Standard enthalpies of formation of ThAl$_2$, ThSi$_2$, ThGe$_2$ and Th$_5$Sn$_3$, determined by high-temperature direct synthesis calorimetry

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Abstract

The standard enthalpies of formation of ThAl$_2$, ThSi$_2$, ThGe$_2$ and Th$_5$Sn$_3$ have been determined by direct synthesis calorimetry at 1473±2 K. The following values of ΔH_f°, in kJ(mol atom)$^{-1}$, are reported: ThAl$_2$, −46.8±2.1; ThSi$_2$, −55.6±2.0, ThGe$_2$, −72.2±1.4, and Th$_5$Sn$_3$, −63.8±2.0. The results are compared with predicted values from the Miedema model and with an earlier calorimetric value for ThSi$_2$. For systematic reasons the results are also compared with earlier values of ΔH_f° for the equivalent Ce and U compounds. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

During the past two decades, this laboratory has carried out systematic studies of the thermochemistry of intermetallic compounds and of some metal–nonmetal compounds. Our work in this field has been summarized recently in several review papers [1–6]. These reviews show that in the past two decades we have determined the standard enthalpies of formation for 273 intermetallic compounds of early transition metals and lanthanide metals with late transition metals and the noble metals [4]. We have also studied a comparable number of compounds of transition metals and lanthanide metals with elements from Groups IIIB and IVB in the periodic table [5].

However, compounds between actinide elements and other elements have not been extensively studied in this laboratory. Until recently, we carried out only one study in this field: An investigation of the thermochemistry of intermetallic compounds of U with Ru, Rh and Pd. Values of ΔH_f° were reported for URu$_3$, URh$_3$ and UPd$_3$ by Jung and Kleppa in 1991 [7]. During the past year we have completed a study of the thermochemistry of alloys of Th with elements from Group VIII [8]. Seven values of ΔH_f°

were reported for ThCo$_5$, ThNi$_5$, ThRu$_2$, ThRh$_3$, ThPd$_3$, ThIr$_2$ and ThPt$_3$. It is a well-known fact that calorimetric studies of actinide compounds are scarce in the literature. Our recent review of the published literature in this field revealed that there are only about 6 calorimetric studies since 1970 [7,9–13]. However, going back about 45 years we found a much larger number of calorimetric studies. This search indicated that the research activity in this field was quite active during 1950s, and even more active during 1960s. Kubaschewski et al. included in their books 'Metallurgical Thermochemistry' (5th Edition) [14] and 'Materials Thermochemistry' (6th Edition) [15] the then available calorimetric values of the standard enthalpy of formation for some actinide compounds, such as, e.g. the intermetallic compounds of Th and U and the borides, aluminides, carbides, silicides, oxides, sulfides, selenides, nitrides, phosphides, arsenides, fluorides, chlorides, bromides, and iodides of these metals. It is worth noting that there is a comprehensive review of the thermodynamic properties of the actinide elements and their compounds published by Chiotti et al. in 1981 [16]. This review included almost all the calorimetric values known at that time.

In contrast to the relatively scarce calorimetric studies, there exist much more published data on the Gibbs free energy of formation for U and Th compounds in the literature [17–61]. These data were mostly obtained by

EMF measurements, but some were also derived from vapor pressure measurements or obtained by other methods.

In the present investigation we carried out calorimetric experiments on six compounds of Th with Al, an element from Group IIIB, and with Si, Ge and Sn, elements from Group IVB (ThAl$_2$, ThSi$_2$, Th$_3$Ge$_2$, Th$_{0.9}$Ge$_2$, ThGe$_2$, and Th$_5$Sn$_3$). However, we found that our experiments were not successful for two of the thorium–germanium compounds. This was due to the formation of too large percentages of other phases than the desired phase. As a result, in this paper we report only on the standard enthalpies of formation for the following four compounds: ThAl$_2$, ThSi$_2$, ThGe$_2$, and Th$_5$Sn$_3$.

2. Experimental details and starting materials

The experiments were all carried out by the direct synthesis method, using a single-unit differential micro-calorimeter, which has been described in detail in an earlier communication [62]. This micro-calorimeter was maintained at 1473±2 K throughout the whole research project. All experiments were conducted in an inert atmosphere of argon gas. In order to eliminate possible traces of oxygen and nitrogen, this gas was purified by passing it through a fused-silica tube filled with pure titanium sponge (~3 mm particles) maintained at about 1173 K. The actual synthesis reactions took place in boron nitride (BN) crucibles.

Calibration of the calorimeter was achieved by dropping pieces of 2 mm diameter high purity copper of known mass from room temperature into the calorimeter at 1473±2 K. The enthalpy of pure copper at this temperature, 46 465 J mol^{-1}, was taken from Hultgren et al. [63]. The calibrations were reproducible within ±1%.

Samples were prepared by mixing two component powders that were accurately weighed according to the appropriate stoichiometry. This mixture was then pressed into 4 mm diameter pellets. The purity of the materials used for the direct synthesis ranged from 99.5% for Si and Al, to 99.8% for Th, to 99.995% for Ge and 99.999% for Sn. The particle sizes were −100 mesh for Th and Sn, −150 mesh for Ge, and −325 mesh for Al and Si. All materials were purchased from Johnson Matthey, ALFA ÆSAR Group (Ward Hill, MA). The germanium powders used for the preparation of ThGe$_2$ was purchased as −10 mesh particles. This powder was ground in an agate mortar and then sifted through a 150 mesh sieve.

Th is a radioactive element and its powder is apt to be oxidized in air, especially in the presence of moisture. The handling of the Th powder used for sample preparation was exactly the same as in our earlier paper [8]. The Th powder was examined by X-ray powder diffraction (XRD) at the beginning of this study. The XRD showed that the Th powder contai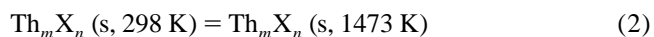ned detectable amount of ThO$_2$. A rough estimate from the relative intensity of the ThO$_2$ 111 peak (100% intensity) on the XRD patterns of the Th powder indicated that this powder might contain as much as about 3–5% of ThO$_2$. However, taking the X-ray penetration depth in the very heavy Th metal into consideration, the XRD patterns obtained may mostly reflect the average composition of the surface layer and a limited depth of the layer underneath the surface of the Th particles. For this reason, we believe that the content of ThO$_2$ in our starting material of Th powder should be less than the rough estimate of 3–5%. The starting material of Th powder was examined again by XRD after the completion of the study. A comparison was made between the XRD patterns obtained before and after the study. It was hard to tell whether there was an increase in the amount of ThO$_2$ in the Th powder.

3. Experimental results

The standard enthalpy of formation of the compound Th$_m$X$_n$ (X stands for a second element) is obtained from the difference between two sets of measurements. In the first set the following reaction takes place in the calorimeter.

$$m\text{Th (s, 298 K)} + n\text{X (s, 298 K)} = \text{Th}_m\text{X}_n \text{ (s, 1473 K)} \quad (1)$$

The products of reaction (1) were reused in a subsequent set of measurements to determine the corresponding heat contents

$$\text{Th}_m\text{X}_n \text{ (s, 298 K)} = \text{Th}_m\text{X}_n \text{ (s, 1473 K)} \quad (2)$$

From Eq. (1) and Eq. (2) we have

$$m\text{Th (s, 298 K)} + n\text{X (s, 298 K)} = \text{Th}_m\text{X}_n \text{ (s, 298 K)} \quad (3)$$

The standard enthalpy of formation is given by

$$\Delta H_f^\circ(\text{Th}_m\text{X}_n) = \Delta H(1) - \Delta H(2)$$

where $\Delta H(1)$ and $\Delta H(2)$ are the enthalpy changes per mole of atoms for Eq. (1) and Eq. (2), respectively.

Table 1 summarizes all the experimental results obtained for the four compounds. The reported values of $\Delta H(1)$ and (2) are averages of four to six individual determinations with standard deviations of δ_1 and δ_2, respectively. If the standard deviation for the calibration is δ_3, the overall uncertainty in the reported standard enthalpy of formation is calculated from $\delta = \sqrt{\delta_1^2 + \delta_2^2 + \delta_3^2}$.

After the measurements, all the reacted samples were examined by XRD. The results of these examinations are listed in Table 2. This table shows that Th$_5$Sn$_3$ does not have a standard XRD pattern listed in the ASTM powder diffraction file. Moreover, there are no standard XRD patterns for other Th–Sn alloys. Using VersaTerm-Pro software, purchased from Abelbeck Software Company and based on the crystallographic data in the Pearson's

Table 1
Observed heats of reaction, average heat contents at 1473 K, and calculated standard enthalpies of formation, in kJ (mol atom)$^{-1a}$

Compound	Melting point (°C)	Structure type	$\Delta H(1)$	$\Delta H(2)$	ΔH_f°
$ThAl_2$	1520	AlB_2	-15.4 ± 0.9 (5)	31.3 ± 1.8 (4)	-46.8 ± 2.1
$ThSi_2$	1850	$ThSi_2$	-23.6 ± 1.5 (6)	31.9 ± 1.2 (5)	-55.6 ± 2.0
$ThGe_2$	1900	$ThGe_2$	-32.0 ± 0.7 (6)	40.2 ± 0.8 (5)	-72.2 ± 1.4
Th_5Sn_3	1780	Mn_5Si_3	-32.4 ± 0.6 (6)	31.5 ± 1.4 (6)	-63.8 ± 2.0

^{a} Numbers in parentheses indicate numbers of experiments averaged.

handbook of crystallographic data [64], the ideal diffraction patterns for Th_5Sn_3, $ThSn_2$, $ThSn_3$ and Th_5Sn_4 were generated. With these XRD patterns and the standard XRD patterns for pure Th and Sn, we established that our Th_5Sn_3 samples did not contain any unreacted elements. However, it might contain less than 10% of Th_5Sn_4 and about 5% of ThO_2.

All the other three compounds have standard XRD patterns. From Table 2 we see that a small amount of ThO_2 was found in all these samples. We believe that most of the ThO_2 found in the reacted samples had its origin in the Th starting materials. However, we cannot completely exclude the possibility that a part of the ThO_2 was formed during the sample preparation. The pre-existing ThO_2 will not participate in the reaction. Hence, it will not contribute to the difference between the enthalpy changes of Eq. (1) and Eq. (2). The probability that additional ThO_2 was formed during the direct synthesis reaction is minimal since the reaction took place in a very good inert atmosphere. It should be noted that the XRD pattern we obtained for our $ThGe_2$ sample did not match the standard XRD pattern for $ThGe_2$, nor the standard XRD patterns for any other Th–Ge compounds. We also did not find any reflections for the component elements (Th and Ge) in this sample. Therefore, we believe that our reacted $ThGe_2$ sample may have a different structure from the one that was used to provide the standard XRD file for this compound.

In addition to ThO_2, a small amount of a second phase was found in our reacted $ThAl_2$ sample. In the same way we found a small amount of an unknown phase in our $ThSi_2$ sample. This is because there were a few weak unknown reflections in the XRD pattern of this sample. Although it is impossible to make a quantitative estimate of the percentage of a second phase in the reacted samples solely based on the XRD patterns, a very rough estimate may be good enough for our purpose. Once we are positive that the total percentage of the phases other than the

desired phase will not be more than 10%, we feel confident in the reported enthalpy values. We believe that the formation of up to 10% of other phases during the direct synthesis reactions will not introduce any significant errors in our reported enthalpy values. This is because the heats of formation of these second phases usually are roughly comparable in magnitude to the heat of formation of the desired phase.

4. Discussion

Fig. 1 shows plots of our experimental results for the standard enthalpies of formation of the four compounds $ThAl_2$, $ThSi_2$, $ThGe_2$ and Th_5Sn_3. All values are given in kJ(mol atom)$^{-1}$. Also plotted in this figure are predicted values calculated from the semi-empirical model of Miedema and co-workers [65]. We found an earlier calorimetric value for the compound $ThSi_2$, -58.0 ± 5.6 kJ(mol atom)$^{-1}$, listed by Kubaschewski et al. in [15] in 1993. This value is plotted along with our own result in Fig. 1. There are no earlier calorimetric values for any of the other three compounds in the literature. From this figure, we see that the Miedema model gives somewhat more exothermic values than we observed for all the four compounds. However, the earlier calorimetric value for $ThSi_2$ is in good agreement with our own value (-55.6 ± 2.0 kJ(mol atom)$^{-1}$).

In Fig. 2 we compare our standard enthalpies of formation for $ThAl_2$, $ThSi_2$, $ThGe_2$ and Th_5Sn_3 with the available calorimetric data for their equivalent Ce and U compounds. The sources of the calorimetric values used for the comparison are as follows: (1) In 1985 Colinet et al. measured ΔH_f° for most of the $REAl_2$ compounds. Their value for $CeAl_2$ was -52.2 kJ(mol atom)$^{-1}$ [66]. (2) In 1993 Kubaschewski et al. listed values of ΔH_f° for UAl_2 and USi_2. They were -32.9 ± 1.8 and -43.2 ± 0.8 kJ(mol

Table 2
Summary of X-ray diffraction examination results

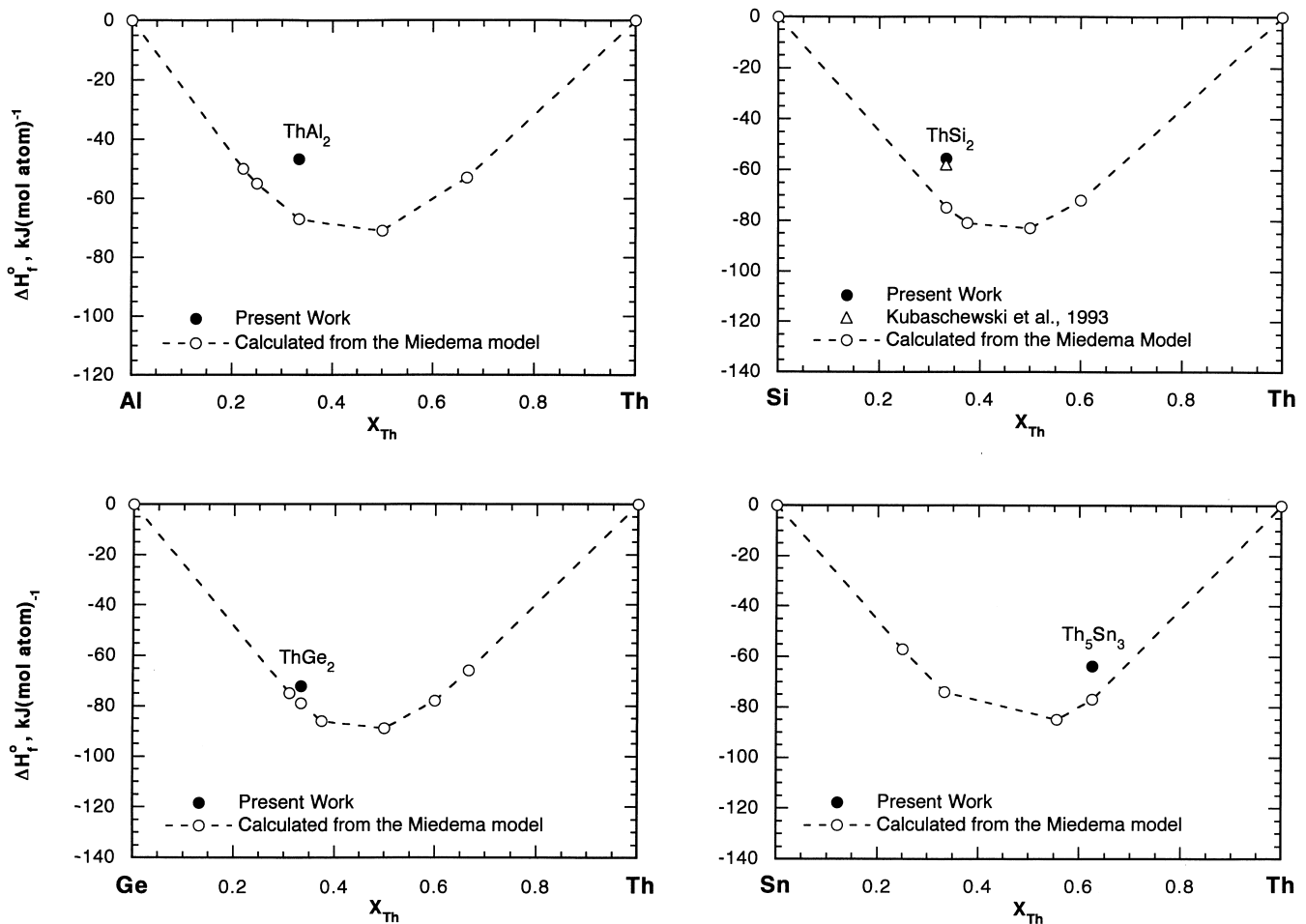| Compound	XRD examination result
$ThAl_2$	No unreacted elements. $ThAl_2$ + minor $ThAl_3$ + ThO_2 (~5%)
$ThSi_2$	No unreacted elements. $ThSi_2$ + three weak unknown peaks + ThO_2 (~5%)
$ThGe_2$	No unreacted elements. $ThGe_2$ + ThO_2 (~5%). (See the text for detail)
Th_5Sn_3	No ASTM file. No unreacted elements. Th_5Sn_3 + minor Th_5Sn_4 + ThO_2 (~5%). (Based on the calculated XRD patterns for alloys in Th–Sn system)

Fig. 1. Standard enthalpies of formation for ThAl$_2$, ThSi$_2$, ThGe$_2$ and Th$_5$Sn$_3$, compared with predicted values from the Miedema model [65], and with an earlier value for ThSi$_2$ given by Kubaschewski et al. [15].

atom)$^{-1}$ [15], respectively. (3) In 1995 Meschel and Kleppa measured ΔH_f^o for CeSi$_2$. Their value was -60.5 ± 2.0 kJ(mol atom)$^{-1}$ [67]. (4) In 1982 Borzone et al. measured ΔH_f^o for Ce$_5$Sn$_3$. Their value was -73.2 kJ(mol atom)$^{-1}$ [68]. These values are all plotted in Fig. 2 for the comparison.

However, in order to consider the systematic variation in magnitude of ΔH_f^o for each of the above four families of compounds we need more information. We found no earlier calorimetric values in the literature for CeGe$_2$, UGe$_2$ and U$_5$Sn$_3$. For CeGe$_2$, we believe that its ΔH_f^o should not be very different from the value for CeGe$_{1.6}$, which was measured by Meschel and Kleppa in 1995 by high-temperature direct synthesis calorimetry [67]. Their value of ΔH_f^o for this compound is -75.6 ± 1.9 kJ(mol atom)$^{-1}$. This value has been plotted in Fig. 2c as an acceptable approximation for CeGe$_2$. For UGe$_2$ we found a non-calorimetric value of ΔH_f^o given by Rand and Kubaschewski, -29.1 kJ(mol atom)$^{-1}$ [69]. This value was derived from the vapor pressure data of Alcock and Grieveson [70], and is plotted in Fig. 2c. For U$_5$Sn$_3$, we found no earlier value. However, we found a non-

calorimetric value of ΔH_f^o for U$_3$Sn$_2$ listed by Rand and Kubaschewski, -32.6 kJ(mol atom)$^{-1}$ [69]. This value was also derived from vapor pressure data [70]. Since the composition of the compound U$_5$Sn$_3$ ($X_U = 0.625$) is very close to the composition of the compound of U$_3$Sn$_2$ ($X_U = 0.600$), we plotted the value of ΔH_f^o(U$_3$Sn$_2$) in Fig. 2d.

From Fig. 2 it is apparent that in all the four cases studied the magnitude of the standard enthalpy of formation decreases only moderately from the Ce compounds to the Th compounds, and then very substantially from the Th compounds to the U compounds.
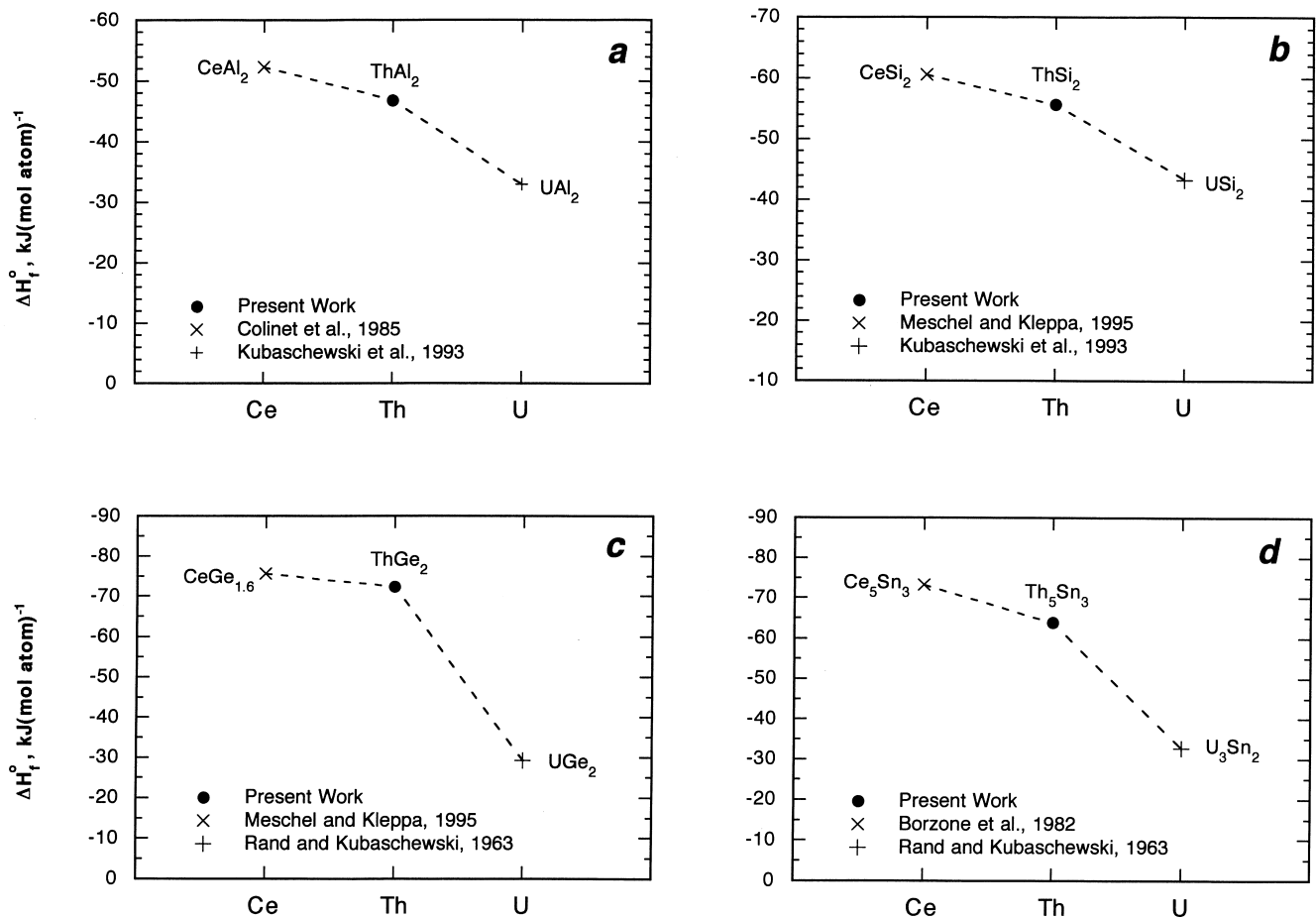
Fig. 2. (a) Standard enthalpy of formation for $ThAl_2$, compared with an earlier value for $CeAl_2$ given by Colinet et al. [66] and with an earlier value for UAl_2 listed by Kubaschewski et al. [15]. (b) Standard enthalpy of formation for $ThSi_2$, compared with an earlier value for $CeSi_2$ given by Meschel and Kleppa [67], and with an earlier value for USi_2 listed by Kubaschewski et al. [15]. (c) Standard enthalpy of formation for $ThGe_2$, compared with an earlier value for $CeGe_{1.6}$, determined by Meschel and Kleppa [67], and with a non-calorimetric value for UGe_2 given by Rand and Kubaschewski [69]. (d) Standard enthalpy of formation for Th_5Sn_3, compared with an earlier value for Ce_5Sn_3, determined by Borzone et al. [68], and with a non-calorimetric value for U_3Sn_2 given by Rand and Kubaschewski [69].

their help with the X-ray diffraction examination of our samples. We are also indebted to Doctor Susan Meschel for the many discussions conducted between us while we carried out the experiments and during the manuscript preparation.

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