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Calorimetric investigations of liquid Ge–Te and Si–Te alloys

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Abstract

The heats of solution of Ge and Te in liquid Ge–Te alloys and of Si in liquid Si–Te alloys were determined in a heat flow calorimeter at different temperatures. The excess enthalpies were calculated from the measured heat effects. In addition the heats of formation of liquid Ge–Te alloys were measured at 1203 K. The associate model was used to describe the thermodynamic functions. Minima of the enthalpy curves in the Ge–Te and Si–Te systems at 1213 K were found at 57 mol.% Te and $-11.2\,\mathrm{kJ\,mol^{-1}}$ and at 62 mol.% Te and $-22.3\,\mathrm{kJ\,mol^{-1}}$ respectively. A temperature dependence of the excess enthalpies was only observed in the Ge–Te system below about 1093 K. The enthalpies of the Si–Te system exhibited an almost linear temperature dependence.

Keywords: Thermodynamics; Calorimetric investigations; Ge–Te alloys; Si–Te alloys

1. Introduction

Ge–Te alloys have promising technological properties, e.g. they are fast switching, reversible, phase-change optical data storing media [1]. For industrial processes thermodynamic data of this system are of interest. The system Ge–Te contains the compound GeTe which melts congruently at 997 K and two eutectics: in the Ge–GeTe part at 49.85 mol.% Te and 993 K, in the GeTe–Te part at 85 mol.% and 658 K. GeTe occurs in three modifications, α-GeTe at ambient and β-GeTe at high temperatures. Below 638 K and at about 51 mol.% Te the γ form appears [2]. Numerous investigations of physical and thermodynamic properties are reported in the literature. The enthalpies of formation from 1028 to 1150 K [3] and heat capacities of liquid and solid alloys were determined by Bergman and Castanet [4,5] and Tsuchiya [6]. The excess enthalpies at 1250 K were measured by quantitative differential thermal analysis by Al'fer et al. [7], the magnetic susceptibilities by Terzieff et al. [8] and viscosities by Herwig and Wobst [9]. Neutron and X-ray diffraction experiments were carried out by Neumann et al. [10,11]. All experiments revealed an anomalous temperature dependence of these properties just above the melting temperature and change to normal behaviour at higher temperatures.

The phase diagram Si–Te contains the compound Si_2Te_3, which melts incongruently at 1158 K, and a eutectic at about 85 mol.% Te and 1158 K [2]. No thermodynamic data of melts are available.

Within a systematic investigation of ternary metal–IVb–Te systems [12], we have measured the heats of solution of Ge in Ge_{i-y}–Te_y alloys, of Si in Si_{1-y} alloys at different temperatures and of Te in liquid Ge_{1-y}–Te_y alloys at 1213 K. Furthermore the heats of formation of liquid Ge_{1-y}–Te_y alloys from solid Ge and liquid Te were determined at 1203 K.

2. Experimental

The measurements were carried out in a heat flow calorimeter which has been described elsewhere [13]. Two different heat effects were measured.

(1) *Heats of solution.* (a) Pieces of solid Ge (99.999%, ABCR) or Si (99%, Schuchardt) were consecutively dropped into a quartz tube (diameter, 15 mm), which was placed in the calorimeter and filled with liquid Ge_{1-y}–Te_y or Si_{1-y}–Te_y alloys. The measured heat effects correspond to the reactions

$$z\mathrm{Ge}(\mathrm{s}, 298\ \mathrm{K}) + (1-z)\mathrm{Ge}_{1-y}\mathrm{Te}_y(\mathrm{l}, T) \xrightarrow{\Delta H^{\mathrm{sol}}} \mathrm{Ge}_{(1-y)(1-z)+z}\mathrm{Te}_{y(1-z)}(\mathrm{l}, T) \tag{1a}$$

or

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$$z\mathrm{Si}(\mathrm{s}, 298\,\mathrm{K}) + (1-z)\mathrm{Si}_{1-y}\mathrm{Te}_y(\mathrm{l}, T) \xrightarrow{\Delta H^{\mathrm{sol}}} \mathrm{Si}_{(1-y)(1-z)+z}\mathrm{Te}_{y(1-z)}(\mathrm{l}, T) \qquad (1\mathrm{b})$$

These experiments were performed at 973, 1033, 1093, 1153 and 1213 K for Ge–Te alloys and at 973, 1093 and 1213 K for Si–Te alloys. (b) Pieces of solid Te (99.999%, Strem Chemicals) were consecutively dropped into liquid Ge_{1-y}–Te_y alloys at 1213 K. The heat effects correspond to the reaction

$$z\mathrm{Te}(\mathrm{s}, 298\,\mathrm{K}) + (1-z)\mathrm{Ge}_{1-y}\mathrm{Te}_y(\mathrm{l}, 1213\,\mathrm{K}) \xrightarrow{\Delta H^{\mathrm{sol}}} \mathrm{Ge}_{(1-y)(1-z)}\mathrm{Te}_{y(1-z)+z}(\mathrm{l}, 1213\,\mathrm{K}) \qquad (1\mathrm{c})$$

(2) *Heats of formation.* A quartz tube (diameter 15 mm) with solid germanium was placed in the calorimeter. A second thin-walled quartz tube (diameter 8 mm) was inserted into the first tube. It was filled with liquid tellurium which was covered by a liquid eutectic mixture of LiCl–KCl. Mixing of the components was achieved by breaking a tip at the end of the inner rube. The heat effect in these experiments was caused by the reaction

$$(1-x)\mathrm{Ge}(\mathrm{s}, 1203\,\mathrm{K}) + x\mathrm{Te}(\mathrm{l}, 1203\,\mathrm{K}) \xrightarrow{\Delta H^{\mathrm{for}}} \mathrm{Ge}_{1-x}\mathrm{Te}_x(\mathrm{l}, 1203\,\mathrm{K}) \qquad (2)$$

These measurements were made at 1203 K in the Ge–Te system between 40 and 81 mol.% Te.

Calibrations were performed before and after each measurement in case of experiments 1a and 1b and after the measurement in case of experiment 2, by dropping Sn (99.999%, Preussag) from ambient temperature (298 K) into a quartz tube (diameter, 6 mm), which was located in the melt.

From the measured heat effects and the enthalpy increments $\Delta H(T\text{–}298\,\mathrm{K})$ of Te, Ge and Si and the enthalpy of melting of Ge, which was assumed to be independent of the temperature, we calculated the excess enthalpies of the reaction

$$(1-x)(\mathrm{Ge\ or\ Si})(\mathrm{l}, T) + x\mathrm{Te}(\mathrm{l}, T) \rightarrow (\mathrm{Ge\ or\ Si})_{1-x}\mathrm{Te}_x(\mathrm{l}, T)$$

The enthalpy increments $\Delta H(T\text{–}298\,\mathrm{K})$ of Ge and Si, which were extrapolated to the measuring temperature, and of Sn and the enthalpy of melting of Ge were taken from Barin [14]. The enthalpy increment $\Delta H(1213\,\mathrm{K}\text{–}298\,\mathrm{K})$ of Te was measured in separate experiments by dropping Te into liquid Te. The value was 45 300 J mol^{-1} (48 860 J mol^{-1} [14]) and is specific for the calorimeter. All procedures were carried out in argon atmosphere. The reproducibility of the cali-

Table 1
Experimental excess enthalpies of liquid Ge–Te alloys, according to reactions (1a) and (1c) at 1213 K

Reaction (1a)				Reaction (1c)			
n_{Ge}	ΔQ (J)	x_{Te}	H^{E} (J mol^{-1})	n_{Te}	ΔQ (J)	x_{Te}	H^{E} (J mol^{-1})
Starting amount: $n_{\mathrm{Te}} = 0.02345$				Starting amount: $n_{\mathrm{Ge}} = 0.02755$			
0.00208	46.7	0.919	−3120	0.00230	69.8	0.077	−1150
0.00298	89.1	0.882	−6030	0.00227	73.4	0.142	−1990
0.00275	89.2	0.750	−7990	0.00278	72.1	0.211	−3370
0.00428	161.8	0.660	−9780	0.00319	78.4	0.277	−4820
0.00226	95.8	0.620	−10290	0.00374	85.7	0.341	−6400
				0.00384	78.4	0.397	−7950
				0.00371	78.3	0.442	−9170
				0.00383	85.9	0.482	−10150
Starting amount: $n_{\mathrm{Te}} = 0.02345$				Starting amount: $n_{\mathrm{Ge}} = 0.02094$			
0.00096	15.1	0.961	−1760	0.00140	48.9	0.062	−640
0.00216	59.8	0.884	−4250	0.00198	60.0	0.139	−1800
0.00337	108.0	0.786	−6960	0.00279	73.1	0.227	−3590
0.00267	90.5	0.723	−8560	0.00320	74.8	0.309	−5520
0.00341	139.5	0.655	−9620	0.00293	67.4	0.370	−7000
Starting amount: $n_{\mathrm{Te}} = 0.01978$				Starting amount: $n_{\mathrm{Ge}} = 0.02066$			
0.00090	11.2	0.956	−2110	0.00154	51.4	0.069	−820
0.00198	54.6	0.873	−4830	0.00228	73.3	0.156	−1960
0.00309	95.6	0.768	−7810	0.00271	71.6	0.240	−3650
0.00326	114.7	0.682	−9810	0.00324	75.0	0.321	−5620
0.00201	79.7	0.638	−10550	0.00351	76.1	0.391	−7480
0.00200	89.0	0.599	−10900	0.00376	79.2	0.452	−9150
0.00201	101.2	0.565	−10870	0.00357	95.6	0.499	−9970
				0.00270	86.8	0.530	−10160
				0.00261	76.7	0.556	−10480

bration was better than ±3%. The total error on the excess enthalpies is estimated to be ±7%.

3. Results

The amounts of the components, heat effects ΔQ, mole fractions and the calculated excess enthalpies for Ge–Te are given in Tables 1 and 2 and for the Si–Te system in Table 3. The enthalpies of formation of liquid Ge–Te alloys are presented in Table 4.

The values of the excess enthalpy and the enthalpy of formation were fitted by means of the associate model [15]. The excess enthalpy per mole atoms is given by

$$H^{\mathrm{E}} = n_{\mathrm{AB}} H^0_{\mathrm{AB}} + \frac{n_{\mathrm{A}} n_{\mathrm{B}}}{n} C^{\mathrm{reg}}_{\mathrm{A,B}} + \frac{n_{\mathrm{A}} n_{\mathrm{AB}}}{n} C^{\mathrm{reg}}_{\mathrm{A,AB}} + \frac{n_{\mathrm{B}} n_{\mathrm{AB}}}{n} C^{\mathrm{reg}}_{\mathrm{B,AB}}$$

with $n = n_{\mathrm{A}} + n_{\mathrm{B}} + n_{\mathrm{AB}}$. The entropy of mixing is given by the equation

$$\Delta S = -R(n_{\mathrm{A}} \ln x_{\mathrm{A}} + n_{\mathrm{B}} \ln x_{\mathrm{B}} + n_{\mathrm{A}_i\mathrm{B}_j} \ln x_{\mathrm{A}_i\mathrm{B}_j}) + n_{\mathrm{A}_i\mathrm{B}_j} \Delta S^0_{\mathrm{A}_i\mathrm{B}_j}$$

in which $\Delta H^0_{\mathrm{A}_i\mathrm{B}_j}$ and $\Delta S^0_{\mathrm{A}_i\mathrm{B}_j}$ are the enthalpy and entropy of formation of the associate $\mathrm{A}_i\mathrm{B}_j$ and $C^{\mathrm{reg}}_{\mathrm{A,B}}$, $C^{\mathrm{reg}}_{\mathrm{A,AB}}$ and $C^{\mathrm{reg}}_{\mathrm{B,AB}}$ the parameters of the regular interactions between the different species.

The numbers n_{AB} of moles in the equilibrium $i\mathrm{A} + j\mathrm{B} \rightleftharpoons \mathrm{A}_i\mathrm{B}_j$ were determined from the mass action law:

$$K_{\mathrm{A}_i\mathrm{B}_j} = \frac{\gamma_{\mathrm{A}_i\mathrm{B}_j} x_{\mathrm{A}_i\mathrm{B}_j}}{(\gamma_{\mathrm{A}} x_{\mathrm{A}})^i (\gamma_{\mathrm{B}} x_B)^j} = \exp\left(\frac{\Delta H^0_{\mathrm{A}_i\mathrm{B}_j} - T\,\Delta S^0_{\mathrm{A}_i\mathrm{B}_j}}{RT}\right)$$

The parameters were derived using the program BINGSS developed by Lukas and colleagues [16,17], assuming "GeTe" associates in the Ge–Te system and "$\mathrm{Si_2Te_3}$" associates in the Si–Te system. These assumptions seem to be justified, because they correspond to the composition of the compounds in the solid state. The parameters used for the calculations with the associate model are given in Table 5. The

Table 2
Excess enthalpies of liquid Ge–Te alloys, according to reaction (1a) at different temperatures

n_{Ge}	ΔQ (J)	x_{Te}	H^{E} (J mol^{-1})	n_{Ge}	ΔQ (J)	x_{Te}	H^{E} (J mol^{-1})
$T = 973$ K				$T = 1033$ K			
Starting amount: $n_{\mathrm{Te}} = 0.01991$				Starting amount: $n_{\mathrm{Ge}} = 0.01959$			
0.00135	16.7	0.937	−2640	0.00151	25.9	0.928	−2760
0.00243	41.0	0.841	−6190	0.00227	50.0	0.838	−5770
0.00263	47.8	0.756	−9130	0.00252	62.0	0.757	−8250
0.00351	73.7	0.667	−11950	0.00288	73.2	0.681	−10450
0.00286	64.5	0.609	−13660	0.00353	103.0	0.606	−12220
0.00204	47.7	0.573	−14670	0.00439	175.6	0.534	−12650
Starting amount: $n_{\mathrm{Te}} = 0.02014$				Starting amount: $n_{\mathrm{Ge}} = 0.02307$			
0.00232	37.5	0.897	−3910	0.00125	19.9	0.949	−2040
0.00296	58.1	0.793	−7470	0.00204	47.6	0.875	−4390
0.00322	65.9	0.703	−10410	0.00261	58.5	0.797	−6990
0.00274	59.1	0.642	−12340	0.00282	71.0	0.726	−9080
0.00262	59.1	0.593	−13830	0.00317	85.8	0.660	−10870
0.00204	37.8	0.559	−15060	0.00385	117.9	0.595	−12280
				0.00416	158.3	0.537	−12810
$T = 1093$ K				$T = 1153$ K			
Starting amount: $n_{\mathrm{Te}} = 0.02324$				Starting amount: $n_{\mathrm{Te}} = 0.02249$			
0.00282	59.6	0.892	−3930	0.00285	64.3	0.887	−4110
0.00413	113.3	0.770	−7500	0.00331	96.1	0.785	−7120
0.00458	140.1	0.668	−10050	0.00312	155.9	0.708	−9260
0.00388	142.1	0.601	−11130	0.00452	128.2	0.620	−11170
				0.00284	128.2	0.575	−11370
Starting amount: $n_{\mathrm{Te}} = 0.02288$							
0.00135	22.8	0.944	−2250	Starting amount: $n_{\mathrm{Te}} = 0.02346$			
0.00224	58.2	0.864	−4730	0.00406	101.1	0.853	−5040
0.00220	60.7	0.798	−6640	0.00454	138.1	0.732	−8380
0.00256	76.1	0.733	−8380	0.00354	117.5	0.659	−10120
0.00288	88.2	0.671	−9920	0.00334	128.5	0.603	−11011
0.00350	123.7	0.609	−11050				
0.00324	138.9	0.560	−11330				

Table 3
Experimental excess enthalpies of liquid Si–Te alloys, according to reaction (1b), at different temperatures

n_{Si}	ΔQ(J)	x_{Te}	H^E (J mol^{-1})
$T = 973$ K			
Starting amount $n_{Te} = 0.02868$			
0.00220	−14.1	0.929	−5320
0.00264	−35.6	0.856	−11330
0.00318	−17.1	0.782	−16730
Starting amount $n_{Te} = 0.02866$			
0.00095	−7.5	0.968	−2450
0.00209	−24.7	0.904	−7560
0.00236	−13.4	0.841	−12160
$T = 1093$ K			
Starting amount $n_{Te} = 0.02625$			
0.00167	−12.6	0.940	−4730
0.00315	4.7	0.845	−11330
0.00287	0.5	0.773	−16390
0.00317	11.7	0.707	−20770
Starting amount $n_{Te} = 0.02665$			
0.00260	−10.6	0.911	−6700
0.00308	−22.6	0.824	−13560
0.00305	8.5	0.753	−18290
0.00262	13.8	0.701	−21590
0.00137	11.5	0.667	−23020
$T = 1213$ K			
Starting amount $n_{Te} = 0.02333$			
0.00142	10.1	0.943	−3860
0.00219	25.6	0.866	−8640
0.00370	4.9	0.761	−16430
0.00332	29.1	0.687	−21240
Starting amount $n_{Te} = 0.02770$			
0.00182	21.4	0.938	−3860
0.00300	35.1	0.862	−8470
0.00388	10.8	0.770	−15270
0.00331	21.6	0.705	−19700
0.00221	32.0	0.667	−21830
0.00246	40.7	0.630	−23850

Table 4
Enthalpies of formation of liquid Ge–Te alloys with respect to solid Ge and liquid Te, according to reaction (2) at 1203 K and calculated excess enthalpies

x_{Te}	H^{for} (J mol^{-1})	H^E (J mol^{-1})
0.404	13500	−8510
0.447	10700	−9710
0.477	9130	−10180
0.484	7820	−11240
0.493	7490	−11270
0.505	6800	−11490
0.506	6780	−11470
0.519	6440	−11340
0.533	5660	−11610
0.548	5110	−11460
0.567	4810	−11200
0.589	4080	−11120
0.601	3870	−10850
0.608	4060	−10430
0.639	3220	−10120
0.721	1790	−8510
0.807	1600	−5530

experimental excess enthalpies and the calculated curves are presented in Fig. 1 for the Ge–Te system and in Fig. 2 for the Si–Te system. Broken lines indicate the crossing of two-phase regions. The minima of the fitted curves at 1213 K are positioned at 57 mol.% Te and −11.2 kJ mol^{-1} in the Ge–Te system and at 62 mol.% Te and −22.3 kJ mol^{-1} in the Si–Te system. These values in the Ge–Te system are less exothermic, compared with those of Al'fer et al. [7] at 1250 K. The calculated concentration dependences of ΔG and the $T\,\Delta S$ term at 1213 K are shown for both systems in Figs. 3 and 4. The enthalpies of formation of Ge–Te melts are depicted in Fig. 5, together with values measured by Bergman and Castanet [3].

4. Discussion

The excess enthalpies of the liquid Ge–Te and Si–Te alloys are exothermic and vary as triangularly shaped functions of the concentration. This unusual behaviour was explained by Wagner [18], who assumed the formation of associates in the liquid state by either ionic or covalent interactions of the components. The composition of these associates is given by the position of the minimum in the enthalpy curve, i.e. GeTe in the Ge–Te and Si_2Te_3 in the Si–Te system.

The enthalpy curve of the Ge–Te system exhibits a positive deviation from the ideal triangular shape in the metal-rich part of the system. This effect is due to endothermic interactions between Ge and the associate GeTe, caused by the different bond character of Ge and the associate. In the Te-rich part of both systems the curves exhibit negative deviations caused by exothermic interactions between the associate and Te. This exothermic effect reflects the tendency of telurium to form polytellurides. These interactions are also indicated by a positive value for C_{A,A_iB_j} in the Ge–Te system and negative values for C_{B,A_iB_j} in both systems (Table 5). Contributions of the latter effects to the excess enthalpies are small, because the enthalpy of formation of the associates is dominating. The calculated mole fractions of the associates are shown in Fig. 6. as a function of the composition of the melt.

A temperature dependence of the excess enthalpies was observed below about 1093 K in the Ge–Te system. Above this temperature the excess enthalpies remain almost constant (Fig. 1). The experimental values from 1093 to 1213 K can be approximated by the same curve, while the excess enthalpies decrease in the range 973–1093 K. This in agreement with the results reported by Castanet and Bergman [4,5], who derived large positive excess heat capacities of melts

Table 5
Interaction parameters of the associate model

	i, j	$\Delta H^0_{A_iB_j}$ (kJ mol^{-1})	$\Delta S^0_{A_iB_j}$ (J mol^{-1} K^{-1})	$C^{reg}_{A,B}$ (kJ mol^{-1})	C^{reg}_{A,A_iB_j} (kJ mol^{-1})	C^{reg}_{B,A_iB_j} (kJ mol^{-1})
Ge–Te	1, 1	−37.6	−17.2	–	13.9	−1.4
Si–Te	2, 3	−150.1	−86.1	–	–	−6.8

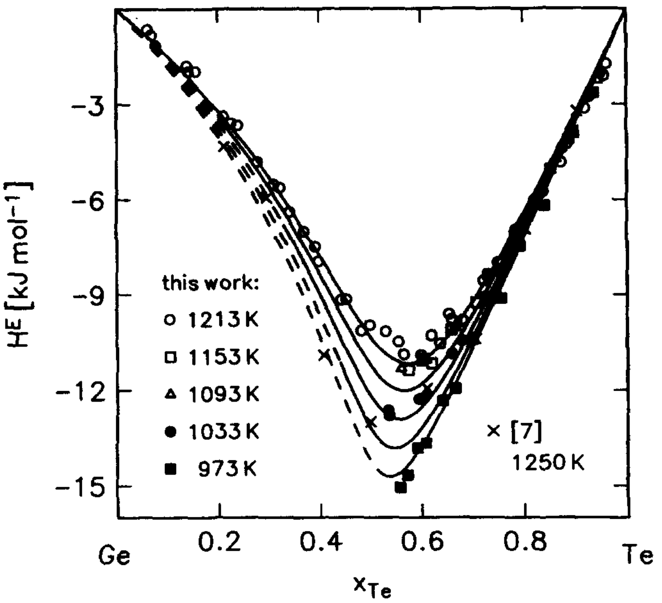

Fig. 1. Experimental (points) and calculated (lines) excess enthalpies of liquid Ge–Te alloys at different temperatures (-----, two-phase region).

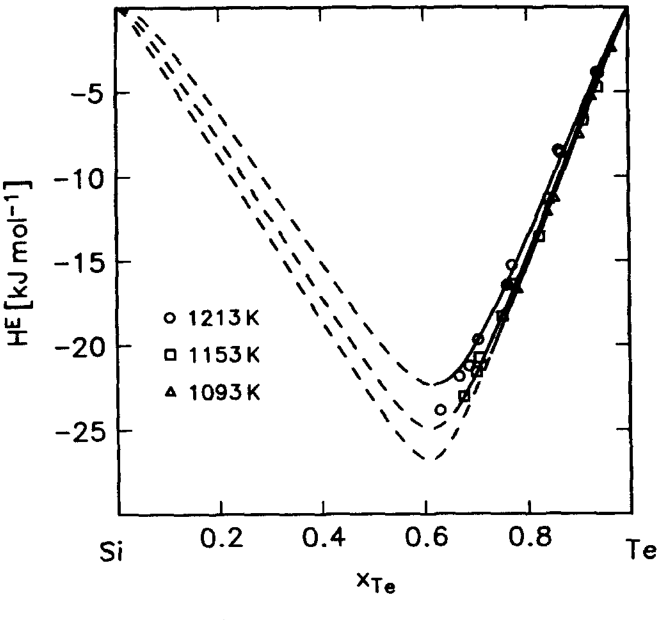

Fig. 2. Experimental (points) and calculated (lines) excess enthalpies of liquid Si–Te alloys at different temperatures (-----, two-phase region).

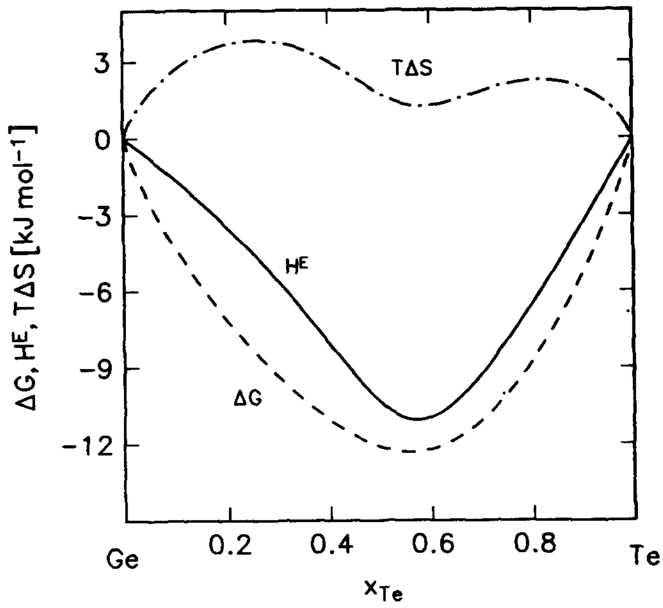

Fig. 3. Calculated thermodynamic functions ΔG, H^E and $T\Delta S$ of the Ge–Te system at 1213 K.

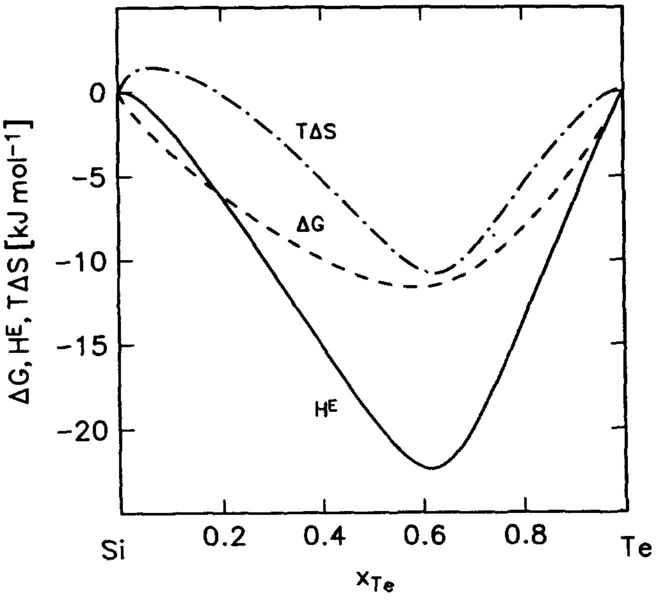

Fig. 4. Calculated thermodynamic functions ΔG, H^E and $T\Delta S$ of the Si–Te system at 1213 K.

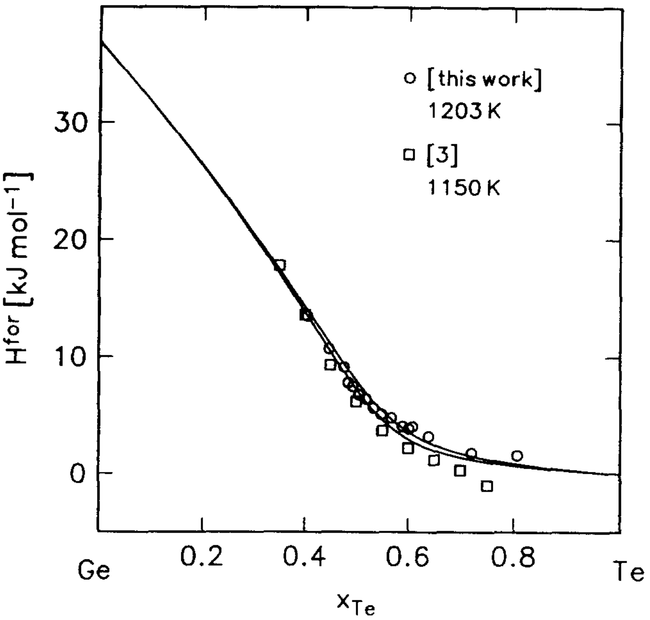

Fig. 5. Experimental (points) and calculated (lines) enthalpies of formation of liquid Ge–Te alloys, referred to solid Ge and liquid Te.

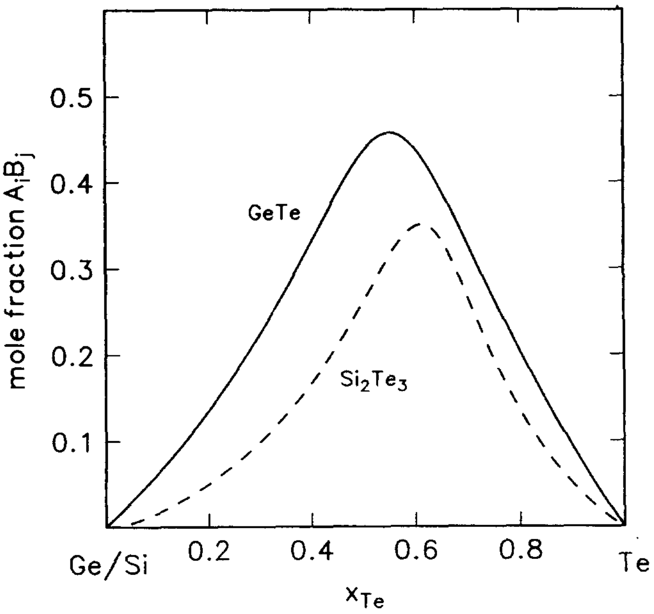

Fig. 6. Calculated concentration dependences of the mole fraction of the associates “GeTe” and “SiTe$_3$” at 1213 K.

above the melting temperature, which decrease with increasing temperatures. Assuming only one type of associate this temperature dependence cannot be described satisfactorily by the associate model. The effect indicates that in melts of the Ge–Te system in addition to formation of associates local order similar to that of the crystalline GeTe is preserved above the melting temperature. These mainly covalent open clusters are destroyed with increasing temperature and a solution with a more metallic bonding is formed [19].

The excess enthalpies of the Si–Te system vary linearly with temperature (Fig. 2). The positive $T\Delta S$ term of the Ge–Te system has a minimum at 50 mol.% Te. The $T\Delta S$ term of the Si–Te system (Fig. 4) exhibits a large negative minimum of the excess entropy at 60 mol.% Te, also revealing the strong tendency to form associates.

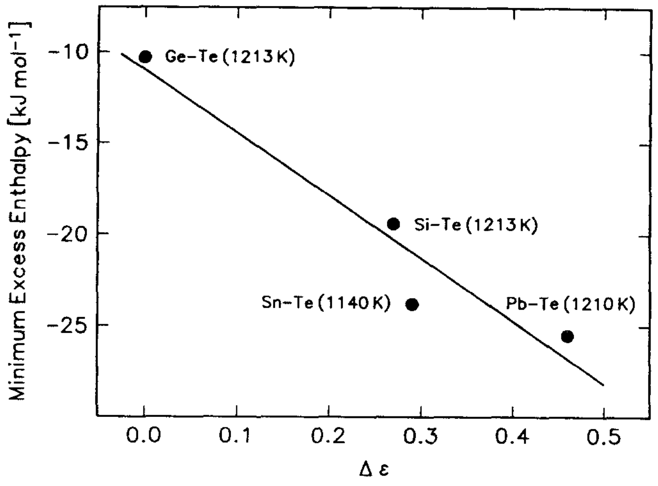

Fig. 7. Minima of the excess enthalpy curves of IVB–Te systems vs. the electronegativity difference $\Delta\epsilon$ of the components.

Associates in the melts of many metal-chalcogen systems have a partically ionic bond character. In Fig. 7 the minima of the excess enthalpies of IVb–Te systems are plotted vs. the difference in the electronegativities [20] of the two components. As in other chalcogenide systems [21] the figure reveals an obvious correlation between the two parameters.

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