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Density measurements of molten solutions of thallium
and tellurium have been made over the temperature range
220 to 650°C and the composition range 31 to 69 atomic
per cent thallium. The Archimedian method was employed
since it is readily adaptable to performing measurements
over wide ranges of temperature and composition with a
high degree of precision.

The density-temperature curves are linear. These
measurements yield the composition dependence of the den-
sity and coefficient of thermal expansion. The purpose
of this work was to obtain information which would sup-
lement known electrical data for liquid thallium-
tellurium. In a particular composition range for which the

liquid exhibits n-type behavior, conversion of the density to the number of atoms per unit volume provides a value which was needed to complete the calculation of the total electron concentration. Also, the average atomic volume demonstrates an unexpected dependence on composition. A discussion of this behavior is presented in terms of known electrical and thermodynamic data.

Density of Molten Solutions
of Thallium and Tellurium

by

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DENSITY OF MOLTEN SOLUTIONS
OF THALLIUM AND TELLURIUM

INTRODUCTION

A number of studies of liquid thallium and tellurium
solutions have been made to determine electrical and
thermodynamic behavior. Thermoelectric measurements by
Cutler and Mallon (1965) have shown that liquid Tl-Te
solutions demonstrate semiconductor or semimetallic be-
havior over a wide range of composition X (atomic per
cent thallium) and temperature T. They observed that a
transition in the electrical conductivity from conduction
by holes to conduction by electrons occurs at a composi-
tion very close to Tl_2Te. In more recent work, Cutler
and Field (1968) have reported the results of an exten-
sive study of the electrical behavior of this liquid al-
loy system in the vicinity of the p-n transition. These
authors show that the p-n transition occurs precisely at
the stoichiometric composition Tl_2Te (X = 66.7). Solu-
tions having a concentration of thallium in the range
$X \geq 66.7$ are n-type, and the rest are p-type. In addi-
tion, a study of the thermodynamic behavior of liquid
Tl-Te solutions by Terpilowski and Zaleska (1963) has
shown that the excess thermodynamic functions of mixing
have large negative values over the composition range.
The enthalpy of mixing and the excess entropy of mixing

have minimum values at a composition of approximately 66.7 atomic per cent thallium. Both the electrical and thermodynamic data provide strong evidence for the formation of Tl_2Te as a compound in the liquid state.

In the present study, measurements of the temperature and composition dependence of the density were made in order to supplement electrical data. Knowledge of the density permits conversion of the concentration from mass per unit volume to the number of atoms per unit volume. For the composition range X > 66.7 Cutler and Field (1968) have shown that the atomic concentration can be used to determine the electron concentration.

In addition, the composition and temperature dependence of the atomic volume permits some interpretation of structural changes in liquid thallium-tellurium. The results appear to show a change in the behavior of the atomic volume near the composition X = 50. This is in contrast to thermodynamic and electrical data mentioned above, which suggests that a change occurs near X = 66.7.

Measurements of the temperature dependence of the density of molten thallium-tellurium solutions were made over the composition range, $31 \leq X \leq 69$, and in the temperature range, 220 to 650 °C. The Archimedian method was selected since this arrangement avoids many of the problems present with other techniques when applied to high temperature investigation of liquid metals.

EXPERIMENTAL METHOD

Background

White (1959) and Bauer (1949) have described a number
of techniques for density determination of various sub-
stances in a liquid state. The type of material and the
conditions of investigation are deciding factors in selec-
tion of the appropriate method. For measurements involv-
ing liquid metals at high temperature, the direct buoyancy
method is generally most satisfactory. Other approaches,
such as pyknometric, volumetric, and pressure methods,
are limited primarily by the effect of temperature on
various parts of the apparatus involved. The pyknometer,
which yields density values from the weight of a given
volume, usually needs recalibration or replacement after
each run, which is conducted at one temperature. A dila-
tometer, which serves to measure the volume of a given
weight, is sometimes hampered by temperature inhomogeneity
and by the requirement of frequent recalibration or re-
placement of the containing vessel. The pressure methods
require remote measurements of liquid levels and correc-
tion for expansion of portions of the apparatus.

The direct buoyancy method requires measurement of
the buoyant force exerted on a reference solid of known
volume while immersed in the liquid sample. This approach
allows investigation over a range of temperatures, does

not require calibration of the containing vessel, and avoids temperature inhomogeneity sometimes present with apparatus requiring visual observation at high temperatures. The accuracy which can be obtained is comparable or superior to that of other methods used for study of molten metals or alloys. In applying this method to density measurements of molten thallium-tellurium solutions there were a number of experimental problems: (1) providing a controlled atmosphere for the sample while measuring the buoyant force from a remote position, (2) fabricating a sufficiently dense, non-corroding plummet, (3) mixing the sample and, (4) measuring the temperature precisely. The solution to these and related problems is presented in the following discussion.

Isolation of the sample

The primary experimental problem was to isolate the molten sample in an inert atmosphere while making remote measurements of the buoyant force with a microbalance positioned in air. Isolation of the sample in a protective encasement was necessary in order to prevent oxidation and escape of vapors. The glass system shown in Figure 1 provided a solution to this problem. A continuous flow of argon entering the upper chamber and passing through two exit ports allowed free access for the plummet suspension while isolating the sample from the external atmosphere.
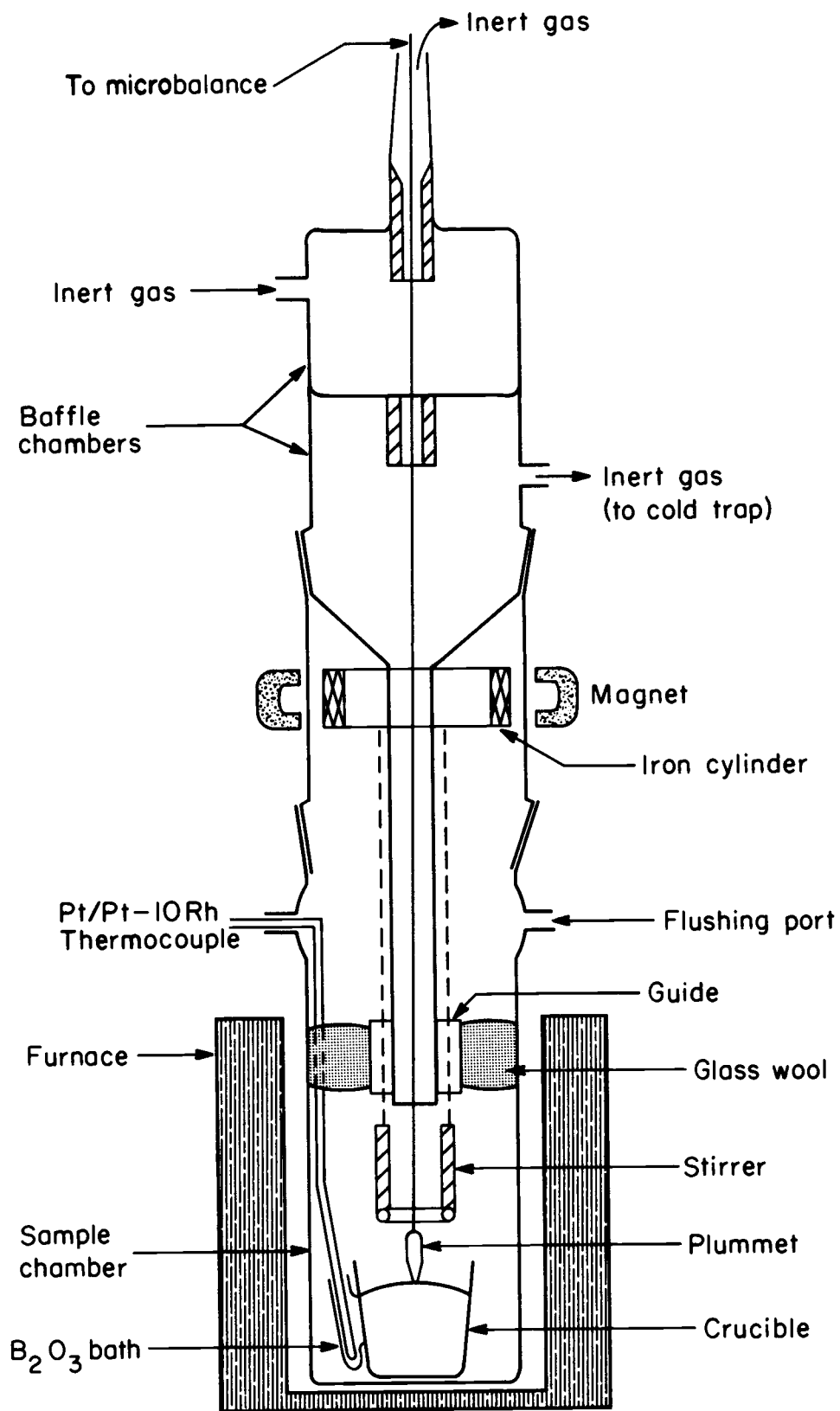
Figure 1. Density measuring apparatus.

The portion of gas going through the lower exit port was directed through a cold trap to condense any metallic vapors. Precise measurement of the viscous force caused by the flow of argon past the suspension was necessary in order to determine the buoyant force accurately. This measurement was performed several times during the course of each run by means of the microbalance.

In the range of investigation $31 < X < 51$ all of the glassware consisted of Pyrex since the low temperature limit of the liquid phase ranges from 200 to 300°C in this region. In this composition region, the density investigation was conducted by beginning with the composition $X = 50.8$ and continuing for successive increases in tellurium concentrations until reaching the composition $X = 31.0$. In order to make measurements in the range $51 < X < 72$, parts of the apparatus were changed to quartz. This allowed measurements to be made over a temperature range at compositions where the liquidus extends to high temperatures.

The microbalance used for remote weighing was a Cahn Gram Electrobalance whose quoted accuracy and precision depended on the electrical range required to measure the weight of the plummet. For the weights used in the present work, the accuracy was typically 0.5 mg. while the precision was 0.1 mg.

Fabrication of the plummet

Due to the chemical activity and high density of molten thallium-tellurium, it was necessary to construct a relatively dense, corrosion resistant plummet. In preliminary efforts, the plummet-suspension assembly consisted of a small tungsten cylinder coated with a thin layer of fused silica attached to a long, thin quartz fiber. The principal problems encountered with this method were the fragility of the suspension fiber and the fact that the liquid thallium-tellurium did not wet quartz. This last problem made submersion of the plummet very difficult because of the added upward force of surface tension. The nonwetting action could be reversed by diffusing a small amount of thallium into the quartz coating. In spite of the excellent surface protection provided by silica, the extreme fragility was a serious disadvantage and another approach became necessary.

A solution to the problem was the use of tungsten oxides which provide a strongly adhering protective layer on tungsten and which are stable up to 700 °C (Kubaschewski and Hopkins, 1953). A protective layer was formed on the tungsten plummet and suspension through controlled oxidation of the surface by first baking the tungsten under vacuum at 800°C for one hour and then allowing a small amount of oxygen to enter while maintaining a temperature

of 600°C for about one-half hour. This procedure was used to fabricate a new plummet and suspension for each composition (with the exception of composition X = 42.0) in order to prevent gradual deterioration of the oxide layer which may have resulted if a single plummet had been used for all runs. Another measure used to maintain the surface protection was limiting each immersion time of the plummet assembly into the melt to only what was needed to measure the buoyant force.

Since most solids are attacked by molten metals to some extent it was necessary to inspect for corrosion of the plummet and suspension. The primary method used to detect corrosion was examination of the buoyancy measurements for systematic effects which would indicate a change in plummet volume. Changes of this kind can be detected by comparing the set of density values recorded for a sequence of increasing temperatures with a corresponding set of values for decreasing temperatures. A small systematic change in density was observed only in the run for X = 48.0. For this composition the maximum difference, which occurred in the low temperature range of buoyancy measurements, represented a change in density of approximately 0.1 per cent. The set of buoyancy measurements recorded for decreasing temperature produced slightly larger density values than those previously recorded for increasing temperature. This direction of change is the

same as would occur for solution attack of the plummet by
the liquid alloy.

In addition, inspection for signs of gross corrosion
during a run was made by visual examination of the suspen-
sion for changes in its diameter. The diameter of the
suspension upon fabrication was approximately 0.002 inches.
Comparison of the suspension diameter above and below the
immersion level was made using a telescope to view the
raised suspension through the glassware. This method did
not indicate the presence of any corrosion of the plummet
or suspension.

Although tungsten oxide provided fairly adequate sur-
face protection, perhaps a more practical approach to the
problem would have been to use evaporation or pyrolytic
deposition of a protective coating on the plummet and
suspension. However, lack of time prevented development
of such a technique.

Mixture of the sample

Mixing of the thallium-tellurium melt was necessary
because of segregation of phases which occurs on freezing.
This was accomplished with a glass stirring ring which
could be moved vertically by the magnetic suspension
assembly shown in Figure 1. The sample was throughly mix-
ed prior to making each set of measurements at a given
composition. Besides providing a homogeneous liquid, the

agitation caused small amounts of oxide on the surface of the melt to collect on the glass surface of the crucible and stirrer, thus reducing errors due to surface contamination.

Temperature measurement

During the first run, X = 50.8, the temperature was measured using a chromel-alumel thermocouple located at the same level as the crucible but outside of the sample chamber. However, the difficulty in attaining thermal equilibrium in this configuration led to the positioning of a Pt_{100}-$Pt_{90}Rh_{10}$ thermocouple inside the chamber for the succeeding runs. Various methods were used to obtain good thermal contact with the crucible for accurate and rapid temperature measurement. The final technique consisted of immersing the thermocouple junction into a fused oxide bath contained in a quartz tube fused to the crucible. B_2O_3 was used for the oxide bath since it has a melting point of 450°C and does not react with the materials involved. For the first run temperature was measured using a Leads and Northrup potentiometer with an accuracy of 0.05 per cent plus 20 microvolts, while a Biddle potentiometer with an accuracy of 0.05 per cent plus 1.6 microvolts was employed for the remaining runs.

Other experimental problems

Glass was selected as the crucible material in order to avoid chemical attack by the molten liquid and consequent contamination of the sample. However, there was a problem of crucible breakage which would tend to occur during the cooling period at the conclusion of a run. It is believed that small amounts of thallium oxide formed a strong bond between the glass vessel and the solidified sample causing the glass to break due to thermal stress which resulted from differing coefficients of expansion of the two materials. In the Te-rich composition range, a heating tape wrapped around the sample chamber allowed for solidification and remelting of the sample without crucible damage. The tape, positioned slightly above the crucible, was used to make sure that the top surface of the sample melted first and froze last. This procedure controlled the stress on that portion of the crucible which would tend to collect oxide contamination. Another feature, intended to reduce breakage, was to include a conical shape in the design of the crucibles.

Another problem occurred in maintaining the same immersion level of the suspension while determining the plummet's volume with a calibrating fluid and during the course of a run when the temperature changes would cause thermal expansion of the suspension and sample.

A change in depth would change the effective volume of immersion and cause an error. The method used to ensure a known depth of immersion involved vertical adjustment of the microbalance stage. With the plummet initially submerged and the microbalance beam in the equilibrium position, the microbalance stage was raised until the effect of surface tension on the emerging plummet caused a radical change of the beam position. Lowering the stage a constant distance from that point allowed immersion to the same level on the 0.002 inch diameter suspension to within an estimated 0.5 mm. Assuming no variation in the diameter of the suspension along its length, the effect of changes in depth of 0.5 mm. would amount to about 2×10^{-5} gm. in the microbalance reading. The magnitude of this weight was beyond the sensitivity of the electrobalance for the electrical range required to weigh the plummet.

Preparation for a run

Final steps before a run included a number of operations. The glassware was carefully cleaned in order to eliminate sources of contamination. The volume of the plummet was determined, using distilled water as a calibrating fluid. Vacuum baking the plummet and glassware removed adsorbed gases and moisture. Preparation of the sample involved removal of an outer oxide layer from the

otherwise commercially pure thallium. The oxide layer was removed from the thallium by allowing the molten metal to pass through a series of glass funnels that were contained in a cell filled with an inert gas. As tellurium does not oxidize readily, carefully weighed amounts were added directly to the crucible. The crucible was then placed inside the sample chamber while it was separated from the upper glassware assembly shown in Figure 1. A second casting operation in an inert atmosphere was then performed to allow the thallium to enter the crucible. The prepared thallium-tellurium sample was quickly frozen to limit the diffusion of small amounts of thallium oxide into the glass vessel and thereby to reduce the possibility of crucible breakage. After attaching the sample chamber to the glass assembly the entire cell was sealed and evacuated before admitting the inert gas. During the initial period of heating, a large argon flow passed through the warm zone to remove any residual moisture from the interior of the cell.

EXPERIMENTAL RESULTS

Buoyancy measurements for liquid thallium-tellurium solutions were made over the composition range, $31.0 < X < 68.8$, and the temperature range, 220 to 650°C. For each composition, the buoyancy measurements permitted determination of the density-temperature relationship and the bulk coefficient of thermal expansion. Adequate description of the density calculation appears in many references, such as Bauer (1949) and White (1959). Using the density-temperature values for each composition, a least squares analysis led to a linear relationship between d and T which may be expressed as

$$d = d_O[1 - \beta_O(T - 500)], \qquad (1)$$

where d_O and β_O are the density and coefficient of volume expansion at the reference temperature 500°C. The values of β_O were determined from the slopes of the density-temperature curves and the densities at 500°C.

From the density, the average atomic volume at 500°C is obtained with the expression

$$v_O = \frac{\overline{M}}{d_O N_O}, \qquad (2)$$

where $\overline{M}$ is the average molecular weight and N_O is Avogadro's number. The average molecular weight is given by

$$\overline{M} = x_{Tl} M_{Tl} + x_{Te} M_{Te}, \qquad (3)$$

where x_i and M_i refer to the atomic fraction and atomic weight of each component, respectively.

Table 1 summarizes the values of d_o, β_o, and v_o for the various compositions along with the corresponding standard deviations for these parameters. The standard deviations indicate that the density was measured with a precision of $\pm$ 0.1 to 0.2 per cent with the exception of the composition X = 50.8. For this run the random errors led to a precision of $\pm$ 0.4 per cent in the density. The values of d_o, β_o, and v_o in Table 1 include an error due to the surface tension of the molten solutions. Due to observed wetting of the suspension by the liquid alloys, the effect of surface tension was to reduce the density

Table 1. Values of density, expansivity, and atomic volume at 500°C for various compositions.

X (at. % Tl)	d_o (gm/cm^3)	β_o x 10^4 (°C)$^{-1}$	v_o x 10^{23} (cm^3/atom)	Temperature Range (°C)
31.0 $\pm$ 0.3	6.772 $\pm$ 0.006	2.00 $\pm$ 0.07	3.71 $\pm$ 0.02	220-500
42.0 $\pm$ 0.3	7.148 $\pm$ 0.005	1.37 $\pm$ 0.05	3.71 $\pm$ 0.02	280-500
48.0 $\pm$ 0.2	7.370 $\pm$ 0.004	1.61 $\pm$ 0.05	3.71 $\pm$ 0.01	320-500
50.8 $\pm$ 0.2	7.587 $\pm$ 0.015	1.34 $\pm$ 0.26	3.65 $\pm$ 0.07	360-500
68.8 $\pm$ 0.2	8.584 $\pm$ 0.005	1.59 $\pm$ 0.06	3.39 $\pm$ 0.01	540-650

values by an amount estimated to be approximately one per cent.

Table 2 lists the estimated surface tension and corrected values of the density at 500°C. These estimates were made by assuming that the surface tension of liquid Tl-Te has a linear dependence on composition. The values for the surface tension of pure thallium and tellurium used for the linear interpolation were taken from Sauerwald, Schmidt, and Pelka (1935) and from Smith and Spitzer (1962), respectively. Although the surface tension of compound containing systems does not generally have a composition dependence that is precisely linear, the surface tension estimations should be accurate to within 50 per cent of their actual values. In view of the small magnitude of the correction, the linear interpolation seems adequate.

Table 2. Estimated surface tension and corrected density values.

X (at. % Tl)	Estimated surface tension (dynes/cm)	Corrected values of d_o (gm/cm^3)
31.0	268	6.847
42.0	301	7.228
48.0	319	7.454
50.8	328	7.674
68.0	382	8.961

Corrections for the effect of surface tension are not included in the values of d, d_o, β_o, and v_o plotted in Figures 2, 3, and 4 which display the temperature and composition dependence of these parameters. The values of d_o, β_o, and v_o which were used in these figures for the pure elements were selected from Lucas and Urbain (1962) for Te and from Schneider and Heymer (1956) for Tl. Figure 2 shows the temperature dependence of the density for the various runs. As can be seen, within the random deviations each set of data appears to have a linear temperature dependence. Further justification for a linear representation of this data will be discussed in terms of the correlation coefficients obtained from the least squares analysis.

Isothermal plots of the density and coefficient of volume expansion appear in Figure 3. The density curve in Figure 3 appears to have a kink near the composition X = 50 indicating a possible change in the liquid structure. The values of β_o shown in the same figure exhibit considerable scatter, making it unreasonable to represent this data with a continuous curve. In Figure 4, the atomic volume appears relatively constant in the Te-rich region, but changes rapidly beyond the composition X = 50. If the values for the atomic volume plotted in Figure 4 were to include corrections for the effect of surface tension based on the estimations given in Table 2, the shape of
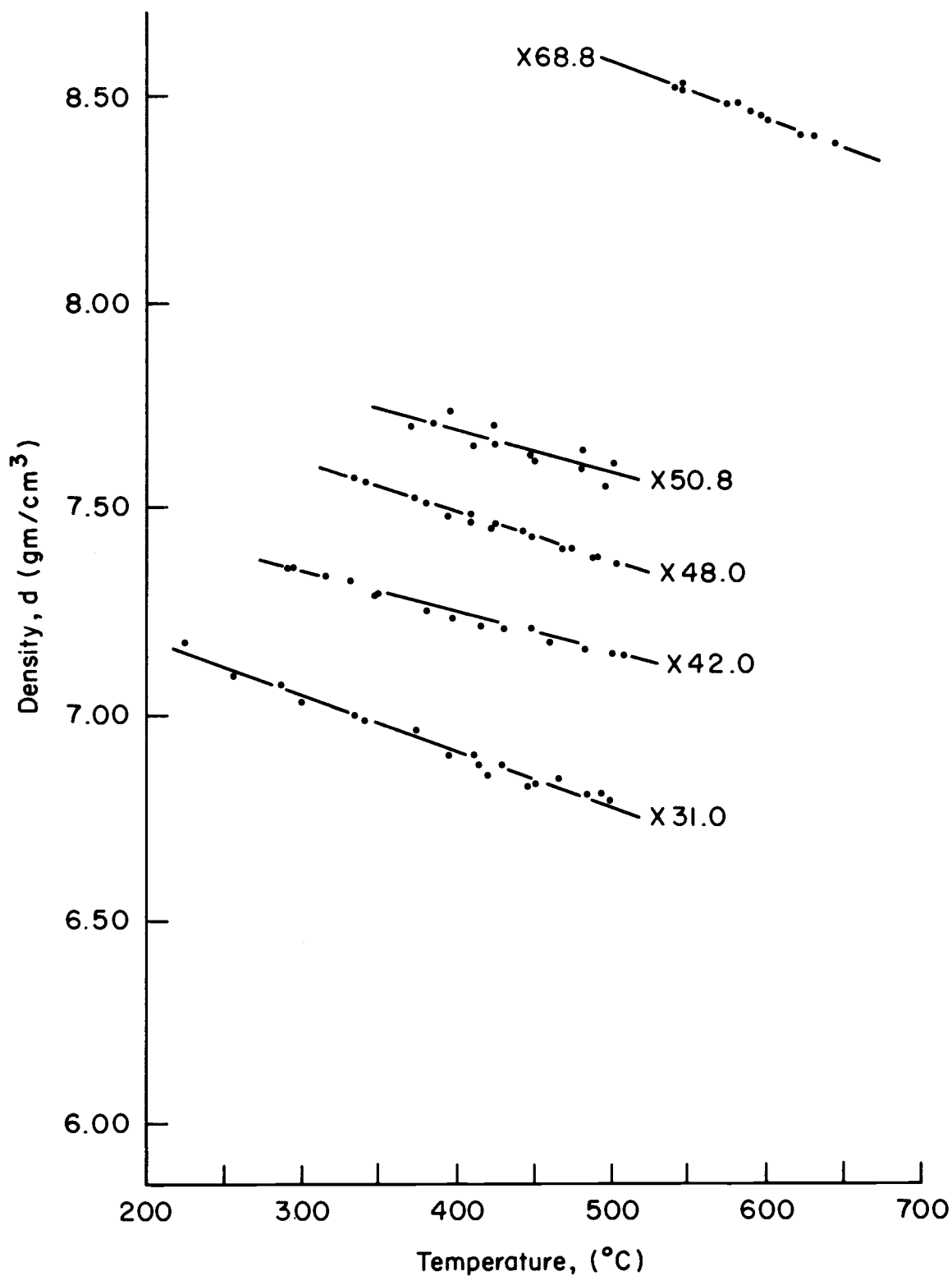
Figure 2. Density versus temperature for various composi-
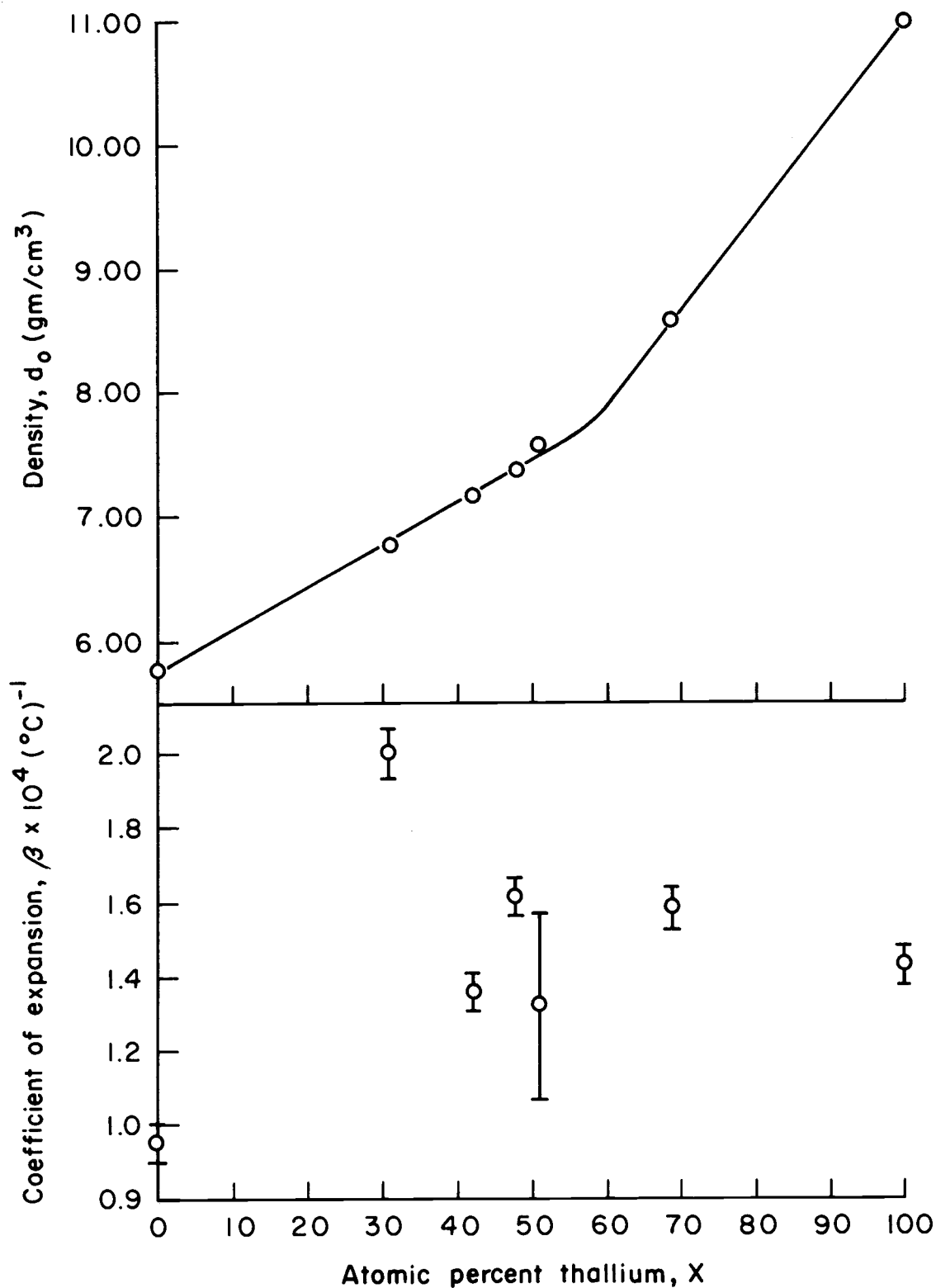tions of thallium-tellurium.

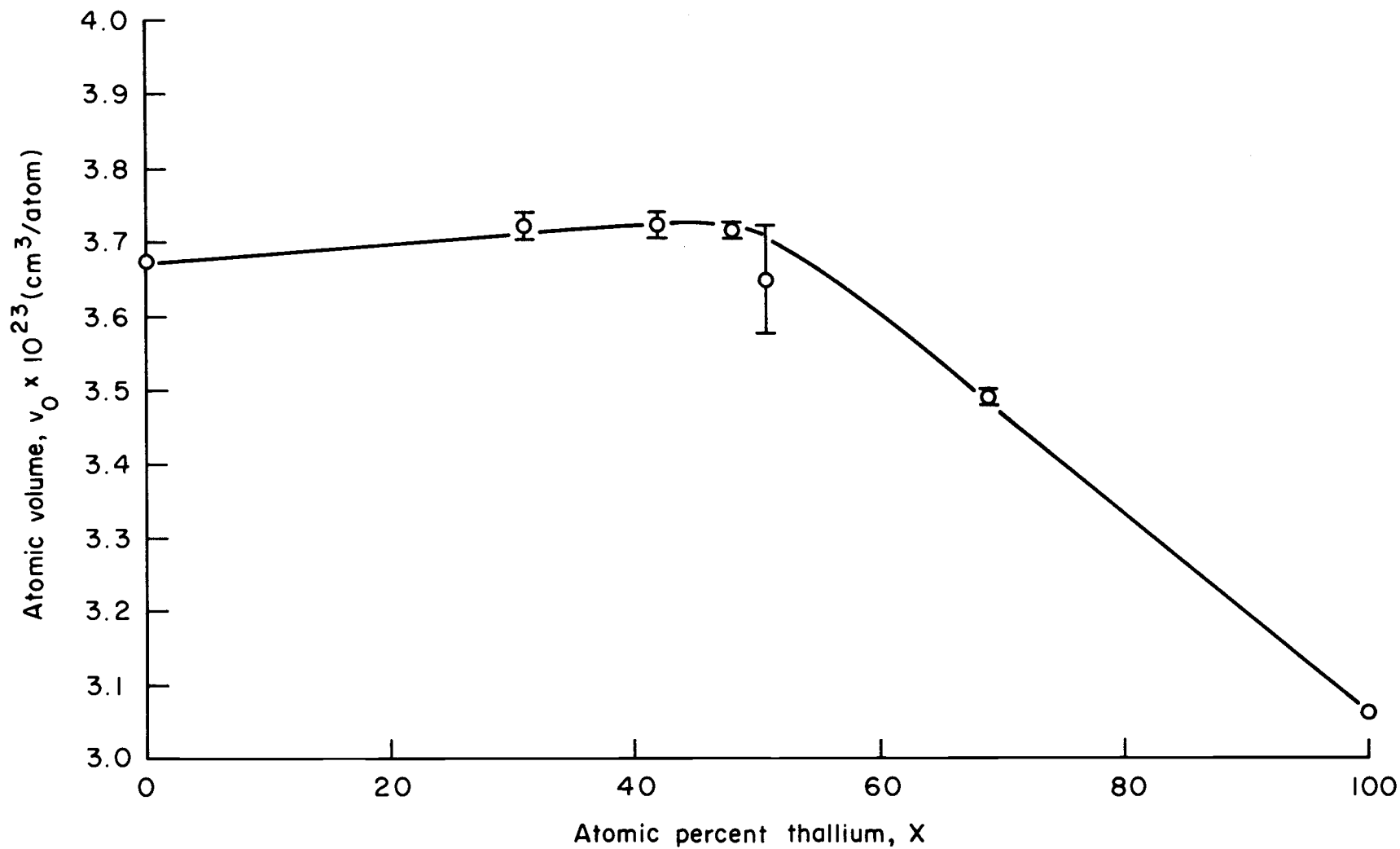Figure 3.   Density and bulk coefficient of volume expansion at 500°C versus composition.

Figure 4. Atomic volume at 500°C for various compositions of thallium-tellurium.

the v_o versus X curve would remain essentially the same
except that the apparent maximum in v_o would disappear.
Instead of increasing above the value for pure Te, the
atomic volume would be nearly constant in the composition
range $0 \leq X \leq 50$.

A standard least squares analysis outlined in Freund
(1962) provided the density-temperature relationships for
the five compositions. The form for the linear regression
assumed accurate measurement of the temperature. The
calculations for the data in Table 1 were performed with
a Hewlett Packard 9100 computer to determine the slopes,
reference coordinates, and their standard deviations. The
program also produced values for the correlation coeffi-
cient r which is used in statistics as a measure of the
degree of linear correlation between two variables. If
r = 0, there is no correlation, while r = ± 1 indicates
a perfect fit. As shown in Table 3, the density-tempera-
ture relationships for four of the five runs have correla-
tion coefficients with values near 0.99, indicating good
linear dependence of d on T. The remaining run X = 50.8,
has a lower degree of correlation 0.85 as expected from
the larger amount of scatter that can be observed in
Figure 2. The values of r for all runs leads to a prob-
ability for linear correlation which is greater than 0.99.

Table 3. Correlation coefficients for the various
          compositions.

X	31.0	42.0	48.0	50.8	68.8
-r	0.99	0.99	0.99	0.85	0.99

DISCUSSION AND CONCLUSION

The primary purpose of the density-temperature meas-
urements for liquid thallium-tellurium was to obtain infor-
mation which would supplement known electrical data.   In
the composition range 66.7 < X < 72, Cutler and Field
(1968) have shown that thallium in excess of the composi-
tion X = 66.7 generates fully ionized donor centers with
the release of three electrons per atom.   Under these con-
ditions the total electron concentration can be calculated
if, in addition, the atomic concentration is known.   The
atomic concentration is obtained by conversion of the den-
sity to the number of atoms per unit volume by means of
Equation 2.   Due to the linear composition dependence of
the total electron concentration, the density-temperature
values of any composition in the range 66.7 < X < 72 are
sufficient for determining the electron concentration
throughout this range.

Measurement of the temperature dependence of the den-
sity over a range of compositions also provides some infor-
mation about the structure of the liquid.   The values for
the coefficients of volume expansion listed in Table 1
are typical of liquid metals.   However, the plot of β_0
versus X in Figure 3 shows that within the margin of error
the coefficient of expansion does not exhibit a discernable
trend in composition dependence.

The absence of a smooth composition dependence for β_0 reflects the variation in the slopes of the density-temperature curves for the different compositions and is probably an indication that systematic errors were present. The systematic effects which may have led to variations in the slopes of the d-T curves would have depended on temperature in a reversible manner because systematic differences were not observed between the density measurements for increasing and decreasing temperatures except for one composition. One type of systematic effect which has a reversible temperature dependence is the surface tension. Variations in the slopes of the d-T curves would have resulted if the surface tension of the pure Tl-Te solutions depended strongly on temperature and if, in addition, appreciable differences in the amounts of surface contamination, such as oxides and impurities were present for the various compositions. However, it is unlikely that these conditions alone would have led to the amount of irregularity which is present in the plot of β_0 versus X in Figure 3, and it therefore seems reasonable to expect that other small, unknown systematic errors were present during the density measurements.

Structural changes in liquid alloys can be inferred from the dependence of the density and atomic volume on composition. The composition dependence of the density at 500°C appearing in Figure 3 shows a change in the slope

near X = 50. However, the true behavior tends to be obscurred by the large difference in the atomic weights of Tl and Te. To interpret structural changes, it is more appropriate to consider the composition dependence of the atomic volume since deviations from linearity will show up more clearly.

The plot of v_o versus X appearing in Figure 4 shows that a rapid change in the average atomic volume begins to occur at approximately the composition corresponding to TlTe. This is interesting since, in view of known electrical and thermodynamic data, a change in structure is expected in the vicinity of the stoichiometric composition Tl_2Te. Electrical measurements reported by Cutler and Field (1968) show that the resistivity approaches a maximum as the composition Tl_2Te is approached from either the tellurium- or thallium-rich direction. This behavior indicates that a preference for unlike-atom associations occurs which leads to the formation of the compound Tl_2Te in the liquid state. A change in the slope of the v_o versus X curve is expected near X = 66.7 since the types of bonding on either side of this boundary should be different. The covalent chain structure which tellurium possesses as a solid is maintained to some extent in the liquid state up to 550-600°C. In the case of thallium-tellurium solutions which are rich in tellurium, it is reasonable to expect that the thallium atoms participate

in the chain structure. However, on the thallium-rich side of the Tl_2Te boundary, the resistivity indicates that the liquid is composed of the compound Tl_2Te plus the excess thallium atoms. Thus, the character of the average atomic volume should differ on either side of X = 66.7.

Figure 5 is an approximate representation of a plot displaying thermodynamic data collected by Terpilowski and Zaleska (1963). The plot shows the behavior of the enthalpy of mixing ΔH and the excess entropy of mixing ΔS^{ex} at 600°C. The large, negative character of both thermodynamic functions over the composition range indicates that the liquid thallium-tellurium possesses a high degree of short range order and a tendency for bonding between Tl and Te. The curve for the enthalpy of mixing exhibits a broad, symmetrical minimum in the approximate range 60 < X < 70, while the curve for the excess entropy of mixing has less symmetry and has a sharper minimum in the same region. The behavior of these curves provides further evidence for the formation of Tl_2Te as a compound in the liquid state and supports the expectation of a singularity in v_o at this composition.

A closer examination of the dissymmetry in the plot of ΔS^{ex} versus X, however, provides the basis for a possible explanation of the observed behavior of v_o. Figure 5 shows that the excess entropy of mixing changes gradually for X $\lesssim$ 50 and then changes rapidly beyond this compo-
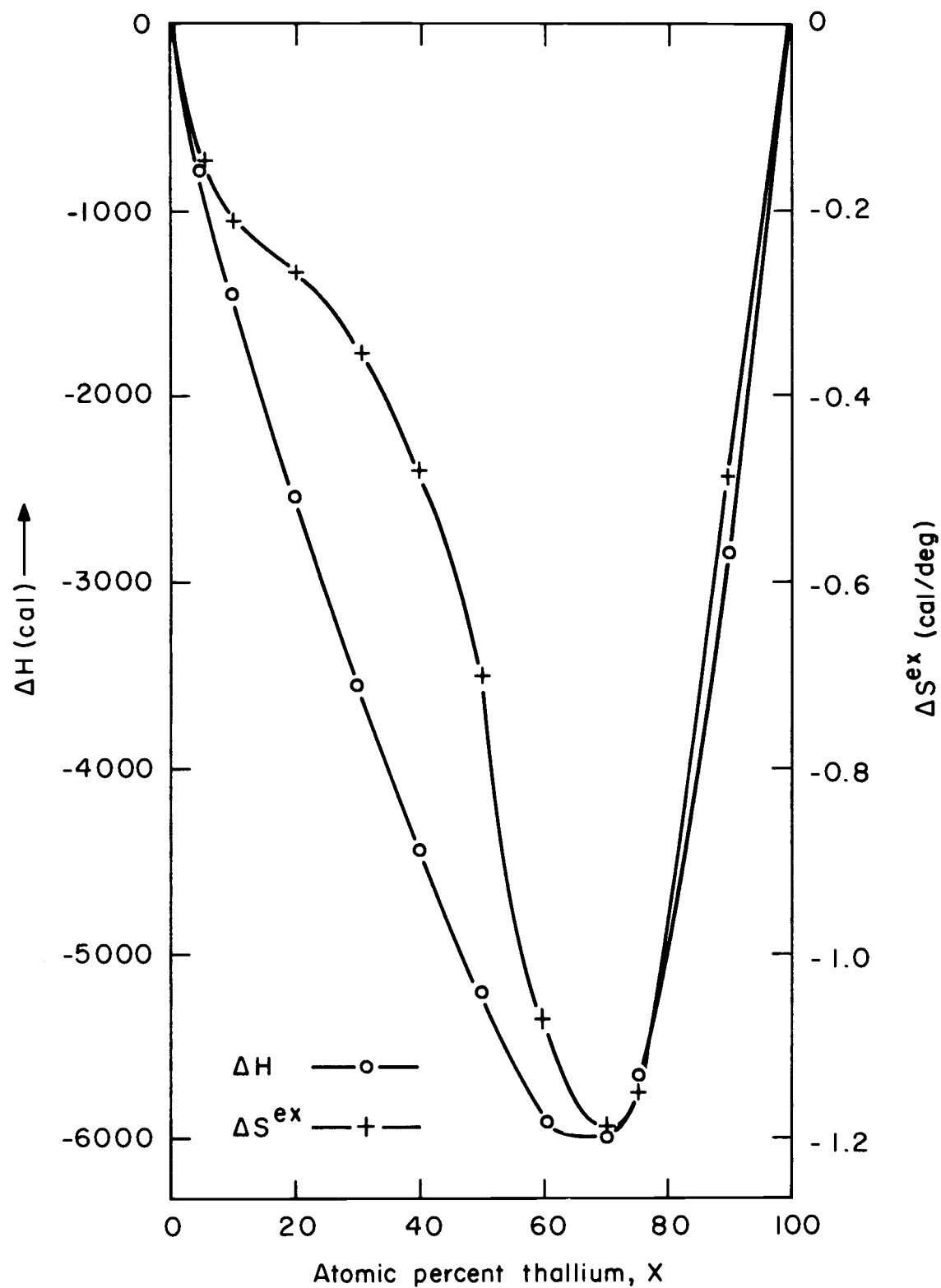
Figure 5. Molar excess enthalpy and entropy of mixing at
600°C versus composition.

sition until the minimum is approached. The change in the slope of the excess entropy curve indicates that there is a more rapid increase in short range order in the region for $X \geq 50$.

Since the curve for the excess entropy reflects the short range order of the liquid, some inferences can be made about the types of bonding that may occur which could account for the behavior of the atomic volume over the composition range. In the region for $X < 50$, the liquid may consist of chains of thallium-tellurium. Since a chain structure is relatively open, the atomic volume should change slowly. As the concentration of thallium atoms exceeds 50 atomic per cent, the additional thallium may become attached to the Tl-Te atoms and form Tl_2Te units. A change in the average atomic volume should then occur since the Tl_2 parts would occupy a smaller volume than the two Te atoms which had been in the original configuration when X was less than 50. As more thallium is added, the formation of Tl_2Te units should continue until reaching the composition $X = 66.7$. Beyond this composition, the original chain structure probably disappears and the liquid consists of the compound Tl_2Te and the excess Tl atoms. Finally, the absence of a change in the slope of the v_o versus X curve at $X = 66.7$ would be explained if the volume occupied by the Tl atoms which became attached to the Tl-Te units

were the same as the volume of the extra Tl atoms be-

yond Tl_2Te.

It must be emphasized that the explanation given for

the observed behavior of the atomic volume is hypotheti-

cal. In order to describe the behavior of v_o with con-

fidence, a more thorough examination of the thallium-rich

composition range is needed. Additional density measure-

ments should be performed to verify the linear behavior

of v_o versus X in the composition range $50 \lesssim X \leq 100$.

Also, scattering experiments would provide data for a

more accurate description of the types of associations

between the atoms.

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