Theoretical evaluation of structure factors and computation of coordination number, compressibility, and self- and mutual-diffusion coefficients of the Hg-Tl alloy

R. V. Gopala Rao and R. Venkatesh

Division of Physical Chemistry, Jadavpur University, Calcutta 700 032, West Bengal, India
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The partial structure factors have been evaluated for the Hg-Tl amalgam. These have been computed with a hard-sphere reference system and a square-well attractive tail as a perturbation. The Libowitz solution of the direct correlation function for the hard-sphere mixtures has been used as a reference. The coordination numbers, the total structure factors, and the compressibilities of the alloy at different concentrations have been computed. The computed total structure factors are found to be in very good agreement with the measured values except at two concentrations. In addition, the thermodynamically important Bhatia-Thoronton correlation functions have been calculated. The cross-correlation function, namely the number-concentration function, did not exhibit any shoulder, thereby indicating no compound formation. The Helfand trajectory principle has been used to evaluate the self- and mutual-diffusion coefficient over the entire range of concentration for which measurements have been made. The ratio $D_{\text{Hg}}/D_{\text{Tl}}$ is found to be constant, thereby indicating that the alloy forms a regular solution. It may be pointed out that the thermodynamic and transport properties have been computed from potential parameters of pure constituents and no experimental data of the alloy are used in these calculations.

I. INTRODUCTION

The unusual properties of liquid amalgams present a problem in explaining the characteristics,[1] effective mass,[2] thermoelectric power,[3] and other properties. An experimental survey of total structure factors at different atomic fractions of Hg was made and the same was investigated in detail by Halder, Metzner, and Wagner.[4]

In the present paper detailed theoretical calculations have been performed using a square-well attractive tail as a perturbation with the hard-sphere potential as a reference system. The partial structure factors are computed within the framework of mean-spherical-model approximation. This approach offers a unique method of evaluation of partial structure factors as generally only two experimental methods are available, namely x-ray-, and neutron-diffraction techniques. Of course, isotope enrichment offers a method of obtaining another set of experimental data, but the complete enrichment of the isotope is rather difficult. Hence, model potential computations give a method of calculation of the three particle structure factors uniquely.

In addition, thermodynamic properties such as compressibilities of the alloy and the number of nearest neighbors can also be determined through these calculations. Further, the partial structure factors can be used in the evaluation of diffusion coefficient through Helfand's linear-trajectory method. The present partial structure factors can also be used to evaluate the resistivities through Faber Ziman's equation.[5]

II. THEORY

Within the framework of the mean-spherical-model approximation (MSMA) the direct correlation function (DCF) can be written as

$$C_{ij}(r)= \begin{cases} C_{ij}^0(r), & 0 < r < \sigma_{ij} & \text{(1a)} \\ -\varepsilon_{ij}/k_B T, & \sigma_{ij} \le r \le \lambda_{ij}\sigma_{ij} & \text{(1b)} \\ 0, & r > \lambda_{ij}\sigma_{ij} & \text{(1c)} \end{cases}$$

where $C_{ij}^0(r)$ is the DCF for a mixture of hard spheres obtained by Lebowitz[6] for the Percus-Yevick equation. Further, σ_{ii}, ε_{ii}, and λ_{ii} are the diameters, potential-energy depth, and breadth, respectively, of the square-well potential of the ith species. The mixed parameters are determined through the use of Lorentz-Berthelot rules.[7]

Fourier transforming the generalized Ornstein-Zernike equation and using convolution theorem, we get

$$h_{ij}(k)=C_{ij}(k)+ \sum_{l=1}^{2} \rho_l C_{il}(k)h_{jl}(k) \ . \tag{2}$$

This equation, upon solving for the $h_{ij}(k)$, gives

$$h_{11}(k)=\{C_{11}(k)[1-\rho_2 C_{22}(k)]+\rho_2 C_{12}^2(k)\}B^{-1}(k) \ , \tag{3}$$

$$h_{22}(k)=\{C_{22}(k)[1-\rho_1 C_{11}(k)]+\rho_1 C_{12}^2(k)\}B^{-1}(k) \ , \tag{4}$$

$$h_{12}(k)=h_{21}(k)=C_{12}(k)B^{-1}(k) \ , \tag{5}$$

where

$$B(k)=[1-\rho_1 C_{11}(k)+\rho_1\rho_2 C_{11}(k)C_{22}(k)$$
$$-\rho_1\rho_2 C_{12}(k)] \ , \tag{6}$$

and the rest of the symbols have already been explained elsewhere.[8] Further, we have the well-known result connecting the partial structure factor and $h_{ij}(k)$ as

$$S_{ij}(k)=\delta_{ij}+(\rho_i\rho_j)^{1/2}h_{ij}(k) \ . \tag{7}$$

TABLE I. Input parameters in calculations of Hg-Tl alloy.

Metal	Temp. (K)	σ_{ii}	ϵ_{ii}/k_B	λ_{ii}
Hg	298	2.80	100.0	1.73
Tl	298	3.08	157.97	1.69

TABLE II. Structural characteristics of the alloy at different at. % Tl.

Comp. (at. % Tl)	Calc.		Expt.	
	k (Å^{-2})	$S(k)$	k (Å^{-2})	$S(k)$
5.0	2.40	2.42	2.29	2.30
8.5	2.40	2.43	2.29	2.27
16.0	2.40	2.43	2.28	2.30
28.6	2.40	2.40	2.28	2.70
40.0	2.30	2.38	2.28	2.44

The total structure factor in terms of partial structure factors can be written as[9]

$$S(k) = \sum_{i=1}^{2} \sum_{j=1}^{2} (C_i C_j)^{1/2} \frac{f_i f_j}{C_1 f_1^2 + C_2 f_2^2} S_{ij}(k) . \quad (8)$$

Here, f_i and f_j are the atomic scattering factors and are taken from the literature;[10] C_i is the atomic fraction of the ith species.

At this juncture it may be pointed out that the parameters used are those of pure metals only and hence no new data are made use of in the evaluation of the partial structure factors. The input parameters used are taken from literature[11,12] and are given in Table I.

The important thermodynamic property—namely, compressibility of the alloy at various concentrations—can be conveniently computed from the Kikwood-Buff equation.[13] The well-known Bhatia-Thoronton[14] thermodynamically important correlation functions—namely, number-number, concentration-concentration, and number-concentration (cross) correlation function—are linearly related to the partial structure factors and are calculated as already explained.[8] Here we take the necessary weight densities for the evaluation of C_1 and C_2 from the literature[15] at various atomic fractions of Hg.

II. COMPUTATION OF COORDINATION NUMBERS

We use the radial distribution functions (RDF's) $g_{ij}(r)$ obtained from total structure factors in calculating the coordination number through the equation given by

$$\eta_{ij} = 4\pi\rho_0 \int_0^{r_{min}} g_{ij}(r) r^2 \, dr . \quad (9)$$

Here, r_{min} is the first minimum of the partial radial distribution function. In an exactly similar way, the total number of nearest neighbors (η^T) has been calculated from the total structure factor.

III. RESULTS AND DISCUSSION

The total structure factors are shown in Fig. 1. The computed partial structure factors $S_{11}(k)$, $S_{22}(k)$, and $S_{12}(k)$ at different at. % (atomic percent) of Tl are shown in Figs. 2-5. It is gratifying to note that the present calculations yield results in agreement with experiment. The calculated peak position is found to be in good agreement with experiment at all concentrations. However, there is a discrepancy in the peak height between experiment and theory at 8.5 at. % of Tl. The structural characteristics of the alloy at various at. % of Tl are given in Table II. It is seen from this that the experimental peak position does not change as the present calculat-

ed results do. Further, there is a disagreement with respect to the peak height at 28.6 at. % Tl. Halder et al.[4] discuss this point at length and they point out that the peak they obtained is diffuse and the eutectic melting point at the composition of 8.5 at. % Tl is −60 °C, while the experiment was performed at 25 °C. Perhaps the large difference in temperature at which the intensities were measured is the cause for the low peak height at this
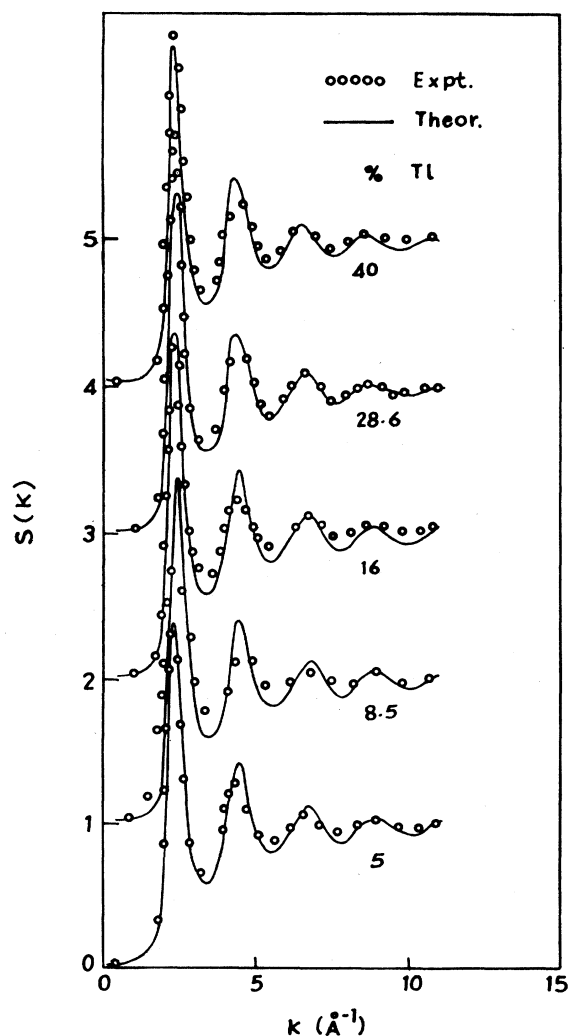

FIG. 1. Total structure factors as a function of wave vector k at different at. % Tl in Hg-Tl alloy.
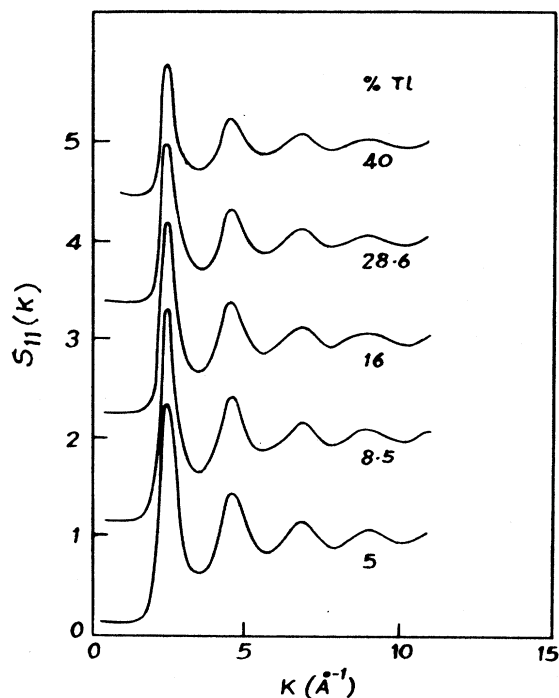
FIG. 2. $S_{11}(k)$ vs k at different at. % Tl.

composition of 8.5 at. % Tl, or since the intensities are diffuse, the experimental measurements may be difficult and, hence, may not be very accurate, or the present theory could not predict correctly the temperature-dependent effect on the peak height. It is well known, as was recently investigated by Wagner himself[16] and vari-
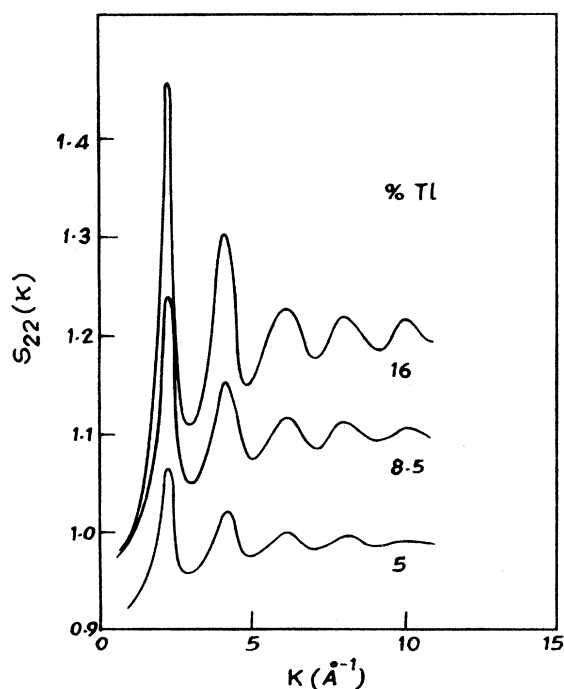
ous other workers,[17,18] that the peak height decreases if the measurements are made much above the melting point. Thus, the higher the temperature above the melting point at which the measurements are made, the lower the peak height.

However, it is very important to note from the experimental results of Halder et al.[4] that the bulk density (Table III) and the numbers of interest neighbors remain the same. Under these circumstances it is difficult to understand the low peak values of $S(k)$, even though one may justify that at this composition the first shell may be large. However, the graphs of Halder et al.[4] show no shift in the first minimum. Hence it is difficult to explain the exact reason for the first peak height at 8.6 at. % Tl. In the same way, it is difficult to explain the high value of the peak height obtained by Halder et al. with 28.6 at. % Tl. The theory gives a lower value.

The Bhatia-Thoronton correlation functions are given in Figs. 6–10. The number-number correlation function is comparatively large and resembles the total structure factor. The concentration correlation functions appear to be small, as shown in the enlarged scale in Figs. 9 and 10. Similar small oscillations were also found in Na-Cs alloy.[19] The cross-correlation function $S_{NC}(k)$ oscillates around zero. No shoulder is observed in $S_{NC}(k)$ as found in Hg-In alloy.[8] Thus $S_{NC}(k)$ does not indicate the formation of a compound in the present alloy. Halder

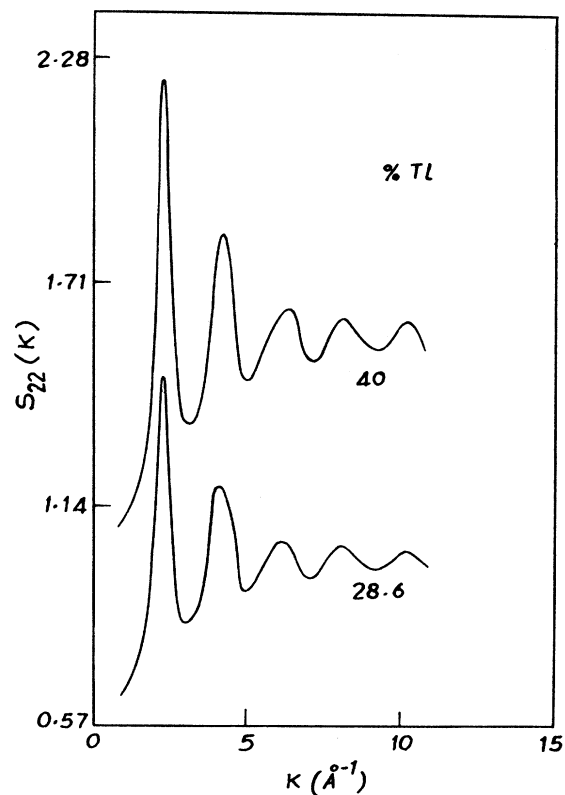FIG. 3. $S_{22}(k)$ vs k at 5, 8.5, and 16 at. % Tl.

FIG. 4. $S_{22}(k)$ vs k at 28.6 and 40 at. % Tl.

TABLE III. Structure factors in the long-wavelength limit and compressibilities at different concentrations of Tl.

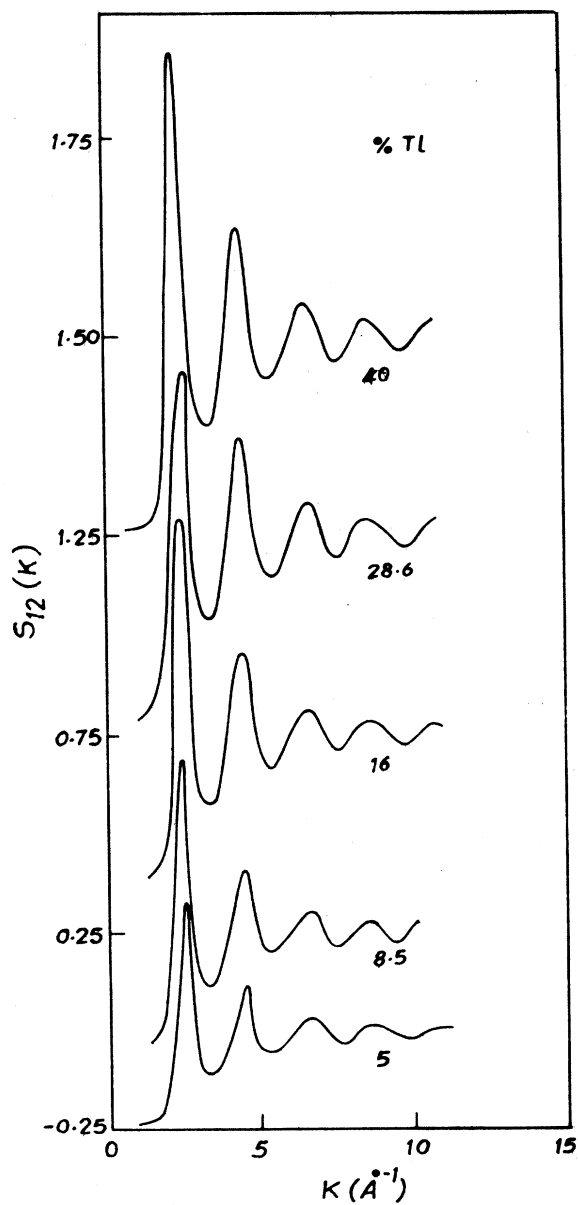| Comp. (at. % Tl)	ρ (Å^{-3})	$S(0)$	χ_T (10^{12} cm^2/dyn)
5.0	0.0403	0.0257	15.51
8.5	0.0400	0.0265	16.12
16.0	0.0396	0.0267	16.42
28.6	0.0386	0.0267	16.83
40.0	0.0378	0.0254	16.36

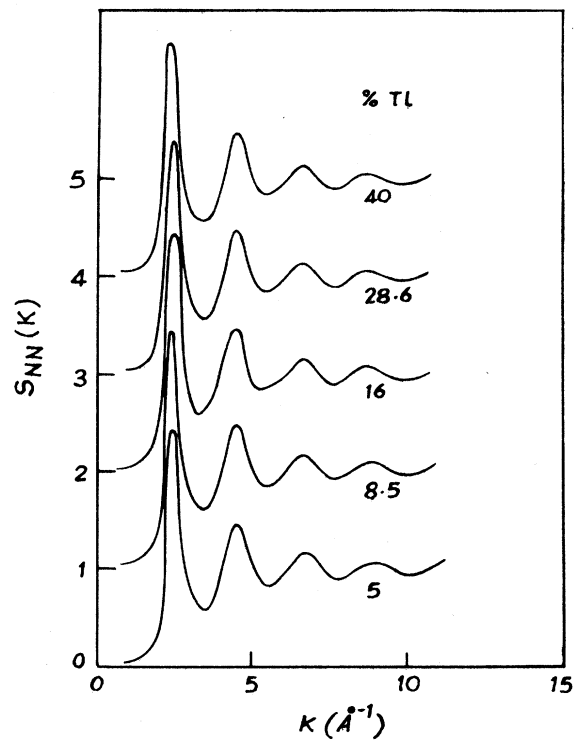FIG. 6. $S_{NN}(k)$ vs k at different at. % Tl.
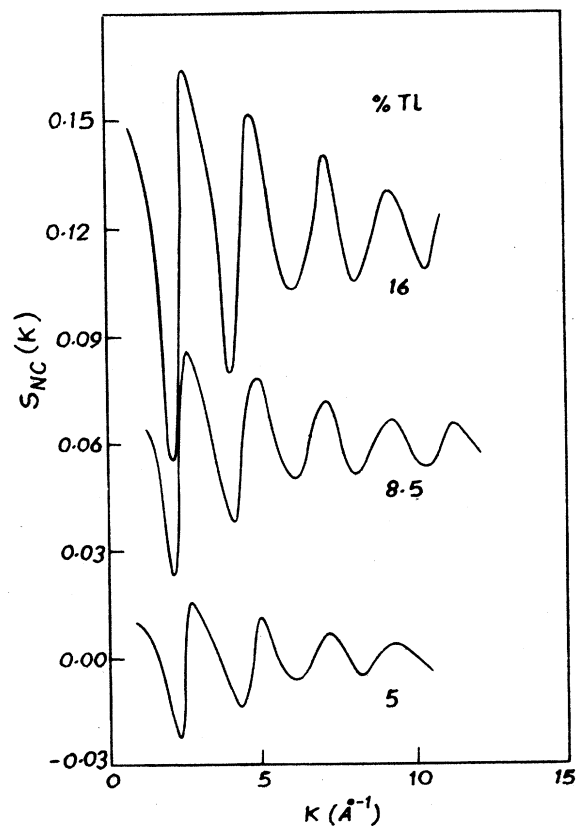

FIG. 5. $S_{12}(k)$ vs k at different at. % Tl.
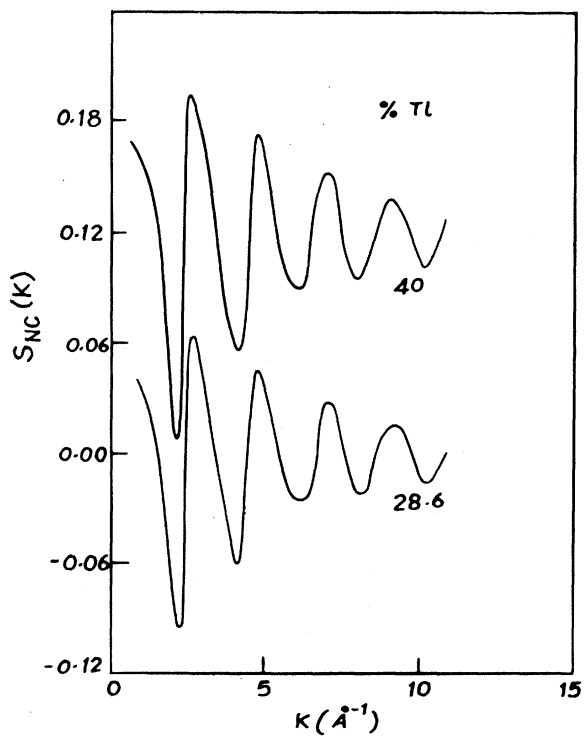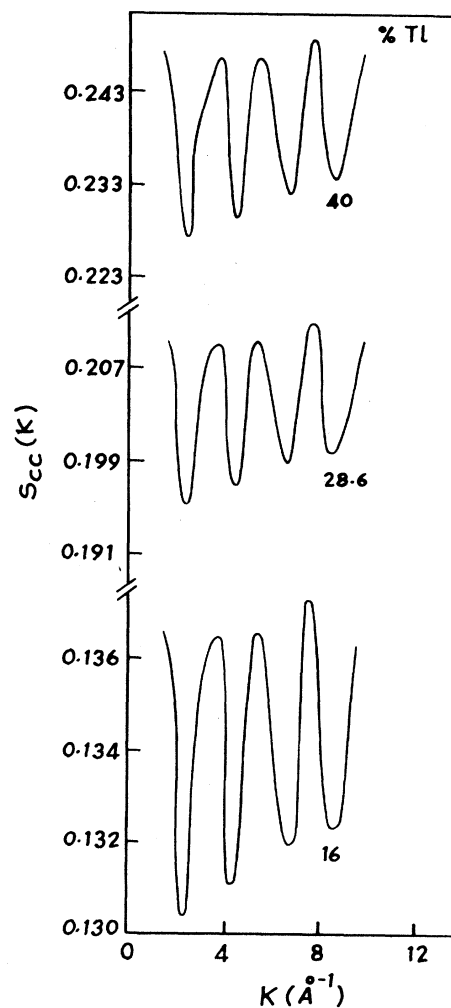

FIG. 7. $S_{NC}(k)$ vs k at 5, 8.5, and 16 at. % Tl.

FIG. 8. $S_{NC}(k)$ vs k at 28.6 and 40 at. % Tl.

FIG. 9. $S_{CC}(k)$ vs k at 5 and 8.5 at. % Tl.

FIG. 10. $S_{CC}(k)$ vs k at 16, 28.6, and 40 at. % Tl.

TABLE IV. Computed values of partial and total coordination numbers.

Comp. (at. % Tl)	η_{11}	η_{22}	η_{12}	η^{T} [a]
5.0	8.1	0.5	2.0	9.2 (10.2)
8.5	7.8	0.9	2.6	11.7 (10.2)
16.0	7.1	1.6	3.4	11.8 (10.3)
28.6	5.9	2.9	4.1	11.9 (11.3)
40.0	4.9	4.1	4.4	12.0 (11.4)

[a]Values in parentheses are those obtained by Halder *et al.* (Ref. 4).

TABLE V. Structural features of partial and total radial distribution functions.

Comp. (at. % Tl)	Particulars of first peak position							
	r_1	$g_{11}(r)$	r_1	$g_{22}(r)$	r_1	$g_{12}(r)$	r_1[a]	$g_{tot}(r)$
5.0	3.02	3.176	3.31	3.312	3.16	3.226	3.04 (3.10)	3.137
8.5	3.02	3.188	3.31	3.326	3.16	3.279	3.04 (3.09)	3.122
16.0	3.02	3.233	3.30	3.375	3.16	3.327	3.07 (3.07)	3.122
28.6	3.02	3.276	3.30	3.426	3.16	3.374	3.10 (3.22)	3.113
40.0	3.02	3.319	3.30	3.476	3.16	3.421	3.14 (3.28)	3.135

[a]Values in parentheses are those obtained by Halder et al. (Ref. 4) for the peak position of $g_{tot}(r)$.

et al.[4] discussed this point at length and they point out that a loose compound formation was reported by Smallman and Frost,[20] and this was supported by Sauerwald and Teske,[21] while a later publication by Sauerwald and Osswald[22] does not at all support the formation of the compound, and this is in conformity with present findings. The compressibilities evaluated from the Kirkwood-Buff equation are tabulated in Table III for different concentrations of Tl in Hg. The compressibility of pure Hg is 6×10^{-12} cm^2/dyn, while that of Tl is 4×10^{-12} cm^2/dyn, whereas the compressibility of the 95-at. % Hg amalgam is found to be 15.5×10^{-12} cm^2/dyn. This may be due to the presence of holes existing in the amalgam and the loose-compound-forming nature of the alloy. Faber[5] also observed the formation of unstable compounds of Hg amalgams. According to Faber, Hg amalgams are complicated by the anomalous behavior which pure Hg displays. This unusual behavior of Hg is exhibited in the case of other properties such as susceptibility, Knight shift, and Hall coefficient.[5] However, from Table III it may be observed that the compressibility smoothly increases from 15.51×10^{-12} to 16.83×10^{-12} cm^2/dyn, and decreases to 16.36×10^{-12} cm^2/dyn, thus showing a very small maxima at 28.6 at. % Tl.

The coordination numbers at different concentrations are given in Table IV. Here, η_{ii} stands for the number of nearest neighbors of the same kind (i.e., first shell), while

TABLE VI. Calculated values of coordination numbers with different equations at different Tl concentrations.

Comp. (at. % Tl)	ρ	η_C^1	η_C^2	η^T	$\eta^{T a}$
5.0	0.0403	9.6	12.2	9.2	10.2
8.5	0.0400	9.5	12.1	11.7	10.2
16.0	0.0396	9.7	12.3	11.8	10.3
28.6	0.0386	9.8	12.4	11.9	11.2
40.0	0.0378	9.9	12.6	12.0	11.4

[a]Halder et al. (Ref. 4).

η_{12} gives the number of nearest neighbors of the second kind with respect to first. In column 5 of Table IV we give the total coordination number, and it may be noted that the agreement between the present calculated values and those of Halder et al. (given in parentheses) is satisfactory and is nearly 12, except at 5 at. % Tl. However, it may be pointed out that α-Tl and β-Tl have hcp and fcc structures, respectively, in the solid state, while Hg has hcp structure. All these structures have 12 nearest neighbors in the solid state. The present calculated results, except that with 5 at. % Tl, have nearly 12 nearest neighbors. Further, the atomic volume of Hg is nearly 14 cm^3/g-at. Thus the volumes and also their compressibilities are nearly equal. Thus it is expected that they mix in all proportions, especially considering the fact that they both have close-packed structures and nearly equal atomic weights. The same conclusions were also drawn by Halder et al.[4] Further, it may be pointed out that we choose $k_{max} = 8.9$ Å^{-1}, while Halder et al. choose two values in Fourier transform of $S(k)$ to $g(r)$. The values that they chose are 7.6 and 12 Å^{-1}. The values of interatomic distance r_1 obtained from the first peak of the RDF evaluated by Fourier inversion of structure factor are in good agreement with those obtained by Halder et al.[4] and are given in Table V.

Furukawa[23] formulated an equation for the coordination number for random close-packed liquid structure as

$$\eta_C^1 = 6\sqrt{2}\rho r_1^3 . \tag{10}$$

The values calculated from Eq. (10) are given in Table VI and are, in general, lower than those obtained from RDF.

However, Halder et al. assumed[4] that Tl has bcc-type packing in the amalgam. Thus, for the coordination number, they obtained an expression as (eight nearest atoms with six next-nearest neighbors)

$$\eta_C^2 = (\tfrac{56}{3}\sqrt{3})\rho r_1^3 . \tag{11}$$

Calculations made on the basis of this equation are also given for comparison in Table VI. Equation (9) gives a coordination number of nearly 12, which therefore points to the fact that the amalgam has a closed-packed struc-

TABLE VII. Computed values of friction constants at various concentrations.

Comp. (at. % Tl)	ξ^{HS} (10^9 g/s)		ξ^S (10^9 g/s)		ξ^{SH} (10^9 g/s)	
	Hg	Tl	Hg	Tl	Hg	Tl
5.0	1.038	1.187	0.417	0.435	0.487	0.698
8.5	1.057	1.203	0.434	0.456	0.517	0.738
16.0	1.079	1.224	0.441	0.470	0.539	0.768
28.6	1.090	1.232	0.452	0.491	0.562	0.799
40.0	1.104	1.246	0.461	0.510	0.586	0.832

ture, except at 5 at. % of Tl, which is close to a bcc-type packing.

IV. CALCULATION OF DIFFUSION COEFFICIENTS

The linear-trajectory principle allows the use of partial structure factors in the evaluation of diffusion coefficients of the constituents of the alloy at different concentrations.[24,25]

The diffusion coefficient according to Einstein's equation is given by

$$D_i = \frac{k_B T}{\xi_i} . \tag{12}$$

Here, ξ_1 is the friction coefficient of the molecules in the liquid and can be written as a sum of three components, namely ξ_1^H, ξ_1^S, and ξ_1^{SH}, which are, respectively, the hard sphere, soft, and cross coefficients of the friction constant and have been explained already.[8]

The contributions of different parts of the friction constants are given in Table VII. The self- and mutual-diffusion coefficients are given in Table VIII. The latter is calculated from the following approximate equation:[26,27]

$$D_{12} = c_1 D_1 + c_2 D_2 + \text{correction terms} . \tag{13}$$

As expected, since repulsion plays a significant part in the liquid structure the hard-sphere contribution can be noticed to be large. It may be pointed out that the masses of both atoms are almost equal, while the size of the Tl atom is greater than that of Hg. Further, another important property that distinguishes Tl is its trivalency. Hence, it may be expected that the mutual attraction among Tl atoms should be comparitively higher than that among Hg atoms and, hence, as the concentration of Tl increases, the friction constant of Tl also increases, thereby decreasing the diffusion coefficient. Further, from

Table VII the soft-hard part of Tl is distinctly higher. This may be due to the superimposed effect of higher mass and higher valency of Tl atoms. This can also be seen from the potential parameters given in Table I. Further, because of its low potential-energy depth, the attractivity of Hg is less and, hence, it has higher diffusivity. For at. % Hg the present value of 2.1×10^{-5} cm^2/s is in satisfactory agreement with that obtained by Mangeldorf,[28,29] who obtained, for pure Hg, 1.6×10^{-5} cm^2/s. In the case of Tl, experimental intensity observations and the present theoretical calculations have been performed up to a concentration of 40 at. % Tl. From Table VIII the values of D_{Tl} may be observed to be decreasing with increasing Tl concentration. A linear extrapolation of these results gives a value for D_{Tl} for 100 at. % Tl as 1.0×10^{-5} cm^2/s while the observed value[30] is 0.99×10^{-5} cm^2/s and these are in good agreement with each other. The present results, which are in agreement with experiment are due to the fact that attractive forces have also been considered, even though the hard-sphere contribution is a major one.

It may be noticed from the last column of Table VIII that the ratio D_{Hg}/D_{Tl} is constant and equal to 1.197×10^{-5} cm^2/s. From the regular-solution theory of Bearman and Jones,[28] this constancy can be expected. A similar observation was also made by Jacucci and McDonald[31] for rare-gas mixtures. Rao and Murthy[32] found for Na-K alloy a constant for the ratio of the diffusion coefficients.

It is found that the diffusion-coefficient ratio is inversely proportional to the square of the atomic diameters,

$$\frac{D_1}{D_2} = \left[\frac{\sigma_2}{\sigma_1} \right]^2 . \tag{14}$$

In the present case the right-hand side is found to give a value of 1.21, which is in excellent agreement with the present average value of 1.20 for the ratio D_1/D_2.

TABLE VIII. Self- and mutual-diffusion coefficients at different at. % Tl.

Comp. (at. % Tl)	D_{Hg} (10^{-5} cm^2/s)	D_{Tl} (10^{-5} cm^2/s)	$D_{Hg\text{-}Tl}$ (10^{-5} cm^{-2}/s)	D_{Hg}/D_{Tl} [from Eq. (15)]	D_{Hg}/D_{Tl}
5.0	2.118	1.772	1.789	1.16	1.195
8.5	2.048	1.715	1.743	1.16	1.194
16.0	1.997	1.670	1.722	1.15	1.196
28.6	1.954	1.630	1.723	1.16	1.199
40.0	1.912	1.589	1.718	1.16	1.203

A more sophisticated equation that can be obtained from regular-solution theory can be written as[32]

$$\frac{D_1}{D_2} = \frac{g_{12}}{g_{11}} \frac{2M_2}{M_1 + M_2} \left[\frac{\sigma_{12}}{\sigma_{11}} \right]^2 . \tag{15}$$

Here, the g_{ij}'s are the contact values of the radial distribution function. These values are also given in Table VIII and the average value is found to be 1.16, which is in excellent agreement with Halfand's result, 1.20.

Thus it may be concluded that Hg-Tl forms a regular solution and the significance of the present work is the evaluation of both thermodynamic, structural, and transport properties completely from the potential-energy parameters of the pure constituents[11,12] determined for the evaluation of their structure factors.

[1]H. A. Davies and J. S. L. Leach, in *The Properties of Liquid Metals—Proceedings of the 2nd International Conference, Tokyo, 1972*, edited by S. Takeuchi (Taylor and Francis, London, 1973), p. 229.

[2]N. F. Mott, Philos. Mag. **13**, 989 (1966).

[3]T. Yoshioka and H. Suzuki, in Ref. 1, p. 513.

[4]N. C. Halder, R. J. Metzger, and C. N. J. Wagner, J. Chem. Phys. **34**, 1259 (1966).

[5]T. E. Faber, *Theory of Liquid Metals* (Cambridge University Press, London, 1972), pp. 164, 456, 514, and 521.

[6]J. L. Lebowitz, Phys. Rev. **133**, A895 (1964).

[7]R. V. Gopala Rao and Ratna Das, Phys. Rev. B **36**, 6325 (1987).

[8]R. V. Gopala Rao and R. Venkatesh, Phys. Rev. B **39**, 3563 (1989).

[9]M. J. Huijben, W. Van Der Lugt, W. A. M. Reimert, J. Th. M. De Hosson, and C. Van Dijk, Physica B+C **97B**, 338 (1979).

[10]*International Critical Tables for X-Ray Crystallography* (Kynoch, Birmingham, 1962), Vol. III, p. 201.

[11]R. V. Gopala Rao and D. Sen, Ind. J. Phys. **51A**, 313 (1977).

[12]R. V. Gopala Rao and A. K. Murthy, Z. Naturforsch. **30a**, 619 (1975).

[13]J. G. Kirkwood and P. Buff, J. Chem. Phys. **19**, 774 (1951).

[14]A. B. Bhatia and D. E. Thornton, Phys. Rev. B **2**, 3002 (1970).

[15]G. Abowitz and R. B. Gordon, Trans. Metall. Soc. AIME **51**, 227 (1963).

[16]C. N. J. Wagner, in *Proceedings of the 3rd International Conference on Liquid Metals, University of Bristol, 1976*, edited by R. Evans and D. A. Greenwood (Institute of Physics, London, 1976), p. 110.

[17]D. Jovic, I. Padureanu, and S. Rapeanu, in Ref. 16, p. 120.

[18]R. Block, J. B. Suck, W. Freyland, F. Hensel, and W. Glaser, in Ref. 16, p. 126.

[19]R. V. Gopala Rao and B. M. Sathpathy, Phys. Status Solidi B **110**, 273 (1982).

[20]R. E. Smallman and B. R. T. Frost, Acta Metall. **4**, 611 (1956).

[21]F. Sauerwald and W. Teske, Z. Anorg. Allgem. Chem. **210**, 247 (1933).

[22]F. Sauerwald and E. Osswald, Z. Anorg. Allgem. Chem. **257**, 195 (1948).

[23]K. Furukawa, Sci. Rep. Tohoku Univ. **12A**, 368 (1960).

[24]E. Helfand, Phys. Fluids **4**, 681 (1961).

[25]R. V. Gopala Rao and A. K. Murth, Ind. J. Pure Appl. Phys. **13**, 158 (1975).

[26]I. Ebbsjo, P. Schoffield, K. Skold, and I. Waller, J. Chem. Phys. C**7**, 3891 (1974).

[27]M. Parrine, M. P. Toshi, and N. H. March, J. Phys. C **7**, 2577 (1974).

[28]P. C. Mangelsdorf, Jr., Met. Soc. Conf. **7**, 428 (1961).

[29]S. G. Epstein, in *Liquid Metals*, edited by Sylvan Z. Beer (Dekker, New York, 1972), pp. 529 and 535.

[30]R. J. Bearman and P. F. Jones, J. Chem. Phys. **33**, 1432 (1960).

[31]G. Jacueci and I. R. McDonald, Physica A **80**, 607 (1975).

[32]R. V. Gopala Rao and A. K. Murthy, Pramana **9**, 587 (1977).