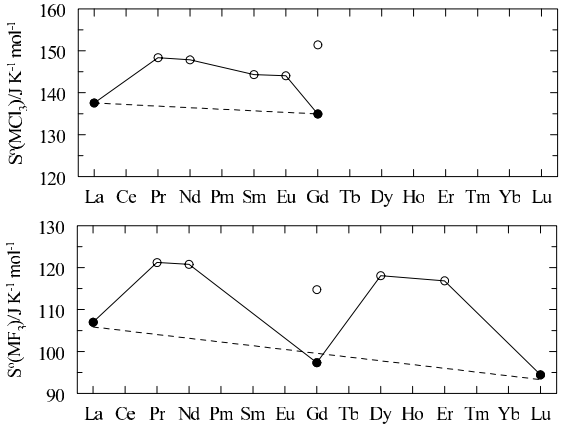

Fig. 2. The variation of the S_{exp} (○) and S_{lat} (●) in the lanthanide trihalides.

3. Results

3.1. Lanthanide compounds

Experimental heat capacity data for a large number of Ln_2O_3, $LnCl_3$ and $Ln(OH)_3$ compounds have been measured systematically by Westrum and coworkers, and of LnF_3 by Flotow et al. using adiabatic low-temperature calorimetry in the 5–350 K range. From these measurements the standard molar entropy at 298.15 K has been derived. In Fig. 1 the experimental values for the lanthanide sesquioxides are plotted as a function of the atomic number. The figure shows that S_{lat} for La_2O_3, Gd_2O_3 and Lu_2O_3 fall approximately on a straight line, in spite of the fact that their crystal structures are different (hexagonal, monoclinic and cubic, respectively). A similar relation is found for the lanthanide fluorides (Fig. 2). The lattice contribution for the other lanthanide compounds is then obtained by inter- or extrapolation of the data. These numbers are shown in Tables 1 and 2.

The excess entropies have been calculated from the energy levels of the Ln^{3+} ions derived from spectroscopic studies of the ions in transparent host crystals as summarised by Dieke et al. [7], Morisson and Leavitt [8] and Carnall et al. [9]. These electronic states are characterised by $(2J + 1)$-fold degeneracies which are removed by the crystalline electric field to result in a number of energy levels in the 0–500 cm^{-1} range. These crystal-field states have been identified for a number of the lanthanide sequioxides, trifluorides and trichlorides and some trihydroxides. The values for S_{exs} thus obtained are listed in Tables 1 and 2. In case the spectroscopic data are missing, S_{exs} is approximated by neglecting the crystal-field splitting and is calculated from the degeneracy of the ground state (and eventual low-lying energy levels)

Table 1
The entropies of the solid lanthanide(III) sesquioxides and hydroxides, in J K^{-1} mol^{-1}

	Calculated[a]			Experimental[b]	
	S_{lat}	S_{exs}	S_{tot}	S_{exp}	Refs.
La_2O_3	127.45	0.00	127.45	127.32	[22]
Ce_2O_3	125.21	13.01	151.23	148.11/148.8 ± 0.4	[23,24]
Pr_2O_3	123.97	18.27	160.51		
Nd_2O_3	122.73	17.43	157.59	158.45	[22]
Pm_2O_3	121.49	18.27	158.03		
Sm_2O_3	120.25	14.42	149.09	150.62	[25]
Eu_2O_3	119.01	9.20	137.41		
Gd_2O_3	117.77	17.29	152.35	152.73	[25]
Tb_2O_3	116.53	21.32	159.17		
Dy_2O_3	115.29	17.86/17.14[c]	151.01/150.65[d]	149.78 ± 0.42	[26]
Ho_2O_3	114.05	21.17	156.39	158.16	[26]
Er_2O_3	112.81	21.21/20.33[c]	155.24/154.80[d]	153.13 ± 0.42	[26]
Tm_2O_3	111.57	16.93/5.20[c]	146.13/139.56[d]	139.75	[13]
Yb_2O_3	110.33	10.72/10.65[c]	131.77/131.74[d]	133.05 ± 0.42	[25]
Lu_2O_3	109.09	0.00	109.09	109.96	[13]
$La(OH)_3$	117.81	0.00	117.81	117.81	[21]
$Ce(OH)_3$	116.60	13.61	130.21		
$Pr(OH)_3$	115.39	17.18	132.57		
$Nd(OH)_3$	114.18	17.44	131.62	129.87	[27]
$Pm(OH)_3$	112.97	18.27	131.24		
$Sm(OH)_3$	111.76	14.90	126.66		
$Eu(OH)_3$	110.55	9.51	120.06	119.88	[21]
$Gd(OH)_3$	109.34	17.29	126.63	126.63	[21]
$Tb(OH)_3$	108.13	20.33	128.46	128.37	[27]
$Dy(OH)_3$	106.92	23.05	129.97		
$Ho(OH)_3$	105.71	22.38	128.09	130.04	[28]
$Er(OH)_3$	104.50	23.05	127.55		
$Tm(OH)_3$	103.29	21.32	124.61		
$Yb(OH)_3$	102.08	17.19	119.37		
$Lu(OH)_3$	100.87	0.00	100.87		

[a] For the sesquioxides $S_{tot} = S_{lat} + 2 \times S_{exs}$.
[b] The uncertainty for the standard entropies derived from the calorimetric measurements has not been given in some cases.
[c] The first value is for the C_2 ions (3/4 of the total), the second value for the C_{3i} ions.
[d] The first value is calculated considering all ions in C_2 site, the second value considering the appropriate contributions of both sites.

of the lanthanide ion. This will lead to a small overestimation of S_{exs} at 298.15 K, which increases when the energy gap of the crystal-field splitting becomes larger. For example, we obtain $S_{exs} = 17.43$ J K^{-1} mol^{-1} at 298.15 K for Nd_2O_3 from the known crystal field levels, whereas we would obtain $S_{exs} = R\ln(10) = 19.14$ J K^{-1} mol^{-1} at 298.15 K from the approximation.

The calculated values for the standard entropy at 298.15 K (S_{tot}) compare well with the experimental values, as shown in Tables 1 and 2. For the trichlorides, trifluorides and trihydroxides the difference $S_{exp} - S_{tot}$ is in the order of 1–2%. For the sesquioxides the difference $S_{exp} - S_{tot}$ is somewhat larger. This is due to the fact that it was neglected that two inequivalent lanthanide sites of different symmetry (C_2 and C_{3i}) exist in the cubic sesquioxides [8]. Because it has been shown that the ground state splitting is different for the C_2 and C_{3i} sites [10–12], a 'fine-tuning' of the calculations is required. Justice et al. [13] and Gruber et al. [14] have listed the crystal fields levels of both sites for a number of lanthanide sesquioxides. In case of Er_2O_3 and Dy_2O_3 the differences in S_{exs} calculated from these data are marginal, but in case of Tm_2O_3 we obtain significantly different values. When these are mixed in the right proportion, the calculated S_{tot} compares well with the experimental value.

3.2. Actinide compounds

In Section 3.1 we have described how the standard entropy of the lanthanide(III) compounds can be derived semi-empirically making use of the reliable sets of experimental values derived from the calorimetric measurements. However, the low-temperature heat capacity data for the actinide(III) compounds are limited to some uranium and plutonium compounds. As a result, S_{lat} cannot be established like in the case of the lanthanide

Table 2
The entropies of the solid lanthanide(III) fluorides and chlorides, in J K^{-1} mol^{-1}

	Calculated			Experimental[a]	
	S_{lat}	S_{exs}	S_{tot}	S_{exp}	Refs.
LaF_3	105.84	0.00	105.84	106.98 ± 0.11	[29]
CeF_3	104.97	13.73	118.70		
PrF_3	104.10	16.75	120.85	121.22 ± 0.12	[30]
NdF_3	103.23	17.24	120.47	120.79 ± 0.12	[31]
PmF_3	102.37	18.27	120.64		
SmF_3	101.50	15.00	116.50		
EuF_3	100.63	9.44	110.07		
GdF_3	99.76	17.29	117.05	114.77 ± 0.22	[32]
TbF_3	98.90	20.07	118.97		
DyF_3	98.03	21.83	119.86	118.07 ± 0.12	[33]
HoF_3	97.16	23.18	120.34		
ErF_3	96.29	22.62	118.91	116.86 ± 0.12	[33]
TmF_3	95.42	19.56	114.98		
YbF_3	94.55	17.29	111.84		
LuF_3	93.69	0.00	93.69	94.83 ± 0.09	
$LaCl_3$	137.57	0.00	137.57	137.57	[34]
$CeCl_3$	136.71	14.71	151.42		
$PrCl_3$	136.28	17.87	154.15	153.30	[34]
$NdCl_3$	135.85	18.30	154.15	153.43	[34]
$PmCl_3$	135.42	17.89	153.31		
$SmCl_3$	134.99	15.27	150.26	150.12	[35]
$EuCl_3$	134.56	9.43	143.99	144.06	[35]
$GdCl_3$	134.13	17.29	151.42	151.42	[35]
$TbCl_3$	133.70	21.15	154.85		
$DyCl_3$	133.27	22.83	156.10		
$HoCl_3$	132.84	23.16	156.00		
$ErCl_3$	132.41	22.60	155.01		
$TmCl_3$	131.98	20.84	152.82		
$YbCl_3$	131.55	15.80	147.35		
$LuCl_3$	131.12	0.00	131.12		

[a] The uncertainty for the standard entropies derived from the calorimetric measurements has not been given in some cases.

compounds. Therefore a reverse approach has been used. The lattice component is derived by subtracting S_{exs} from the available experimental values. It is then assumed that the variation in S_{lat} of the actinide compounds is parallel to that in the lanthanide compounds. The values thus calculated are listed in Table 3.

Only in case of the actinide trifluorides two experimental determinations of $S°(298.15\ K)$ are known, UF_3 and PuF_3. Unfortunately details of the UF_3 measurements have not been published [15]. For that reason our calculation of S_{lat} of the actinide trifluorides is based on PuF_3 only. The value we then obtain for UF_3 is 6.69 J K^{-1} mol^{-1} higher than the experimental value, which is close to $R\ln(g_0)$ (for $g_0 = 2$, the ground state degeneracy in the uranium trihalides), suggesting that the experimental value may not include the excess entropy associated with the antiferromagnetic transition in the 0–10 K range. Evidence for this is derived from a reanalysis of the experimental data, which were extracted from the graph in [15]. Integration of the C_p data, extrapolated to 0 K using a C_p/T vs. T^2 method, gives an almost identical result as originally reported. This, of course, affects our calculation for the trichlorides significantly, as details for UCl_3 are also lacking [15]. It is believed that an $R\ln(2)$ correction is necessary for the UCl_3 value, likewise confirmed by the re-analysis of the experimental data.

The electronic states of the An^{3+} ions in the trichloride compounds are well known and the data used for the calculation of S_{exs} have been taken from the review by Carnall [16], who summarised the data of $AnCl_3$ and An:$LaCl_3$ optical spectroscopic studies. Less information is available for the trifluorides and the sequioxides, and for some of these compounds the crystal field splitting has been estimated (U, Pu) or neglected (Np).

4. Discussion

The present results for the actinide(III) compounds can be compared to previous estimates presented by Westrum and Grønvold [1] for the sesquioxides, Moskin

Table 3
The entropies of solid actinide(III) oxides and halides, in J K^{-1} mol^{-1}

	Calculated			Experimental	
	S_{lat}	S_{exs}	S_{tot}	S_{exp}	Ref.
Pu_2O_3	134.88	14.07	163.02	163.02 ± 0.65[a]	[32]
Am_2O_3	133.64	0.00	133.64		
Cm_2O_3	132.40	17.29	166.98		
UF_3	113.18	16.97	130.15	129.22 ± 0.50[a]	[15]
NpF_3	112.31	18.27	130.58		
PuF_3	111.44	14.67	126.11	126.11 ± 0.36	[36]
AmF_3	110.58	0.00	110.58		
CmF_3	109.71	17.29	127.00		
BkF_3	108.84	21.32	130.16		
CfF_3	107.97	23.05	130.02		
UCl_3	147.40	16.50	163.90	163.90 ± 0.50[a]	[15]
$NpCl_3$	146.97	18.27	165.24		
$PuCl_3$	146.60	14.78	161.38		
$AmCl_3$	146.22	0.00	146.22		
$CmCl_3$	145.85	17.29	163.14		
$BkCl_3$	145.47	19.16	164.63		
$CfCl_3$	145.19	22.11	167.20		

[a] Includes an addition of $R\ln(2)$ to the experimental value reported in [15] for the excess entropy associated with the antiferromagnetic transition below 10 K which was very likely not taken into account in [15] (see text for explanation).

[20] for the sesquioxides and trihalides, and by Fuger et al. [17] for the trihalides (see Table 4). The latter values are also adopted with minor changes in the NEA-TDP project [2,3,18].

In 1962 Westrum and Grønvold [1] estimated the standard entropies of the lanthanide and actinides sesquioxides in a systematic manner using an approach similar to that used in the present study. Their value for Cm_2O_3 is in reasonable agreement with our estimate (Table 4) but the value for Am_2O_3 deviates considerably. This is not suprising as the experimental basis (both calorimetric as spectroscopic) was very limited at that time. Ackermann et al. [19] suggested $S°(298.15\ K) = 150\ J\ K^{-1}\ mol^{-1}$ for Am_2O_3 based on an analysis of vaporization data of americium oxide dissolved in plutonium oxides. The more recent estimate of this quantity given in the NEA-TBP [2], is $(160 \pm 15)\ J\ K^{-1}\ mol^{-1}$, being taken close to the experimental value for plutonium sesquioxide.

Moskin [20] used a Latimer-type approach to estimate the standard entropies of the actinide compounds, but details of his calculations are lacking. The results are

Table 4
Comparison of the estimates of the standard molar entropies $S°(298.15\ K)$ of solid actinide(III) oxides and halides, in J K^{-1} mol^{-1}

	Westrum and Grønvold[a]	Moskin[b]	IAEA[c]	NEA-TDP[d]	This study[e]
Am_2O_3	158.2	131.8		160 ± 15	133.6 ± 5.0
Cm_2O_3	160.7	144.3			167.0 ± 5.0
NpF_3			125 ± 4	124.9 ± 2.0	130.6 ± 3.0
AmF_3		113.0	128 ± 4	127.6 ± 5.0	110.6 ± 3.0
CmF_3		119.2			127.0 ± 3.0
$NpCl_3$			162 ± 8	160.4 ± 4.0	165.2 ± 6.0
$PuCl_3$		141.0	164 ± 4	161.7 ± 3.0	161.4 ± 6.0
$AmCl_3$		149.4	165 ± 6	164.8 ± 6.0	146.2 ± 6.0
$CmCl_3$		155.6	166 ± 8		163.1 ± 6.0

[a] Ref. [1].
[b] Ref. [20].
[c] Ref. [17].
[d] Refs. [2,18].
[e] The overall uncertainty of the values from the present work is estimated to be $\pm 5\ J\ K^{-1}\ mol^{-1}$ for the sesquioxides and $\pm 3\ J\ K^{-1}\ mol^{-1}$ for the trifluorides, and $\pm 6\ J\ K^{-1}\ mol^{-1}$ for the trichlorides.

significantly lower than those of Westrum and Grønvold and show, in contrast, good agreement for Am_2O_3 and not for Cm_2O_3. For the trihalides the agreement between our results and those of Moskin is good, but this seems to be forfituous as the results for the uranium and plutonium compounds do not agree at all.

Fuger et al. [17] estimated the standard entropies of the actinide trihalides in a systematic way for the IAEA series on the chemical thermodynamics of actinide elements and compounds. At that time the unpublished entropy values of UF_3 and UCl_3 were the only experimental values available. These values, for which we have presented doubt that they may be too low due to the neglect of the S_{exs} in the 0–10 K range, were used by Fuger at al. as starting point. They applied a 'spin-only' correction, which means that $R\ln(n_f/4)$ is added to the entropy of UX_3 for each extra f electron (n_f being the number of f electrons in the transuranium ion). However, this assumption implies a steady increase of $S°(298.15\ K)$, which is not consistent with the experimental observations for the lanthanide compounds. As is evident from Table 4, the most prominent difference for the trihalides also occurs for the americium compounds (about 18 J K^{-1} mol^{-1}).

The remarkably low(er) entropy values found in our calculations for the americum compounds (compared to the previous estimates but also in the actinide series) are due to the fact that S_{exs} is zero because the $^7F_0$ ground state of Am^{3+} is non-degenerate and the first excited level $^7F_1$ does not contribute at 298.15 K. For example, the $^7F_1$ level is about 2750 cm^{-1} above the ground state in $AmCl_3$ [16]. This is different from the iso-electronic Eu^{3+} ions for which the $^7F_1$ levels in the trichloride are observed at 355.05 and 405.27 cm^{-1}, and also the $^7F_2$ levels around 1000 cm^{-1} [8] contribute to S_{exs}. The calculated value for S_{exs} of the europium(III) compounds is between 9.2 and 9.5 J K^{-1} mol^{-1}, and is consistent with the experimental result for $Eu(OH)_3$ and $EuCl_3$, the only Eu(III) compounds for which low-temperature heat capacity measurements in the 10–350 K range have been made [21].

5. Conclusion

The standard entropies ($S°(298.15\ K)$) of the lanthanide(III) compounds can be described with reasonable accuracy as the sum of the lattice entropy S_{lat}, derived empirically from the values for the La, Gd and Lu compounds, and the excess entropy S_{exs} calculated from the crystal field levels. A good agreement between calculated and experimental values has been obtained. The standard entropies of some actinide(III) compounds have been estimated using this approach. The results show significant differences when compared previous estimates, in particular for the americium compounds. This can be explained by the unique electronic configuration of the Am^{3+} ion which was not taken into account in the previous studies. Of course, experimental confirmation of our results is highly recommended.

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