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Molten salt EMF cell measurements on U–Ga alloys

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Abstract

The Gibbs free energies of formation of UGa_3, UGa_2 and U_2Ga_3 were determined by using high temperature molten salt galvanic cell measurements. Three cells were assembled and EMFs were measured. The typical cell is as follows: U(s)//UCl_3 in LiCl–KCl (eutectic)(l)//U–Ga (two phase alloy), where pure uranium is used as one of the electrodes and the other electrode is a two phase U–Ga alloy. The two phase alloys used as the electrodes in the cells I, II and III were $\langle UGa_3\rangle + \{Ga\}$, $\langle UGa_3\rangle + \langle UGa_2\rangle$ and $\langle UGa_2\rangle + \langle U_2Ga_3\rangle$, respectively. The Gibbs free energy of formation of UGa_3, UGa_2 and U_2Ga_3 with respect to α-uranium and liquid gallium in the temperature range 644–988, 751–947 and 800–950 K were determined to be $-44.1 + 0.016T$, $-46.0 + 0.017T$ and $-46.3 + 0.017T$ kJ gatom^{-1}, respectively. The partial molar excess Gibbs free energy of uranium in liquid gallium at 816 K was determined to be -75.5 ± 1.8 kJ gatom^{-1} and the partial molar enthalpy of uranium in liquid gallium in the saturated solution at 816 K to be -103.2 ± 1.8 kJ gatom^{-1}.

Keywords: Thermodynamics; Gibbs free energy; Galvanic cell; EMF; U–Ga system

1. Introduction

Thermodynamic properties of actinide–gallium alloys are of interest in the pyrochemical processes for the recovery of actinides from spent molten salts by equilibrating them with liquid Ca–Ga alloys [1]. Thermodynamic properties such as activities and chemical potentials are especially useful for understanding the stability of actinide–gallium intermetallic compounds which could be formed when the actinide chlorides or oxides are reduced from the molten salt phase to an alloy phase. Hence, in this study the Gibbs free energies of formation of the intermetallic compounds of U–Ga system were determined by using molten salt galvanic cell measurements.

There are three intermetallic compounds, UGa_3, UGa_2 and U_2Ga_3 in the U–Ga system [2]. The Gibbs free energy of formation of UGa_3 has been determined by Johnson and Feder [3] and Lebedev et al. [4] by molten salt galvanic cell measurements. Alcock et al. [5] have measured the vapour pressures of gallium over gallium metal as well as U–Ga alloys by a weight loss Knudsen effusion method and derived the Gibbs free energies of formation of the intermetallic compounds UGa_3, UGa_2 and U_2Ga_3. Chiotti et al. [6] have reviewed the thermodynamic data of the U–Ga system and they have computed the free energies of formation of all the three intermetallic compounds. Gardie et al. [7] have measured the thermodynamic activity of gallium in the U-rich region by a Knudsen cell mass spectrometric method. There exists large

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disagreement among the thermodynamic data for UGa_3 determined by vapour pressure measurements and the molten salt EMF method. The only data available for UGa_2 is that of Alcock et al. [5]. The data available for U_2Ga_3 are those of Alcock et al. [5] and Gardie et al. [7]. We have earlier determined the enthalpies of formation of intermetallic compounds UGa_3 and UGa_2 at 298.15 K by calorimetry [8].

2. Experimental

2.1. Materials

Uranium of nuclear grade purity and gallium (99.99%) from Nuclear Fuel Complex, Hyderabad were used for the preparation of alloys. Anhydrous LiCl (>99%) from M/s. Fluka, Switzerland, analytical grade KCl (99.8%) from M/s. Glaxo Laboratories, India and anhydrous $CdCl_2$ (99%) from M/s. Merck, Germany were used for preparing the molten salt electrolyte.

2.2. Preparation of the electrolyte salt

The salt mixture of LiCl–KCl was purified by passing a stream of HCl gas at 673 K [9] through the mixture and melting the salt under an HCl atmosphere and cooling the salt. The salt was again remelted at 673 K under an HCl atmosphere. The purified salt was loaded with 10 wt.% UCl_3 by equilibrating the salt containing $CdCl_2$ with uranium metal in an alumina crucible at 707 K under argon atmosphere inside an argon atmosphere glove box. The amount of uranium used for equilibration was in excess of the stoichiometric amount in order to ensure that no $CdCl_2$ remained in the electrolyte salt. The salt thus prepared was stored in an argon atmosphere glove box until use.

2.3. Preparation of electrodes

U–Ga alloys were prepared by arc melting the component metals followed by annealing and characterised by X-ray diffraction as was done for the preparation of single-phase intermetallic compounds, as described in our earlier publication [8]. The metals/alloys used as the electrodes were contained in tantalum cups (6-mm diameter and 10–15-mm height) which were welded to tantalum wires of 1.5-mm diameter to serve as electrode leads.

2.4. EMF measurement procedure

A schematic diagram of the EMF measurement system is shown in Fig. 1. The degassed EMF cell assembly was taken inside an argon atmosphere glove box. The metal and alloys used as electrodes were loaded in the tantalum cups, which were closed by crimping. The electrolyte salt was taken in a recrystallised alumina crucible, which was heated above the melting point of the salt. The tantalum cups containing the alloys were lowered into the molten salt and cooled. Then, the cell was assembled, taken out of the glove box and suspended into a Kanthal wire wound, resistance furnace. The temperature of the furnace was controlled within ±1 K by using a PID controller. The cell temperature was measured using a chromel–alumel thermocouple, kept inside an alumina thermowell which was kept immersed in the electrolyte salt. Purified argon gas was continuously passed through the cell at a very low flow rate. The cell was heated to the desired temperature and EMF measurements were done. After equilibrium was attained, as indicated by stable EMF reading, the EMF was measured using a high impedance electrometer. The EMF value at a given temperature was measured, while increasing and decreasing the temperature of the cell. The reversibility of the cell was verified by shorting the two electrodes by using a conducting wire and ascertaining that the EMF reaches the original value after some time.

Three different cells were used in these studies. In all of them, pure uranium was used as the reference electrode. The sample electrodes used in the three cells had the following compositions: Ga-6 at.% U, Ga-30 at.% U and Ga-36 at.% U. The first alloy corresponds to a two-phase alloy $(\{Ga\} + \langle UGa_3\rangle)$, the second one to $(\langle UGa_2\rangle + \langle UGa_3\rangle)$ and the third to $(\langle UGa_2\rangle + \langle U_2Ga_3\rangle)$ in the temperature range of measurement. The Gibbs free energy of formation of UGa_3 was determined from the EMF measurements of the following cell:

$$U(s)//UCl_3 \text{ in } LiCl-KCl_{(eut)}(l)//\langle UGa_3\rangle + \{Ga\}$$

(Cell I)

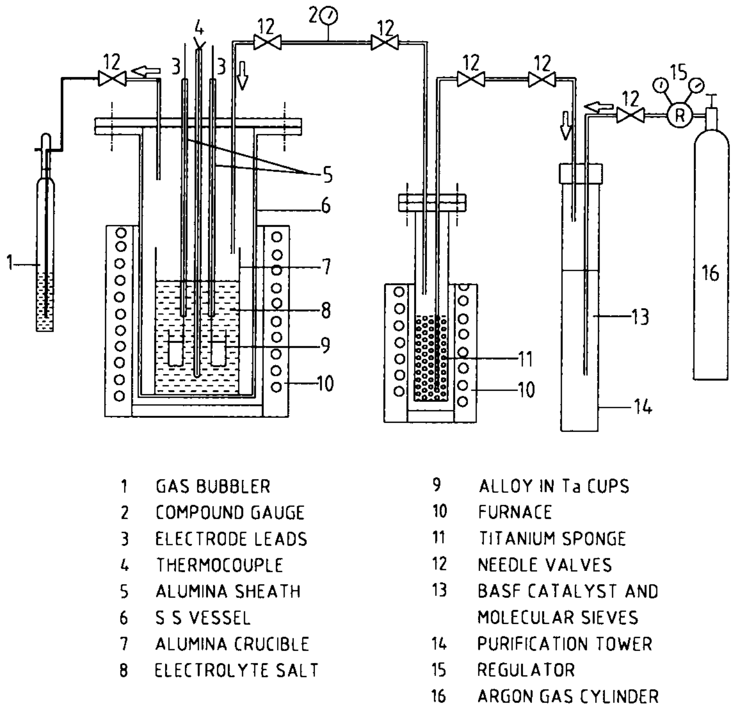

Fig. 1. Schematic diagram of EMF cell set-up.

in the temperature range 644–988 K. The Gibbs free energy of formation of UGa_2 was determined from EMF of the cell

$U(s)//UCl_3$ in $LiCl{-}KCl_{(eut)}(l)//\langle UGa_3\rangle + \langle UGa_2\rangle$ (Cell II)

in the temperature range 751–947 K. The Gibbs free energy of formation of U_2Ga_3 was determined from EMF measurements of the following cell:

$U(s)//UCl_3$ in $LiCl{-}KCl_{(eut)}(l)//\langle UGa_2\rangle + \langle U_2Ga_3\rangle$ (Cell III)

in the temperature range 800–950 K.

In the first cell, about 3–4 h were required to attain the initial equilibrium. But, once the initial equilibrium was attained, only 1 h was required to attain equilibrium at a new temperature, when the temperature was changed. In the case of cells II and III where two solid phases are present, the time required for attainment of equilibrium was longer. For these cells, initial equilibrium was attained only after about 12 h and for further changes in temperature about 3–4 h were required for attaining equilibrium.

The error on the EMF values has been estimated to be ±1 mV. The errors on the free energies of formation of the intermetallic compounds have been computed by using this uncertainty and the error on the free energies of formation of other compounds, used for computations.

3. Results

The EMF values of cells I, II and III are given in Tables 1–3, respectively. The EMF values of the cells were found to be reproducible at a given temperature, during heating as well as cooling cycles. The variation of the EMF of cell I with temperature is shown in Fig. 2. The least square regression analysis of the EMF data of the cell I yields:

$$E(V) = (0.609 \pm 0.003) - (0.220 \pm 0.004)10^{-3}T \quad (1)$$

Table 1
Measured EMF values of cell I

Serial No.	Temperature (K)	EMF (V)
1	654	0.4651[a]
2	678	0.4583[a]
3	683	0.4571[a]
4	709	0.4523[a]
5	735	0.4469[a]
6	758	0.4407[a]
7	785	0.4383[b]
8	785	0.4361[a]
9	811	0.4332[b]
10	811	0.4304[a]
11	836	0.4246[a]
12	836	0.4278[b]
13	861	0.4182[b]
14	861	0.4176[b]
15	863	0.4223[b]
16	886	0.4129[b]
17	888	0.4127[b]
18	913	0.4070[b]
19	938	0.4030[b]
20	962	0.3964[b]
21	988	0.3894[b]

[a] Cooling.
[b] Heating.

Table 2
Measured EMF values of cell II

Serial No.	Temperature (K)	EMF (V)
1	751	0.1395[a]
2	776	0.1400[b]
3	800	0.1314[b]
4	825	0.1312[b]
5	839	0.1281[b]
6	850	0.1293[a]
7	851	0.1274[a]
8	875	0.1266[a]
9	876	0.1257[b]
10	899	0.1266[a]
11	900	0.1224[b]
12	901	0.1254[a]
13	923	0.1228[a]
14	924	0.1228[b]
15	947	0.1202[b]
16	900	0.1235[b]
17	899	0.1248[a]

[a] Cooling.
[b] Heating.

Table 3
Measured EMF values of cell III

Serial No.	Temperature (K)	EMF (V)
1	800	0.1227[a]
2	802	0.1226[b]
3	825	0.1205[a]
4	839	0.1200[a]
5	851	0.1186[b]
6	875	0.1183[b]
7	899	0.1173[b]
8	900	0.1158[a]
9	923	0.1161[b]
10	924	0.1156[a]
11	947	0.1142[a]
12	950	0.1125[a]

[a] Heating.
[b] Cooling.

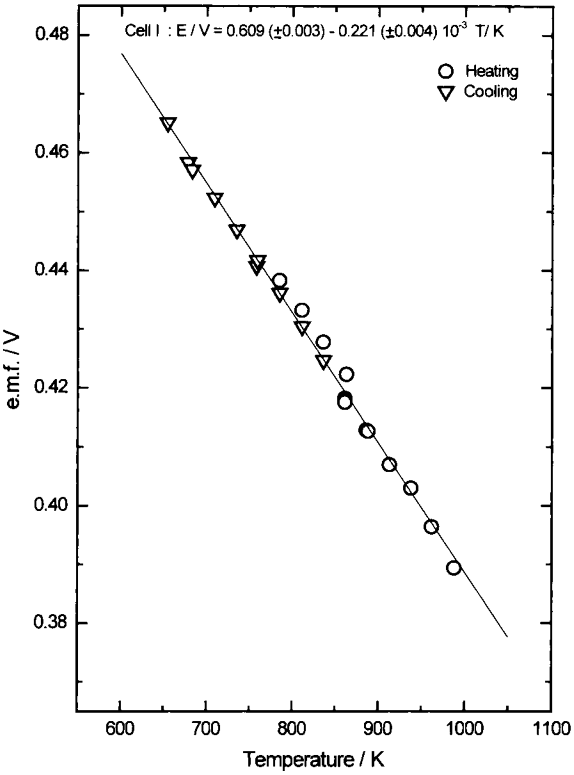

Fig. 2. Variation of EMF of the cell I with temperature.

The cell EMF, E, the partial molar Gibbs free energy of uranium, $\overline{G}_{\mathrm{U}}$, and the standard Gibbs free energy of formation of UGa$_3$, $\Delta G^0_{\mathrm{f,UGa_3}}$, from α-uranium and pure liquid gallium are related by the equations

$$\overline{G}_{\mathrm{U}} - G^0_{\mathrm{U}} = -zFE \tag{2}$$

and

$$\Delta G^0_{\mathrm{f,UGa_3}} = -zFE + 3RT\ln a_{\mathrm{Ga}} \tag{3}$$

where G^0_{U} is the free energy of uranium in the reference state (α-uranium), $z = 3$ the number of electrons participating in the electrode reaction, F the Faraday constant and a_{Ga} the activity of gallium in the saturated solution. In the uranium–gallium system, the saturated solutions were sufficiently dilute that the term $RT\ln a_{\mathrm{Ga}}$ was estimated to the required accuracy by assuming that the solutions were ideal with respect to gallium. The standard Gibbs free energy of formation of UGa$_3$, $\Delta G^0_{\mathrm{f,UGa_3}}$, with reference to α-uranium and liquid gallium is expressed as

$$\Delta G^0_{\mathrm{f,UGa_3}} = -176.36 + 0.0632T(\pm 1.8)\,\mathrm{kJ\,mol^{-1}} \tag{4}$$

The enthalpy and entropy of formation of UGa$_3$ are, thus, $-176.4\,\mathrm{kJ\,mol^{-1}}$ and $63\,\mathrm{J\,mol^{-1}\,K^{-1}}$, respectively.

The EMF versus temperature plot for cell II is shown in Fig. 3 and it yields

$$E(V) = (0.211 \pm 0.006) - (0.097 \pm 0.007)10^{-3}T \tag{5}$$

The standard Gibbs free energy of formation of UGa$_2$ was calculated using the following equation:

$$\Delta G^0_{\mathrm{f,UGa_2}} = \tfrac{1}{3}(2\,\Delta G^0_{\mathrm{f,UGa_3}} - zFE_{\mathrm{II}}) \tag{6}$$

with the Gibbs free energy of formation of UGa$_3$ computed using cell I and the EMF of the cell II (E_{II}) which gives

$$\Delta G^0_{\mathrm{f,UGa_2}} = -138.0 + 0.052T(\pm 2.9)\,\mathrm{kJ\,mol^{-1}} \tag{7}$$

The enthalpy and entropy of formation of UGa$_2$ are, thus, $-138.0\,\mathrm{kJ\,mol^{-1}}$ and $52\,\mathrm{J\,mol^{-1}\,K^{-1}}$, respectively.

The EMF versus temperature of cell III is shown in Fig. 4 and it yields

$$E(V) = 0.169(\pm 0.003) - (0.058 \pm 0.004)10^{-3}T \tag{8}$$

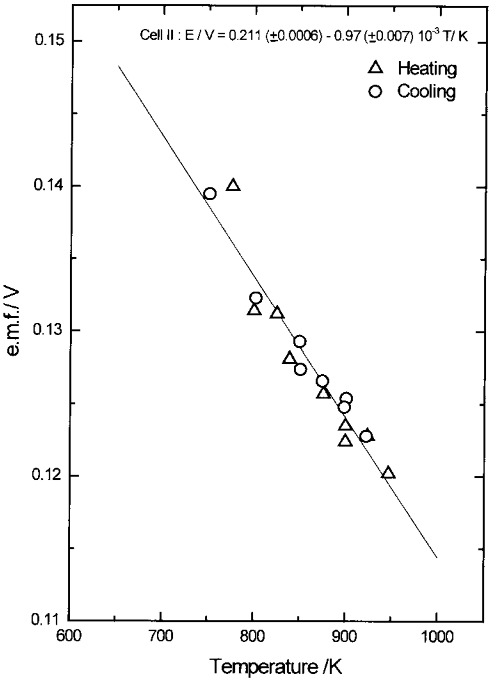

Fig. 3. Variation of EMF of the cell II with temperature.

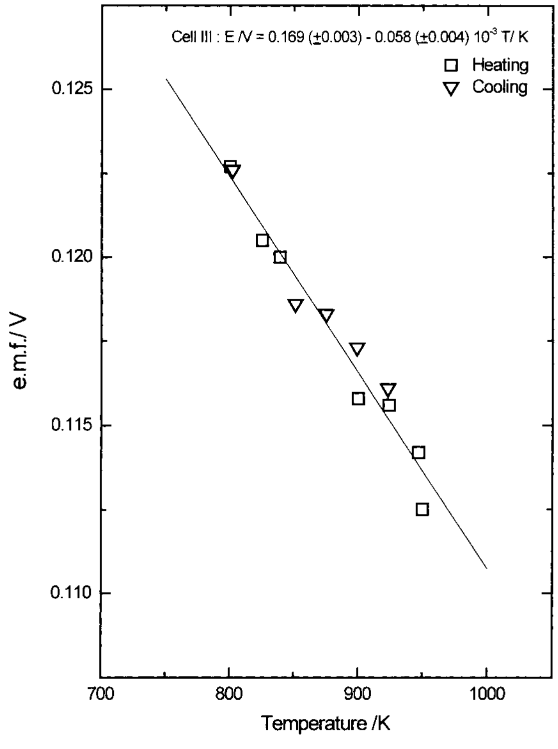

Fig. 4. Variation of EMF of the cell III with temperature.

The standard Gibbs free energy of formation of U_2Ga_3 was calculated using the following equation:

$$\Delta G^0_{\mathrm{f,U_2Ga_3}} = \tfrac{1}{2}(3\,\Delta G^0_{\mathrm{f,UGa_2}} - zFE_{\mathrm{III}}) \tag{9}$$

with the Gibbs free energy of formation of UGa_2 computed using cell II and the EMF of the cell III, (E_{III}) which gives

$$\Delta G^0_{\mathrm{f,U_2Ga_3}} = -231.4 + 0.086T(\pm 4.5)\,\mathrm{kJ\,mol^{-1}} \tag{10}$$

The enthalpy and entropy of formation of U_2Ga_3 are, thus, $-231.4\,\mathrm{kJ\,mol^{-1}}$ and $86\,\mathrm{J\,mol^{-1}\,K^{-1}}$, respectively.

The partial molar excess Gibbs free energy of the uranium in the saturated solution can be computed from the EMF data of cell I and the variation of the solubility of uranium in liquid gallium [10]. The partial molar excess Gibbs free energy calculated is given by

$$\Delta \overline{G}^{\mathrm{xs}}_{\mathrm{U}} = -103.2 \pm 0.034T(\pm 1.8)\,\mathrm{kJ\,gatom^{-1}}$$

The partial molar enthalpy and the partial molar excess entropy of uranium in gallium in the temperature range 693–922 K are $-103.2\,\mathrm{kJ\,gatom^{-1}}$ and $33.8\,\mathrm{J\,gatom^{-1}\,K^{-1}}$, respectively.

4. Discussion

The Gibbs free energies of formation of UGa_3 with reference to α-uranium and liquid gallium computed from our EMF measurements are shown in Table 4 along with the values reported by others. These are also plotted in Fig. 5 as a function of temperature. Lebedev et al. [4] have reported a change in slope of EMF versus temperature plot at 920 K which they attributed to a polymorphic transition in UGa_3. They have reported two equations for the temperature dependence of EMF: one for temperatures below 920 K and the other for the higher temperatures. The lines Lebedev-a $(-42.1 + 0.014T)$ and Lebedev-b $(-47.3 + 0.020T)$ indicated in Fig. 5 are for the temperature range 692–942 and 920–1084 K, respectively. However, as noted by Chiotti et al. [6], the direction of the change in slope is opposite to that

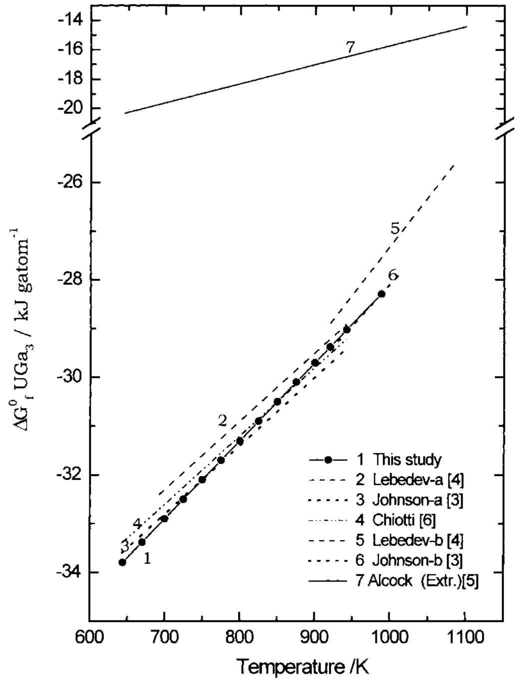

Fig. 5. Variation of $\Delta G^0_f(UGa_3)$ with temperature.

Table 4
Gibbs free energy of formation of UGa_3

Compound	ΔG^0_f (kJ gatom^{-1})	Method	T (K)	Reference
UGa_3 (w.r.t. α-U, liquid Ga)	$-44.1 + 0.016T$ (K) (±0.5)	EMF	644–988	This study
	$-42.1 + 0.014T$ (K)	EMF	696–942	Lebedev et al. [4]
	$-42.6 + 0.014T$ (K)	EMF	643–942	Johnson and Feder [3]
	$-28.7 + 0.013T$ (K)	Vapour pressure	1280–1530	Alcock et al. [5]
	$-42.4 + 0.014T$ (K)	Assessment	298–942	Chiotti et al. [6]
UGa_3 (w.r.t. α-U, liquid Ga)	$-30.5(\pm 0.5)$	EMF	850	This study
	-30.2	EMF	850	Lebedev et al. [4]
	-30.5	EMF	850	Johnson and Feder [3]
	-17.6 (extrapolated)	Vapour pressure	850	Alcock et al. [5]
	-30.3	Assessment	850	Chiotti et al. [6]

Table 5
Gibbs free energy of formation of UGa_2

Compound	ΔG_f^0 (kJ gatom^{-1})	Method	T (K)	Reference
UGa_2 (w.r.t. α-U, liquid Ga)	$-46.0 + 0.017T$ (K) (± 1.0)	EMF	751–947	This study
	$-28.6 + 0.010T$ (K)	Vapour pressure	1280–1530	Alcock et al. [5]
	$-46.9 + 0.012T$ (K)	Assessment	298–942	Chiotti et al. [6]
UGa_2 (w.r.t. α-U, liquid Ga)	-31.6 (± 1.0)	EMF	850	This study
	-20.1 (extrapolated)	Vapour pressure	850	Alcock et al. [5]
	-36.7	Assessment	850	Chiotti et al. [6]

normally expected for such a transition besides being too large to be accounted by the α to β transition in the uranium electrode, if any. Hence, Chiotti et al. [6] have taken into account only the lower temperature equation of Lebedev et al. [4] for temperatures up to 942 K, the α to β transition temperature of uranium. We have also used only this recommended equation for comparison. In the case of the data of Johnson and Feder [3] who had given a four term equation for the Gibbs free energy of formation of UGa_3, Chiotti et al. [6] observed that the equation shows a large change in curvature below 942 K implying a large ΔC_p value for temperatures below 942 K. The two fitted lines Johnson-a $(-42.6 + 0.014T)$ and Johnson-b $(-62.0 + 0.086T - 0.000088T^2 + 0.000000036T^3)$ are shown in Fig. 5 for the temperature range 643–942 and 920–1013 K, respectively. Chiotti et al. [6] have recommended a two term equation for the Gibbs free energies of formation data of Johnson and Feder [3] which is shown in Table 4 and used for comparison. As can be seen from Table 4 and Fig. 5, our values for the Gibbs free energy of formation of UGa_3 obtained from the molten salt galvanic cell measurements agree very well with the data of Johnson and Feder [3] and Lebedev et al. [4]. The mean of the values of Lebedev et al. and Johnson and Feder has been recommended by Chiotti et al. [6], which is also shown in Table 4. We have not observed any change in slope at 920 K as reported by Lebedev et al. [4]. No change of slope was observed also at 942 K, the transformation temperature of $U(\alpha) \rightarrow U(\beta)$ which could be due to the enthalpy of the transition being low as well as to the limited number of experimental data above 942 K in our study. The Gibbs free energy of formation of UGa_3 at 850 K obtained in this study is also compared with those at 850 K computed from literature data in Table 4. The value obtained by this study is in very good agreement with the values from the other EMF measurements, except the extrapolated data of Alcock et al. from vapour pressure measurements The latter are unreliable for the reasons suggested by the authors themselves, i.e., that there could be errors in their vapour pressure data due to the possible presence of volatile gallium oxide species.

The present data for the Gibbs free energy of formation of UGa_2 are slightly less negative than the recommended value of Chiotti et al. [6] as shown in Table 5. The Gibbs free energy of formation of UGa_2 of Alcock et al. [5], which is much less negative than the present value, is considered unreliable for reasons cited earlier. The Gibbs free energy of formation of U_2Ga_3 obtained by the present galvanic cell measurements is shown along with the literature data in Table 6. The Gibbs free energies of formation of U_2Ga_3 at 1250 K computed from the expressions given in Table 6 are also compared in the same table. Since the temperature ranges of the vapour pressure measurements by Gardie et al. [7] as well as by Alcock et al. [5] are much higher than that of the present study, our data were extrapolated to 1250 K for comparison. It can be seen that our free energy data is in very good agreement with the data of Gardie et al. [7]. Our data is more negative than that of Alcock et al. [5] but less negative than the assessed value of Chiotti et al. [6]. It has to be noted that our Gibbs free energy of formation is in agreement with that of Gardie et al. [7], who have used a different technique for measurements at high temperature.

The partial molar excess Gibbs free energy and the partial molar enthalpy of uranium in liquid gallium obtained from the present study are compared with the literature data in Table 7. The partial molar enthalpy of

Table 6
Gibbs free energy of formation of U_2Ga_3

Compound	ΔG_f^0 (kJ gatom^{-1})	Method	T (K)	Reference
U_2Ga_3 (w.r.t. α-U, liquid Ga)	$-46.3 + 0.017T$ (K) (±1.0)	EMF	800–950	This study
	$-20.5 + 0.004T$ (K)	Vapour pressure	1280–1530	Alcock et al. [5]
	$-47.3 + 0.010T$ (K)	Assessment	298–942	Chiotti et al. [6]
	$-35.3 + 0.010T$ (K)	Vapour pressure	1100–1670	Gardie et al. [7]
U_2Ga_3 (w.r.t. γ-U, liquid Ga)	−24.3 (±1.0) (extrapolated)	EMF	1250	This study
	−17.1	Vapour pressure	1250	Alcock et al. [5]
	−33.6 (extrapolated)	Assessment	1250	Chiotti et al. [6]
	−22.4 (±0.8)	Vapour pressure	1250	Gardie et al. [7]

Table 7
Partial molar excess Gibbs free energy and partial molar enthalpy of U in liquid Ga in the saturated solution

$-\Delta\overline{G}_U^{xs}$ (kJ gatom^{-1})	$-\Delta\overline{H}_U$ (kJ gatom^{-1})	T (K)	Method	Reference
–	95.5 ± 4.3[a]	1408	Ga solution calorimetry	Previous work [8]
75.5 ± 1.8	103.2 ± 1.8	816	EMF	This study
74.5	95.4	805	EMF	Lebedev et al. [4]
73.6	119.7	823	EMF	Johnson and Feder [3]
74.5	96.3	808	Assessment	Chiotti et al. [6]

[a] At infinite dilution.

uranium in the saturated solution computed from EMF measurements is only slightly more exothermic than the partial molar enthalpy of uranium at infinite dilution obtained form solution calorimetry [11]. This shows that the activity coefficient of uranium does not vary much from infinite dilution to the saturation region and is only weakly dependent on concentration. The present value for partial molar enthalpy in the saturated solution is more exothermic than that of Lebedev et al. [4] and the assessed value of Chiotti et al. [6], but less exothermic than that of Johnson and Feder [3]. The partial molar excess Gibbs free energy computed from this study is in very good agreement with the previous data of Lebedev et al. [4], Johnson and Feder [3] and Chiotti et al. [6].

5. Conclusions

The Gibbs free energy of formation of UGa_2 and U_2Ga_3 were determined for the first time by using molten salt galvanic cell measurements.

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