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Memorandum of Project MICHIGAN

# THE PHASE DIAGRAM FOR THE BINARY SYSTEM CADMIUM-TELLURIUM

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Robert L. Hess  
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The Phase Diagram for the Binary System Cadmium-Tellurium<sup>1</sup>

## ABSTRACT

The phase diagram for the cadmium-tellurium system has been redetermined by measuring the thermal behavior of representative compositions in the system by the method of differential thermal analysis. The experimental details and the resulting data are described and discussed. Thermodynamic analysis shows that the system CdTe-Te appears to form an ideal solution. The latent heat of fusion of CdTe is estimated to be 10,700 cal/gm mol. The system Cd-CdTe is an elementary solution and has a relatively large excess partial molar entropy of solution and a large partial molar enthalpy of solution. These partial molar qualities are independent functions of composition, but not of temperature. The eutectic compositions have been determined as  $10^{-6}$  atom-fraction Te and about 0.99 atom-fraction Te. The validity of this work vis-a-vis that of other investigators is discussed.

## 1

## INTRODUCTION

Cadmium telluride is receiving considerable scientific attention because of its interesting semiconductor properties (References 1-5). The Semiconductor Materials Research Laboratory of The University of Michigan has prepared samples of this material in conjunction with studies on ternary semiconductor compounds in the binary system CdTe-In<sub>2</sub>Te<sub>3</sub>. These materials are being measured to ascertain their utility as sensors in infrared systems for battlefield surveillance.

The results obtained in this work showed a serious discrepancy in the widely published melting point of CdTe. In the original work done on this system, Kobayashi (Reference 6) found

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The authors are indebted to Dr. C. D. Thurmond for providing a preprint of his paper referred to in this memorandum.

that CdTe is the only compound formed between these elements, and reported that the two eutectics between the pure elements and CdTe occur very close to the Cd and Te ends of the diagram. The original data indicate that the melting point of CdTe would be in the range of 1041°C to 1050°C, the values commonly reported in the literature (References 7, 8, 9).

More recently, Lawson, Nielsen, Putley, and Young (Reference 4) have reported a value of 1106°C, without describing the details of their measuring technique. DeNobel (Reference 5) has published values for several additional points on the liquidus line in the Cd-Te system, and represents the melting point of CdTe to be 1090°C. Visual observations of the first-to-freeze temperatures were made at each composition reported. In working with this compound in the binary system CdTe-In<sub>2</sub>Te<sub>3</sub>, Thomassen and Mason (Reference 10) observed recently that the melting point of CdTe is 1098 ± 3°C.

In view of these discrepancies, we have redetermined the phase diagram for the Cd-Te system. A series of samples was prepared in which the Cd-Te compositions were varied over the range from 1 mol % Te to 98.7 mol % Te, using the purest available elements for their preparation. The latent heat transitions in each sample were determined by using the method of differential thermal analysis. On the basis of these experiments and subsequent thermodynamic analysis, our work appears to be more precise than that of previous investigators, and has special theoretical significance.

The system CdTe-Te appears to be an ideal solution. The latent heat of fusion of CdTe from these measurements is estimated as 10,700 cal/gm mol. The system Cd-CdTe is an elementary solution, and has a relatively large excess partial molar entropy of solution and a large partial molar enthalpy of solution. These partial molar quantities are independent functions of composition, but not of temperature. The eutectic compositions have been estimated as 10<sup>-6</sup> atom-fraction Te and about 0.99 atom-fraction Te.

## 2

## EXPERIMENTAL PROCEDURES

The experimental operations comprise three categories: the sample preparation, the DTA (differential thermal analysis) equipment and procedures, and the preliminary interpretation of the data.

## 2.1. SAMPLE PREPARATION

The samples for thermal analysis were prepared by weighing stoichiometric amounts of the pure elements (99.99% Cd and 99.999% Te from American Smelting and Refining Company) into



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specially cleaned, clear, fused-silica tubes, and sealing them off under moderate (below  $10^{-4}$  mm Hg) vacuum. The samples were reacted by heating to temperatures well above the liquidus in a rotating, rocking furnace, and were rapidly cooled or air-quenched to room temperature to obtain a homogeneous sample. In many instances the samples wet the fused-silica tube and induced cracks and fractures in the containers during cooling. Only those samples which remained bright, shiny, and unoxidized after fusion were processed further. Each fused sample was crushed as much as possible (which was impossible with the high-cadmium samples) and 15 gm of the material was resealed into a new, specially cleaned, clear, fused-silica tube equipped with a concentric thermocouple well in the bottom end about 1 inch deep.

## 2.2. EQUIPMENT AND PROCEDURES

A schematic diagram of the DTA apparatus is shown in Figure 1. The power input to the DTA furnace is controlled by means of an automatically programmed motor-driven autotransformer. The system is purged with dry nitrogen in order to prevent corrosion of the nickel sample holder, which is provided with three sample wells spaced at  $120^\circ$  intervals. Two specimens may be measured at one time, the third well being occupied by the reference material. The sample temperature is measured by means of a chromel-alumel thermocouple, the output of which is measured on a Leeds and Northrup precision potentiometer. The output of the differential thermocouple (Figure 2) is amplified and recorded using a Leeds and Northrup Speedomax recorder. The thermocouples were calibrated against standards of indium ( $156^\circ\text{C}$ ), lead ( $327^\circ\text{C}$ ), and silver ( $961^\circ\text{C}$ ) and found to agree within  $1^\circ\text{C}$ .

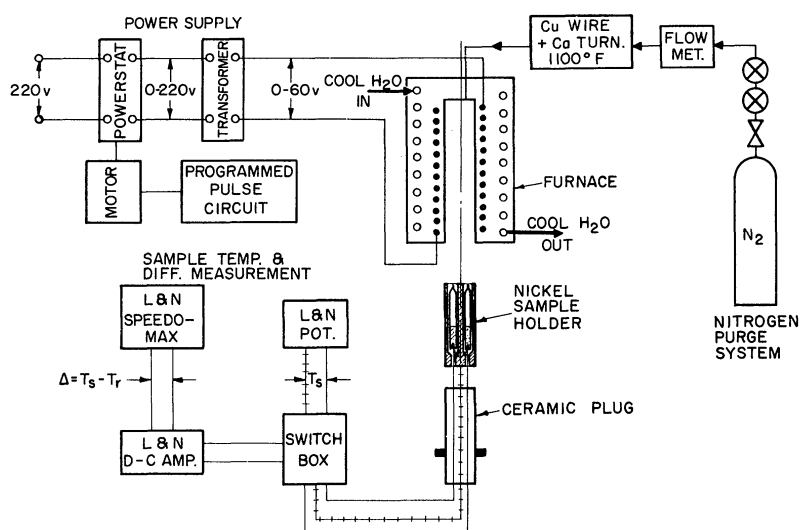


FIGURE 1. SCHEMATIC DIAGRAM OF DTA EQUIPMENT

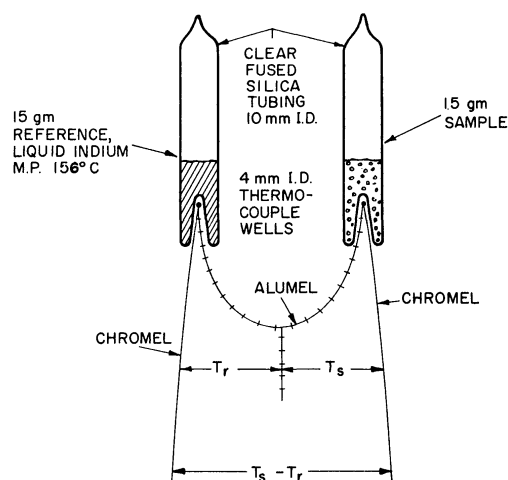


FIGURE 2. DIAGRAM OF DIFFERENTIAL THERMOCOUPLES AND SAMPLE TUBES

The samples were heated and cooled inside the nickel block using a liquid-indium standard, at a rate of about  $2.5^{\circ}\text{C}/\text{min}$  from room temperature to a maximum temperature well above that of the highest transition, and back to room temperature. Although the DTA sample tubes sometimes cracked at low temperatures as the sample contracted around the thermocouple well, they were not wet significantly by the samples, and no apparent oxidation occurred. The sample temperature and the differential emf (electromotive force) were both measured as functions of time, and the data were replotted to show the differential emf as a function of sample temperature.

### 2.3. PRELIMINARY DATA INTERPRETATION

In interpreting the experimental differential curves, the liquidus line was chosen on the heating curves as the point where the differential completed its last deviation. The presence of chunks and large crystals in many of the samples gave rise to erratic deviations in the region of the liquidus, so that the liquidus was relatively difficult to establish. In some instances, considerable supercooling occurred on the cooling cycle, as was evidenced by a large initial differential emf which sometimes actually induced an increase in the sample temperature. In those cases where the initial cooling emf was moderate and/or the heating curves were difficult to interpret, the cooling curves were used to establish the liquidus lines. These results are indicated in Table I. The data of deNobel (Reference 5) and Kobayashi (Reference 6) are summarized in Table II.

TABLE I. EXPERIMENTAL DATA FOR THE Cd-Te PHASE DIAGRAM

Sample Number	Composition Atom %		Maximum Fusion Temperature (°C)	Eutectic (°C)	Liquidus Temperatures		
	Te = y	Cd = 1 - y			Heat (°C)	Cool (°C)	"Chosen" (°C)
---	0	100	---	---	---	---	---
376	1.0	99.0	980	(333)	730	---	321
380	3.0	97.0	970	(340)	845	---	730
379	10.0	90.0	975	322	940	808	808
384	25.0	75.0	1050	323	1000	895	895
385	40.0	60.0	1100	325	1035	963	963
402	45.0	55.0	1150	325	1089	1015*	1035
354	50.0	50.0	1200	---	1100	1067	1067
362	50.0	50.0	1200	---	1100	1099	1098
389	54.0	46.0	1100	---	1097	1096	1098
383	62.5	37.5	1100	450	1075	1052*	1075
375	75.0	25.0	1000	450	1000	950*	1000
382	85.0	15.0	900	450	885	860*	885
381	95.0	5.0	750	449	735	755	755
388	98.2	1.8	850	450	640	602	602
396	98.7	1.3	800	448	480	---	480
006	100	0	---	450	480	---	480
G-1	100	0	---	---	456	452	454
			---	---	454	---	454

\*Considerably supercooled

TABLE II. SUMMARY OF PHASE-DIAGRAM DATA  
AVAILABLE FROM THE LITERATURE

Composition		Eutectic Temperature (°C)	Liquidus Temperature (°C)	Reference
Te = y	Cd = 1 - y			
0	100	322	322	6
0.88	99.12	322	692	6
8.94	91.06	322	---	6
8.94	91.06	---	885	5
18.10	81.90	322	---	6
18.10	81.90	---	940	5
27.45	72.55	321.5	---	6
27.45	72.55	---	974	5
37.00	63.00	321	---	6
37.00	63.00	---	983	5
41.92	58.08	---	1006	5
46.95	53.05	321	---	6
46.95	53.05	---	1036	5
50.07	49.93	---	1090	5
51.90	48.10	---	1068	5
52.10	47.90	366.5	1041.5	6
52.4	47.6	383.5	1032.5	6
56.97	43.03	---	1024	5
60.5	39.5	408	1001	6
67.2	32.8	---	926	5
69.5	30.5	422.5	893	6
77.95	22.05	---	840	5
79.7	20.3	437	815	6
85.8	14.2	437	741.5	6
93.7	6.3	437	604	6
97.2	2.8	437	506.5	6
100	0	437	437	6

## THERMODYNAMIC THEORY OF SOLID-LIQUID EQUILIBRIUM

From thermodynamic analysis of the data, it is possible to obtain an estimate of the latent heat of fusion of CdTe, to establish the form of the liquidus lines on a theoretical basis, to predict the eutectic compositions, and to gain some insight as to the nature of the liquid regions. A more detailed derivation of the thermodynamic equations is presented in the appendix.

For purposes of calculation, the diagram can be considered to be comprised of two separate binaries: (I) the system Cd-CdTe, and (II) the system CdTe-Te.

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The liquidus curve for a binary system which exhibits ideal solution behavior can be represented by the Schroeder-vanLaar equation (Reference 11).

$$\ln x = -\frac{\Delta H_f}{R} \left( \frac{1}{T_f} - \frac{1}{T} \right) \quad (1)$$

where  $x$  = mol fraction of component A in solution at T°K

$T_f$  = melting point of component A (°K)

$R$  = gas constant, 1.987 cal/gm mol °K

$\Delta H_f$  = latent heat of fusion of component A (cal/gm mol)

This equation has been derived by assuming that the system is in equilibrium, that component B does not form a mixed crystal with component A, and that the change in specific heat between liquid and solid ( $\Delta C_p$ ) for component A is negligible or zero. For an ideal solution the values of  $\Delta H_f$  derived from the T vs. x data should be substantially independent of composition.

In nonideal solutions the departures from ideality are characterized by an activity coefficient,  $\gamma$ . By analyzing the liquidus data for a large number of binary systems containing germanium and silicon, Thurmond and Kowalchik (Reference 12) have postulated a new class of solutions, called elementary solutions, wherein the relative partial molar excess entropy and the relative partial molar enthalpy of mixing exhibit a particular dependence on concentration, but are not direct functions of temperature. The relationship between the activity coefficients and the excess thermodynamic functions can be represented by an equation in the form

$$\alpha = \frac{RT \ln \gamma}{(1-x)^2} = \frac{\overline{\Delta H}}{(1-x)^2} - \frac{T \overline{\Delta S}^E}{(1-x)^2} = a - bT \quad (2)$$

where  $a = \frac{\overline{\Delta H}}{(1-x)^2} = \text{constant}$

$b = \frac{\overline{\Delta S}^E}{(1-x)^2} = \text{constant}$

$\overline{\Delta H} = a(1-x)^2$  = partial molar excess enthalpy of mixing of component A in elementary solutions

$\overline{\Delta S}^E = b(1-x)^2$  = partial molar excess entropy of mixing of component A in elementary solutions

$\gamma$  = activity coefficient at composition x

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Since the activity coefficients in the ideal solutions are equal to unity, the partial molar excess enthalpy and entropy are zero.

3.1. CALCULATION OF THE LATENT HEAT OF FUSION,  $\Delta H_f$

Using those liquidus data points from each DTA run which were considered to be the most reliable ("chosen" values), an estimate for the latent heat of fusion of CdTe at each data point was made, using Equation 1. The compound CdTe was chosen as component A. Cadmium was taken as component B in the system I, and tellurium was taken as component B in system II. Define the atom fraction of Te in Cd as  $y$ . Hence in system I

$$x_{CdTe} = 2y \tag{3}$$

and in system II

$$x_{CdTe} = 2(1 - y) \tag{4}$$

Figure 3 presents the results of the calculations on the basis of atom-fraction Te,  $y$ , in Cd. Calculations made from the data of deNobel (Reference 5) and Kobayashi (Reference 6) are also presented for comparative purposes, Kobayashi's data having been calculated on the basis of his reported melting point of 1042°C. On the Te-rich half of the system, the values of  $\Delta H_f/R$  appear to be substantially independent of composition, and ideal solution behavior seems to be indicated.

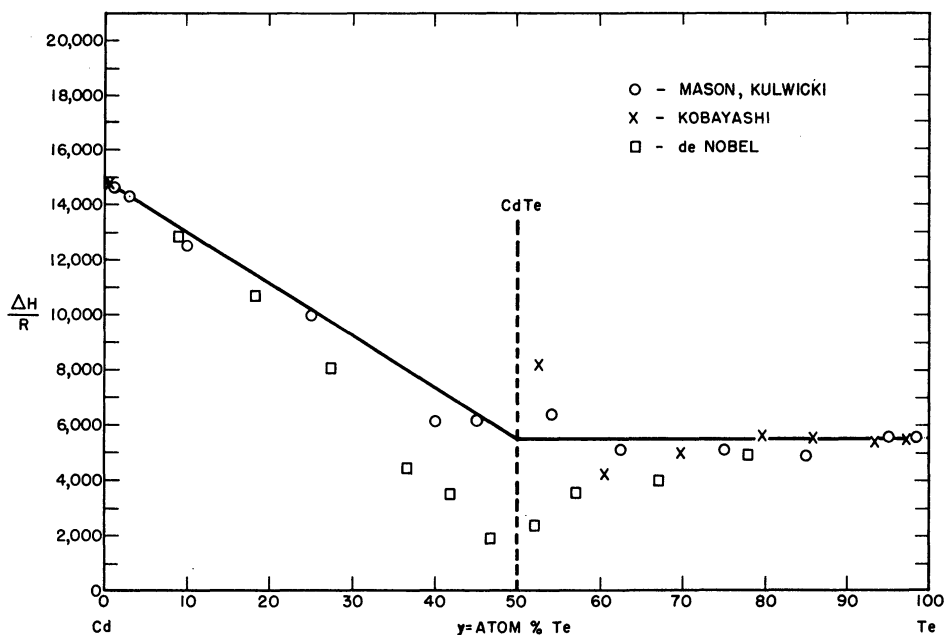


FIGURE 3. PLOT OF  $\Delta H/R$  VS.  $y$ , DERIVED FROM EXPERIMENTAL DATA AND EQUATION 1.  $\circ$  = Mason, Kulwicki;  $\times$  = Kobayashi;  $\square$  = deNobel.

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On the Cd-rich half of the diagram, the derived values of  $\Delta H_f/R$  are not independent of composition, and the solution is nonideal. At the composition CdTe the two segments of the curve apparently intersect at approximately the point where  $\Delta H_f/R = 5400$ , or  $\Delta H_f = 10,700$  cal/gm mol.

3.2. THEORETICAL FORMS OF THE LIQUIDUS LINES

By using the value of  $\Delta H_f$  estimated above, a quantitative measure of the nonideality of both binary systems can be obtained by calculating the activity coefficients and the  $\alpha$ -functions defined in Equation 2. A plot of  $\alpha$  vs. T is shown in Figure 4. Here again the data of deNobel and Kobayashi are presented for the larger values of (1 - x) where they fall within the domain shown on the figure. For very small values of (1 - x) the evaluation of the liquidus temperature becomes quite critical, and small errors are magnified greatly in calculating the  $\alpha$ -function. This is apparent from the measurement at 45 atom % Te, where a change of 2°C in the experimental liquidus temperature changes  $\alpha$  by almost 50%.

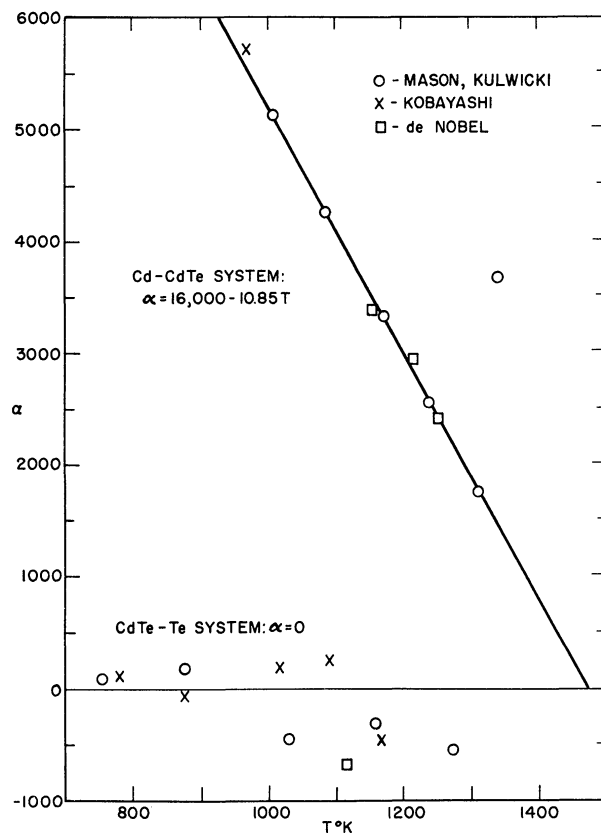


FIGURE 4. PLOT OF  $\alpha$  VS. T, FOR EVALUATION OF CONSTANTS a AND b IN EQUATION 2. ○ = Mason, Kulwicki; X = Kobayashi; □ = deNobel.

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From these calculations it again appears that the system (II) CdTe-Te is an ideal solution. As Te is dissolved in liquid CdTe there are no significant heat or excess entropy effects on mixing, and the liquidus curve can be approximated by the expression

$$T = \frac{\Delta H_f}{\Delta S_f - R \ln x} = \frac{10,700}{7.80 - 1.987 \ln x} \quad (5)$$

where  $x = 2(1 - y) =$  mol fraction of CdTe in Te

$$\Delta S_f = \Delta H_f / T_f = \text{entropy of fusion, cal/gm mol } ^\circ\text{K}$$

From the inspection of Figure 4 it is also apparent that the system I (Cd-CdTe) forms an elementary solution, with  $\alpha = 16,000 - 10.85T$ ; i. e.,  $\overline{\Delta H} = 16,000(1 - x)^2$  cal/gm mol and  $\overline{\Delta S}^E = 10.85(1 - x)^2$  cal/gm mol  $^\circ\text{K}$

The liquidus curve in this region can be approximated by the expression

$$T = \frac{\Delta H_f + a(1 - x)^2}{\Delta S_f + b(1 - x)^2 - R \ln x} = \frac{10,700 + 16,000(1 - x)^2}{7.80 + 10.85(1 - x)^2 - 1.987 \ln x} \quad (6)$$

where  $x = 2y =$  mol fraction of CdTe in Cd.

### 3.3. CALCULATION OF EUTECTIC COMPOSITIONS

The experimental melting point of CdTe ( $1371^\circ\text{K}$ ), the experimentally measured eutectic temperature on the Te side ( $723^\circ\text{K}$ ), and the derived latent heat of fusion (10,700 cal/mol) were used in Equation 1 to predict the eutectic composition in the system II (CdTe-Te). The predicted value of 0.986 atom-fraction Te in Cd indicates that the eutectic should be distinguishable from pure Te. The same relationship can also be used to calculate the eutectic composition from the properties of pure Te. From its melting point ( $727^\circ\text{K}$ ) and latent heat of fusion (3230 cal/gm mol) (Reference 13) the eutectic composition is calculated to be  $x = 0.014$ ; hence  $y = 0.993$ .

An experimental check was carried out using a sample containing 0.987 atom-fraction Te in Cd. The check was made on the presumption that the eutectic point predicted from the CdTe was more accurate than that predicted from the Te. It was found that the measurement was almost indistinguishable from that made at 0.982 atom-fraction Te. This discrepancy apparently arises because the liquidus curve rises so steeply on the left side of the eutectic that slight inhomogeneities in the sample tend to give erratic results. We conclude that the eutectic lies at approximately 0.99 atom-fraction Te.



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In order to calculate the eutectic composition on the Cd side from Equation 6 the mol fraction of CdTe at the eutectic can be approximated as zero in all the terms except the logarithmic factor. Since the experimental eutectic temperature is known (595°K), the eutectic composition is calculated to be  $10^{-6}$  atom-fraction Te in Cd, which is experimentally indistinguishable from pure Cd. However, our data would seem to indicate that the solidus transition temperature (322°C) is higher than the melting point of Cd (321°C), and a peritectic pattern may be present.

The liquidus curves and eutectic points derived above are plotted on Figure 5 along with all the experimental data from Table I, deNobel (Reference 5), and Kobayashi (Reference 6). The liquidus temperatures calculated from Equations 5 and 6 at the experimental compositions are also tabulated in Table I.

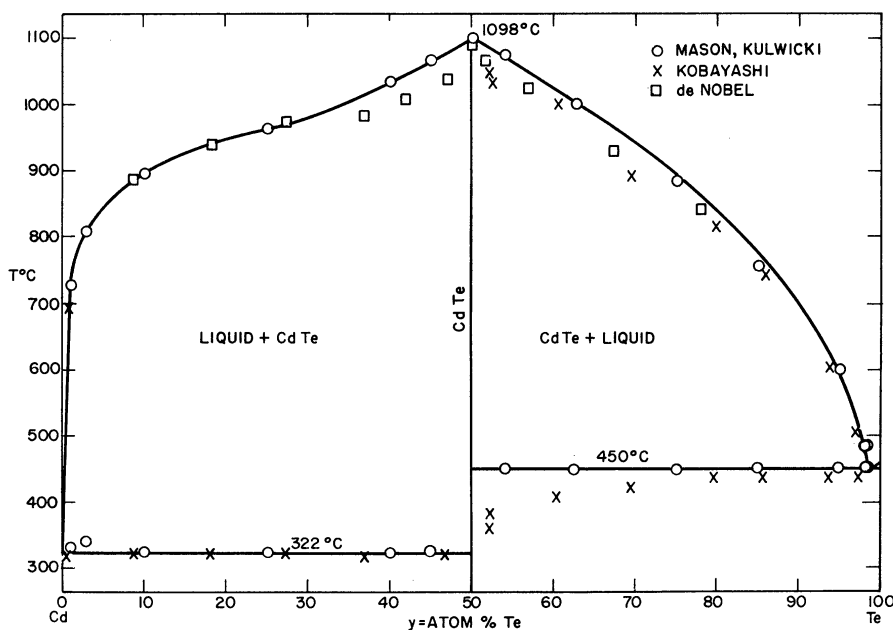


FIGURE 5. PHASE DIAGRAM FOR THE BINARY SYSTEM Cd-Te. The liquidus curves are drawn using Equations 3 and 4. ○ = Mason, Kulwicki; X = Kobayashi; □ = deNobel.

3.4. SLOPE AND CURVATURE OF THE LIQUIDUS LINES

The abrupt change in slope and curvature of the liquidus lines at the melting point of CdTe is also of interest. For system II (CdTe-Te), the slope is obtained by differentiating Equation 5.

$$\frac{\delta T}{\delta x} = \frac{\Delta H_f R}{x(\Delta S_f - R \ln x)^2} = \frac{10,700R}{x(7.80 - R \ln x)^2} \tag{7}$$

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As  $x \rightarrow 1$ ,  $\ln x \rightarrow 0$ , and

$$\left. \frac{\delta T}{\delta x} \right|_{x \rightarrow 1} = \frac{10,700R}{7.80^2} = 349.5 \quad (8)$$

For the system I (Cd-CdTe), the slope is obtained by differentiating Equation 6.

$$\frac{\delta T}{\delta x} = \frac{\left[ \Delta S_f + b(1-x)^2 - R \ln x \right] \left[ -2a(1-x) \right] - \left[ \Delta H + a(1-x)^2 \right] \left[ -2b(1-x) - R/x \right]}{\left[ \Delta S_f + b(1-x)^2 - R \ln x \right]^2} \quad (9)$$

As  $x \rightarrow 1$ ,  $(1-x) \rightarrow 0$  and  $\ln x \rightarrow 0$ , giving

$$\left. \frac{\delta T}{\delta x} \right|_{x \rightarrow 1} = \frac{\Delta H_f R}{x(\Delta S_f)^2} \quad (10)$$

which is identical with Equation 8. Hence the slope of the liquidus curve is discontinuous at the composition CdTe but symmetrical on either side as  $x$  approaches unity.

The curvature of the liquidus curve is given by the second derivative of the  $T$  vs.  $x$  expressions. For the system II (CdTe-Te) which approximates an ideal solution, differentiation of Equation 7 gives

$$\frac{d^2 T}{dx^2} = \frac{\Delta H_f R}{(\Delta S_f - R \ln x)^2} \left( \frac{2R}{\Delta S_f - R \ln x} - 1 \right) \frac{1}{x} \quad (11)$$

which for the system under discussion as  $x \rightarrow 1$  (pure CdTe) becomes

$$\left. \frac{d^2 T}{dx^2} \right|_{x \rightarrow 1} = \frac{\Delta H_f R}{\Delta S_f^2} \left( \frac{2R}{\Delta S_f} - 1 \right) = \frac{10,700 \times 1.987}{7.8^2} \left( \frac{3.975}{7.8} - 1 \right) = -171 \quad (12)$$

The moderately large negative value of the second derivative indicates that the liquidus curve should be convex upwards as  $x$  approaches unity from the Te side as shown on the right half of the diagram; i. e., as  $x$  increases the slope is decreasing.

In contrast, however, for the system I (Cd-CdTe) the expression for the second derivative of the elementary-solution curve is obtained by differentiation of Equation 9.

$$\frac{d^2 T}{dx^2} = \frac{2a - RT/x^2}{D} + \frac{2R^2 T + 4RTb x(1-x) - 4Rax(1-x) - 8abx^2(1-x)^2}{D^2 x^2} \quad (13)$$

where D is the denominator in Equation 6.

For the special case wherein  $x \rightarrow 1$ , then  $D \rightarrow \Delta S_f = 7.80$ , and  $T \rightarrow T_f = 1371^\circ\text{K}$ . This gives

$$\left. \frac{d^2T}{dx^2} \right|_{x \rightarrow 1} = \frac{2\alpha}{\Delta S_f} + \frac{RT_f}{\Delta S_f} \left( \frac{2R}{\Delta S_f} - 1 \right) \quad (14)$$

From Figure 4 or Equation 2 we can find  $\alpha = 1110$ , whence

$$\left. \frac{d^2T}{dx^2} \right|_{x_{\text{CdTe}} \rightarrow 1} = \frac{2220}{7.80} + \frac{1.987 \times 1371}{7.80} \left( \frac{3.975}{7.8} - 1 \right) = 113 \quad (15)$$

The positive second derivative indicates that the slope is positive and increasing, thus giving the observed curvature to the liquidus curve as  $x$  approaches unity from the Cd side. In fact, in order for the second derivative of the elementary-solution liquidus curve to be negative at the melting point of component A, it is necessary that

$$\frac{a}{\Delta H_f} + \frac{R^2}{\Delta S_f^2} < \frac{b}{\Delta S_f} + \frac{R}{2\Delta S_f} \quad (16)$$

4

DISCUSSION OF RESULTS

The verisimilitude of the results obtained from this work can be discussed from three different points of view: the validity of the experimental work, the accord between theory and experiment, and the theoretical insights which the results give toward a better comprehension of the structure of the liquid.

4.1. VALIDITY OF THE EXPERIMENTAL WORK

The large discrepancy between the melting point of CdTe reported by Kobayashi (Reference 6) and later investigators can be attributed to three factors; (a) the purity of the Te used in preparing the compounds, as indicated by the melting points (Kobayashi's  $437^\circ\text{C}$  vs.  $454^\circ\text{C}$  in this work), (b) Kobayashi's use of cooling curves only, instead of combining results from both heating and cooling curves, (c) unequal volatilization of the elemental constituents, particularly Cd, since Kobayashi's samples were open to the atmosphere instead of being sealed.

A close comparison of our data with that of deNobel (Reference 5) indicates that the agreement is excellent below about 25 atom % Cd. Above this range, our liquidus temperatures are

consistently higher than those reported by deNobel. In order to explain this discrepancy, we suggest that the error lies in deNobel's method of measurement. If the thermocouple and support rod in deNobel's experimental systems extract heat from the sample holder, it is conceivable that CdTe would freeze out on the bottom of the sample holder before solidification became apparent by visual observation on the surface of the sample. The liquid remaining after partial solidification and precipitation of CdTe (component A) would be more rich in component B (either Cd or Te, depending on which element was in excess over the stoichiometry of CdTe). The remaining liquid then would freeze at a temperature lower than the true liquidus temperature defined by the sample composition in the absence of partial segregation. By using thermal means to sense the onset of solidification, we feel that the effects of temperature gradients in our equipment are thereby significantly decreased.

In the absence of a detailed description of the method used by Larson, Nielsen, Putley, and Young (Reference 4), we cannot account for the discrepancy between  $1098 \pm 3^\circ\text{C}$  and  $1106^\circ\text{C}$ , which we feel is significant.

#### 4.2. ACCORD BETWEEN THEORY AND EXPERIMENT

On the Te half of the diagram the average discrepancy in our data between the observed and derived liquidus temperature is about  $10^\circ\text{C}$ . The solubility of excess Te in CdTe may account for this discrepancy. It is also probable that a slight excess entropy and enthalpy of mixing are associated with this system which would alter the shape of the liquidus curve slightly. However, since the plot of the data in Figure 4 fails to show any clear trend, and in the absence of information on the solubility of Te in CdTe, perfect solution behavior has been assumed in deriving the theoretical liquidus curve.

On the Cd half of the diagram, the average discrepancy between the observed and derived liquidus temperatures is only about  $1^\circ\text{C}$ . This unusually good agreement not only substantiates the conclusion that this system is an elementary solution, but also gives an example for the existence of elementary solutions between elements and compounds. At the same time the validity of the derived value for  $\Delta H_f$  is indirectly substantiated.

#### 4.3. THEORETICAL SIGNIFICANCE

In attempting to interpret the physical significance of the relative partial molar enthalpy and the relative partial molar excess entropy, we can consider first the CdTe-Te system. The absence of substantial energy and entropy effects suggests that the environment of the Cd atoms is not changed appreciably as excess molten Te is added to molten CdTe.

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However, as excess molten Cd is added to molten CdTe, the Cd atoms form additional bonds with the Te atoms. The Te atoms are subsequently surrounded by a much larger average number of Cd atoms. There is a discontinuity in the solution behavior on either side of the compound CdTe, and the slope of the liquidus curve is discontinuous at that point. A. M. G. D.

## Appendix

DERIVATION of THERMODYNAMIC EQUATIONS  
FOR LIQUIDUS CURVES

## 1. INTRODUCTION

This appendix describes the theoretical foundations for the conclusions of this memorandum. First to be developed will be the appropriate thermodynamic equations, serving also as a means of review for those readers not immediately concerned with the formal study of thermodynamics; then the definitions will be reviewed both for ideal solutions (Reference 11) and the various other classes of nonideal solutions that have been defined (Reference 11), including elementary solutions (Reference 12).

## 2. DERIVATION of GIBBS-HELMHOLTZ EQUATION

Since the system was maintained at constant volume and there is no pressure-volume work effect in the system, the thermodynamic quantity describing the equilibrium condition will be the thermodynamic work function,  $A$ , rather than the free energy,  $F$ .

Starting from elementary thermodynamics, consider first the following definitions for a system of constant total mols,  $N$ :

$$A = E - TS \quad (A-1)$$

where  $A$  = thermodynamic work function

$E$  = internal energy

$S$  = entropy

$T$  = absolute temperature

Hence,

$$dA = dE - T dS - S dT \quad (A-2)$$

But since, from the First Law

$$T dS = dE + P dV \quad (A-3)$$

substitution of Equation A-3 into Equation A-2 gives

$$dA = -P dV - S dT \quad (A-4)$$

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Defining A as a function of T and V gives

$$dA = \left(\frac{\delta A}{\delta V}\right)_{T, N} dV + \left(\frac{\delta A}{\delta T}\right)_{V, N} dT \quad (A-5)$$

from which, by comparing coefficients between Equations A-4 and A-5 we find

$$\left(\frac{\delta A}{\delta V}\right)_{T, N} = -P \quad (A-6)$$

and

$$\left(\frac{\delta A}{\delta T}\right)_{V, N} = -S \quad (A-7)$$

From Equations A-1 and A-7 we find

$$A = E + \left(\frac{\delta A}{\delta T}\right)_{V, N} T \quad (A-8)$$

Division by  $T^2$  and rearrangement of Equation A-8 gives

$$-\frac{E}{T^2} = \frac{1}{T} \left(\frac{\delta A}{\delta T}\right)_{V, N} - \frac{A}{T^2} = \left[\frac{\delta}{\delta T} \left(\frac{A}{T}\right)\right]_{V, N} \quad (A-9)$$

This is the Gibbs-Helmholtz equation for a fixed-composition system at constant volume.

2.1. CHEMICAL POTENTIAL IN MULTICOMPONENT OPEN SYSTEMS. In an open system where compositions and masses may vary, the thermodynamic work function at equilibrium may be expressed in the general form

$$A = f(T, V, n_1, n_2, \dots, n_i, \dots) \quad (A-10)$$

By using Equations A-6 and A-7 the change in work function in a differential displacement from equilibrium in an open system can be shown to be

$$dA = -P dV - S dT + \left(\frac{\delta A}{\delta n_1}\right)_{T, V, n_2, \dots} dn_1 + \dots + \left(\frac{\delta A}{\delta n_i}\right)_{T, V, n_1, \dots} dn_i + \dots \quad (A-11)$$

At constant temperature and volume the partial differentials in Equation A-11 are expressions for the chemical potential. Hence the chemical potential can be defined in a variety of ways.

$$\begin{aligned} \mu_i &= \left( \frac{\delta F}{\delta n_i} \right)_{T, P, n_1, \dots} = \left( \frac{\delta E}{\delta n_i} \right)_{S, V, n_1, \dots} = \left( \frac{\delta H}{\delta n_i} \right)_{S, P, n_1, \dots} \\ &= -T \left( \frac{\delta S}{\delta n_i} \right)_{E, V, n_1, \dots} = \left( \frac{\delta A}{\delta n_i} \right)_{T, V, n_1, \dots} \end{aligned} \quad (A-12)$$

From Equation A-12 we find

$$\mu_i = \left( \frac{\delta A}{\delta n_i} \right)_{T, V, n_1, \dots} \quad (A-13)$$

2.2. CLOSED CONSTANT VOLUME SYSTEM. The Gibbs-Helmholtz equation for a closed system can be written in terms of partial molar quantities at constant temperature and volume.

For a system of definite composition, Equation A-11 can be integrated with the aid of Equation A-13 to give

$$A = \mu_1 n_1 + \mu_2 n_2 + \dots + \mu_i n_i + \dots \quad (A-14)$$

By maintaining temperature and volume constant in the system during an equilibrium change in phase, then  $dA = 0$ , and

$$0 = \mu_1 dn_1 + \mu_2 dn_2 + \dots + \mu_i dn_i + \dots \quad (A-15)$$

for any phase. Differentiation of Equation A-13 with respect to  $T$  at constant total mols  $N$  gives

$$\left( \frac{\delta \mu_i}{\delta T} \right)_{N, V} = \left( \frac{\delta^2 A}{\delta n_i \delta T} \right)_{N, V} \quad (A-16)$$

Differentiation of Equation A-7 with respect to  $n_i$  gives

$$-\left( \frac{\delta S}{\delta n_i} \right)_{V, N} = \left( \frac{\delta^2 A}{\delta T \delta n_i} \right)_{V, N} = -\bar{S}_i \quad (A-17)$$

But Equations A-16 and A-17 are equal, hence

$$-\bar{S}_i = \left( \frac{\delta \mu_i}{\delta T} \right)_{V, N} \quad (A-18)$$



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Differentiating Equation A-1 with respect to  $n_i$  gives

$$\left(\frac{\delta A}{\delta n_i}\right)_{T, V, n_1, \dots} = \left(\frac{\delta E}{\delta n_i}\right)_{T, V, n_1, \dots} - T \left(\frac{\delta S}{\delta n_i}\right)_{T, V, n_1, \dots} \quad (A-19)$$

Defining  $\left(\frac{\delta E}{\delta n_i}\right)_{T, V, n_1, \dots} = \bar{E}_i$  and substituting from Equations A-13 and A-17 we find

$$\mu_i = \bar{E}_i - T\bar{S}_i \quad (A-20)$$

Substitutions from Equation A-18 and rearrangement gives

$$-\mu_i + \left(\frac{\delta \mu_i}{\delta T}\right)_{V, N} T = -\bar{E}_i \quad (A-21)$$

Dividing by  $T^2$  gives the desired result,

$$\frac{-\bar{E}_i}{T^2} = \frac{1}{T} \left(\frac{\delta \mu_i}{\delta T}\right)_{V, N} - \frac{\mu_i}{T^2} = \left[\frac{\delta}{\delta T} \left(\frac{\mu_i}{T}\right)\right]_{V, N} \quad (A-22)$$

2.3. FUGACITY AND ACTIVITY. The thermodynamic equations for liquid and solid systems are derived from concepts established for gaseous systems through a consideration of the behavior of the fugacity and activity in the various phases. For 1 mol of a constituent of an ideal gas mixture in a closed system with partial pressure  $P_i$ ,  $\mu_i = \mu^0(T) + RT \ln P_i$ :

$$\left(\frac{\delta \mu_i}{\delta P}\right)_{T, N} = \bar{V}_i = \frac{RT}{P_i} \quad (A-23)$$

For a system containing 1 mol of an ideal gas, with mol fraction of constituent  $i$  equal to  $x_i$  at constant temperature and composition at constant total pressure  $P$ ,

$$\mu_i = \mu_i(T, P) + RT \ln x_i \quad (A-24)$$

In general, to accommodate nonideal systems the partial pressure is replaced by the fugacity. Hence

$$\delta \mu_i = RT d \ln f_i \quad (A-25)$$

Integration gives

$$\mu_i = RT \ln f_i + C \quad (A-26)$$

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To define the constant of integration, we can define a reference state as the pure constituent in the gas phase at the same temperature under one atmosphere pressure or its own vapor pressure (if a condensable vapor, with vapor pressure below one atmosphere) such that it is substantially a perfect gas.

Hence,

$$\mu_i - \mu_i^O = RT \ln f_i/f_i^O = RT \ln a_i \quad (A-27)$$

where  $a_i = f_i/f_i^O = \text{activity} = \text{fugacity/fugacity of standard state}$ . Rearranging and differentiating with respect to temperature at constant volume and composition gives

$$\frac{d}{dT} \left( \frac{\mu_i}{T} \right)_{V,N} - \frac{d}{dT} \left( \frac{\mu_i^O}{T} \right)_{V,N} = R \left( \frac{d \ln a_i}{dt} \right)_{V,N} \quad (A-28)$$

Substitution from Equation A-22 gives

$$\left( \frac{d \ln a_i}{dT} \right)_{V,N} = \frac{E_i^O - \bar{E}_i}{RT^2} \quad (A-29)$$

### 3. CHANGE OF STATE

These basic thermodynamic concepts can now be utilized to investigate the phenomena accompanying changes in state. It is necessary to apply these definitions to the particular situations of interest.

If we consider a phase change of a pure constituent from solid to the reference state in the vapor, then in Equation A-29 the various terms evaluated for this special case become

$$E_i^O = E_v^O = \text{internal energy of pure vapor in standard state} \quad (A-30)$$

$$\bar{E}_i = E_i^S = \text{internal energy in solid state} \quad (A-31)$$

then

$$E_i^O - E_i^S = \Delta E_{sv} = \text{internal energy of sublimation} \quad (A-32)$$

If instead of the preceding change of state we consider a phase change from liquid to the vapor reference state, then the various terms in Equation A-29 become

$$E_i^O = E_v^O = \text{internal energy of pure vapor in standard state} \quad (A-33)$$

$$\bar{E}_i = E_i^L = \text{internal energy in liquid bath} \quad (A-34)$$

hence,

$$E_1^O - E_1^L = \Delta E_v = \text{internal energy of vaporization} \quad (\text{A-35})$$

3.1. TWO COMPONENT CONSTANT VOLUME SYSTEM. Consider now a closed system of three phases (gas, liquid, solid) and two components. At equilibrium from Equation A-15 we must have

$$\mu_1^g dn_1^g + \mu_1^L dn_1^L + \mu_1^S dn_1^S + \mu_2^g dn_2^g + \mu_2^L dn_2^L + \mu_2^S dn_2^S = 0 \quad (\text{A-36})$$

Also the total mass of each component must be constant, hence

$$dn_1^g + dn_1^L + dn_1^S = 0 \quad (\text{A-37})$$

$$dn_2^g + dn_2^L + dn_2^S = 0 \quad (\text{A-38})$$

These relationships can be satisfied for all variations in  $n_1$ ,  $n_2$  only if

$$\mu_1^g = \mu_1^L = \mu_1^S \quad (\text{A-39})$$

$$\mu_2^g = \mu_2^L = \mu_2^S \quad (\text{A-40})$$

3.2. SOLID-LIQUID EQUILIBRIA. When the temperature of a closed solid-liquid system is changed slightly, then the activities of the constituents also change slightly. However, under equilibrium conditions the activity in the liquid remains equal to the activity in the solid, although the magnitude of the activities can change as a result of the temperature change. Applying Equations A-27 and A-39 to constituent 1 this result can be expressed as

$$\left( \frac{\delta \ln a_1^L}{\delta T} \right)_{V, x_1^L} dT + \left( \frac{\delta \ln a_1^L}{\delta x_1^L} \right)_{T, V} dx_1^L = \left( \frac{\delta \ln a_1^S}{\delta T} \right)_{V, x_1^S} dT + \left( \frac{\delta \ln a_1^S}{\delta x_1^S} \right)_{T, V} dx_1^S \quad (\text{A-41})$$

After rearrangement and substitution from Equation A-29,

$$\left( \frac{\delta \ln a_1^L}{\delta x_1^L} \right)_{V, T} dx_1^L - \left( \frac{\delta \ln a_1^S}{\delta x_1^S} \right)_{T, V} dx_1^S = \left( \frac{E_1^L - E_1^S}{RT^2} \right) dT \quad (\text{A-42})$$

where  $E_1^L - E_1^S = \text{internal energy of fusion of component 1} = \Delta E_f^O$ .

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3.3. IDEAL SOLUTIONS. For an ideal solution the fugacity of the constituent is equal to the product of the mol fraction times the fugacity of the pure substance

$$f_i = x_i f_i^O \quad \text{or} \quad a_i = x_i$$

Hence by definition

$$\left( \frac{\delta \ln a_1^L}{\delta x_1^L} \right)_{V, T} = \frac{1}{x_1^L} \quad (\text{A-43})$$

and

$$\left( \frac{\delta \ln a_1^S}{\delta x_1^S} \right)_{V, T} = \frac{1}{x_1^S} \quad (\text{A-44})$$

Substituting into Equation A-42 finally gives

$$\left( \frac{\delta \ln x_1^L / x_1^S}{\delta T} \right)_{T, V} = \frac{\Delta E_f^O}{RT^2} \quad (\text{A-45})$$

If component 1 is considered as the solute, which separates as a pure constituent on freezing from the solution (hence  $x_1^S = 1$ ), then integration of Equation A-45 from the solute fusion temperature,  $T_f$ , and composition  $x_1^L = 1$ , to the equilibrium-solution temperature  $T$  and composition  $x_1^L$  gives

$$\int_1^{x_1^L} d \ln x_1^L = \frac{\Delta E_f^O}{R} \int_{T_f}^T \frac{dT}{T^2} \quad (\text{A-46})$$

$$\ln x_1 = \frac{\Delta E_f}{R} \left( \frac{1}{T_f} - \frac{1}{T} \right) \quad (\text{A-47})$$

assuming  $\Delta E_f^O$  to be independent of temperature and  $(C_v^L - C_v^S) = 0$ . If the difference in heat capacities at constant volume is not zero, then an additional integral is introduced into Equation A-46 which should produce another term in Equation A-47. In most cases of practical interest, the heat capacity difference is neglected. Not only are the calculations simplified, but also the precise data are often not available to make the corrections. In any event, the consistency of the results obtained by ignoring this factor give tacit evidence that it is negligible.

3.4. NONIDEAL SOLUTIONS. Most solutions are not ideal solutions. However, the deviation from ideality can be lumped into a single parameter, the activity coefficient, such that

$$a_i = \gamma_i x_i \quad (\text{A-48})$$

Under these circumstances, Equation A-47 would be rewritten as

$$\ln \gamma_1 x_1 = \frac{\Delta E_f^{\circ}}{R} \left( \frac{1}{T_f} - \frac{1}{T} \right) \quad (\text{A-49})$$

As the mol fraction approaches unity, then by definition the activity also approaches unity and equation A-49 can be rearranged to

$$\ln \gamma_1 + \ln x_1 = \frac{\Delta E_f^{\circ}}{R} \left( \frac{1}{T_f} - \frac{1}{T} \right) \quad (\text{A-50})$$

In accordance with the required boundary limitations on  $\gamma$ , it is possible to express  $\ln \gamma$  as a function of mol fraction by

$$\ln \gamma = \sum_{n=1}^{\infty} C_n (1-x)^n \quad (\text{A-51})$$

which satisfies the relationship that

$$\gamma \rightarrow 1 \quad \text{as} \quad x \rightarrow 1.$$

For nonelectrolytic solutions, the Gibbs-Duhem equation can be used to show that  $C_1 = 0$ . Hence

$$\ln \gamma = \sum_{n=2}^{\infty} C_n (1-x)^n \quad (\text{A-52})$$

The simplest concentration dependence obtainable from Equation A-52 is given by the first term, or we can let

$$\ln \gamma = C_2 (1-x)^2 \quad (\text{A-53})$$

By defining the partial molar free energy of the solute in solution as

$$F_L - F_L^{\circ} = \overline{\Delta F} = RT \ln \gamma x = \overline{\Delta H} - T \overline{\Delta S} \quad (\text{A-54})$$

the relative partial molar entropy  $\overline{\Delta S}$  can be expressed as the sum of two terms, the ideal entropy of mixing  $\overline{\Delta S}^i$  and an excess entropy of mixing  $\overline{\Delta S}^E$

$$\overline{\Delta S} = \overline{\Delta S}^i + \overline{\Delta S}^E = \overline{\Delta S}^E - R \ln x \quad (\text{A-55})$$

Substitution of Equation A-55 into Equation A-54 gives

$$\ln \gamma = \frac{\overline{\Delta H}}{RT} - \frac{\overline{\Delta S}^E}{R} \quad (\text{A-56})$$

where  $\overline{\Delta H}$  = relative partial molar enthalpy of mixing of the solute

$\overline{\Delta S}^E$  = relative partial molar excess entropy of mixing of the solute

Several types of nonideal solutions have been defined according to the various dependencies of  $\ln \gamma$ .

3.4.1. Regular Solutions. Regular solutions have been defined as those solutions wherein the partial molar excess entropy of mixing is zero, but the partial molar enthalpy of mixing is not zero. In particular, if

$$\overline{\Delta H} = a_r (1 - x)^2 \quad (\text{A-57})$$

and

$$\overline{\Delta S}^E = 0$$

then a strictly regular solution has been defined.

3.4.2. Athermal Solutions. Athermal solutions have been defined as solutions wherein the partial molar excess entropy of mixing is finite, whereas the partial molar enthalpy of mixing is zero. A particular example of this type of behavior could be represented by the relationship

$$\overline{\Delta S}^E = b_a (1 - x)^2 \quad (\text{A-58})$$

$$\overline{\Delta H} = 0$$

This definition is of value predominantly for conceptual purposes.

3.4.3. Elementary Solutions. In general, both the partial molar enthalpy and partial molar excess entropy need not be zero. However, the particular manner in which these properties depend on temperature and composition is of considerable importance.

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If the dependence is as defined below, the resulting solutions have been called "Elementary Solutions"(Reference 12).

$$\overline{\Delta H} = a(1 - x)^2 \quad (A-59)$$

$$\overline{\Delta S}^E = b(1 - x)^2 \quad (A-60)$$

Substitution of Equations A-59 and A-60 into Equation A-56 gives

$$\ln \gamma = \left( \frac{a}{RT} - \frac{b}{R} \right) (1 - x)^2 \quad (A-61)$$

Equation A-61 can be rearranged to give a function  $\alpha$ , which varies linearly with temperature.

$$\frac{RT \ln \gamma}{(1 - x)^2} = \alpha = a - bT \quad (A-62)$$

As noted in the text of the memorandum, the function  $\alpha$  was calculated for the system I (Cd-CdTe) and found to be  $\alpha = 16,000 - 10.85 T$ . Elementary solution behavior is thus indicated for this system.

3.5. LIQUIDUS CURVES. The dependency of T on x can be obtained from Equation A-50 to define the liquidus curves explicitly. The various dependencies of  $\ln \gamma$  for ideal, regular, athermal, and elementary solutions must be substituted appropriately.

For ideal solutions

$$T_i = \frac{\Delta H_f}{\Delta S_f - R \ln x} \quad (A-63)$$

For strictly regular solutions

$$T_r = \frac{\Delta H_f + a_r (1 - x)^2}{\Delta S_f - R \ln x} \quad (A-64)$$

For strictly athermal solutions

$$T_a = \frac{\Delta H_f}{\Delta S_f + b_a (1 - x)^2 - R \ln x} \quad (A-65)$$

For elementary solutions

$$T_e = \frac{\Delta H_f + a(1 - x)^2}{\Delta S_f + b(1 - x)^2 - R \ln x} \quad (A-66)$$

3.6. THE SLOPES OF THE LIQUIDUS CURVES. The slopes of the various types of liquidus curves can be obtained by taking the derivatives of the equations in Section 3.5 with respect to the composition variable  $x$ .

For ideal solutions

$$\frac{dT_i}{dx} = \frac{\Delta H_f R}{x(\Delta S_f - R \ln x)^2} = \frac{RT_i}{x(\Delta S_f - R \ln x)} \quad (A-67)$$

where  $T_i$  is defined by Equation A-63.

For strictly regular solutions

$$\frac{dT_r}{dx} = \frac{(RT_r) - 2a_r(1-x)x}{x(\Delta S_f - R \ln x)} \quad (A-68)$$

where  $T_r$  is defined by Equation A-64.

For strictly athermal solutions

$$\frac{dT_a}{dx} = \frac{RT_a + 2b_a(1-x)x}{x[\Delta S_f + b_a(1-x)^2 - R \ln x]} \quad (A-69)$$

where  $T_a$  is defined by Equation A-65.

For elementary solutions

$$\frac{dT_e}{dx} = \frac{RT_e - 2\alpha(1-x)x}{x[\Delta S_f + b(1-x)^2 - R \ln x]} \quad (A-70)$$

where  $T_e$  is defined by Equation A-66 and  $\alpha$  is defined in Equation A-62.

3.7. THE CURVATURES OF THE LIQUIDUS CURVES. The curvatures of the various types of liquidus curves can be obtained by taking the derivatives of the slopes with respect to the composition variable  $x$ .

For ideal solutions

$$\frac{d^2 T_i}{dx^2} = \frac{RT_i}{(\Delta S_f - R \ln x)} \left[ \frac{2R}{(\Delta S_f - R \ln x)} - 1 \right] \frac{1}{x^2} \quad (A-71)$$



For strictly regular solutions

$$\frac{\delta^2 T_r}{\delta x^2} = \frac{RT_r}{(\Delta S_f - R \ln x)} \left[ \frac{2R}{(\Delta S_f - R \ln x)} - 1 \right] \frac{1}{x^2} + \frac{2a_r}{(\Delta S_f - R \ln x)} \left[ 1 - \frac{2R(1-x)}{x(\Delta S_f - R \ln x)} \right] \quad (\text{A-72})$$

For strictly athermal solutions

$$\frac{\delta^2 T_a}{\delta x^2} = \left[ \frac{RT_a + 2b(1-x)x}{\Delta S_f + b(1-x)^2 - R \ln x} \right]^2 \frac{1}{x^2} + \frac{RT_a + 2b(1-x)x}{[\Delta S_f + b(1-x) - R \ln x]} \left[ \frac{R(2-T_a)}{\Delta S_f + b(1-x) - R \ln x} - 1 \right] \quad (\text{A-73})$$

For elementary solutions

$$\frac{\delta^2 T_e}{dx^2} = \frac{2\alpha - RT_e/x^2}{[\Delta S_f + b(1-x)^2 - R \ln x]} = \frac{2R^2 T_e - 4\alpha R(1-x)x - 8\alpha b(1-x)^2 x^2}{x[\Delta S_f + b(1-x) - R \ln x]^2} \quad (\text{A-74})$$

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THE PHASE DIAGRAM FOR THE BINARY SYSTEM CADMIUM-TELLURIUM by Donald R. Mason and Bernard M. Kulwicksi.  
Memo. of Proj. MICHIGAN. Apr 60. 28 p. incl. illus. 2 tables, 13 refs.  
(Memo. no. 2900-139-R)  
(Contract DA-36-039 SC-78801)

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